

On Droplet Interactions and Suspension Flow

by

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Cover: schematics of a thermal exhaust port of the Death Star.

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If the scientist had an infinity of time at their disposal,
it would be sufficient to say to them,
“Look, and look *carefully*.”

Henri Poincaré

On droplet interactions and suspension flow

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Abstract

Micron to millimetre sized droplets, precisely generated or sustained in controlled environment, have great potential in myriads of engineering applications functioning as the basic element to assemble metamaterials, deliver drugs, host surfactant, reduce friction and damp turbulence. The interaction of droplets from pairwise to collective levels is the most important factor in controlling these processes, yet little is known about the detailed mechanisms in various nonideal conditions. The present thesis combines a number of studies aiming to elucidate the physical principles of droplet interactions and suspension flow using both high- and low-fidelity numerical simulations.

We first study flow-assisted droplet assembly in microfluidic channels, seeking to harness the droplet interactions to produce photonic bandgap materials. A novel interface-correction level set/ghost fluid method (ICLS/GFM) is developed to directly simulate liquid droplets under depletion forces. Comparing to previous methods, ICLS/GFM conserves the global mass of each fluid using a simple mass-correction scheme, accurately computes the surface tension and depletion forces under the same framework, and has subsequently been applied to investigate the droplet clustering observed in a microfluidic experiment. Our simulations, supported by theoretical derivations, suggest that the observed fast self-assembly arises from a combination of strong depletion forces, confinement-mediated shear alignments of the droplets, and fine-tuned inflow conditions of the microchannel. However, the interplay of these 3D effects negates a simple droplet interaction model of parametric dependence, rendering the design of microfluidic chips for photonic crystal fabrications elusive in practice.

The next focus of the thesis is the implementation of a minimal discrete-element lubrication/contact dynamics model for simulation of dense particle suspensions. Two main ingredients of the model include (i) a frame-invariant, short-range lubrication model for spherical particles, and (ii) a soft-core, stick/slide frictional contact model activated when particles overlap. Physically, since contact interactions dominate at high particle concentrations, we expect the methodology to be applicable for probing the jamming of non-spherical particles and the rheology of foams as well.

Finally, we include two miscellaneous studies concerning the slippage property of liquid-infused surfaces and droplets statistics in a homogeneous turbulent shear flow. Overall, results of these simulations provide detailed flow visualisations and qualitative dependence of the target functional on various governing

parameters, facilitating experimental and theoretical investigations to design more robust drag-reducing surfaces and predict droplet distributions in emulsions.

Key words: droplets, suspension, multiphase flow, microfluidics, soft matter, rheology, depletion force, level set, ghost fluid, discrete element.

Droppinteraktioner och suspensionsflöden

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Sammanfattning

Mikro- till millimeter stora droppar, exakt genererade eller hållna i en kontrollerad miljö, har stor potential i många olika tekniska tillämpningar. De representerar en grundläggande teknik vid uppbyggnad av metamaterial, transport av läkemedel i kroppen, som bärare av ytaktivt medel, vid minskning av friktion och dämpning av turbulens. Växelverkan mellan droppar från parvis till kollektivnivå är den viktigaste faktorn för att kontrollera dessa processer; ändå är lite känt om de detaljerade mekanismerna vid olika icke-ideala förhållanden. I denna avhandling kombineras ett antal studier som syftar till att belysa de fysikaliska principerna för dropp-växelverkningar och suspensionsflöden med numeriska simuleringar av högre och lägre noggrannhet.

Vi studerar först flödesassisterad droppmontering i mikrofluidkanaler och försöker utnyttja dropp-växelverkningar för att producera fotoniska bandgapmaterial. En ny interface-correction level set/ghost fluid method (ICLS/GFM) är utvecklad för att direkt simulera vätskedroppar under inverkan av utarmningskrafter. Jämfört med tidigare metoder bevarar ICLS/GFM den totala massan för varje fluid med hjälp av ett enkelt masskorrigeringsschema, och beräknar exakt ytspänningen och utarmningskrafterna under samma omständigheter. Detta tillämpas sedan för att undersöka droppklustering, något som observerats i mikrofluidiska experiment. Våra simuleringar, med stöd av teoretiska härledningar, antyder att den observerade snabba klusteringen uppstår på grund av en kombination av starka utarmningskrafter och inneslutningsförmedlade skjukvrafter på dropparna samt finjusterade inflödesförhållanden för mikrokanalen. Men samspelet mellan dessa 3D-effekter omöjliggör en enkel parameterberoende dropp-växelverkningsmodell vilket gör att utformningen av mikrofluidiska chips för fotonisk kristallfabrikation är svår i praktiken.

Nästa fokus i avhandlingen är implementeringen av en minimal diskretelementsmörjnungs-/kontaktdynamikmodell för simulerings av tät partikelsuspensioner. Två huvudingredienser i modellen är (i) en referensram-invariant smörjmodell med kort räckvidd för sfäriska partiklar, och (ii) en stick/slip friktionskontaktmodell med mjuk kärna som aktiveras när partiklar överlappar varandra. Eftersom kontakt-växelverkningar domineras fysikaliskt vid höga partikelkoncentrationer, förväntar vi oss att metodologin också är tillämplig för att undersöka inklämning av icke-sfäriska partiklar och reologi för skum.

Slutligen inkluderar vi också två studier rörande glidningsegenskaper hos vätske-bemängda ytor och droppstatistik i ett homogen turbulent skjuvflöde.

Sammantaget ger resultaten av dessa simuleringar detaljerade flödesvisualiseringar, kvalitativt beroende av målfunktionen på olika reglerande parametrar, underlättar, experimentellt och teoretiskt, utformningen av mer robusta dragreducerande ytor samt förutsäger droppfördelningar i emulsioner.

Nyckelord: droppar, suspension, flerfasflöde, mikrofluidik, mjukt material, reologi, utarmningskraft, nivåuppsättning, spökvätska, diskret element.

Preface

This thesis summarizes a selection of studies on droplet interactions and suspension flow that may find applications in microfluidics, soft matter, and rheology. A brief introduction of the basic physical concepts and numerical methods is presented in Part I, followed by seven journal articles or preprints in Part II, see below. For consistency, all papers are formatted in the *jfm* style as the rest of the thesis. Their contents remain faithful to the original publications.

Paper 1. Z. GE, J-CH. LOISEAU, O. TAMMISOLA, L. BRANDT, 2018. *An efficient mass-preserving interface-correction level set/ghost fluid method for droplet suspensions under depletion forces.* *J. Comput. Phys.* **353**, 435–459.

Paper 2. Z. GE, O. TAMMISOLA, L. BRANDT, 2019. *Flow-assisted droplet assembly in a 3D microfluidic channel.* *Soft Matter.* **15**, 3451–3460.

Paper 3. I. FOUXON, Z. GE, L. BRANDT, A. LESHANSKY, 2017. *Integral representation of channel flow with interacting particles.* *Phys. Rev. E.* **96** (063110).

Paper 4. I. FOUXON, B. RUBINSTEIN, Z. GE, L. BRANDT, A. LESHANSKY. *The theory of hydrodynamic interaction of two spheres in wall-bounded shear flow.* Under review for *Phys. Rev. Fluids*.

Paper 5. M.E. ROSTI, Z. GE, S.S. JAIN, M.S. DODD, L. BRANDT, 2019. *Droplets in homogeneous shear turbulence.* *J. Fluid Mech.* **876**, 962–984.

Paper 6. Z. GE, H. HOLMGREN, M. KRONBICHLER, L. BRANDT, G. KREISS, 2018. *Effective slip over partially filled microcavities and its possible failure.* *Phys. Rev. Fluids.* **3** (054201).

Paper 7. Z. GE, L. BRANDT, 2020. *Implementation note on a minimal discrete-element lubrication/contact dynamics model for dense suspensions.* Technical Report.

March 2020, Stockholm
Zhouyang Ge

Division of work between authors

The main thesis advisor is Professor Luca Brandt (LB). Associate Professor Outi Tammisola (OT) acts as the co-advisor.

Paper 1. LB directed the research. Z. Ge (ZG) and J-Ch. Loiseau developed, implemented and validated the methods. OT oversaw the entire project. ZG wrote the paper with inputs from all other authors.

Paper 2. Following Paper 1, ZG continued the research, performed the simulation and wrote the paper with inputs from the rest of the authors.

Paper 3. A. Leshansky (AL) directed the research. I. Fouxon (IF) derived the theory. ZG performed the simulation. IF wrote the paper with inputs from the rest of the authors.

Paper 4. AL directed the research. IF constructed the theory. B. Rubinstein performed the multipole expansion simulation. ZG performed the Navier-Stokes simulation. IF wrote the paper with inputs from all other authors.

Paper 5. LB conceived the project. M.E. Rosti (MER) implemented the method and performed the simulation. MER and ZG performed the droplet statistics and scaling analyses. S.S. Jain visualised the flow. M.S. Dodd analysed the turbulent kinetic energy budget. MER wrote the paper with inputs from the rest of the authors.

Paper 6. G. Kreiss initiated the project. H. Holmgren implemented the code and performed the preliminary Stokes simulation. M. Kronbichler simulated the phase field equations. ZG performed the final Stokes simulation and wrote the paper with inputs from the rest of the authors.

Paper 7. LB led the research. ZG implemented the method and wrote the report with input from LB.

Conferences

In addition to journal publications, part of the work in this thesis were presented in the following conferences and workshops. The presenting author is underlined.

Z. GE, L. BRANDT. *Simulation of the self-assembly of colloidal particles in a microchannel.* 7th Summer school of Complex Motions in Fluids. Zenderen, Twente, Netherlands, June 2016.

Z. GE, O. TAMMISOLA, J.CH. LOISEAU, L. BRANDT. *Direct numerical simulation of the self-assembly of colloidal particles in a micro-channel.* 1st International Conference on Multiscale Applications of Surface Tension. Brussel, Belgium, September 2016.

Z. GE, L. BRANDT. *Do self-assembly colloidal droplets behave like droplets?.* 69th Annual Meeting of the APS Division of Fluid Dynamics. Portland, Oregon, USA, November 2016.

Z. GE, O. TAMMISOLA, L. BRANDT. *A fast mass-preserving interface-correction level set/ghost fluid method for colloidal suspensions under depletion forces.* 3rd International Conference on Numerical Methods of Multiphase Flows (ICNMMF-III). Tokyo, Japan, June 2017.

Z. GE, H. HOLMGREN, M. KRONBICHLER, L. BRANDT, G. KREISS. *Effective slip over partially filled microcavities and its possible failure.* 12th European Fluid Mechanics Conference. Vienna, Austria, September 2018.

Z. GE, O. TAMMISOLA, L. BRANDT. *Flow-assisted droplet assembly in a 3D microfluidic channel.* 71st Annual Meeting of the APS Division of Fluid Dynamics. Atlanta, Georgia, USA, November 2018.

Z. GE, O. TAMMISOLA, L. BRANDT. *Flow-assisted droplet assembly in a 3D microfluidic channel.* Scattering and Dynamics of Flowing Soft Material. Lund, Sweden, December 2018.

Z. GE, O. TAMMISOLA, L. BRANDT. *Flow-assisted droplet assembly in a 3D microfluidic channel.* Colloidal Science & Metamaterials (CSM). Paris, France, February 2019.

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Part I

Overview

CHAPTER 1

Introduction

Droplets, one of the most basic forms of fluids, are abundant and ubiquitous in nature. From the falling rain drops to the rising aerosols, from the deforming embryo to the rolling tears, micron to millimetre sized droplets are an integral part of our everyday experience, conditioning and nourishing *life*.

Droplets are also important in many artificial and engineering applications. In material science, technicians can now routinely generate large amount of droplets in massively parallelised microfluidic channels (Tabeling 2005); these liquid drops can serve as microcontainers, encapsulating functional substances, and subsequently be manipulated and scaffolded into larger structures with desired properties. In life science, droplet-based microfluidic devices have already been used to automate genome sequencing and, in the future, may enable more effective drug development by building synthetic test platforms more faithful to the human body than the conventional Petri dish (Squires & Quake 2005; Sontheimer-Phelps *et al.* 2019). In transportation, combustion engines rely on the breakup of liquid fuels into tiny drops to enhance mixing, improve combustion efficiency, and reduce pollutant emissions. Even in publications, printed posters, newsletters or books, including this thesis, would not have the same quality without the precise control of pico-litre (10^{-9} mL) ink droplets.

All the above-mentioned processes or technologies depend on the presence and motion of droplets. Understanding the physics and hydrodynamics of their interactions in a variety of conditions thus has a great practical importance. Traditionally, the study of liquid-embedded systems are mostly conducted by experimenters; fluid mechanics, as most other disciplines of physics, is primarily an experimental science after all. However, the last few decades have also observed a phenomenal paradigm shift in many research areas owing to the exponential growth of computer power and continuous development of numerical algorithms. Fluid dynamics is now in the vanguard of applying large-scale computations in resolving natural and engineering problems and is, in turn, pushing the boundary of computer science. Comparing to experimental measurements, computer simulations are non-invasive and offer the entire information that can be readily visualised up to model errors and the numerical precision. Physical parameters can also be freely adjusted to test conditions that are not easy to implement in lab experiments.

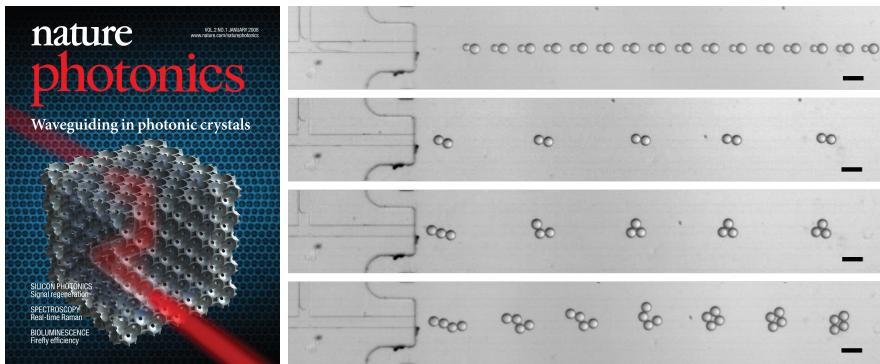


Figure 1.1: (left) Cover photo of the January 2008 issue of *Nature Photonics*, showing an artistic rendering of a light beam passing through a 3D photonic crystal. © Springer Nature. (right) Self-assembly of two to four droplets in a microfluidic chip; scale bar, $100\ \mu\text{m}$. Images courtesy of Dr. Bingqing Shen.

Motivated by these reasons, the present work embarks on a number of problems that originates from material fabrication, surface engineering and fluid transport. We develop and utilise both high- and low-fidelity numerical models to simulate the underlying processes with the aims of complementing experimental studies and elucidating their physical mechanisms. The common ground at the bottom of these enquiries is that the processes all entail interactions of droplets in an ambient fluid or are governed by the dynamics near a fluid interface. Their specific backgrounds are given below (see also Chapter 4).

1.1. Fabricating photonic crystals

Photonic crystals (PhC) are materials patterned with a periodicity in dielectric constant and show great potential for building sophisticated optical circuitry that can route, filter, store or suppress optical signals; see Figure 1.1 (left). They resemble semiconductors in that photons can be manipulated in PhC in a similar fashion as electrons are controlled in silicon-based semiconductors. Furthermore, it has been theoretically predicted that PhC can exhibit a complete photonic bandgap (PBG), *i.e.* a range of frequencies over which light cannot propagate in any direction within the material (Yablonovitch 1987). This unique property provides the basis for numerous optoelectronic applications, including low-threshold lasers, low-loss waveguides, on-chip optical circuitry and fibre optics (Meade *et al.* 1994; Joannopoulos *et al.* 1997; Rinne *et al.* 2008).

Despite the theoretical promise, fabricating photonic crystals with 3D bandgaps remains a practical challenge in the colloidal community (Velev & Gupta 2009; Sacanna & Pine 2011; Shen *et al.* 2016). The issue is twofold. On the one hand, ideal PhC has the structure of a diamond cubic, which can have up

to 40% bandgap opening (Ho *et al.* 1990).¹ Therefore, for a long time, the holy grail for photonic researchers has been designing a protocol that can position colloidal droplets in a diamond cubic with alternating dielectric constants. In practice, however, diamond crystals are extremely difficult to assemble because of the large gaps in between; specifically, a diamond cubic of equal spheres has at most 34% packing fraction, while a face-centered cubic can reach 74%. This makes PBG hard to achieve in 3D considering its sensitivity to structural defects. On the other hand, even if one does aim for the diamond structure, current techniques relying on Brownian motions and “patchy particles” are too slow to build PhC in the range of visible light (Yi *et al.* 2013). In some sense, droplets assembled this way are assembled by chance, which favors anisotropic structures with a higher rotational entropy (Meng *et al.* 2010).

In the last six years, an alternative approach has emerged – an approach based on the experiments of Shen *et al.* (2014, 2016), which proposes to use high-throughput microfluidic channels to first produce elementary clusters of droplets with controlled morphologies (trimers, diamond, tetrahedrons, etc.) then solidify and stack them into larger photonic crystals with a finite bandgap; see Figure 1.1 (right). The advantages of this *flow-assisted* assembly strategy are the potentially large production rate and robust control. The remaining questions include (i) why the droplets self-organize into those clusters and, if we understand that, (ii) how to harness this self-assembly mechanism to build photonic materials. The answers are discussed in *Paper 1-4*.

1.2. Modelling dense particle suspensions

Dense suspensions are mixtures of particles in a fluid medium where the former strongly interact via short-range lubrication or non-hydrodynamic forces. There are a number of practical or theoretical reasons to model such systems, which are perhaps best described by the following paragraph quoted from the KITP program on *Physics of Dense Suspensions* held in the spring of 2018:²

“Dense, or high-solid-loading, suspensions of particulates in liquids are found in a range of applications across many industrial sectors and in geophysical flows. The size range of the particles may vary from the colloidal (say, 10 nm to 10 μm , where Brownian motion dominates) to the granular (say, 10 μm to 10 mm). At low solid loading, suspensions are typically fluid-like, but at high loadings, their behavior is complex and diverse. In particular, a number of flow transitions can occur as the applied stress or shear rate increases – from solid to flowing, from a low- to high-viscosity state, or from flowing to jammed. There is great practical interest in unraveling the origins of these and other non-linear flow phenomena in suspensions spanning the whole range of particle sizes. Such advances will form the basis of a predictive science of suspensions in diverse contexts, from cement mixing to advanced additive manufacturing. At the same time, elucidating the physical basis for such

¹The percentage corresponds to the portion of frequencies that propagation of light is forbidden.

²URL: <https://www.kitp.ucsb.edu/activities/suspensions18>. Accessed on 2020-01-22.

behavior will be challenging, and is of great fundamental interest to a large community of soft matter and statistical physicists interested in non-equilibrium phenomena.”

In partial response to the challenge, we numerically explore dense suspensions under different imposed flow conditions and geometric constraints. In particular, we study particles in the noncolloidal regime, and ask: (i) how to minimally characterise the microstructural evolution in an unsteady flow. Candidate answers include the complex viscosity, particle diffusivity, suspension fabric, size distribution of local clusters, structure factor, etc. (Brady & Bossis 1988; Torquato & Stillinger 2003; Ness *et al.* 2017). (ii) Can qualitatively different global behaviours, such as a rate-dependent rheology vs. a rate-independent rheology, occur to the same suspension under different flow protocols (Ge *et al.*, in preparation)? And (iii) how does the particle geometry affect the rheology and jamming of a suspension at fixed volume fractions? Solving these problems requires a numerical methodology that is both accurate enough to test different physical hypotheses and efficient enough to generate data at a wide range of parameters. The implementation detail of one such algorithm is documented in *Paper 7*.

Thesis structure. In the remaining of the thesis, an extended overview on droplet interactions and suspension flow is first summarized from previous studies in the literature. This is the *Part I* of the thesis and contains four more chapters. In Chapter 2, we formulate the microhydrodynamics mathematically and spotlight the essential physics for one, two and more particles in various flow fields or geometries. These are the prerequisites for understanding droplet interactions in more complicated conditions. In Chapter 3, we introduce two categories of numerical methodologies that either solve the governing equations directly or approximate the solution in reduced order. Each category is then divided into several classes, which have further variations and are mentioned succinctly. These are followed by a short detour in Chapter 4, where two additional topics studied alongside the main subject are motivated and described. Finally, Chapter 5 concludes the first part of the thesis, leading us to *Part II*, where the original results are collected in the form of papers.

CHAPTER 2

Microhydrodynamics

This chapter summarizes the mathematical formulation and theoretical basis of microhydrodynamics and particle interactions. By *particle*, we loosely mean a solid or liquid body of micron to millimetre in size, which may also be referred to as *droplet* for abuse of language. As preliminaries, most of the contents below are given without any proof. The interested reader is encouraged to read Batchelor (1967); Happel & Brenner (1983); Pozrikidis (1992); Kim & Karrila (2013); Graham (2018) for greater details.

To begin with, recall the incompressible *Navier-Stokes equations*

$$\nabla \cdot \mathbf{u} = 0, \quad (2.1a)$$

$$\rho \left(\frac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{u} \right) = -\nabla p + \mu \nabla^2 \mathbf{u} + \mathbf{f}, \quad (2.1b)$$

relating the flow velocity \mathbf{u} with the pressure p in the presence of some force density \mathbf{f} , where ρ and μ are the density and dynamic viscosity of the fluid. Eqs. (2.1), together with appropriate boundary conditions, govern the dynamics of a Newtonian fluid that satisfies¹

$$\boldsymbol{\sigma} = -p \mathbf{I} + \mu \left(\nabla \mathbf{u} + (\nabla \mathbf{u})^T \right), \quad (2.2)$$

where $\boldsymbol{\sigma}$ is the stress tensor, and \mathbf{I} the identity matrix.

Depending on the bounding material, conditions on the velocity or stress may be prescribed as boundary conditions. In the case of a solid wall, it is usually the *no-slip* velocity condition; while in the case of a liquid interface, the following stress condition shall be satisfied,

$$(\boldsymbol{\sigma}_+ - \boldsymbol{\sigma}_-) \cdot \mathbf{n} = \gamma \kappa \mathbf{n} - \nabla \gamma, \quad (2.3)$$

where \mathbf{n} denotes the unit normal vector across the liquid interface (from the $-$ side to the $+$ side), γ the surface tension coefficient, and κ the curvature of the interface (e.g. $\kappa = 2/a$ for a sphere of radius a). The above equation states that both normal and tangential stresses can be discontinuous across an interface.

¹The present thesis only concerns Newtonian fluids as the solvent, though, combined with particles, the suspension may exhibit non-Newtonian bulk behaviours. See Section 2.4.

Eq. (2.1b) can be further non-dimensionalized, using the reference length L , velocity U , time T , pressure $P \equiv \mu U/L$, and force density $F \equiv P/L$ scales, into

$$\text{Re} \left(\text{Sr}^{-1} \frac{\partial \hat{\mathbf{u}}}{\partial \hat{t}} + \hat{\mathbf{u}} \cdot \hat{\nabla} \hat{\mathbf{u}} \right) = -\hat{\nabla} \hat{p} + \hat{\nabla}^2 \hat{\mathbf{u}} + \hat{\mathbf{f}}, \quad (2.4)$$

where the hat signifies dimensionless quantities, and Re and Sr represent the *Reynolds* and *Strouhal* numbers, defined as

$$\text{Re} = \frac{\rho U L}{\mu}, \quad \text{Sr} = \frac{U T}{L}. \quad (2.5)$$

A capillary number can also be defined to non-dimensionalize the surface tension appearing in Eq. (2.3),

$$\text{Ca} = \frac{\mu U}{\gamma}. \quad (2.6)$$

2.1. Stokes flow and its symmetries

When both Re and ReSr^{-1} are small, Eq. (2.4) reduces to the *Stokes equation*,

$$-\nabla p + \mu \nabla^2 \mathbf{u} + \mathbf{f} = \mathbf{0}, \quad (2.7)$$

or, equivalently

$$\nabla \cdot \boldsymbol{\sigma} + \mathbf{f} = \mathbf{0}. \quad (2.8)$$

Physically, the Stokes equation describes systems that are characterised by small length scales or are in slow motions, which are typically encountered in microfluidic or biological applications. For instance, the Reynolds number based on a $10 \mu\text{m}$ droplet traveling at a speed of $10 \mu\text{m}/\text{s}$ in water ($\rho = 10^3 \text{ kg/m}^3$, $\mu = 10^{-3} \text{ Pa}\cdot\text{s}$) is 10^{-4} ; and if there is no imposed frequency, the Strouhal number will be 1. Manifestly, both $\text{Re} \ll 1$ and $\text{ReSr}^{-1} \ll 1$.

Mathematically, the removal of the nonlinear term in Eq. (2.1b) dramatically simplifies the solution. In general, the Stokes flow has the following symmetries and properties.

1. *Linearity*, which makes analytical solutions possible in simple cases and the superposition principle applicable in general. For example, if (\mathbf{u}_1, p_1) is a solution to $\mathbf{f} = \mathbf{f}_1$ and (\mathbf{u}_2, p_2) is a solution to $\mathbf{f} = \mathbf{f}_2$, then $(\alpha \mathbf{u}_1 + \beta \mathbf{u}_2, \alpha p_1 + \beta p_2)$ is a solution to $\mathbf{f} = \alpha \mathbf{f}_1 + \beta \mathbf{f}_2$.
2. *Reversibility*, or *time reversal symmetry*. This is a consequence of the linearity and entails that, if the forcing changes from \mathbf{f} to $-\mathbf{f}$, the flow should reverse, *i.e.* $(\mathbf{u}, p) \rightarrow (-\mathbf{u}, -p)$. While seemingly redundant, the reversibility symmetry can be very powerful in making *reductio ad absurdum* type-of arguments, see e.g. the classical note of Purcell (1977).
3. *Stress equilibrium*, which states that any force exerted within the fluid is transmitted instantaneously to the boundary or, if there is no boundary, to infinity (Graham 2018).

4. *Lorentz reciprocal identity*, which relates two solutions, $(\mathbf{u}', \boldsymbol{\sigma}')$ and $(\mathbf{u}'', \boldsymbol{\sigma}'')$ (they may differ by boundary conditions), of the Stokes equation by,

$$\begin{aligned} \int_V \left(\mathbf{u}' \cdot (\nabla \cdot \boldsymbol{\sigma}'') - \mathbf{u}'' \cdot (\nabla \cdot \boldsymbol{\sigma}') \right) dV = \\ \int_S \left(\mathbf{u}' \cdot (\boldsymbol{\sigma}'' \cdot \mathbf{n}) - \mathbf{u}'' \cdot (\boldsymbol{\sigma}' \cdot \mathbf{n}) \right) dS, \end{aligned} \quad (2.9)$$

where V is some enclosed volume with surface S . This identity is often useful in analytical calculations.

5. *Minimum dissipation principle*. This variational principle asserts that the flow minimizing the energy dissipation rate subject to incompressibility under certain boundary conditions is the Stokes flow under the same boundary conditions. Turbulent flow, on the contrary, is very dissipative.

2.2. Single particle in unbounded Stokes flow

When particles are present in a fluid, the coupled dynamics is often significantly more complex than a single phase Stokes flow. Theoretical analyses are thus limited to a handful of simple problems. However, even in a simplified condition, an analytical solution often provides us useful physical intuitions that may be applied to more complicated cases. Below, we list some elementary solutions of the Stokes equation in its simplest setup, *i.e.* a single particle in unbounded Stokes flow.

Imagine the force density \mathbf{f} in Eq. (2.7) is singularly supported at the origin, *i.e.* $\mathbf{f}(\mathbf{x}) = \mathbf{F}\delta(\mathbf{x})$, δ being the Dirac delta. This might represent a reasonable approximation of a small sedimenting particle viewed from a large distance. Then, the solution of the Stokes equation due to this forcing can be written as

$$\mathbf{u}(\mathbf{x}) = \mathbf{G}(\mathbf{x}) \cdot \mathbf{F}, \quad (2.10)$$

where

$$\mathbf{G}(\mathbf{x}) = \frac{1}{8\pi\mu} \left(\frac{\mathbf{I}}{|\mathbf{x}|} + \frac{\mathbf{x}\mathbf{x}}{|\mathbf{x}|^3} \right), \quad (2.11)$$

is the free-space Green's function, also known as the *Stokeslet* or *Oseen tensor*.

Now, if the particle has a finite radius a and the background fluid has a linear velocity \mathbf{u}^∞ in the absence of the particle, we can perform a Taylor expansion of \mathbf{G} and integrate Eq. (2.10) over the particle surface to obtain a solution of the following form (in index notation)²

$$u_i(\mathbf{x}) - u_i^\infty(\mathbf{x}) = -G_{ij}(\mathbf{x})F_j^d + \frac{\partial G_{ij}}{\partial x_l}(\mathbf{x})D_{jl} + \mathcal{O}\left(\frac{a^3}{|\mathbf{x}|^3}\right), \quad (2.12)$$

where \mathbf{F}^d denotes the total drag force exerted on the particle by the fluid, and \mathbf{D} is an integral expression representing a force dipole ($\sim \mathbf{F}\mathbf{h}$, \mathbf{h} being a separation vector). For purposes of analytical calculations, it is customary to

²In this simple example, we require the point \mathbf{x} to be far from the particle so that the terms involving the Green's function can be taken outside the integral, see Graham (2018).

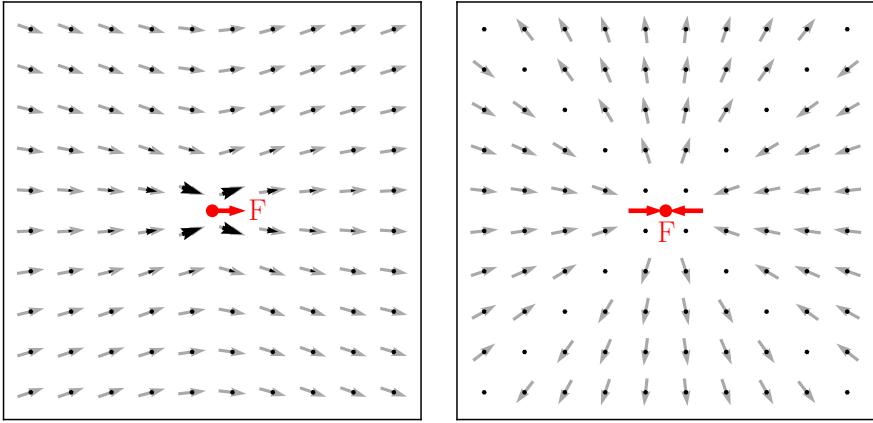


Figure 2.1: Flow fields due a Stokeslet (left) and a stresslet (right) centered at the origin (red dot). The black arrows display the velocity magnitude, while their directions are also indicated by the gray arrows. The flow fields are both axi- and fore-aft symmetric. Note the difference in the velocity magnitude of the two flows.

decompose \mathbf{D} into a traceless symmetric part \mathbf{S} , called the *stresslet*, and an antisymmetric part \mathbf{R} , called the *rotlet*, *viz.*

$$\mathbf{S} = \mathbf{D}^S - \frac{1}{3} \text{Tr}(\mathbf{D}) \mathbf{I}, \quad (2.13a)$$

$$\mathbf{R} = \mathbf{D}^A, \quad (2.13b)$$

where $D_{ij}^S = (D_{ij} + D_{ji})/2$ and $D_{ij}^A = (D_{ij} - D_{ji})/2$.

The second term on the right-hand side of Eq. (2.12) can then be expressed as

$$\frac{\partial G_{ij}}{\partial x_l}(\mathbf{x}) D_{jl} = T_{lij}^{STR}(\mathbf{x}) S_{jl} + T_{lij}^{ROT}(\mathbf{x}) R_{jl}, \quad (2.14)$$

where

$$T_{ijk}^{STR}(\mathbf{x}) = -\frac{1}{8\pi\mu} \frac{3x_i x_j x_k}{|\mathbf{x}|^5}, \quad (2.15a)$$

$$T_{ijk}^{ROT}(\mathbf{x}) = -\frac{1}{8\pi\mu} \frac{\delta_{jk} x_i - \delta_{ij} x_k}{|\mathbf{x}|^3}. \quad (2.15b)$$

Eq. (2.12) is the *multipole expansion* of the disturbance velocity, $\mathbf{u} - \mathbf{u}^\infty$. One immediate physical implication is that a forced particle (e.g. a rising bubble) will generate a disturbance that decays as $1/|\mathbf{x}|$, while a force-free particle (e.g. a self-propelled bacterium) has a $1/|\mathbf{x}|^2$ decay thus leaving less trace at the same distance. See Figure 2.1 for illustrations.³

³The rotlet flow is simply a rigid-body rotation centered at the origin.

Finally, for a sphere of radius a , translating with velocity \mathbf{U} and rotating with angular velocity $\boldsymbol{\Omega}$, its total drag, torque, and stresslet are given by the *Faxén's laws*,

$$\mathbf{F}^d = 6\pi\mu a \left(\left(1 + \frac{a^2}{6} \nabla^2 \right) \mathbf{u}^\infty - \mathbf{U} \right), \quad (2.16a)$$

$$\mathbf{T}^d = 8\pi\mu a^3 (\boldsymbol{\omega}^\infty - \boldsymbol{\Omega}), \quad (2.16b)$$

$$\mathbf{S} = \frac{20}{3}\pi\mu a^3 \left(1 + \frac{a^2}{10} \nabla^2 \right) \mathbf{E}^\infty, \quad (2.16c)$$

where $\mathbf{E}^\infty = (\nabla \mathbf{u}^\infty + (\nabla \mathbf{u}^\infty)^T)/2$ is the rate-of-strain tensor.

2.3. Droplet interactions in shear flow

The preceding section presents the most basic solutions of the Stokes equation in the presence of one particle. In practice, however, it is rarely the case particles or droplets exist in isolation. Particles tend to abound; and they often interact with each other through various types of hydrodynamic or non-hydrodynamic mechanisms, giving rise to a multitude of phenomena in natural or engineering applications. For example, soap bubbles are always in a cluster due to the interfacial capillary attraction, while the same underlying principle can be applied to manipulate droplet motions on a surface to aid liquid handling (Cira *et al.* 2015). To offer at least some partial insights on the hydrodynamics, we highlight recent theoretical developments of droplet interactions in bulk microflow, starting from the simplest case of two particles immersed in unbounded simple shear flow.

2.3.1. Pair interactions in unbounded flow

Among the early studies of suspension hydrodynamics, Batchelor & Green (1972a) solved completely the dynamics of two rigid spheres of arbitrary sizes subject to a background fluid motion whose velocity at infinity is a linear function of position, *i.e.* $\mathbf{u}^\infty \sim \mathbf{E}^\infty \cdot \mathbf{x} + \boldsymbol{\omega}^\infty \times \mathbf{x}$ as $|\mathbf{x}| \rightarrow \infty$. The solution entails the relative velocity of the two sphere centers and their force dipole strengths (related to the effective viscosity of the suspension, see Section 2.4), and is given in terms of several geometric parameters functions of the spheres' relative position and size ratio. In the special case of two equal-sized spheres suspended in simple shear flow, the relative velocity can be integrated numerically to yield a trajectory map as in Figure 2.2. Here, two types of trajectories can be identified: (i) an *open* trajectory where one particle approaches the other particle from one side, overtakes it, and continues in the same direction to infinity; and (ii) a *closed* trajectory where two particles stay bound and rotate around each other permanently regardless of how far they may separate temporarily. It was noted by the authors that, under such ideal conditions (*e.g.* smooth particle surface), the separatrix between open and closed trajectories encloses an infinite volume. Furthermore, if the two particles are once aligned in the flow direction, they will form a closed loop forever.

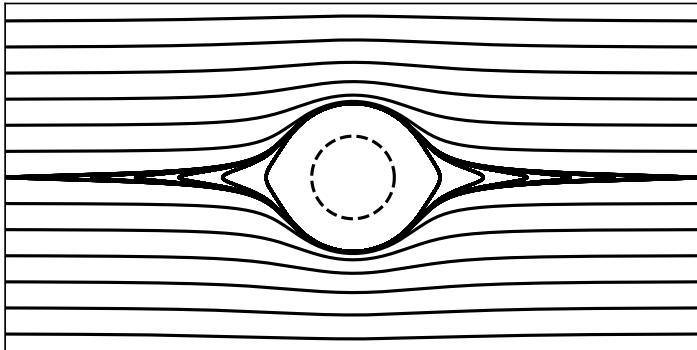


Figure 2.2: Trajectory map of two equal spheres in unbounded simple shear flow. The dashed circle indicates the reference sphere, while the solid lines depict the relative trajectories of the second sphere in all planes of axisymmetry, which include the shear plane.

Zinchenko (1983, 1984) considered the problem when the two particles are “fluid spheres”. Specifically, he calculated the scalar parameters introduced in Batchelor & Green (1972a) for two identical droplets, and showed that they are functions of the relative position and viscosity ratio, λ , defined as the ratio between the droplet and the solvent viscosities. For $\lambda = 0.25$ to 20, the phase diagram in simple shear flow has similar topological features to the case of rigid particles,⁴ *i.e.* an open region and a closed region, though its asymptotic limit depends on the viscosity ratio and short-range non-hydrodynamic interactions.⁵

For the above analysis to hold for liquid droplets, an additional constraint must be introduced, *i.e.* $\text{Ca} \ll 1$. This is usually the case for microfluidic applications. For instance, recall the case considered in the beginning of Section 2.1, and suppose it is an oil droplet traveling in water under a relatively large local shear rate of $\dot{\gamma} = 10^3 \text{ s}^{-1}$. The surface tension coefficient would be around 0.05 N/m, depending on surfactants and the temperature, resulting in $\text{Ca} = \mu(a\dot{\gamma})/\gamma \sim 10^{-4}$. Clearly, the capillary number is small enough for the droplet to remain spherical in the flow.

We note that, in both cases, the relative particle velocity is proportional to the shear rate of the underlying flow. That is, doubling the shear will result in twice as fast relative motions between the particles, though the relative velocity remains much smaller than the absolute velocity in the lab frame (typically

⁴Different types of trajectories were also hypothesized, which may only exist in the case of liquid drops. See Zinchenko (1984) for details.

⁵In the original work of Zinchenko, he used the DLVO theory, which includes the van der Waals attraction and the electrostatic repulsion, for droplets in close contact. Other type of forces, such as the depletion attraction (cf. *Papers 1 & 2*), are also possible and may lead to different pair interactions.

two orders-of-magnitude smaller). Hence, these are truly slow, but long-range, interactions.

When particles are at close contact, say the surface gap $h \ll a$, with a being the particle radius. Then, the hydrodynamic interaction will be dominated by the lubrication term. Specifically, the lubrication force diverge as a/h in the longitudinal direction, and as $\log(a/h)$ in the transverse directions (Jeffrey & Onishi 1984; Jeffrey 1992). Qualitatively, lubrication functions as a dashpot that prevents incoming particles from colliding and separating particles from overshooting. These will be further discussed and demonstrated in Section 3.2 and *Paper 7*.

2.3.2. Pair interactions under weak confinement

When boundaries cannot be neglected in a binary encounter, how will the particle interactions be modified from the Batchelor & Green (1972a) (BG) solution? Before answering this question, observe that walls may be mathematically replaced with carefully chosen reflection images of the original particles such that the boundary conditions along the walls remain unchanged (Blake 1971; Liron & Mochon 1976). Therefore, we expect pair interactions in bounded flow to differ *qualitatively* from the unbounded ones.

Beginning with the simplest case where only one wall is remotely present, Fouxon *et al.* (2019) recently derived the relative velocity of two rigid particles as a sum of the BG's velocity and a wall-correction term (*Paper 4*). The solution is constructed from an boundary integral representation of channel flow using multipole expansions similar to Eq. (2.12), see Fouxon *et al.* (2017) (*Paper 3*) for details. Figure 2.3 illustrates the main results. Here, the lines depicts the relative trajectories of the second particle in the shear plane, where the velocity is in the x direction and increases with z . The wall is located at $z = -z_0$. Remarkably, there exist two types of closed trajectories (in blue) and two types of open trajectories (in red). Close to the origin, a degenerate of the BG-type closed trajectory is singly connected to an open trajectory at an unstable saddle point at distance r_s from the origin. The latter is not admissible in BG, as the particles swap their z coordinates after a binary encounter rather than returning to the original positions. Such open trajectories have been previously termed “swapping trajectories” and recognized as a wall-induced cross-streamline particle migration mechanism, explaining the anomalously large self-diffusivity observed in a dilute suspension measurement (Zarraga & Leighton 2002; Zurita-Gotor *et al.* 2007). Our derivation and numerical simulations independently verify this result.

Extending further away from the reference particle, we identify a new class of closed trajectories along which the second particle rotates around a neutral equilibrium center. Note that, despite a distant wall, the trajectories remain axisymmetric with respect to the z axis. Thus, all points on the circle $x^2 + y^2 = r_c^2$ are stationary configurations. Slightly moving away from the center point, the second particles will revolve perpetually as it is convected

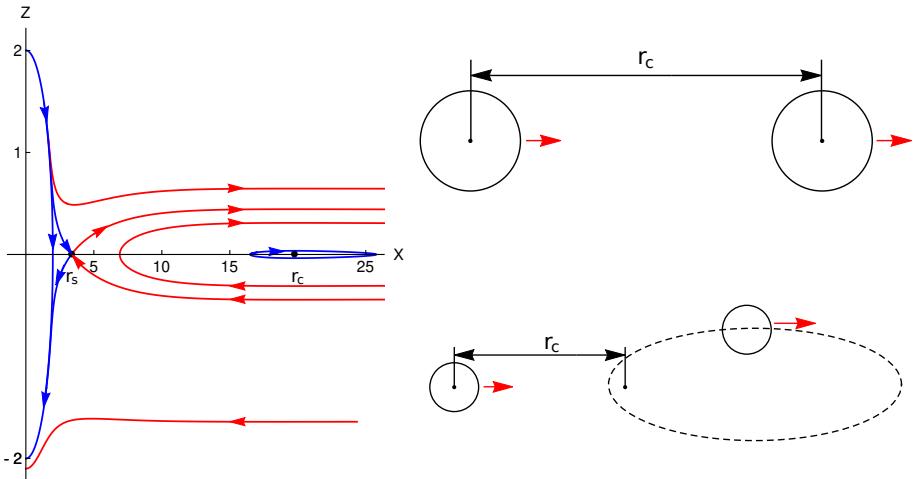


Figure 2.3: (left) Trajectory map of two equal spheres in simple shear flow bounded by a distant wall. The distance between the reference particle (located at the origin) and the wall is $z_0 = 5$ (in units of particle radius, same for the labels of the plot). r_s and r_c denote the saddle point and the neutral equilibrium point, respectively. (right) Schematic illustrations of the stationary configuration (upper) and the dancing trajectory (lower).

by the flow; hence, we call them the “dancing trajectories” (see Figure 2.3, right). The positions of these critical points are approximately related to the wall separation alone. Specifically, $r_s = (32z_0^3/15)^{1/5}$ and $r_c \approx 4z_0$. Overall, the wall is a singular perturbation to the free-space particle interaction.

A few final remarks are in order before we proceed to more confined geometries. Firstly, in a dilute suspension of inertialess particles in Stokes flow, up to leading order in the particle-particle and particle-wall separations, the trajectory of one particle is the same as the fluid streamline resulting from the rest of the particle(s)/wall(s). Therefore, a comparison of the particle and fluid velocities has at least a qualitative meaning, or even quantitatively if the separations are large (Zurita-Gotor *et al.* 2007).⁶ This can be more readily seen from the opposite statement, *i.e.* if the particle trajectories are available, then the flow field can be qualitatively constructed using the particles as tracers.

Secondly, the effect of a rigid boundary on particle interactions can sometimes be intuitively predicted by image analyses utilising basic solutions of the Stokes equation. For example, Zurita-Gotor *et al.* (2007) performed a relatively simple analysis to explain the flow reversal pattern (*i.e.* the swapping trajectories) by calculating the scattering flow. As sketched in Figure 2.4, the wall

⁶This, of course, will be invalid if there are sufficient symmetry-breaking short-range interactions, see e.g. *Paper 2*.

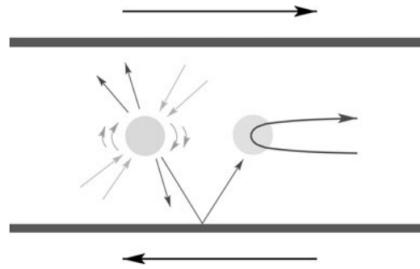


Figure 2.4: Schematic of the wall reflection of the stresslet flow (cf. Figure 2.1) induced by one particle and its qualitative effect on the other particle. Reproduced with permission from Zurita-Gotor *et al.* (2007), © Cambridge University Press.

reflection of the stresslet flow (the particles are force-free) produces a vertical velocity component consistent with what is required for the position swapping (see Zurita-Gotor *et al.* (2007) for the detailed calculations). This flow, despite being very small, is what causes the second particle to be shielded from the first one.

Thirdly, we note that these observed particle interactions may be superimposed pairwise to infer the collective dynamics of a group of particles in similar conditions. To illustrate this point, we simulated two rigid particles in a periodic channel (thus mimicking a 1D crystal) and observed a crossover from permanent separation of the particle pair (for $0.32 \lesssim z_0/H \lesssim 0.68$) to a trapping state (for $z_0/H \lesssim 0.32$ or $z_0/H \gtrsim 0.68$), where H is the channel height;⁷ see Figure 2.5. Qualitatively, the former results from cross-stream migrations that resemble the BG open trajectories, while the latter is due to position swappings with neighbouring particles strongly influenced by the wall. See Beatus *et al.* (2006); Janssen *et al.* (2012); Uspal *et al.* (2013) for a few more analogous and vivid demonstrations.

2.3.3. *Droplet interactions in quasi-two-dimensional geometries*

The last two decades has witnessed a boom of studies on hydrodynamic interactions in strongly confined geometries owing to the development of microfluidics (Cui *et al.* 2002, 2004; Thlusty 2006; Davit & Peyla 2008; Beatus *et al.* 2017). At a first glance, increasing the confinement seems to complicate the problem as the boundary effects become less negligible, see e.g. Liron & Mochon (1976); Fouxon *et al.* (2019). However, by returning to the physical origin of the equations, one can surprisingly gain a useful insight applicable to a wide range of geometries and particle sizes (Diamant *et al.* 2005). Below, we exemplify two such conditions,

⁷The computational setup and notations are the same as the case presented before, cf. Figures 2.3 and 2.4.

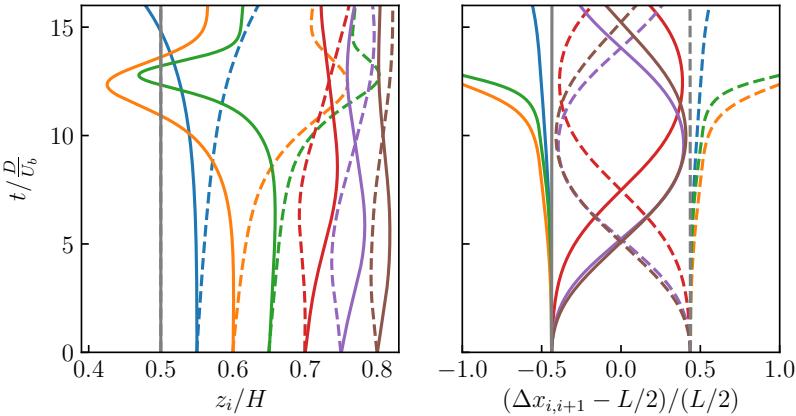


Figure 2.5: Evolution of a particle chain under various initial vertical positions (indicated by the color). The periodic channel has a height of $H = 6$ and a length of $L = 8$, both in units of particle radius. The solid and dashed lines denote the up- and downstream particles in the two-particle system, respectively. For $0.32 \lesssim z_0/H \lesssim 0.68$ (only a half plane is shown due to symmetry), the lines diverge (see the right panel) indicating a permanent separation.

one in which the interacting droplets are confined between two rigid planes, the other between free surfaces. The solutions are simplified by focusing on the far-field interactions and realizing that the problem is *quasi-two-dimensional* (q2D).

Consider the flow between two parallel plates where the gap H is much smaller than the other two dimensions (say x and y); such geometry is commonly known as the *Hele Shaw cell*. Then, from the lubrication theory, the velocity of the flow within the thin layer is approximately prescribed as

$$\mathbf{u}(x, y, z) \approx -\frac{z(H-z)}{2\mu} \nabla p(x, y), \quad (2.17)$$

where the vertical components of the velocity and pressure are neglected, though \mathbf{u} remains z -dependent. Hence, the flow is quasi-two-dimensional.

Under such conditions, the velocity field is irrotational and satisfies the boundary conditions of zero normal components at a rigid boundary in the horizontal plane (Batchelor 1967). That is, a moving droplet (shaped as a pancake in q2D) shall have zero mass flux through its edge, not no-slip. With these requirements, it can be shown that the flow created by the drop far from its surface is proportional to

$$\mathbf{B}^s(\mathbf{x}) \approx -\frac{\alpha H}{\mu} \left(\frac{\mathbf{I}}{|\mathbf{x}|^2} - \frac{2\mathbf{x}\mathbf{x}}{|\mathbf{x}|^4} \right), \quad \text{for } |\mathbf{x}| \gg H, \quad (2.18)$$

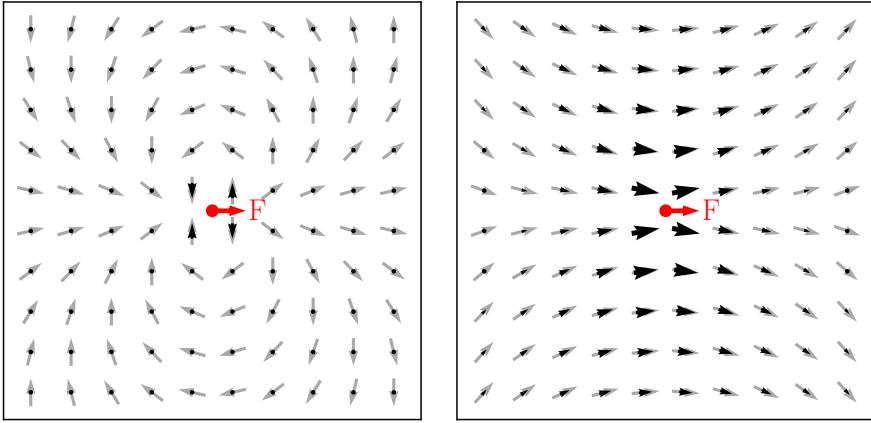


Figure 2.6: Flow fields due to a moving particle in a Hele-Shaw channel (left) and in a soap film (right). Both systems are quasi-two-dimensional.

where α is a dimensionless prefactor depending on the confinement (Diamant 2009).

Physically, the above equation describes a 2D *mass dipole* that displaces the surrounding fluid as the droplet moves. Mathematically, it is the q2D Oseen tensor that approximates the far-field velocity due to forcing on a moving droplet, *i.e.* $\mathbf{u} \sim \mathbf{B}\mathbf{F}$. Specifically, \mathbf{u} decays with the distance as $1/|\mathbf{x}|^2$, which is faster than the free-space Stokeslet flow, but surprisingly slower than the flow caused by a moving particle near a single plate (Cui *et al.* 2004). Heuristic arguments based on conservation laws have been proposed to explain these scaling differences, see Diamant (2009) for details.

To see the flow structure more clearly, we plot the dipolar flow resulting from a horizontal force in Figure 2.6 (left). Here, the most salient feature is the negative transverse coupling of the droplet motion, *i.e.* droplets exert “antidrag” on one another when moving perpendicular to their connecting line (Cui *et al.* 2004). This pattern, however, can also be reversed if the droplets are driven by the flow. In that case, droplets lag behind the bulk flow and the net effect of the dipolar flow is to drag a neighbouring droplet from the behind to the front, or from the middle to the side; see Beatus *et al.* (2006) for schematic illustrations. We note that the magnitude of typical dipolar flow are very small comparing to the driven flow in a dilute suspension. In the limit of $a/H \rightarrow 0$, where a is the particle radius, the prefactor in Eq. (2.18) is analytically found to be $\alpha = 3/(32\pi) \approx 0.030$ (Liron & Mochon 1976). The measured value at $a/H \approx 0.45$ is $\alpha \approx 0.019$ (Diamant *et al.* 2005). However, if the droplet nearly blocks the entire channel cross-section, the resulting flow will be plug-like and has a constant velocity (Beatus *et al.* 2017).

An interesting variation of the aforementioned q2D system is to change the boundary from solid plates to free surfaces. Physically, this could be realized by trapping colloidal particles within a soap film using optical tweezers (Di Leonardo *et al.* 2008).

The Oseen tensor describing the long-range interactions within such a system is given as (Di Leonardo *et al.* 2008; Diamant 2009)

$$\mathbf{B}^f(\mathbf{x}) \approx \frac{1}{4\pi\mu H} \left[(\ln(1/\kappa r) - 1) \mathbf{I} + \frac{\mathbf{x}\mathbf{x}}{|\mathbf{x}|^2} \right], \quad \text{for } |\mathbf{x}| \gg H, \quad (2.19)$$

where κ^{-1} is a cutoff length necessitated by the lateral system size or finite liquid inertia; *i.e.* the radial velocity component vanishes for $|\mathbf{x}| \geq \kappa^{-1}$. Upon visualisation of the resulting flow field (Figure 2.6, right), it is obvious that particle interactions have a much longer range in free-standing liquid films. Furthermore, the transverse motions remain strongly coupled at distances where the longitudinal mode has mostly decayed. The slow logarithmic decay comparing to the dipolar flow in a Hele Shaw cell is physically resulting from the momentum conservation at the liquid boundaries, as opposed to momentum absorption by the rigid plates (Diamant 2009). This difference may have implications to protein transport in biological membranes (Saffman & Delbrück 1975).

Finally, we emphasise that the above analyses are strictly limited to far-field interactions as a consequence of the dimensionality reduction. To consider near-field interactions, the full Navier-Stokes or Stokes equations often have to be solved to accurately resolve the detailed flow fields near the droplet, see e.g. Zhu & Gallaire (2016); Ge *et al.* (2019). In general, q2D analyses are applicable if the system contains a limiting length scale smaller than any other relevant distances, and if a long time evolution is of interest. 3D calculations, however inconvenient, cannot be avoided in most other situations.

2.4. Bulk properties and suspension rheology

When the number of particles is large within a suspension, analytical calculations of their relative motions can quickly become forbidden due to the complexities of the N -body problem.⁸ This is because, despite the fact that we know how to calculate the interaction at the individual level, the collective dynamics are often in regimes that are far from equilibrium, for which there is no established theoretical framework to rely on. Even if we are primarily interested in averaged properties of a suspension, *a priori* predictions can seldom be made as the *microstructure*, *i.e.* the spatial arrangement of each suspending particle, both determines and is determined by the bulk motion in a dynamic way (Brady & Bossis 1988). Nevertheless, in the following, we will introduce a few macroscopic properties of particle suspensions and show how they can be related to certain measurable or computable system parameters. The purpose is to provide a basic

⁸Theoretically, $N = 3$ is the lower bound to break the time reversal symmetry.

physical understanding and, in particular, the rheological characterizations of a particulate system.

2.4.1. Sedimentation velocity

For a sedimenting suspension, the average velocity is given by

$$\langle \mathbf{U} \rangle = \langle \mathbf{M} \rangle \cdot \mathbf{F}, \quad (2.20)$$

where \mathbf{M} is the *mobility tensor* that relates the overall forcing \mathbf{F} , such as gravity, to the particle velocity \mathbf{U} , cf. Eq. (2.11); and $\langle \cdot \rangle$ denotes an average over all particles followed by an average over all configurations. Instantaneously, \mathbf{M} depends only on the particle configuration instead of the velocity (Durlofsky *et al.* 1987); its diagonal blocks represent the single-body drag coefficients, while its off-diagonal blocks embody the hydrodynamic interactions between the particles. In addition, for rigid particles undergoing purely hydrodynamic interactions, \mathbf{M} is symmetric and positive definite due to the dissipative nature of the system (Graham 2018). This property facilitates the proper inversion of \mathbf{M} to produce the many-body approximation to the resistance matrix as required in some reduced-order computational models, see Section 3.2.

2.4.2. Diffusion tensor

The N -particle diffusion tensor \mathbf{D} can be generalized from the Einstein relation (Einstein 1905) and is given by

$$\mathbf{D} = k_B T \mathbf{M}, \quad (2.21)$$

where k_B is the Boltzmann constant, T is the temperature, and \mathbf{M} is again the mobility tensor. That is, diffusion and mobility have the same qualitative meaning for a particulate system.

For non-Brownian suspensions, several particle diffusivities may be defined, though the motion of a particle is typically not diffusive except at short and long times (Brady & Bossis 1988). In the short-time limit, the self-diffusivity \mathbf{D}_0^s can be taken as the diagonal of \mathbf{D} per particle and average over all particles; whereas in the long-time limit, the self-diffusivity \mathbf{D}_∞^s is usually defined with the mean-square displacement, *viz.*

$$\mathbf{D}_\infty^s = \lim_{t \rightarrow \infty} \frac{\langle (\mathbf{x}_i - \mathbf{x}_{i,0})^2 \rangle_i}{2t}, \quad (2.22)$$

where \mathbf{x}_i is the position of particle i at $t \rightarrow \infty$, $\mathbf{x}_{i,0}$ is its starting position, and $\langle \cdot \rangle_i$ is an average over i . Note that self-diffusivities are vectors of the same size of the dimensionality of the space, d . For an isotropic suspension, it can also be expressed as a scalar by averaging the components of \mathbf{D}^s over d .

2.4.3. Stress tensor

One of the central tasks of rheology is to determine the bulk stress tensor $\langle \boldsymbol{\Sigma} \rangle_i$ of a flowing material, expressed as (Batchelor 1970, 1977)

$$\langle \boldsymbol{\Sigma} \rangle_i = -\boldsymbol{\Pi} + 2\mu \boldsymbol{E}^\infty + \frac{N}{V} \left(\langle \boldsymbol{S}^H \rangle_i + \langle \boldsymbol{S}^B \rangle_i + \langle \boldsymbol{S}^A \rangle_i \right), \quad (2.23)$$

where V is the enclosing volume, N is the number of particles, and \boldsymbol{S} 's denote the stresslets due to various particle interactions, including the hydrodynamic (H), Brownian (B), and any other applied (A) forces, as written. Apart from the viscous stress tensor, $-\boldsymbol{\Pi} + 2\mu \boldsymbol{E}^\infty$, due to the underlying flow, the rest of the terms in Eq. (2.23) are the particle contribution. In general, these stress components are all related to the suspension microstructure. In the case of the hydrodynamic stresslet, it can be computed as

$$\langle \boldsymbol{S}^H \rangle_i = -\langle \boldsymbol{R}_{SU} \cdot (\boldsymbol{U} - \boldsymbol{U}^\infty) - \boldsymbol{R}_{SE} : \boldsymbol{E}^\infty \rangle_i, \quad (2.24)$$

where \boldsymbol{R}_{SU} and \boldsymbol{R}_{SE} are configuration-dependent resistance matrices that resemble \boldsymbol{M}^{-1} . See Section 3.2.1 and *Paper 7* for further details.

Suppose that the bulk stress tensor has been determined, then a number of *effective* rheological properties can be readily extracted as follows. Specifically, we focus on non-Brownian, rigid, neutrally buoyant spheres suspended in a Newtonian fluid with viscosity μ undergoing a simple shearing motion in the xy plane with shear rate $\dot{\gamma}$.

1. Suspension viscosity $\eta_s = \Sigma_{xy}/(\mu\dot{\gamma})$.

When particles are added to a fluid, the increase of the suspension viscosity is linear with the shear rate if there is no other associated time scale in the system (Hinch 2011). Moreover, dimensional analysis indicates that there is only one independent variable, the volume fraction of the particles, ϕ . For very dilute suspensions (typically up to volume fraction $\phi \approx 0.05$), Einstein (1906, 1911) has shown that $\eta_s = 1 + 2.5\phi$, assuming the particles only perturb the surrounding flow but do not interact with each other. Including pair interactions, Batchelor & Green (1972b) calculated the suspension viscosity up to the quadratic order. For semi-dilute suspensions (up to $\phi \approx 0.15$) in simple shear flow, it follows $\eta_s = 1 + 2.5\phi + 5\phi^2$. As the concentration continues increasing, analytical calculations of the stress tensor become difficult due to coupled hydrodynamic interactions and contact forces. Plenty of empirical relationships have been proposed, such as that of Eilers (1941), $\eta_r = \mu[1 + (5\phi/4)/(1 - \phi/\phi_J)]^2$, where ϕ_J is the jamming fraction. The latter may depend on the particle shape and friction coefficient. For example, random close packing of frictionless sphere predicts $\phi_J \approx 0.64$, while in experiments suspensions usually jam under 60% volume fraction. See Guazzelli & Pouliquen (2018); Morris (2020) for detailed reviews on the rheology of dense suspensions.

2. Particle pressure $\eta_n = -\text{Tr}(\boldsymbol{\Sigma})/(3\mu\dot{\gamma})$.

Reminiscent of the osmotic pressure in a molecular fluid, particles in a suspension can generate an isotropic stress resulting from their hydrodynamic and other types of interactions (Brady 1993). Defined as the minus one-third the trace of the first moment of the surface stress acting on the particle, Jeffrey *et al.* (1993) derived the analytical expressions for the cases of one and two particle(s) in linear flow. In the former case, a Faxén law for the pressure moment is given as $S = 4\pi a^3 p^\infty$, where p^∞ is the ambient pressure due to the point force, cf. Eq. (2.16). As the particle concentration increases, η_n tends to diverge similarly to the suspension viscosity. In simple shear flow, a possible constitutive relationship is suggested as $\eta_n = [\phi/(\phi_J - \phi)]^2$ (Boyer *et al.* 2011).

3. Normal stress differences $N_1 = \boldsymbol{\Sigma}_{xx} - \boldsymbol{\Sigma}_{yy}$ and $N_2 = \boldsymbol{\Sigma}_{yy} - \boldsymbol{\Sigma}_{zz}$.

When the suspension is non-dilute, normal stresses can deviate from each other as a result of the anisotropic microstructure or particle collision. This leads to the definitions of the first and second normal stress differences as above, where x , y , z denote the flow, velocity-gradient, and negative vorticity directions, respectively. N_1 and N_2 are usually normalized by the modulus of the shear stress, $|\boldsymbol{\Sigma}_{xy}|$, since they also diverge as $\phi \rightarrow \phi_J$. The exact relationship between N_1 or N_2 and ϕ have not been clearly found. The overall picture is that N_1 is mostly of hydrodynamic origin, while N_2 is negative and large in magnitude due to particle collisions, which happen more frequently in the xy plane (Guazzelli & Pouliquen 2018). See Seto & Giusteri (2018) for further discussions.

CHAPTER 3

Numerical Methods

The previous chapter depicts the physical pictures of droplet interactions and suspension flow in a few simplified conditions. In the current chapter, we peek in the technical details of various numerical methods when theoretical analyses are insufficient and the full equations have to be simulated to describe the coupled fluid and particle dynamics. First, we discuss standard Navier-Stokes solvers along with different interface-tracking/capturing methods. Simulations adopting these approaches intend to resolve all spatial and temporal scales within the system, and sometimes qualify as *direct numerical simulations* (DNS). Then, we discuss a special class of methods that solves the particle dynamics without directly computing the flow, thus is much faster, but becomes invalid beyond the Stokes flow approximation. Depending on the level of simplifications, these may fall under the category of *coarse-grained* dynamics. In view of the large amount of numerical methods and the constant development of computational fluid dynamics (CFD) in the last several decades, the list of methods is by no means complete. For more comprehensive surveys of available numerical techniques for multiphase flow problems, see Prosperetti & Tryggvason (2007); Rosti *et al.* (2019).

3.1. Interface-resolved one-fluid methods

The incompressible Navier-Stokes equations, Eqs. (2.1), can be solved numerically using the classical *projection method* (Chorin 1968) by repeatedly executing the following steps,

$$\mathbf{u}^* = \mathbf{u}^{(n)} + \Delta t \left(\frac{3}{2} \mathbf{R}\mathbf{U}^{(n)} - \frac{1}{2} \mathbf{R}\mathbf{U}^{(n-1)} \right), \quad (3.1a)$$

$$\nabla^2 p^{(n+1)} = \frac{\nabla \cdot \mathbf{u}^*}{\Delta t}, \quad (3.1b)$$

$$\mathbf{u}^{(n+1)} = \mathbf{u}^* - \Delta t \nabla p^{(n+1)}, \quad (3.1c)$$

where

$$\mathbf{R}\mathbf{U}^{(n)} = -\mathbf{u}^{(n)} \cdot \nabla \mathbf{u}^{(n)} + \nabla^2 \mathbf{u}^{(n)} + \mathbf{f}^{(n)}. \quad (3.2)$$

In the above, superscripts (n) , $*$, and $(n+1)$ denote the discrete time-levels in step of Δt ; and ρ and μ are assumed to be 1 for simplicity. The flow field, \mathbf{u} , is updated in time via a fractional-step procedure, where the first sub-step,

Eq. (3.1a), utilises the second-order Adams-Bashforth (AB2) scheme. Other temporal integration schemes, e.g. the Runge-Kutta (RK) or Crank-Nicolson (CN) methods, can also be used or mixed-and-used depending on the numerical stability and desired accuracy. In moderate-Reynolds-number systems, AB2 is typically favored over other schemes as it is easy to implement (comparing to implicit schemes such as CN) and relatively fast (comparing to high-order schemes such as third-order RK).

In order to evaluate the spatial derivatives in Eqs. (3.1), the flow domain should be discretized so that either finite difference or weighted residual methods can be applied. In essence, finite difference methods express the derivative as a truncated Taylor series expansion, while weighted residual methods assume that the solution can be represented analytically and they are usually formulated as one of the following better-known variants, including finite volume, finite element, or spectral methods (Fletcher 1991). Loosely speaking, finite difference and finite volume schemes are easier to implement on a structured grid; finite element methods are more flexible on non-Cartesian grids; and spectral methods have the highest order-of-accuracy.

Depending on the numerical condition and specific implementation, solving Eqs. (3.1) can be costly. The most computationally intensive part in a projection method is usually Eq. (3.1b), where a Poisson equation for pressure must be solved numerically at each time step. Consequently, much work has gone into developing fast or iterative Poisson solvers that take advantage of fast Fourier transforms, Gauss elimination, or multigrid methods, see Buzbee *et al.* (1970); Swarztrauber (1977); Brandt (1977); Wesseling (2000) for some early examples.

The above entails the most basic algorithm for single-phase flow. When particles or droplets are present, the flow is *multiphase* and additional equations need to be introduced to describe the dispersed phase that modifies the underlying flow. Below, we give a brief overview of some interface-tracking/capturing methods that have been developed along this direction. The common feature of these methods is that they follow the motion of finite-size particles without deforming the original grid where the fluid variables are defined on. Hence, they can all be referred to as “*one-fluid*” methods.

3.1.1. *Front-tracking methods*

Using discrete marker points to specify and track particle motions, front-tracking type-of methods are perhaps the most intuitive approach for modelling finite-size particles and have been used in multiphase flow simulations since the inception of super computers (Daly 1969; Viecelli 1969; Peskin 1972; Glimm 1982; Unverdi & Tryggvason 1992; Tryggvason *et al.* 2001; Pozrikidis 2001; Muradoglu & Tryggvason 2014). The general methodology is to (i) prescribe the particle location by distributing a set of Lagrangian points on the particle surface (see Figure 3.1), (ii) advect them using the local flow velocity, and (iii) update the fluid properties and compute the extra forces at the updated particle position.

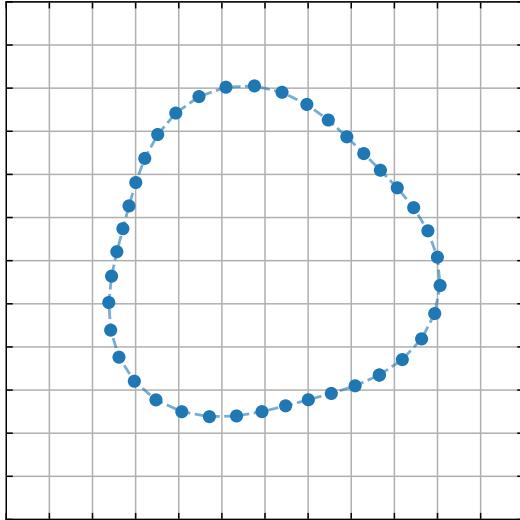


Figure 3.1: Marker points on a triangular droplet.

In the case of liquid droplets, (ii) and (iii) can be written as

$$\frac{d\mathbf{X}_l}{dt} = \mathbf{u}(\mathbf{X}_l), \quad (3.3)$$

$$\mathbf{f} = \gamma \kappa \mathbf{n} \delta(\mathbf{X}_l), \quad (3.4)$$

where \mathbf{X}_l , $l = 1, 2, \dots, N_L$ denote the positions of the Lagrangian points, \mathbf{u} the local flow velocity, \mathbf{f} the forcing term, γ the surface tension coefficient, κ the interface curvature, \mathbf{n} the interface normal vector, and δ the Dirac delta function. Eq. (3.4) corresponds to the normal stress jump condition due to surface tension, cf. Eq. (2.3). No-slip boundary conditions on solid particles can be represented in similar ways.

Despite the conceptual simplicity, there are several technical details that require careful treatment when implementing front-tracking methods. First, due to deformation, breakup or coalescence of the particle, or simply due to the non-uniform flow, the Lagrangian points may need to be redistributed, added, or deleted from time to time to ensure numerical stability and accuracy. This is particularly cumbersome in a parallelised implementation. Second, advection of the marker points requires interpolation of the velocity field, as the moving grid is generally non-aligned with the background fixed mesh. Third, the internal boundary conditions (no-slip or surface tension) are to be constraint on the Eulerian mesh, thus a “spreading” procedure is necessary to feed the particle dynamics back to the flow. This is usually done with a smeared delta function.

With these in mind, now we describe a specific class of front-tracking methods.

Algorithm 1: A multi-direct forcing immersed boundary method for the coupled Navier-Stokes/Newton-Euler equations.

```

//time marching
for  $n = 0, 1, \dots, N$  do
     $\mathbf{u}_{ijk}^0 = \mathbf{u}_{ijk}^{(n)}$ ,  $\mathbf{RU}_{ijk}^0 = \mathbf{RU}_{ijk}^{(n)}$ ,  $\mathbf{RU}_{ijk}^{-1} = \mathbf{RU}_{ijk}^0$  //fluid variables
     $\mathbf{X}_l^0 = \mathbf{X}_l^{(n)}$ ,  $\mathbf{U}_p^0 = \mathbf{U}_p^{(n)}$ ,  $\mathbf{\Omega}_p^0 = \mathbf{\Omega}_p^{(n)}$  //particle motions
     $\mathbf{F}_p^0 = \mathbf{F}_p^{(n)}$ ,  $\mathbf{T}_p^0 = \mathbf{T}_p^{(n)}$ 
    //the RK3 loop
    for  $q = 1, 2, 3$  do
         $\mathbf{u}_{ijk}^* = \mathbf{u}_{ijk}^{q-1} + \Delta t \left( -(\alpha_q + \beta_q) \nabla P_{ijk}^{q-3/2} + \alpha_q \mathbf{RU}_{ijk}^{q-1} + \beta_q \mathbf{RU}_{ijk}^{q-2} \right)$ 
         $\mathbf{u}_{ijk}^{**,-1} = \mathbf{u}_{ijk}^*$ 
        //the mutli-direct forcing loop
        for  $s = 0, 1, \dots, N_s$  do
            //the Lagrangian particle loop
            for  $l = 1, 2, \dots, N_l$  do
                 $\mathbf{U}_l^{**,-s-1} = \sum_{ijk} \mathbf{u}_{ijk}^{**,-s-1} \delta_p(\mathbf{x}_{ijk} - \mathbf{X}_l^{q-1}) \Delta x \Delta y \Delta z$ 
                 $\mathbf{F}_l^{q-1/2,s} = \mathbf{F}_l^{q-1/2,s-1} + \frac{\mathbf{U}_p^{q-1} - \mathbf{U}_l^{**,-s-1}}{\Delta t}$ 
            end
             $\mathbf{f}_{ijk}^{q-1/2,s} = \sum_l \mathbf{F}_l^{q-1/2,s} \delta_p(\mathbf{x}_{ijk} - \mathbf{X}_l^{q-1}) \Delta V_l$  //spreading
             $\mathbf{u}_{ijk}^{**,-s} = \mathbf{u}_{ijk}^* + \Delta t \mathbf{f}_{ijk}^{q-1/2,s}$ 
        end
         $\nabla^2 \tilde{P} = \frac{1}{(\alpha_q + \beta_q) \Delta t} \nabla \cdot \mathbf{u}^{**,-N_s}$  //projection
         $\mathbf{u}^q = \mathbf{u}^{**,-N_s} - (\alpha_q + \beta_q) \Delta t \nabla \tilde{P}$  //correction
         $P^{q-1/2} = P^{q-3/2} + \tilde{P}$ 
        //integration of Newton-Euler
        Evaluate  $\mathbf{F}_p^q$  and  $\mathbf{T}_p^q$  from the current particle configuration
         $\mathbf{U}_p^q = \mathbf{U}_p^{q-1} - \frac{\Delta t}{V_p} \sum_l \mathbf{F}_l^{q-1/2,N_s} \Delta V_l + \frac{(\alpha_q + \beta_q) \Delta t}{2V_p} (\mathbf{F}_p^q + \mathbf{F}_p^{q-1}) + \dots$ 
         $\mathbf{X}^q = \mathbf{X}^{q-1} + \frac{(\alpha_q + \beta_q) \Delta t}{2} (\mathbf{U}_p^q + \mathbf{U}_p^{q-1})$ 
         $\mathbf{\Omega}_p^q =$ 
         $\Omega_p^{q-1} - \frac{\Delta t}{I_p} \sum_l \mathbf{r}_l^{q-1} \times \mathbf{F}_l^{q-1/2,N_s} \Delta V_l + \frac{(\alpha_q + \beta_q) \Delta t}{2I_p} (\mathbf{T}_p^q + \mathbf{T}_p^{q-1}) + \dots$ 
    end
     $\mathbf{u}_{ijk}^{(n+1)} = \mathbf{u}_{ijk}^3$ ,  $\mathbf{RU}_{ijk}^{(n+1)} = \mathbf{RU}_{ijk}^3$ 
     $\mathbf{X}_l^{(n+1)} = \mathbf{X}_l^3$ ,  $\mathbf{U}_p^{(n+1)} = \mathbf{U}_p^3$ ,  $\mathbf{\Omega}_p^{(n+1)} = \mathbf{\Omega}_p^3$ 
     $\mathbf{F}_p^{(n+1)} = \mathbf{F}_p^3$ ,  $\mathbf{T}_p^{(n+1)} = \mathbf{T}_p^3$ 
end

```

Immersed boundary methods. Pioneered by Peskin (1972), immersed boundary (IB) methods were originally used for modelling cardiac mechanics and associated blood flow, and have ever since been continuously modified and

refined for multiphase flow simulations involving moving boundaries (Fadlun *et al.* 2000; Uhlmann 2005; Mittal & Iaccarino 2005; Pinelli *et al.* 2010; Breugem 2012; Favier *et al.* 2014). Here, the governing equations for the particle motion are the Newton-Euler equations,

$$m_p \frac{d\mathbf{U}_p}{dt} = \int_s \boldsymbol{\sigma} \cdot \mathbf{n} dA + \mathbf{F}_p, \quad (3.5a)$$

$$\mathbf{I}_p \frac{d\boldsymbol{\Omega}_p}{dt} + \boldsymbol{\Omega}_p \times (\mathbf{I}_p \boldsymbol{\Omega}_p) = \int_s \mathbf{r} \times (\boldsymbol{\sigma} \cdot \mathbf{n}) dA + \mathbf{T}_p, \quad (3.5b)$$

where \mathbf{U}_p and $\boldsymbol{\Omega}_p$ denote the translational and angular velocities of particle p ; m_p and \mathbf{I}_p are its mass and moment-of-inertia tensor in the body frame (scalar for spheres); and \mathbf{F}_p and \mathbf{T}_p stand for the total force and torque, such as collision or electrostatic repulsion, exerted on the particle center-of-mass (the viscous stress contributions are excluded as they are represented by the integral terms).

The feedback of the particle on the fluid flow are embedded in a diffuse body-forcing term, \mathbf{f} . Depending on whether it is incorporated into the continuous equations before or after discretisation, IB methods can be broadly divided into continuous forcing and direct forcing. Regardless of the approach, the spreading of the Lagrangian force onto the Eulerian mesh is realized with regularized Dirac delta functions, which may take different forms as long as $\sum_{ijk} \delta_p(\mathbf{x}_{ijk} - \mathbf{X}_l) \Delta x \Delta y \Delta z = 1$ is satisfied for all \mathbf{X}_l .

To illustrate the overall algorithm, a pseudo code corresponding to the direct forcing IB method used in *Papers 3 & 4* for spherical particles is provided in Algorithm 1. Here, the code is structured in three nested loops: an outer time-marching loop advancing the unsteady solution, an intermediate third-order Runge-Kutta (RK3) loop integrating the fluid velocity along with the particle motion, and an inner loop for multi-direct forcing iterations. The notation is similar to previous formulations, cf. Eqs. (3.1) and (3.3), with minor omissions of terms or exceptions, such as the definition of **RU** (it does not include \mathbf{f} in Algorithm 1). The objective is to give an overall picture of the complexity of the code. For full details, see Breugem (2012).

3.1.2. Front-capturing methods

Instead of meshing and tracking the moving boundary on a separate grid, an alternative approach is to capture the particle motion on the same grid at the expense of an additional function. Such function may correspond to the local volume fraction of a dispersed phase, or measure a geometric quantity of the interface away from the particle. Conceptually, it specifies the multiphase flow in a full Eulerian manner, and is sometimes referred to as *Eulerian-Eulerian* methods (front-tracking methods would be called *Eulerian-Lagrangian* methods). Many numerical methods fall under the Eulerian-Eulerian framework. In the following, we describe three of them (see Figure 3.2). As always, a brief historical background is given first, followed by the definition of the “front-capturing”

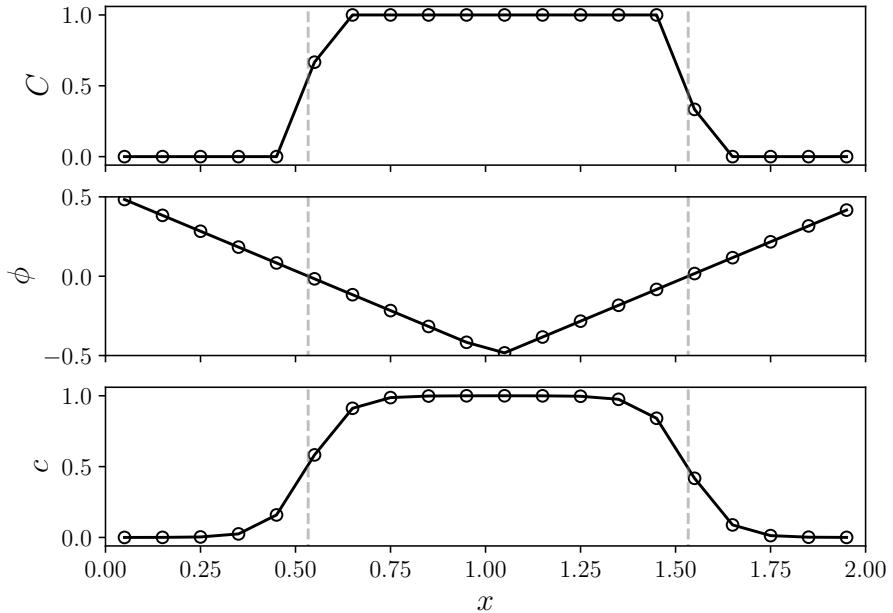


Figure 3.2: Comparison of the three front-capturing methods for a 1D droplet of radius 0.5, centered at $x = 1.033$. Markers indicate the nodal values of the representative functions. Dashed lines depict the boundary of the droplet. (upper) The volume-of-fluid representation, where $C = 1$ inside the droplet. (middle) The level set representation, where $\phi > 0$ outside the droplet. (lower) The phase field representation, where the profile is given by $c = 0.5[1 - \tanh(\phi/\epsilon)]$, with $\epsilon = 0.1$.

function and its governing equation. We also highlight the major advantages and disadvantages of each method on a general basis. Finally, when coupling these front-capturing methods with a flow solver, the internal boundary conditions need to be imposed near the interface and, in principle, can be treated in exactly the same way as in front-tracking methods, cf. Eq. (3.4). We skip these details and refer the reader to Brackbill *et al.* (1992); Fedkiw *et al.* (1999); Lalanne *et al.* (2015); Popinet (2018); Gibou *et al.* (2018); Ge *et al.* (2018) for further discussions.

Volume-of-fluid methods. First proposed in the 1970s, volume-of-fluid (VOF) methods were among the oldest interface-capturing methods for modelling free-surface flow (DeBar 1974; Nichols & Hirt 1975; Hirt & Nichols 1981), and they remain as being the more popular methods for multiphase flow simulations within the CFD community (Scardovelli & Zaleski 1999; Popinet 2003; Pilliod & Puckett 2004; Xiao *et al.* 2005; Ii *et al.* 2012; Aniszewski *et al.* 2019). The

principle idea is, first, introducing a cell-averaged volume fraction, C_k , defined as

$$C_k = \frac{1}{\Delta V_k} \int_{\Omega_k} H(\mathbf{x}) dV, \quad (3.6)$$

where ΔV_k is the volume of computational cell k , and $H(\mathbf{x})$ is a phase indicator function, given as

$$H(\mathbf{x}_k) = \begin{cases} 1 & \text{if } \mathbf{x} \text{ is in the reference fluid,} \\ 0 & \text{otherwise,} \end{cases} \quad (3.7)$$

then, obtaining the temporal variation of the local volume fraction by solving the following equation,

$$\frac{\partial C_k}{\partial t} + \int_{\partial\Omega_k} (\mathbf{u} \cdot \mathbf{n}_k) H(\mathbf{x}) dS = 0, \quad (3.8)$$

where \mathbf{n}_k denote the six normal vectors (in a 3D Cartesian mesh) of surfaces $\partial\Omega_k$, and \mathbf{u} is the local divergence-free velocity.

Note that, in the above formulations, the *total* volume of each fluid phase is conserved by construction. For many engineering calculations, this is the minimum requirement for a long-time simulation to be meaningful and makes VOF appealing in a wide range of industrial problems, such as predicting the impact of ocean waves splashing on the ship hull or designing spray nozzles to enhance fuel mixing in engines. On the other hand, to correctly capture the local interface motion, the volume flux in Eq. (3.8) must be calculated accurately, which requires certain information of the local interface *geometry*, such as the normal direction and curvature. This further requirement is not straightforward knowing only the cell-averaged volume fraction (see Figure 3.2 for a very simplified illustration), therefore, many efforts have been devoted to developing methods that either reconstruct the interface in an extra step (these methods are called geometric VOF) or represent the interface in certain ways such that the volume flux can be computed directly (these methods are called algebraic VOF). See Mirjalili *et al.* (2017) for a recent overview of the different methods.

Level set methods. Another way to model interface evolution is to use level set (LS) methods (Sethian 1999). Originally devised by Osher & Sethian (1988), LS methods encompass a broad class of algorithms and numerical schemes for “front propagation” with curvature-dependent speeds. The mathematical machinery is rooted in the boundary-value and initial-value partial-differential-equations (PDE) perspective on moving interfaces.¹ Its applications include geometry, computer vision, fluid mechanics, combustion, tissue growth, path optimization, etc. In terms of fluid mechanics, specifically, the last three decades

¹It may seem similar to VOF methods in the sense that the both methods are Eulerian and the governing equations can be written in a similar way. However, the level set method embeds the *geometry* of an interface, while VOF has the *volume* embedding. This, together with that level set is Lipschitz-continuous and VOF is discontinuous, makes the two approaches fundamentally different. See Figure 3.2.

has witnessed continuous numerical development in applying level set methods to multiphase flow, see e.g. Mulder *et al.* (1992); Sussman *et al.* (1994); Sussman & Puckett (2000); Enright *et al.* (2002); Olsson & Kreiss (2005); Marchandise *et al.* (2007); Desjardins *et al.* (2008); Desjardins & Pitsch (2009); Aanjaneya *et al.* (2013); Luo *et al.* (2015b); Ge *et al.* (2018). See also Gibou *et al.* (2018) for a recent review.

The level set function, ϕ , is classically defined as the signed distance to the interface Γ ,

$$\phi(\mathbf{x}, t) = \text{sgn}(\mathbf{x})|\mathbf{x} - \mathbf{x}_\Gamma|, \quad (3.9)$$

where $\text{sgn}(\mathbf{x})$ is a sign function equal to 1 or -1 depending on which side of the interface the observation point \mathbf{x} lies, and \mathbf{x}_Γ denotes the point on Γ that has the shortest distance to \mathbf{x} . That is, the interface can be parameterized by ϕ as $\Gamma = \{\mathbf{x} \mid \phi(\mathbf{x}, t) = 0\}$. The interface position is then advected according to the following level set equation,

$$\frac{\partial \phi}{\partial t} + F|\nabla \phi| = 0, \quad (3.10)$$

where F is a speed function. From a PDE perspective, the above equation asserts that the entire level set evolution can be obtained by integrating $\phi(\mathbf{x}, t = 0)$ forward in time. In other words, Eq. (3.10) is an initial value problem without boundary conditions.

Geometrically, the normal and the curvature of the interface can be readily derived from the level set as below,²

$$\mathbf{n} = \frac{\nabla \phi}{|\nabla \phi|}, \quad (3.11a)$$

$$\kappa = \nabla \cdot \mathbf{n}. \quad (3.11b)$$

When solving the level set equation for multiphase flow, the most natural choice for F is the local flow velocity projected in the normal direction, *i.e.* $F = \mathbf{u} \cdot \mathbf{n}$, reducing Eq. (3.10) to

$$\frac{\partial \phi}{\partial t} + \mathbf{u} \cdot \nabla \phi = 0. \quad (3.12)$$

However, any non-uniform flow will distort the LS field, making Eq. (3.9) invalid after a few advection steps and making the numerical solution unstable. To remedy this problem, it is customary to perform an infrequent level set “reinitialization” to reshape ϕ into a distance function while preserving the interface location (Sussman *et al.* 1994). Various techniques have been developed to do so by e.g. using fast marching methods (Sethian 1999), or solving a time-dependent Hamilton-Jacobi equation (Sussman & Fatemi 1997; Russo & Smereka 2000). It is also possible to avoid this additional step by modifying F such that the gradient of ϕ is always fixed, see e.g. Adalsteinsson & Sethian (1999).

²Some authors express the curvature as $\kappa = -\nabla \cdot \mathbf{n}$. This is merely due to different sign conventions for the level set.

Algorithm 2: The interface-correction level set/ghost fluid method.

```

//time marching
for  $n = 1, 2, \dots, N$  do
    //level set advection
     $\phi^1 = \phi^{(n)} + \Delta t \cdot AD(\phi^{(n)})$ 
     $\phi^2 = \frac{3}{4}\phi^{(n)} + \frac{1}{4}\phi^1 + \frac{1}{4}\Delta t \cdot AD(\phi^1)$ 
     $\phi^{(n+1)} = \frac{1}{3}\phi^{(n)} + \frac{2}{3}\phi^2 + \frac{2}{3}\Delta t \cdot AD(\phi^2)$ 
    Calculate  $\rho^{(n+1)}$ ,  $\mu^{(n+1)}$ ,  $\mathbf{n}$ , and  $\kappa$  using  $\phi^{(n+1)}$ 
    //correct and reinitialize the level set every  $N_i$  steps
    if  $n \bmod N_i = 0$  then
        Mass-correction: advect with  $AD(\phi) = (\delta V / \delta t)(\kappa \delta(\phi) / A_f)$ 
        Reinitialization: advect with  $AD(\phi) = S(\phi)(|\nabla \phi| - 1)$ 
    end
    //flow solver (AB2)
     $\mathbf{RU}^{(n)} = -\mathbf{u}^{(n)} \cdot \nabla \mathbf{u}^{(n)} + \frac{1}{Re} \left( \frac{1}{\rho^{(n+1)}} \nabla \cdot [\mu^{(n+1)} (\nabla \mathbf{u}^{(n)} + (\nabla \mathbf{u}^{(n)})^T)] \right)$ 
     $\mathbf{u}^* = \mathbf{u}^{(n)} + \Delta t \left( \frac{3}{2} \mathbf{RU}^{(n)} - \frac{1}{2} \mathbf{RU}^{(n-1)} \right)$ 
     $\nabla^2 p^{(n+1)} = \nabla_g^2 [p]_\Gamma + \nabla \cdot \left[ \left( 1 - \frac{\rho_0}{\rho^{(n+1)}} \right) \nabla_g \hat{p} \right] + \frac{\rho_0}{\Delta t} \nabla \cdot \mathbf{u}^*$  //FastP*-GFM
     $\mathbf{u}^{(n+1)} = \mathbf{u}^* - \Delta t \left[ \frac{1}{\rho_0} \nabla_g p^{(n+1)} + \left( \frac{1}{\rho^{(n+1)}} - \frac{1}{\rho_0} \right) \nabla_g \hat{p} \right]$  //correction
     $\mathbf{RU}^{(n-1)} = \mathbf{RU}^{(n)}$ 
end

```

At this point, it is important to mention a critical numerical issue of classical level set methods. The problem is, even if Eq. (3.12) is solved on an incompressible velocity field, there is no guarantee that the numerical solution will conserve the total enclosed volume; in practice, there is often significant “mass loss”. The reason for this problem is a combination of numerical dissipation, under-resolution, and lack of built-in mass conservation properties in the numerical scheme. Plenty of methods have thus been proposed to overcome such issues by using higher-order numerical schemes, coupling the LS with VOF or lagrangian particles, imposing additional mass constraints, etc. See Ge *et al.* (2018) (*Paper 1*) for a brief overview.

Finally, we summarize the numerical algorithm of the interface-correction level set/ghost fluid method of *Paper 1* in Algorithm 2. Here, the level set field is integrated in time with a total-variation-diminishing third-order Runge-Kutta scheme; the spatial derivatives, AD , are computed with either fifth-order upstream-central (HOU) or weighted essentially non-oscillatory (WENO) schemes. The mass correction and reinitialization are treated similarly. In the flow solver, the pressure Poisson equation is split into a constant-coefficient part and a variable-coefficient part to utilise the fast Fourier transform. The surface tension is incorporated in the same equation using the ghost fluid method (denoted by $[\cdot]_\Gamma$). The full algorithm is presented in details in *Paper 1*. Note the high-level simplicity of the level set method comparing to the immersed boundary method (Algorithm 1).

Phase field methods. The last class of methods that we will describe is derived from theories of capillarity and critical phenomena. Here, by introducing a so-called order parameter, a single-component fluid with varying density or a binary fluid with two immiscible components are modelled by a phase field separated by an interface of finite thickness (Anderson *et al.* 1998). Historically, the proposition of a finite-width interface dates back to as early as Poisson (1831). Since then, various theories have been developed by Maxwell, Gibbs, Lord Rayleigh, van der Waals, etc.; see e.g. van der Waals (1893); Cahn (1961). We note that the physical notion of finite width is particularly relevant for fluids that are near their critical point, or for processes involving moving contact lines, breakup or coalescence, where a sharp interface description will be faced with singularities (Zhang & Mohseni 2018; Eggers 1997). For this reason, phase field methods are also regarded as *diffuse interface* methods.

Putting aside its physical origin in capillary and kinetic theories, phase field methods have recently attracted a renewed interest in the numerical modelling of twophase flow (Jacqmin 1999, 2000; Badalassi *et al.* 2003; Ding *et al.* 2007; Shen & Yang 2010; Dong & Shen 2012; Wang *et al.* 2015; Kronbichler & Kreiss 2017; Mirjalili *et al.* 2020). There are several ways to formulate the governing equation for the interface evolution. One popular version is the following Cahn-Hilliard equation,

$$\frac{\partial c}{\partial t} + \mathbf{u} \cdot \nabla c = \tilde{m} \nabla^2 \psi, \quad (3.13)$$

where c is a dimensionless order parameter smoothly varying from $+1$ in one fluid to -1 in the other within a thickness of ϵ , \tilde{m} is the mobility coefficient, and ψ is the fluid chemical potential, given as

$$\psi = \frac{3\tilde{\sigma}\epsilon}{4} \left(\frac{2}{\epsilon^2} (c^3 - c) - \nabla^2 c \right), \quad (3.14)$$

with $\tilde{\sigma}$ being the surface tension. Clearly, there is a distinction between phase field methods and VOF or LS based on the equations alone – the diffusion term appearing in the transport of phase field within the interfacial region requires proper boundary conditions on c , whereas VOF or LS are purely hyperbolic, cf. Eqs. (3.8) and (3.12). This, together with the fourth derivative of c resulting from the chemical potential expression, both leads to a number of desired properties and imposes certain numerical constraints of phase field methods. See Mirjalili *et al.* (2020) for a recent discussion.

Finally, we note that the interface thickness often loses its physical meaning in typical engineering twophase flow simulations. For example, the thickness of a water-oil interface may be estimated from the mean free path of the water/oil molecules. According to the molecular theory, it is $\sim 3 \text{ \AA}$ in room environment. Now, even if we consider a microfluidic flow where the droplet radius is $\sim 3 \mu\text{m}$, there is still a 10^4 difference in the length scale, meaning that one droplet needs $\mathcal{O}(10^4)$ grid points in each spatial direction – a clearly unrealistic resolution. Therefore, interface thickness (see Figure 3.2) in phase field simulations bears only numerical meaning.

3.2. Meshless particle-based methods

In particle-based methods, the central equation to solve is

$$\mathbf{m} \cdot \frac{d\mathbf{U}}{dt} = \mathbf{F}^H + \mathbf{F}^A, \quad (3.15)$$

where \mathbf{m} is a generalized mass/moment-of-inertia matrix, \mathbf{U} is the particle translational/rotational velocity vector, and \mathbf{F} represent the force/torque vectors owing to hydrodynamics (denoted with superscript H) or any applied interactions (denoted with superscript A) that may include contact, electrostatic, van der Waals, or magnetic forces, etc.³ Note that, for N particles in 3D, both \mathbf{U} and \mathbf{F} have dimension $6N$, while \mathbf{m} has size $6N \times 6N$.

If the particle inertia is negligible, the left-hand side of Eq. (3.15) goes to zero, resulting in $\mathbf{F}^H \approx -\mathbf{F}^A$ and quasi-static particle dynamics. If the fluid inertia is also negligible, the Navier-Stokes equations reduces to the Stokes equations and \mathbf{F}^H may be obtained without even explicitly solving the fluid motions. In the following, we examine two such simplified techniques. One is more accurate and holds for any particulate system in general, albeit computationally expansive. This is the *Stokesian dynamics* method. The other is derived from the former, but with additional features to model particle contact; hence, it is better suited for dense suspensions. We call the latter *hybrid lubrication/granular dynamics* method.

3.2.1. Stokesian dynamics

To put it in context, consider the following integral representation of the velocity field in Stokes flow (Ladyzhenskaya 1963),

$$u_i(\mathbf{x}) - u_i^\infty(\mathbf{x}) = -\frac{1}{8\pi\mu} \sum_{\alpha=1}^N \int_{S_\alpha} G_{i,j}(\mathbf{x}, \mathbf{x}_0) f_j(\mathbf{x}_0) dS_{x_0}, \quad (3.16)$$

where $\mathbf{u}^\infty(\mathbf{x})$ is the velocity at position \mathbf{x} in the absence of particles, S_α is the surface of particle α , $G(\mathbf{x}, \mathbf{x}_0)$ is the Stokeslet due to a point force at \mathbf{x}_0 (cf. Eq. 2.11), $f(\mathbf{x}_0)$ denotes its density, and the summation is over all N particles.

The local velocity \mathbf{u} can be obtained numerically by dividing the surface of each particle into M elements and resolving the linear system of equations for the force densities \mathbf{f} subject to the total force and torque conditions. This approach belongs to a broad class of *boundary integral methods* (Pozrikidis 1992), which are highly accurate and computationally efficient, since the discretisation is required only on the particle surface instead of in the flow domain, cf. Section 3.1. More specifically, the number of unknowns are $N_E = (3M + 6)N$, i.e. three components of force density for each element, and six translational and angular velocity components per particle. The computational time depends on the algorithm and boundary conditions. In the best case, it is $\mathcal{O}(N_E \log N_E)$;

³In principle, stochastic forces can also be included in the general formulation, though its numerical treatments demand special care such that the fluctuation-dissipation theorem is satisfied.

while it can be as large as $\mathcal{O}(N_E^3)$ if using direct solution methods (Graham 2018).

Now, instead of directly evaluating the integral over all particle surfaces, the method of Stokesian dynamics (Durlofsky *et al.* 1987) rewrites the right-hand side of Eq. (3.16) as a multipole expansion, *viz.*

$$-\frac{1}{8\pi\mu} \sum_{\alpha=1}^N \int_{S_\alpha} \dots \approx \mathbf{u}_F + \mathbf{u}_S + \mathbf{u}_T + \mathbf{u}_Q + \mathbf{u}_O, \quad (3.17)$$

where \mathbf{u}_F , \mathbf{u}_S , \mathbf{u}_T , \mathbf{u}_Q , and \mathbf{u}_O denote the flow due to the total forces, stresslets, total torques, quadrupoles, and octupoles, respectively. The last two moments are included to account for the finite particle size. They are important both for reducing the truncation error, which to the leading order scales as $\mathcal{O}(r^{-6})$, where r is the distance between the observation point and the center of a sphere, and in maintaining the positive definiteness of the mobility matrix (Durlofsky *et al.* 1987), shown right below. Note that, each particle is associated with 11 unknown variables (6 velocity components and 5 stresslet components since the stresslet is symmetric and traceless), which is usually much less than $3M + 6$ as required by the boundary integral method.

Under this approximation, Eq. (3.17) can then be combined with the Faxén's formulae for spheres (cf. Eq. 2.16) to obtain a relationship between particle velocities and forces, written in matrix form as

$$\begin{pmatrix} \mathbf{U} - \mathbf{U}^\infty \\ -\mathbf{E}^\infty \end{pmatrix} = \mathcal{M}^\infty \cdot \begin{pmatrix} \mathbf{F}^H \\ \mathbf{S}^H \end{pmatrix}, \quad (3.18)$$

where \mathcal{M}^∞ is the far-field approximation to the so-called “grand mobility” matrix that contains the structure of all hydrodynamic interactions per particle configuration.

Furthermore, the grand mobility matrix may be inverted to obtain the “grand resistance” matrix \mathcal{R} , transforming Eq. (3.18) to

$$\begin{pmatrix} \mathbf{F}^H \\ \mathbf{S}^H \end{pmatrix} = \mathcal{R} \cdot \begin{pmatrix} \mathbf{U} - \mathbf{U}^\infty \\ -\mathbf{E}^\infty \end{pmatrix}, \quad (3.19)$$

where \mathcal{R} may be partitioned as

$$\mathcal{R} = \begin{pmatrix} \mathbf{R}_{FU} & \mathbf{R}_{FE} \\ \mathbf{R}_{SU} & \mathbf{R}_{SE} \end{pmatrix}. \quad (3.20)$$

If \mathcal{R} is known, the particle velocity can be straightforwardly obtained from

$$\mathbf{U} = \mathbf{U}^\infty + \mathbf{R}_{FU}^{-1} \cdot (-\mathbf{F}^A + \mathbf{R}_{FE} : \mathbf{E}^\infty). \quad (3.21)$$

Note that \mathbf{F}^H has been replaced with $-\mathbf{F}^A$.

There is one more crucial step in the Stokesian dynamics method. In the above construction of \mathcal{M}^∞ , lubrication interaction is not fully accounted for due to the omission of higher-order multipole moments (hence the superscript

∞). To correctly include all contributions from the lubrication, \mathcal{R} is calculated as

$$\mathcal{R} = (\mathcal{M}^\infty)^{-1} + \mathcal{R}_{2B} - \mathcal{R}_{2B}^\infty, \quad (3.22)$$

where \mathcal{R}_{2B}^∞ denotes the far-field parts of lubrication already embedded in \mathcal{M}^∞ , and the subscript $2B$ indicates that lubrication is mainly a two-body interaction. See Durlofsky *et al.* (1987); Brady & Bossis (1988) for more detailed discussions.

Finally, the solution procedure of the Stokesian dynamics method can be summarized as follows.

1. Construct the grand mobility matrix \mathcal{M}^∞ from the suspension microstructure.
2. Obtain the grand resistance matrix \mathcal{R} by inverting \mathcal{M}^∞ and adding the lubrication correction as in Eq. (3.22).
3. Compute the particle velocity according to Eq. (3.21).
4. Update the particle position using a temporal integration scheme, such as the Euler's method $\mathbf{x}(t + \Delta t) = \mathbf{x}(t) + \mathbf{U}\Delta t$.
5. Stop if the desired time step has been reached; otherwise, go back to Step 1.

3.2.2. Hybrid lubrication/granular dynamics

If the particle volume fraction is very high, say $\phi > 40\%$, the far-field, many-body hydrodynamic interaction will be less important than the pairwise, short-range lubrication force due to particle crowding. This suggests a further simplification of the mobility matrix by dropping all additional low-moment terms apart from the lubrication (Ball & Melrose 1997), *i.e.*

$$\mathcal{R} = \mathcal{R}_{2B}. \quad (3.23)$$

Specifically, \mathcal{R}_{2B} is composed of blocks of second-, third-, and fourth-rank tensors that relate the force, torque, and stresslet acting on one particle to its neighbour (Kim & Karrila 2013). In the case of unequal spheres, the results have been extensively calculated by Jeffrey & Onishi (1984); Jeffrey (1992) using the twin multipole expansion method. The detailed algebra is rather cumbersome. Here, we simply note that the components are essentially functions of the particles' velocities, size ratio, and surface gap (h). The diverging behaviour is strongest for the lubrication force in the normal direction ($F^{L,n} \sim 1/h$), whereas in the tangential directions, as well as for the lubrication torque, the divergence is in weaker form $\sim \log(1/h)$. See *Paper 7* for the detailed expressions.

While the hydrodynamic interactions have been further truncated, another mode of interaction needs to be supplemented. For concentrated suspensions, it is known that lubrication interaction alone is not sufficient to reproduce certain experimental observations, such as the discontinuous shear thickening of cornstarch solutions (Morris 2020). On the other hand, granular dynamics, in which particles interact with each other via collision and friction forces, is often adopted for modelling of suspensions in dry media (Campbell & Brennen 1985). The hybrid of the two approaches, where both lubrication and granular

dynamics are considered, has recently emerged as a new simulation method and led to improved physical understandings of many phenomena in dense particle suspensions (Trulsson *et al.* 2012; Seto *et al.* 2013; Mari *et al.* 2014; Cheal & Ness 2018; Ness *et al.* 2018).

In a hybrid lubrication/granular dynamics method, particles interact via short-range lubrication and contact forces in a pairwise additive fashion.⁴ There are several models to describe frictional contacts between particles. The currently popular one is the stick/slide friction model employing springs and dashpots (Luding 2008), which for the case of spheres reads

$$\mathbf{F}_{i,j}^C = -k_n \mathbf{h}_{ij} - \gamma_n \mathbf{U}_{ij,n} - k_t \boldsymbol{\xi}_{ij}, \quad (3.24a)$$

$$\mathbf{T}_{i,j}^C = a_i k_t (\mathbf{n}_{ij} \times \boldsymbol{\xi}_{ij}), \quad (3.24b)$$

where $\mathbf{h}_{ij} = h_{ij} \mathbf{n}_{ij}$ denotes the signed normal surface gap between particles i and j (positive when overlapping), $\boldsymbol{\xi}_{ij}$ the signed tangential stretch (see *Paper 7* for details), k_n the normal spring constant, γ_n the damping constant, and k_t the tangential spring constant. Furthermore, the contact forces need to satisfy the Coulomb's law of friction, *i.e.* $|\mathbf{F}_{tan}^C| \leq \mu_c |\mathbf{F}_{nor}^C|$, with μ_c being the friction coefficient.

The above expressions for the contact forces and torques can be directly added in the \mathbf{F}^A tuple in Eq. (3.15) and be solved similarly to the Stokesian dynamics method. From a computational standpoint, the present method does not require invert of the mobility matrix and the grand resistance matrix composed of pair interactions is sparse; hence a much less numerical cost. Alternatively, the particle dynamics can also be obtained by explicitly solving Eq. (3.15). That is, computing all forces and torques to get the acceleration, then integrating in time once to get the velocity and twice to get the position. Similar techniques have been called dissipative particle dynamics or Brownian dynamics, depending on the context (Hoogerbrugge & Koelman 1992; Groot & Warren 1997). Provided that the particle Reynolds number remains much less than unity, this is equivalent to the quasi-static matrix inversion approach.

⁴Analytically, a diverging lubrication would prevent particle contact, though that neither agrees with everyday experience nor holds beyond the continuum limit in the presence of surface roughness.

CHAPTER 4

Miscellaneous

In this chapter, we describe two additional topics that were investigated during the course of the present work. As side projects, they share with the rest of the thesis in that the flow involved are multiphase and characterised by relatively small length scales; hence, they can be, and have been, studied using similar numerical methods. Beyond that, there is no further connection. In the following, we first give the physical motivation of each problem, then summarise the overall understanding and research methodology prior to our study. The new results are reported in *Paper 5 & 6*.

4.1. Surface effective slippage

The last several decades of development in material science and nanotechnology has greatly enhanced our ability to probe, pattern, fabricate and modify mechanical structures with chemical treatments in nano-to-micron length scales. Superhydrophobic surfaces (SHS) (Figure 4.1), which perfectly repel water due to gas entrapment within the roughness, have been studied extensively in both natural and laboratory settings (Neinhuis & Barthlott 1997; Onda *et al.* 1996), and has led to what was called a “gold rush” to search and design surfaces with extreme non-wettability (Bocquet & Lauga 2011). Such properties may not only be utilised to reduce drag in liquid or marine vessel transport (Watanabe *et al.* 1999; Ou *et al.* 2004; Choi & Kim 2006; Lee *et al.* 2016), but also has the potential to achieve self-cleaning, anti-fouling, or enhanced heat transfer functionalities (Blossey 2003; Roach *et al.* 2008). In addition, its alternative – liquid-infused surfaces (LIS) – which replace the air cushion with a liquid layer, has also attracted increasing attention due to its robustness (Solomon *et al.* 2014; Rosenberg *et al.* 2016; Wexler *et al.* 2015). Plenty of applications have been conceived with great excitement.

From the fluid mechanics perspective, questions invoked by these findings naturally include, (i) what mechanism leads to the drag reduction, and (ii) how it can be optimised given other constraints. The constraints involve surface chemistry, contamination, degradation, fabrication defects, wetting, etc., making the problem truly multiscale and multiphysics. Basic research along this line, including *Paper 6*, is motivated to resolve at least part of these issues.

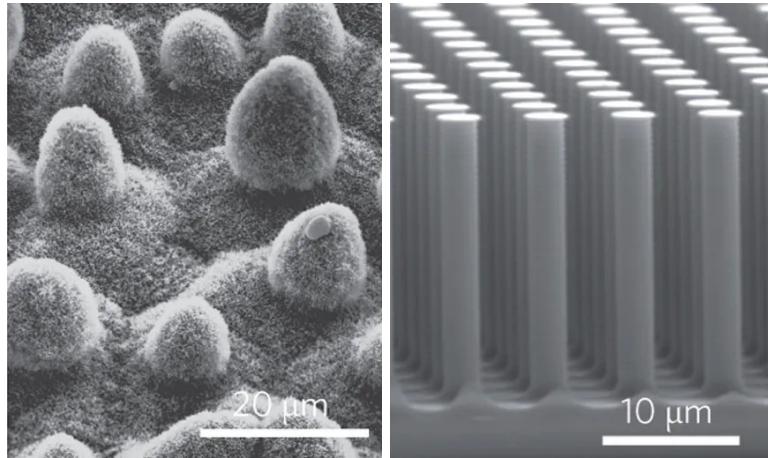


Figure 4.1: (left) Hierarchical structure of a lotus leaf (*Nelumbo nucifera*). (right) Patterned micro-pillars. Image adapted with permission from Bocquet & Lauga (2011), © Springer Nature.

Theoretically, the resistance of a solid surface to fluid flow can be characterised by the *slip length*, λ_s , defined as

$$\mathbf{u}_s = \lambda_s \left(\nabla \mathbf{u} + (\nabla \mathbf{u})^T \right) \cdot \mathbf{n}, \quad (4.1)$$

where \mathbf{u} is the flow velocity, \mathbf{u}_s the slip velocity on the solid surface, and \mathbf{n} the normal vector pointing from the solid to the fluid. In the case of a planar simple shear flow, it can be reduced to

$$u_s = \lambda_s \dot{\gamma}, \quad (4.2)$$

where $\dot{\gamma} = \partial u / \partial y$ is the shear rate. In most situations, λ_s is a constant material property with a value on the order-of-magnitude of the molecular interaction range (Thompson & Troian 1997). However, even with a no-slip boundary condition, many of the aforementioned studies have demonstrated a large measurable slippage over SHS. Close inspection of the surface structure suggests that it is the hairy pattern combined with its hydrophobic property that yields the apparent slip. This is beautifully depicted in Figure 4.1, which shows the natural meadow-looking roughness on a lotus leaf and an artificial array of micron-scale pillars.

To quantify the slippage analytically, one can define an *effective slip length*, λ_e , as

$$\lambda_e = \frac{\bar{u}(H)}{\dot{\gamma}} - H, \quad (4.3)$$

where $\bar{u}(H)$ is the average streamwise velocity at a small distance H above the substrate. Physically, λ_e is equal to the distance below the surface at which

the velocity would extrapolate to zero. Under this simple definition, many studies have been conducted to obtaining theoretical expressions of the effective slip for two-dimensional longitudinal or transverse grooves (Lauga & Stone 2003; Sbragaglia & Prosperetti 2007; Davis & Lauga 2009; Ng & Wang 2009; Schönecker *et al.* 2014; Nizkaya *et al.* 2014; Crowdy 2017*a,b*). The general procedure is to solve the Stokes equations in a periodic domain subject to a mixture of no-slip and finite-slip boundary conditions. To facilitate analytical calculations, some simplifications may have to be made, such as a static interface shape and the pinning of the contact line. In general, different geometry- and viscosity-ratio-dependent expressions for λ_e have been found, depending on the configuration.

While these solutions are helpful in guiding the optimal surface design, they neglect an important ingredient – wetting of the fluid. One reason that superhydrophobic or liquid-infused surfaces are still mostly limited to laboratory testings at present is the various failure modes, often associated with wetting in some form. Specifically, SHS is known to be sensitive to external pressure under which the entrapped bubble can collapse (Bocquet & Lauga 2011); whereas LIS is prone to shear-driven failures, *i.e.* drainage of the lubricant from the surface grooves (Wexler *et al.* 2015). Therefore, the *dynamics* is equally, if not more, important comparing to the static slippage property. Unfortunately, precise control of wetting in experiments is still very challenging due to sensitivity to surface conditions and instrument inaccuracies (Liu *et al.* 2019), let alone the longstanding theoretical disputes (de Gennes 1985). In light of these difficulties, numerical studies with controlled parameters in non-ideal situations are expected to bridge the gap between analytical calculations and experimental findings. *Paper 6* is one such effort.

4.2. Droplets in turbulence

Turbulence is the unsteady flow generated from the Navier-Stokes equations at large Reynolds numbers. As it prevails in a broad range of length and time scales, predicting the statistics of droplets or bubbles in turbulence is of great practical importance. Take the size distribution for example. In the ocean, air bubbles entrained in breaking waves enhance the air-sea gas flux, affect aerosol production, scavenge biological surfactants, and create ambient noise; knowing their creation mechanism and distribution in size can thus greatly improve our understanding of the nature and climate (Deane & Stokes 2002). In the atmosphere, vapor condenses on particles and forms droplets; the droplet size distribution is the defining characteristics of a cloud, controlling how it interacts with electromagnetic radiation and how fast precipitation will form (Shaw 2003). In emulsions or colloidal suspensions, the distribution of the dispersed phase determines the stability, rheology, and area available for physical or chemical processes that occur at the interface (Boxall *et al.* 2012). This is directly related to our daily life, as many processed food and cosmetic products are complex fluids in one way or another.

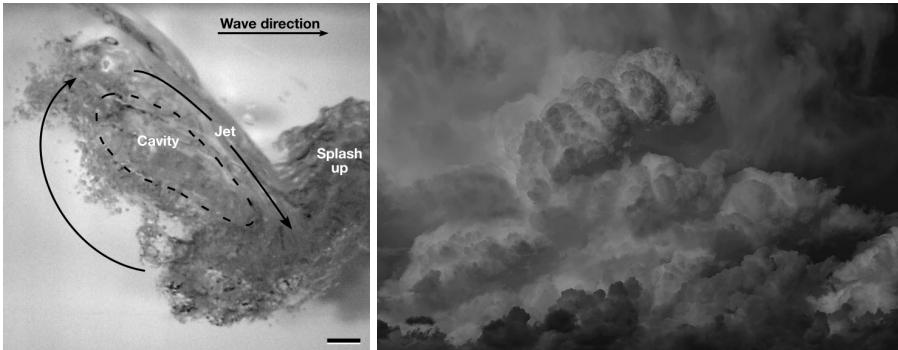


Figure 4.2: (left) A lab-generated breaking wave crest, showing bubble entrainment. Scale bar, 1 cm. Image adapted with permission from Deane & Stokes (2002), © Springer Nature. (right) Nimbostratus clouds taken at Karlskrona, Sweden. © Jari Hytönen.

Despite the ubiquity and importance, detailed numerical simulations on droplet-turbulence interactions are surprisingly scarce. It is not until the last few years that computational studies start to appear in the literature, e.g. Perlekar *et al.* (2012); Skartlien *et al.* (2013); Komrakova *et al.* (2015); Scarbolo *et al.* (2015); Dodd & Ferrante (2016). Here, the advantages of performing these large-scale simulations are dual. On the one hand, interface-resolved, high-resolution simulations reveal a unprecedented level of details, offering arguably the full droplet and flow statistics within the afforded system. These results can then be used as benchmarks to test theoretical scalings and empirical models of the droplet distribution (Hinze 1955; Deane & Stokes 2002). On the other hand, as simulation methods for multiphase flow are continuously being developed, the current results will serve as verification for later simulations. Based on these reasons, we performed direct numerical simulations of two immiscible and incompressible fluids in a homogeneous turbulent shear flow under various Weber numbers and initial conditions (*Paper 5*). The choice of this base flow is rationalized by its natural turbulence sustaining mechanism and the relevance to the logarithmic layer of wall-bounded turbulent flow, see e.g. Pumir (1996); Sekimoto *et al.* (2016). We admit that modelling of turbulence is difficult even by itself, let alone in the presence of droplets. Nevertheless, numerical studies like the one we conducted shall provide additional information to future modellers and shed new lights on the droplet-turbulence interactions.

CHAPTER 5

Summary

The investigations presented in this thesis are motivated by the interest in understanding droplet interactions and suspension flow that originates from a number of applications, including material science, microfluidic technology and transport of complex fluids. The general physical picture and underlying mathematical formulation are described in Chapter 2, where we introduce elementary properties of the Stokes flow and its approximate solutions in the presence of liquid droplets or solid particles in a few simple cases. These are the fundamentals of microhydrodynamics, essential for developing a physical intuition and making back-of-the-envelope estimates when faced with more complicated engineering problems. Unfortunately, sometimes a problem may be too complicated to be solved analytically, or we may simply be misguided by our intuitions. In those cases, numerical simulations offer a relatively inexpensive means to visualise fluid motions and disentangle the various effects. Chapter 3 thus provides a mini-review of some currently popular methods that can be applied to simulate coupled multiphase systems. This is the main subject of the present work – we develop methods, implement algorithms, and simulate droplets or particles in fluids to reproduce experiments or verify theories. The specific aim may differ by applications; what remain the same are the numerical approach to problem solving as well as a physics-oriented search for new principles driven by curiosity.

The detailed results of the present work are given in the next part of the thesis in the format of journal articles or technical reports. Among them, papers 1–4 entail droplet interactions, papers 5 & 6 are miscellaneous, and paper 7 concerns suspension flow. Separate retrospective summaries of each paper are given in the beginning of *Part II*. A diagrammatic categorization is also illustrated in Figure 5.1. Overall, the common limitations of the conducted studies are that simulations still fall much behind experiments in terms of physical complexity – which could sometimes be justified, but it is still not the same – and the lack of simple, quantitative models that can be extracted from the results. In general, our numerical work provides additional evidences to conclude qualitatively, but a new set of simulations or experiments may have to be performed if precise predictions need to be made in different situations.

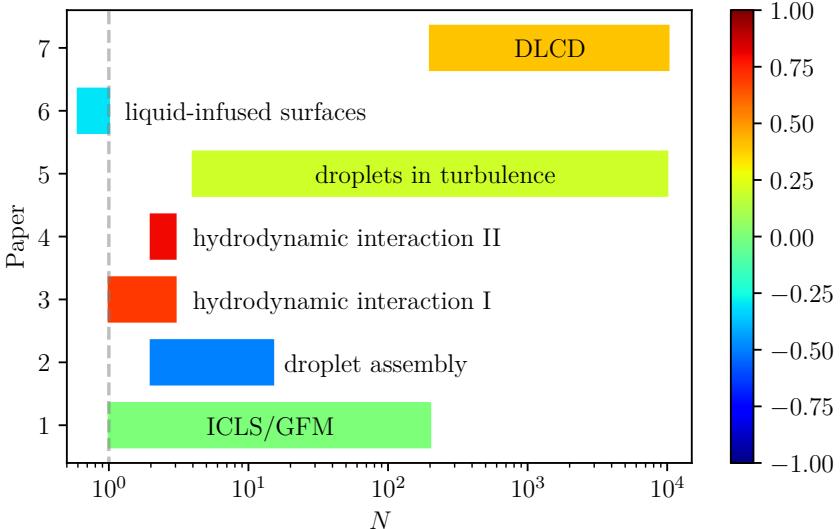


Figure 5.1: Diagrammatic representation of the papers in *Part II* parameterized by the number of droplets (N) that are or could potentially be studied, and by the distance (indicated by color) to being purely experimental (−1) or purely theoretically (1); 0 corresponds to purely numerical. The papers are labelled with short titles. Note that *Paper 6* does not have a meaningful definition of N .

Finally, as an outlook, we summarise the lessons learned during all the projects and name a few open questions worth further investigation:

- Fabricating photonic crystals using flow-assisted droplet assembly could potentially achieve a moderate bang gap and be scaled up to meet industrial needs. The rest of the task is in engineering. Current focus of the photonic community seems to be shifted to searching for disordered hyperuniform materials that are easier to make and still possess bandgaps (Torquato 2018; Ricouvier *et al.* 2017).
- For multiphase flow simulations, classical level set methods are not mass conserving, thus many efforts (including the present work) have been spent to improve the accuracy. Despite all the progress, it remains a challenge to devise a general-purpose solver that can be applied to any multiphase flow problems without tweaking, given the large separation of length scales and inherent singularities. In retrospect, the simulation of droplet assembly in microfluidic environments could be done with available open-source software instead of inventing a new method. However, the latter process is rewarding for its own sake, and the resulting

method may be adapted to study plenty of other unforeseen applications, such as tissue/tumor growth.

- Wetting is important in many practical applications; but assumptions made in many numerical work are often too crude to be trusted fully. Careful and rigorous simulations, along with accurate experiments, are needed to develop better models for processes involving interfaces moving on solid surfaces.
- Dense particle suspension is a very exciting research front, offers an immense parameter space to explore, and is related to both applied and fundamental questions. For example, questions like how particle geometry affects jamming, do active particles jam in the same way as passive particles, how do particle interactions and the medium topology come into play in long time limits (relevant to certain disease progression, e.g. Alzheimer's disease), etc. are all unknown today. Investigating these problems requires development of new computational models and collaboration with specialists in different research areas, and may inspire new pathways to advance life science and biology in the future.

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Part II

Papers

Summary of the papers

Paper 1

An efficient mass-preserving interface-correction level set/ghost fluid method for droplet suspensions under depletion forces

In this first work, we develop and implement an interface-correction level set/ghost fluid method (ICLS/GFM) for high-fidelity simulations of liquid droplets in an immiscible carrier fluid. The main novelty is a global mass correction scheme that is conceptually simple, relatively straightforward to program, and efficient since the scheme does not need to be executed at every time step. The method works best for well-resolved droplets as validated in the paper; however, extra care must be taken when applying it otherwise. In the same paper, we also formulate a hydrodynamic model for surfactant-induced depletion forces between neighbouring drops, removing the need to simulate suspending surfactants, hence further improving the computational efficiency. The overall methodology can be readily used to study droplet interactions in liquid flows, and may be extended to simulate multiphase flows in porous media in general.

Paper 2

Flow-assisted droplet assembly in a 3D microfluidic channel

In this paper, we perform a series of simulations, using methods developed in *Paper 1*, to examine the droplet interaction in increasingly realistic conditions, mimicking a microfluidic experiment. The main objective is to elucidate the physical mechanism that leads to the experimentally observed droplet clustering, which had been explained by a dipolar interaction theory. Contrary to previous belief, we find that dipolar interactions alone cannot account for the fast droplet dynamics; a number of 3D effects must be included to faithfully reproduce the observation. This conclusion is supported by theoretical derivations of particle interactions in the Stokes flow (*Papers 3 & 4*), implying that designing optimal microfluidic channels to directly produce large droplet crystals remains difficult in practice.

Paper 3

Integral representation of channel flow with interacting particles

In this part one of a two-part theoretical work on droplet interactions, we consider the far-field interaction of spherical droplets or particles traveling in the Poiseuille flow. A boundary integral representation of the disturbance flow is derived, showing that, to leading order, the flow is dipolar and proportional to a weighted integral of the stress and flow at the particle surface. We tabulate the dipole moment under various confinement and particle initial positions using direct numerical simulations of the Navier-Stokes equation, then introduce and solve the equation of motion that describes hydrodynamic interactions in the special case of two and three particles.

Paper 4

The theory of hydrodynamic interaction of two spheres in wall-bounded shear flow

In the second part of the two-part theoretical work on droplet interactions in shear flow, we revisit the classical work of Batchelor & Green (1972a), where two spherical particles interact hydrodynamically in a unbounded linear flow. Using the boundary integral formulation derived in *Paper 3*, we show that the inclusion of a distant wall results in qualitatively different particle trajectories demonstrative of the wall's singular perturbation. Specifically, the distance at which the wall significantly alters the particles interaction scales as $z_0^{3/5}$, with z_0 being the initial distance between the particle and the wall. The phase portrait of the particle relative paths admits domains of closed (dancing) and open (swapping) trajectories, apart from those that also exist in unbounded shear flows. We postulate that this is topologically stable and verified it by direct numerical simulations of the Navier-Stokes equations. Qualitatively, the distant wall is the third body that changes the global topology of the phase portrait of two-particle interaction.

Paper 5

Droplets in homogeneous shear turbulence

This paper reports a simulation study of liquid droplets immersed in another density- and viscosity-matched fluid (5% volume fraction) in homogeneous shear turbulent flow at a shear Reynolds number equal to 15 200. Detailed fluid motions are visualised, showing intense deformation, breakup, and coalescence of the droplets as the flow evolves. We examine the flow and droplet statistics, including Taylor-microscale Reynolds numbers, turbulent kinetic energy budget, droplet size distributions, etc., under various surface tensions and initial droplet diameters. The overall results suggest that the dispersed phase acts as a sink of the turbulent kinetic energy for the carrier fluid, and the maximal droplet sizes are predicted by the Hinze scaling despite of droplet coalescence and a mean shear.

Paper 6

Effective slip over partially filled microcavities and its possible failure

In this paper, we combine two separate simulation methods (phase field and level set methods) to numerically study the transverse flow over a 2D model liquid-infused surface (LIS). Computationally, we introduce and verify a multiscale numerical framework for wetting modelling by locally solving the Cahn-Hilliard equations near the moving contact line within a larger Stokes system. Physically, we conduct a parametric study of the performance of the LIS, quantified by its effective slip length and potential drainage. Among the many parameters (the capillary number, viscosity ratio, contact angle, etc.) explored, we identify that filling fraction is the most sensitive parameter for the effective slip; the effects of other parameters are generally intertwined but weakened if the surface groove is not fully filled. Furthermore, we report a shear-driven failure mode associated with lubricant drainage, that is worse at higher lubricant viscosities if the lubricant is less viscous than the outer fluid, but disappears if the lubricant is more viscous. Overall, our study may serve as a conceptual guide for LIS design complementing experimental tests.

Paper 7

Implementation note on a minimal discrete-element lubrication/contact dynamics model for dense suspensions

This report documents the implementation detail of a minimal discrete-element lubrication/contact dynamics (DLCD) model intended for simulation of dense particle suspensions in shear flows. The method is meshless, explicit, and solves directly the particle dynamics in the overdamped limit by balancing conservative and dissipative interactions at the pair level. Potential limitations include omissions of fluid/particle inertia and long-range hydrodynamic interactions. The implementation has been tested for $200 \sim 500$ particles under volume fractions from 40% to 64%, showing jamming transition, continuous and discontinuous shear thickening that are in excellent agreement with literature results.

1

Paper 1

An efficient mass-preserving interface-correction level set/ghost fluid method for droplet suspensions under depletion forces

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Aiming for the simulation of colloidal droplets in microfluidic devices, we present here a numerical method for two-fluid systems subject to surface tension and depletion forces among the suspended droplets. The algorithm is based on an efficient solver for the incompressible two-phase Navier-Stokes equations, and uses a mass-conserving level set method to capture the fluid interface. The four novel ingredients proposed here are, firstly, an interface-correction level set (ICLS) method; global mass conservation is achieved by performing an additional advection near the interface, with a correction velocity obtained by locally solving an algebraic equation, which is easy to implement in both 2D and 3D. Secondly, we report a second-order accurate geometric estimation of the curvature at the interface and, thirdly, the combination of the ghost fluid method with the fast pressure-correction approach enabling an accurate and fast computation even for large density contrasts. Finally, we derive a hydrodynamic model for the interaction forces induced by depletion of surfactant micelles and combine it with a multiple level set approach to study short-range interactions among droplets in the presence of attracting forces.

Key words: Multiphase flow, Level set method, Ghost fluid method, Droplet, Depletion force

1. Introduction

In the field of colloidal science, much progress has been made on the synthesis of elementary building blocks (Fig. 1) mimicking molecular structures to elaborate innovative materials, e.g. materials with complete three dimensional band gaps (Xia *et al.* 2000; Velev & Gupta 2009; Li *et al.* 2011; Sacanna & Pine 2011). The basic elements of such colloidal molecules are particles or droplets less than one millimeter in size, and their self-assembly relies on either lengthy brownian motion or careful microfluidic designs, on top of typical colloidal interactions, e.g. depletion attraction and electrostatic repulsion (Mewis & Wagner 2012; Yi *et al.* 2013; Shen *et al.* 2016). Regardless of the approach, however, questions remain

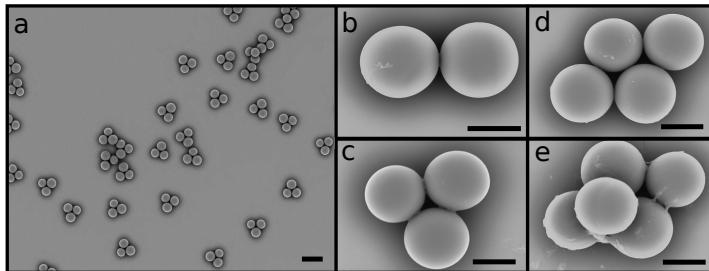


Figure 1: Self-assembled colloidal clusters. a) Electron micrograph of a suspension of triplet clusters. Scale bar, $30 \mu\text{m}$. b-e) Close up of doublet, triplet, quadruplet, and quintuplet clusters. Scale bars, $10 \mu\text{m}$. Further details are available in Shen *et al.* (2016), photograph courtesy of Dr. Joshua Ricouvier.

why the colloidal particles/droplets undergo certain path to organize themselves and how such process can be controlled and optimized. Since full data are not yet accurately accessible from experiments in such miniature systems, computer simulations will be useful to provide supplemental information.

Scaling down to microscale appears first to be a convenience for the numerical simulations of multicomponent and multiphase systems as the non-linear Navier-Stokes (NS) equations can be reduced to the linear Stokes equations. This allows the use of boundary integral methods (BIM) (Pozrikidis 1992), e.g. most recently the GGEM-based BIM (Kumar & Graham 2012; Zhu *et al.* 2014) solving the Stokes equations in general geometries. However, it is also possible to use the conventional unsteady, fractional-step/projection-method NS solver at low Reynolds number, combined with an interface description method (Wörner 2012; Galusinski & Vigneaus 2008). The latter approach is more versatile, probably less difficult to implement, and enjoys a rich literature of standard numerical techniques. Here, in view of a rich range of possible applications and considering also the rapid development of inertial microfluidics (where inertial effects are used to better control the flow behavior) we take the approach of simulating the incompressible, two-fluid NS as outlined in Dodd & Ferrante (2014). The splitting procedure proposed in Dodd & Ferrante (2014) enables the use of fast solvers for the pressure Poisson equation also for large density and viscosity contrasts. The remaining choice then is to be made among the available interface-description methods.

Generally, there are two categories of methods to resolve an interface in a NS solver, *i.e.* front-tracking methods and front-capturing methods. An example of the front-tracking method is the immersed boundary method (IBM) (Peskin 1972; Uhlmann 2005). Using Lagrangian points in a moving frame, IBM can offer a high interface resolution without the need to deform the underlying mesh in the fixed frame. However, the coupling of the two meshes relies on

a regularized delta function, which introduces certain degrees of smearing. Moreover, large interface deformation requires frequent mesh rearrangement; and topology changes may have to be handled manually. These constraints make IBM typically more expensive and less appealing for droplet simulations.

Front-capturing methods, on the other hand, are Eulerian and handle topology changes automatically; they are therefore easier to parallelize to achieve higher efficiency. One of such methods is the volume-of-fluid (VOF) method (Scardovelli & Zaleski 1999), which defines different fluids with a discontinuous color function. The main advantage of VOF is its intrinsic mass conservation. It suffers however from inaccurate computations of the interface properties, e.g. normals and curvatures. This makes it less favorable for simulations of microfluidic systems where surface tension is the dominant effect and requires accurate modelling.

Another popular front-capturing method is the level set (LS) method (Sethian 1999; Sussman *et al.* 1994). Contrary to VOF, LS prescribes the interface through a (Lipschitz-)continuous function which usually takes the form of the signed distance to the interface. Under this definition, normals and curvatures of the interface can be readily and accurately computed. However, the problem when simulating incompressible flows is that mass loss/gain may occur and accumulate because the LS function embeds no volume information. In addition, errors can also arise from solving the LS advection equation and/or the reinitialization equation, a procedure commonly required to reshape the LS into a distance function. Therefore, additional measures have to be taken to ensure mass conservation.

Many different approaches have been proposed to make LS mass-conserving, which can be classified into the following four methodologies. The first approach is to improve the LS discretization and reinitialization so that numerical errors are reduced. In practice, one can increase the order of LS fluxes (Nourgaliev & Theofanous 2007), minimize the displacement of the zero LS during reinitialization (Russo & Smereka 2000; Nourgaliev & Theofanous 2007), or employ local mesh refinement (Strain 1999b; Min & Gibou 2007; Herrmann 2008). By doing so, mass loss can be greatly reduced, although the LS function is still inherently non-conservative. The second remedy couples the LS with a conservative description (e.g. VOF) or Lagrangian particles. For example, the hybrid particle level set method (Enright *et al.* 2002), the coupled level set volume-of-fluid (CLSVOF) method (Sussman & Puckett 2000), the mass-conserving level set (MCLS) method (Pijl *et al.* 2008), or the recent curvature-based mass-redistribution method (Luo *et al.* 2015b). With varying level of coupling, these methods can usually preserve mass really well; the drawback is that the complexity and some inaccuracy (due to interpolation, reconstruction, etc.) of the other method will be imported. The third approach improves mass conservation by adding a volume-constraint in the LS or NS formulation. Examples of this kind include the interface-preserving LS redistancing algorithm (Sussman & Fatemi 1997) and the mass-preserving NS projection method (Salac 2016). Finally, one

can also smartly modify the definition of the LS, such as the hyperbolic-tangent level set (Olsson & Kreiss 2005), to reduce the overall mass loss.

With the physical application of colloidal droplets in mind, and using ideas from some of the above-mentioned methods, we heuristically propose an interface-correction level set (ICLS) method. The essential idea of ICLS is to construct a normal velocity supported on the droplet interface and use it in an additional LS advection to compensate for mass loss, in a way similar to inflating a balloon. Because no coupling with VOF or Lagrangian particles is required, the simplicity and high accuracy of the original LS method is preserved, yet the extra computational cost of this procedure is negligible.

Provided a mass-preserving level set method, the coupled flow solver must also accurately compute the surface tension, a singular effect of the normal stress on the interface. This is particularly important for microfluidic systems; as surface tension scales linearly with the dimension, it decays slower than volumetric forces (e.g. gravity) when the size of the system reduces. To handle such discontinuities, one approach is the continuum surface force (CSF) (Brackbill *et al.* 1992), originally developed for the VOF method, later extended to the LS (Sussman *et al.* 1994). Although easy to implement, CSF effectively introduces an artificial spreading of the interface by regularizing the pressure difference, and it can become erroneous when two interfaces are within its smoothing width. A second, non-smearing approach is the ghost fluid method (GFM). Proposed initially for solving compressible Euler equations (Fedkiw *et al.* 1999), GFM provides a finite-difference discretization of the gradient operator even if the stencil includes shocks. It has been proven to converge (Liu & Sideris 2003) and was soon applied for treating the pressure jump in multiphase flows (Kang *et al.* 2000). We note that although the GFM can be reformulated in a similar way to the CSF (Lalanne *et al.* 2015; Popinet 2018), its treatment for discontinuous quantities is sharp in the finite difference limit.

Several implementation options of the GFM were suggested in Kang *et al.* (2000); Lalanne *et al.* (2015); Desjardins *et al.* (2008). Here, we follow the methodology of Desjardins *et al.* (2008), *i.e.* using the GFM for the pressure jump due to surface tension while neglecting the viscous contribution. As will be discussed later, this choice is especially suitable for microfluidic applications where the capillary effect is strong. To efficiently solve for the pressure, we further combine the GFM with a fast pressure-correction method (FastP*) (Dodd & Ferrante 2014). Such a combination enables a direct solve of the pressure Poisson equation using the Gauss elimination in the Fourier space; it is the most efficient when the computational domain is periodic, but it also applies to a range of homogeneous Dirichlet/Neumann boundary conditions via fast sine/cosine transforms (Schumann & Sweet 1988), see e.g. a recent open-source distribution (Costa 2017). Using a second-order accurate, grid-converging interface curvature estimation, we will show that the coupled ICLS/NS solver can handle large density/viscosity contrasts and converges between first and second order in both space and time.

Finally, a unique challenge to the simulation of colloidal droplets is the modeling of near-field interactions. It is known that two or more colloids can interact via dispersion, surface, depletion, and hydrodynamic forces (Mewis & Wagner 2012). Apart from the hydrodynamic forces which is determined directly from the NS, and the dispersion forces which arise from quantum mechanical effects, the depletion and surface forces must be modelled. These forces can be either attraction or repulsion and are typically calculated from the gradient of a potential. Based on colloidal theory, we propose a novel hydrodynamic model for the depletion force in the framework of the ICLS/NS solver. Our method relies on two extensions: *i*) extending the single level set (SLS) function to multiple level set (MLS) functions; and *ii*) extending the GFM for computation of the gradient of depletion potential. MLS has the benefits that each droplet within a colloidal cluster can be treated individually, is allowed to interact with the other droplets, and is guarded from its own mass loss. MLS also prevents numerical coalescence of droplets when they get too close. The computational complexity, proportional to the number of MLS functions (l) and the number of cells in each dimension (N), is higher than SLS. However, we note that many techniques exist to reduce the CPU cost and/or memory consumption if lN^d ($d = 2$ or 3) is large. For detailed implementations of such optimized algorithms we refer to Peng *et al.* (1999); Nielsen & Museth (2006); Brun *et al.* (2012). In the present paper, we will demonstrate the self-assembly of colloidal droplets using one droplet per MLS function.

The paper is organized as follows. In Sec. 2, the governing equations for the incompressible, two-phase flow are briefly presented. In Sec. 3, the classical signed-distance LS methodology together with some commonly used numerical schemes is discussed. We then introduce the ICLS method in Sec. 4, starting from the derivation ending with a demonstration. We further provide a geometric estimation of the interface curvature tailored to the GFM in Sec. 5. The complete ICLS/NS solver is outlined in Sec. 6, including a detailed description of the implementation and three examples of validation. In Sec. 7, we propose a MLS/GFM-based method for the modeling of near-field depletion potential. Finally, we summarize the overall methodology in Sec. 8.

2. Governing equations for interfacial two-phase flow

The dynamics of the incompressible flow of two immiscible fluids is governed by the Navier-Stokes equations, written in the non-dimensional form

$$\nabla \cdot \mathbf{u} = 0, \quad (1a)$$

$$\frac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{u} = \frac{1}{\rho_i} \left(-\nabla p + \frac{1}{Re} \nabla \cdot [\mu_i (\nabla \mathbf{u} + \nabla \mathbf{u}^T)] \right) + \frac{1}{Fr} \mathbf{g}, \quad (1b)$$

where $\mathbf{u} = \mathbf{u}(\mathbf{x}, t)$ is the velocity field, $p = p(\mathbf{x}, t)$ is the pressure field, and \mathbf{g} is a unit vector aligned with gravity or buoyancy. ρ_i and μ_i are the density and

dynamic viscosity ratios of fluid i ($i = 1$ or 2) and the reference fluid. These properties are constant in each phase and subject to a jump across the interface, which we denote as $[\rho]_\Gamma = \rho_2 - \rho_1$ for density and $[\mu]_\Gamma = \mu_2 - \mu_1$ for viscosity. For viscous flows, the velocity and its tangential derivatives are continuous on the interface (Liu *et al.* 1994). However, the pressure is discontinuous due to the surface tension and the viscosity jump, *i.e.*

$$[p]_\Gamma = \frac{1}{We} \kappa + \frac{2}{Re} [\mu]_\Gamma \mathbf{n}^T \cdot \nabla \mathbf{u} \cdot \mathbf{n}, \quad (2)$$

where κ is the interface curvature, and \mathbf{n} is the normal to the interface. If the surface tension coefficient, $\tilde{\sigma}$, varies on the interface the tangential stress is also discontinuous. In this paper, we assume constant and uniform $\tilde{\sigma}$. In Eqs. (1b) and (2), Re , We , and Fr are, respectively, the Reynolds, Weber, and Froude numbers, defined as

$$Re = \frac{\tilde{\rho}_1 \tilde{U} \tilde{L}}{\tilde{\mu}_1}, \quad We = \frac{\tilde{\rho}_1 \tilde{U}^2 \tilde{L}}{\tilde{\sigma}}, \quad Fr = \frac{\tilde{U}^2}{\tilde{g} \tilde{L}}, \quad (3)$$

where \tilde{U} , \tilde{L} , $\tilde{\rho}_1$, $\tilde{\mu}_1$, and \tilde{g} denote the reference dimensional velocity, length, density, dynamic viscosity, and gravitational acceleration. Note that $\rho_1 = 1$ and $\mu_1 = 1$ (*i.e.* we define fluid 1 as the reference fluid).

3. Classical level set methodology

In the level set framework, the interface Γ is defined implicitly as the zero value of a scalar function $\phi(\mathbf{x}, t)$, *i.e.* $\Gamma = \{\mathbf{x} \mid \phi(\mathbf{x}, t) = 0\}$. Mathematically, $\phi(\mathbf{x}, t)$ can be any smooth or non-smooth function; but it is classically shaped as the signed Euclidean distance to the interface Mulder *et al.* (1992); Sussman *et al.* (1994), *viz.*

$$\phi(\mathbf{x}, t) = sgn(\mathbf{x}) |\mathbf{x} - \mathbf{x}_\Gamma|, \quad (4)$$

where \mathbf{x}_Γ denotes the closest point on the interface from nodal point \mathbf{x} , and $sgn(\mathbf{x})$ is a sign function equal to 1 or -1 depending on which side of the interface it lies. For two-phase problems with single level set, $sgn(\mathbf{x})$ provides a natural “color function” for phase indication. Furthermore, with this definition, geometric properties such as the unit normal vector, \mathbf{n} , and the local mean curvature, κ , can be conveniently computed as

$$\mathbf{n} = \frac{\nabla \phi}{|\nabla \phi|}, \quad (5)$$

$$\kappa = -\nabla \cdot \mathbf{n}. \quad (6)$$

3.1. Advection

The motion of a fluid interface is governed by the following PDE

$$\frac{\partial \phi}{\partial t} + \mathbf{u} \cdot \nabla \phi = 0, \quad (7)$$

where \mathbf{u} is the flow velocity field. Despite of its simple form, obtaining an accurate and robust solution to Eq. (7) is challenging. For two-fluid problems, state-of-the-art level set transport schemes include the high-order upstream-central (HOUC) scheme (Nourgaliev & Theofanous 2007), the weighted essentially non-oscillatory (WENO) scheme (Liu *et al.* 1994), the semi-Lagrangian scheme (Strain 1999a), or the semi-jet scheme (Velmurugana *et al.* 2016). Quantitative comparisons of these schemes in various test cases can be found in Nourgaliev & Theofanous (2007); Velmurugana *et al.* (2016). We note that the choice of the scheme is case-dependent, *i.e.* depending on the smoothness of the overall level set field or the stiffness of Eq. (7). For flows involving moderate deformations, HOUC is usually sufficient and most efficient. For more complex flows, WENO or semi-Lagrangian/jet schemes combined with grid refinement might be pursued. In the present study, we use either HOUC5 or WENO5 (5 denotes fifth-order accuracy) to evaluate $\nabla\phi$.

For the temporal discretization of Eq. (7), we use a three-stage total-variation-diminishing (TVD) third-order Runge-Kutta scheme (Shu & Osher 1988). Denoting $f(\phi) = -\mathbf{u} \cdot \nabla\phi$, it updates ϕ from time level n to $n+1$ in three sub-steps

$$\begin{cases} \phi^1 = \phi^n + \Delta t \cdot f(\phi^n) \\ \phi^2 = \frac{3}{4}\phi^n + \frac{1}{4}\phi^1 + \frac{1}{4}\Delta t \cdot f(\phi^1) \\ \phi^{n+1} = \frac{1}{3}\phi^n + \frac{2}{3}\phi^2 + \frac{2}{3}\Delta t \cdot f(\phi^2). \end{cases} \quad (8)$$

Finally, we note that Eq. (7) does not need to be solved in the entire computational domain, as only the near-zero values are used to identify the interface and compute its curvature. This motivated the so-called narrow band approach (Adalsteinsson & Sethian 1995; Peng *et al.* 1999), which localizes the level set to the interface using index arrays. Combined with optimal data structures (Nielsen & Museth 2006; Brun *et al.* 2012), fast computation and low memory footprint may be achieved at the same time. In our implementation, we store all the level set values while only update those in a narrow band, *i.e.* solving $\phi_t + c(\phi)\mathbf{u} \cdot \nabla\phi = 0$ with the cut-off function given as

$$c(\phi) = \begin{cases} 1 & \text{if } |\phi| < \gamma \\ 0 & \text{otherwise,} \end{cases} \quad (9)$$

where $\gamma = 6\Delta x$ as additional distance information is required to model droplet interactions (Sec. 7). This is equivalent to Peng *et al.* (1999) with a simplified $c(\phi)$.

Zalesak's disk. The Zalesak's disk (Zalesak 1979), *i.e.* a slotted disc undergoing solid body rotation, is a standard benchmark to validate level set solvers. The difficulty of this test lies in the transport of the sharp corners and the thin slot, especially in under-resolved cases. The initial shape should not deform under solid body rotation. Hence, by comparing the initial level set field and that after one full rotation one can characterise the degree of accuracy of a numerical solver. Here, the parameters are chosen so that a disk of radius 0.15,

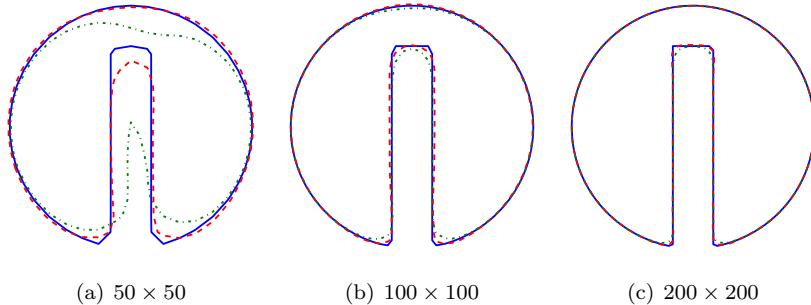


Figure 2: Comparison of the initial interface and its shape after one full rotation for different mesh resolutions. Solid lines depict the initial interface. Two different schemes have been used to evaluate the gradients, namely HOUC5 (dashed lines) and WENO5 (dash-dotted line).

slot width of 0.05 is centered at $(x, y) = (0, 0.25)$ of a $[-0.5, 0.5] \times [-0.5, 0.5]$ box. The constant velocity field is given as

$$u = -2\pi y, \quad v = 2\pi x. \quad (10)$$

Three different mesh resolutions have been considered, namely 50×50 , 100×100 and 200×200 . Fig. 2 depicts the shape of the interface after one full rotation of the disk, solving Eq. (7) only. Along with the results of the HOUC5 scheme (red dashed line), the shape of the interface obtained using the WENO5 scheme (green dash-dotted line) is also reported in this figure. Both schemes yield good results on fine grids, but HOUC5 clearly outperforms WENO5 on the coarsest mesh considered here.

3.2. Reinitialization

Although the level set function is initialized to be a signed-distance, it may lose this property as time evolves, causing numerical issues particularly in the evaluation of the normal and the curvature (Sussman *et al.* 1994). In order to circumvent these problems, an additional treatment is required to constantly reshape ϕ into a distance function, *i.e.* $|\nabla\phi| = 1$. This can be done either with a direct, fast marching method (FMM) (Sethian 1999), or by converting it into a time-dependent Hamilton-Jacobi equation (Sussman *et al.* 1994)

$$\frac{\partial\phi}{\partial\tau} + S(\phi_0)(|\nabla\phi| - 1) = 0, \quad (11)$$

where τ is a pseudo-time, and $S(\phi_0)$ is a mollified sign function of the original level set, usually defined as

$$S(\phi_0) = \begin{cases} -1 & \text{if } \phi_0 < -\Delta x \\ 1 & \text{if } \phi_0 > \Delta x \\ \frac{\phi_0}{\sqrt{\phi_0^2 + \Delta x^2}} & \text{otherwise.} \end{cases} \quad (12)$$

Comparing with FMM, the second approach allows the use of higher order schemes (e.g. WENO5) and is easy to parallelize; hence, it has been a much more popular choice. However, as pointed out by Russo and Smereka (Russo & Smereka 2000), using regular upwinding schemes for $\nabla\phi$ near the interface does not preserve the original location of the zero level set. This can lead to mass loss, especially if the level set is far from a distance function and Eq. (11) needs to be evolved for long time. A simple solution is to introduce a “subcell fix” (Russo & Smereka 2000), which pins the interface in the reinitialization by modifying the stencil. Beautifully as it works in redistancing the level set, this method is however only second order accurate and thus not well-suited for evaluating curvature. Its fourth order extension (du Chéné *et al.* 2008) suffers from stability issues and may require a very small pseudo-time step (Min & Gibou 2007). Based on these observations, in this paper we solve Eq. (11) using the classical WENO5 (Liu *et al.* 1994) and the same SSP-RK3 (Shu & Osher 1988). The reinitialization is not performed at every physical time step, but depends on the advection velocity. In our applications, it typically requires one to two iterations of Eq. (11) per ten to a hundred time steps.

Distorted elliptic field. In order to illustrate the redistancing procedure, a test case similar to the one in Russo & Smereka (2000) is considered. Define the initial level set as

$$\phi(x, y, 0) = f(x, y) \left(\sqrt{\left(\frac{x^2}{4} + \frac{y^2}{16} \right)} - 1 \right),$$

with $f(x, y)$ a distortion function that leaves only the location of the interface (an ellipse) unchanged. The initial condition is displayed in Fig. 3(a), where the shape of the ellipse is depicted as the thick blue line; the red dashed lines depict iso-contours of ϕ ranging from -1 to 1. Clearly, this initial condition is far from being equidistant. However, as $\phi(x, y, \tau)$ is evolved under Eq. (11), it eventually converges towards a signed-distance function as seen in Fig. 3(b) and (c).

4. Interface-correction level set (ICLS) method

It is known that classical level set methods lead to mass loss when applied to multiphase flows, partially because there is no underlying mass conservation in the level set formalism, partially because of the reinitialization procedure. Such mass loss can sometimes be reduced or even removed by using the various approaches listed in Sec. 1, e.g. the CLSVOF method (Sussman & Puckett

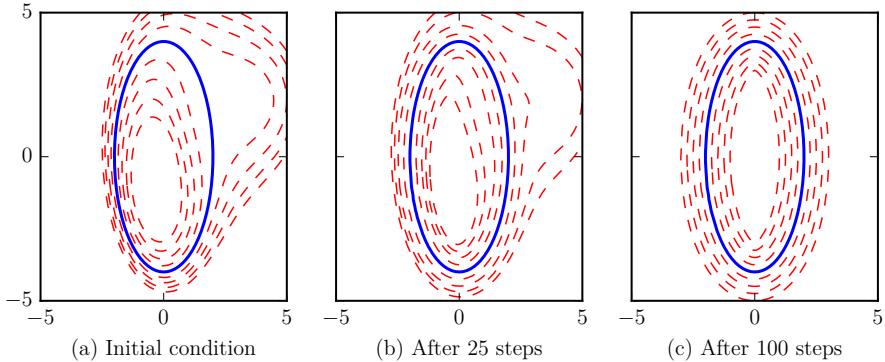


Figure 3: Illustration of the reinitialization procedure. The shape of the ellipsoid is depicted as the thick solid line. The dashed lines then depict iso-contours of $\phi(x, y)$ ranging from -1 to 1 by increments of 0.25 .

2000) or the hybrid particle level set method (Enright *et al.* 2002). However, doing so often makes the level set schemes complicated to implement and less efficient. To maintain the simplicity of the original level set method, we propose an alternative approach to conserve mass by performing small corrections near the interface. Because such corrections are done by directly solving a PDE (same as Eq. (7)), the proposed method is straightforward to implement in both 2D and 3D. Meanwhile, because the correction does not need to be performed at every time step, the additional cost is also negligible. Below, we first present the derivation of the correction-velocity, then we demonstrate the mass conservation with an example.

Let Γ divide a domain into two disjoint subsets Ω_1 (e.g. a droplet) and Ω_2 (e.g. the ambient fluid), and V denote the volume of Ω_1 (Fig. 4). Without loss of generality, we let $\phi < 0$ in Ω_1 , and $\phi > 0$ in Ω_2 . The rate of change of V can be written as the integral of a normal velocity \mathbf{u}_c defined on Γ (Salac 2016), *i.e.*

$$\int_{\Gamma} \mathbf{n} \cdot \mathbf{u}_c d\Gamma = \frac{\delta V}{\delta t}, \quad (13)$$

where \mathbf{n} is the outward-pointing normal from the interface Γ . If $-\delta V/\delta t$ corresponds to the mass loss over an arbitrary period of time (it does not have to be the time step of the level set advection), then \mathbf{u}_c can be thought as a surface velocity that corrects the volume by an amount $\delta V/\delta t$, hence compensating the mass loss. In other words, if \mathbf{u}_c is known, then the following PDE can be solved,

$$\frac{\partial \phi}{\partial t} + \mathbf{u}_c \cdot \nabla \phi = 0, \quad (14)$$

after which the mass loss accumulated over δt is removed.

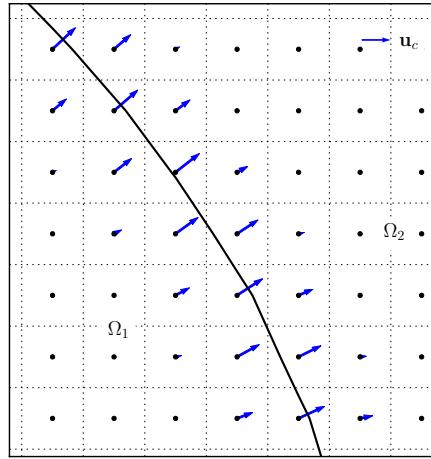


Figure 4: 2D illustration of the mass correction. The solid line represents the interface. The arrows indicate the normal correction-velocity located at cell centers of the grid.

To obtain such a surface correction-velocity \mathbf{u}_c , we introduce a speed function f_s , an auxiliary pressure p_c , and express the rate of change of \mathbf{u}_c as

$$\frac{d\mathbf{u}_c}{dt} = -f_s \nabla p_c. \quad (15)$$

Here, p_c can be imagined as a non-dimensional correction-pressure in Ω_1 . If $f_s = 1$, the physical interpretation of Eq. (15) is analogous to the inflation of a balloon by δV under pressure p_c over time Δt . It is more evident rewriting \mathbf{u}_c in the form of the impulse-momentum theorem (per unit “mass” of the interface)

$$\mathbf{u}_c = - \int_0^{\Delta t} \nabla p_c dt, \quad (16)$$

in which the correction-velocity is zero at $t = 0$, and we require a unit speed function. In general, substituting Eq. (16) into Eq. (13) results in

$$\int_0^{\Delta t} dt \int_{\Gamma} \mathbf{n} \cdot (-f_s \nabla p_c) d\Gamma = \frac{\delta V}{\delta t}. \quad (17)$$

In order for ∇p_c to be compatible with \mathbf{u}_c , p_c has to be differentiated at the interface. Using a 1D regularized Heaviside function of ϕ , such as

$$H_{\epsilon}(\phi) = \begin{cases} 1 & \text{if } \phi > \epsilon \\ \frac{1}{2} \left[1 + \frac{\phi}{\epsilon} + \frac{1}{\pi} \sin\left(\frac{\pi\phi}{\epsilon}\right) \right] & \text{if } |\phi| \leq \epsilon \\ 0 & \text{otherwise,} \end{cases} \quad (18)$$

with $\epsilon = 1.5\Delta x$ the half smoothing width, the correction-pressure and its gradient in Eq. (17) can be conveniently written as

$$p_c = (1 - H_\epsilon(\phi))p_0, \quad (19)$$

and

$$\int_{\Gamma} \nabla p_c = - \int_{\Gamma} \delta_\epsilon(\phi) \nabla \phi p_0, \quad (20)$$

where $\delta_\epsilon(\phi)$ is the derivative of $H_\epsilon(\phi)$, and p_0 is a constant. Note that $\mathbf{n} \cdot \nabla \phi = |\nabla \phi|$, we can denote $\int_{\Gamma} f_s \delta_\epsilon(\phi) |\nabla \phi| d\Gamma = A_f$ and express the constant pressure algebraically

$$p_0 = \frac{\delta V}{\delta t} \frac{1}{A_f \Delta t}, \quad (21)$$

by substituting Eq. (20) into (17), and approximating the time integration to first order, *i.e.* $\int_0^{\Delta t} A_f dt = A_f \Delta t$. Finally, Eqs. (15) (20) and (21) can be combined to give

$$\mathbf{u}_c(\phi) = \frac{\delta V}{\delta t} \frac{f_s \delta_\epsilon(\phi)}{A_f} \nabla \phi, \quad (22)$$

or

$$\mathbf{u}_c(\phi) = \frac{\delta V}{\delta t} \frac{f_s}{A_f} \nabla H_\epsilon(\phi). \quad (23)$$

Once \mathbf{u}_c is found, Eq. (14) can be solved for one time step to correct the mass loss. Here, we have required a bounded support for \mathbf{u}_c , *i.e.* $\mathbf{u}_c = \mathbf{0}$ for $|\phi| \geq \epsilon$ (see Fig. 4). There are two benefits of spreading the surface velocity. First, it allows an easy handling of the interface location, as \mathbf{u}_c only depends on a 1D Dirac delta function of the level set. The choice of $\delta_\epsilon(\phi)$ can also be different from the trigonometric form implied from Eq. (18); however, we prove in Appendix A that the discretization error of $\int_{\Gamma} \mathbf{n} \cdot \mathbf{u}_c d\Gamma$ is always zero, independent of $\delta_\epsilon(\phi)$. The important point here is we spread the *correction-velocity* rather than the *interface*. The interface remains sharp, as it is implicitly represented by the level set function. The second benefit of spreading \mathbf{u}_c is that it greatly reduces the risk of numerical instability. As \mathbf{u}_c is supported on a 2ϵ band around the interface, the maximal nodal value of \mathbf{u}_c scales with $1/\epsilon$. In our tests, we have never found its non-dimensional value to exceed 1. Therefore, the CFL conditions imposed by Eq. (14) is satisfied as long as we use the same temporal scheme (e.g. RK3) for solving Eq. (7) and Eq. (14). Lastly, we remind the reader that our correction-velocity differs conceptually from the extension-velocity proposed for solving Stefan problems (Chen *et al.* 1997; Adalsteinsson & Sethian 1999). The extension-velocity by design will keep the level set a distance function; while the design principle here is to preserve the global mass. This distinction is clear comparing the construction procedures of the two velocities.

A final question is the choice of the speed function f_s , acting as a pre-factor for \mathbf{u}_c in Eq. (22) or (23). To the best of the authors knowledge, there is no

simple, universally-valid criteria for such corrections. Two possible ways are

$$f_s \equiv \begin{cases} 1 & \text{uniform speed} \\ \kappa(\phi) & \text{curvature-dependent speed.} \end{cases} \quad (24)$$

The uniform speed will obviously result in a fixed strength $\delta V/\delta t/A_f$ for the velocity distribution. In the case of a static spherical droplet, this is the ideal choice for f_s , since the droplet should remain a sphere. In more general cases, when a fluid interface is subject to deformations or topological changes, a curvature-dependent speed may be more appropriate. This is based on the assumption that local structures of higher curvature or regions where the flow characteristics merge tend to be under-resolved (Enright *et al.* 2002); hence, they are more prone to mass losses. Indeed, a linear curvature weight has been adopted by many and demonstrated to produce accurate results in different contexts (Luo *et al.* 2015b; Aanjaneya *et al.* 2013). Furthermore, κ/A_f reduces to $1/A_f$ when the curvature is uniform. Therefore, we can rewrite Eq. (23) using a curvature-dependent speed

$$\mathbf{u}_c(\phi) = \frac{\delta V}{\delta t} \frac{\kappa(\phi)}{A_f} \nabla H_\epsilon(\phi). \quad (25)$$

Clearly, this correction-velocity is larger in highly curved parts, and smaller in flatter parts. It thus includes “local” information while maintaining “global” mass conservation. Standard central-difference discretization applies, where the components of \mathbf{u}_c can be obtained at either the cell faces or cell centers. The computation of $\kappa(\phi)$ is crucial and will be presented in the next section. We stress that such a curvature-dependence is not unique. In principle, one can choose different weight-functions, and validate the choice based on the specific applications. Practically, the difference is expected to be negligible since the mass loss remains small (typically around 10^{-5}) at each correction step.

After correcting the level set on a 2ϵ band around the interface, a reinitialization step is required to redistribute the values within the entire narrow band (2γ). The two procedures can be readily combined, since it is not necessary to perform mass correction at every time step. Also, because the formalism is cast in a level set frame, generalization from 2D to 3D is trivial. Comparing with other mass-preserving methods, the additional computational cost of ICLS is small. This is due to the simple algebraic expression of \mathbf{u}_c (Eq. (25)), and only one solve of Eq. (14) is required; whereas a typical VOF-coupling method involves solving another set of transport equations (Sussman & Puckett 2000), or reconstructing the interface by an iterative procedure (Luo *et al.* 2015b).

In summary, the ICLS method proceeds by performing the following steps:

1. Advect ϕ^n from time t^n to t^{n+1} with Eq. (7), using the flow velocity \mathbf{u}^n .
2. If reinitialization will be executed (otherwise, go to step 3):
 - (a) Perform mass correction with Eq. (14), using \mathbf{u}_c from Eq. (25).
 - (b) Reinitialize ϕ^{n+1} with Eq. (11).
3. Exit the level set solver.

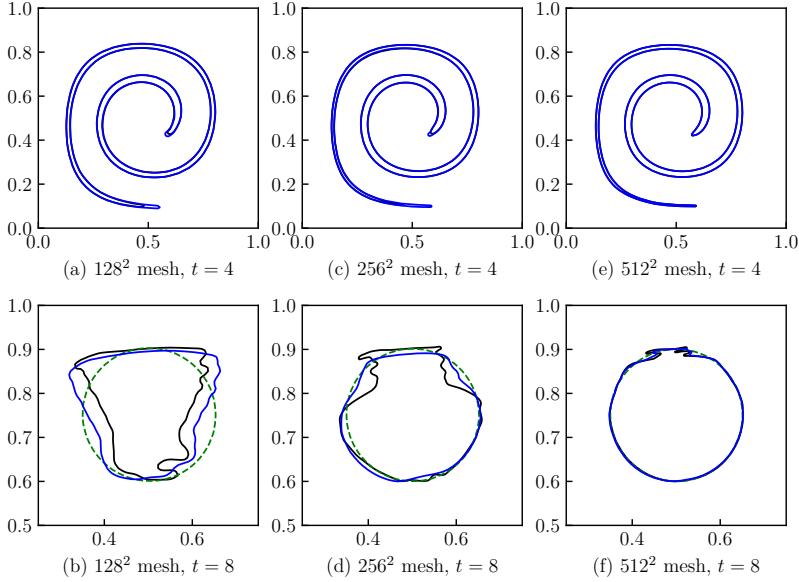


Figure 5: Interface at $t = 4$ and $t = 8$ for different meshes. The solid black lines indicate simulations without mass correction, the solid blue lines indicate simulations with the current mass correction method, the green dashed lines in (b)(d)(f) indicate the original circle. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Deforming circle. To assess the performance of ICLS on mass conservation, we test the standard benchmark of a circle deformed by a single vortex. Here, the circle of radius 0.15 is initially centered at $(x, y) = (0.5, 0.75)$ of a $[0, 1] \times [0, 1]$ box. The velocity is imposed directly and can be obtained from the stream function

$$\psi(x, y, t) = \frac{1}{\pi} \sin^2(\pi x) \sin^2(\pi y) \cos\left(\frac{\pi t}{T}\right),$$

where T is traditionally set to 8. Under this flow, the circle will be stretched to maximum at $t = T/2$ and rewound to its initial condition at $t = T$. Although formulated simply, accurately transporting the interface without mass loss is a difficult task.

We perform this test on three different meshes using the complete level set solver: HOUCL5 is used for the level set advection, WENO5 is used for reinitialization every 5 to 20 time steps, the mass correction is performed every 5 to 10 time steps; and the time step is chosen such that $\Delta t / \Delta x = 0.32$. Fig. 5 shows the shapes of the filament/circle at $t = 4$ and $t = 8$ at various resolutions. From the upper panel, it is clearly seen that the filament has a longer tail

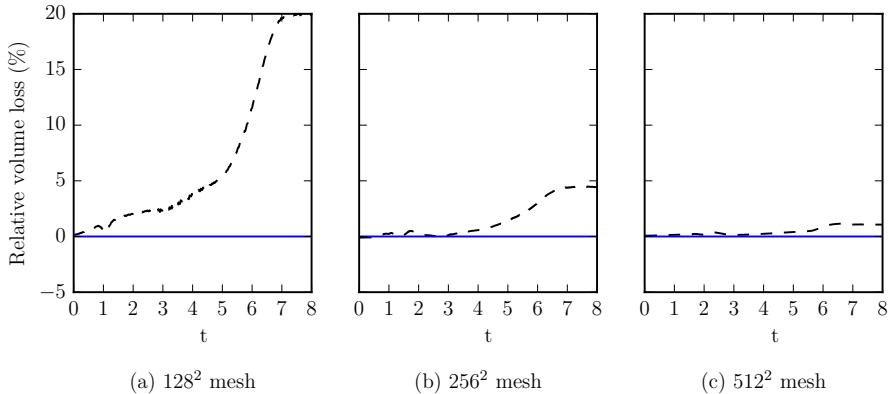


Figure 6: Relative volume loss for three different meshes. Dashed lines indicate simulations without mass correction; solid lines indicate simulations with mass correction.

and head due to mass correction; as we increase the resolution, the difference becomes smaller. The lower panel of Fig. 5 depicts the final shapes, ideally the initial circle if the motion is totally passive. Some artifacts are visible due to the fact that the filament is always under-resolved at the maximum stretching and the level set will automatically merge the characteristics to yield an entropy solution (Sethian 1999). We note that the final outcome can be tuned by modifying the frequency of the reinitialization/mass correction, a trade-off between the appearance and the mass loss. However, the objective here is to demonstrate the mass conservation enforced by ICLS, which is clearly illustrated in Fig. 6. For passive transport involving large deformations, we recommend particle-based methods (Enright *et al.* 2002). Examples of droplets/bubbles in physical conditions using ICLS will be shown in the validations (Sec. 6.5) and applications (Sec. 7) below.

5. Curvature computation

Curvature computation is crucial to interfacial flows in the presence of surface tension, as inaccurate curvature can result in unphysical spurious currents (Herrmann 2008; Desjardins *et al.* 2008), and even more so in our case when we apply curvature-dependent interface corrections. In this section, we first briefly describe the calculation of cell-center curvatures; *i.e.*, the curvature evaluated at the same nodal position as the level set function. Then, we introduce a geometric approach for the estimation of interface curvatures corresponding to the zero level set. The second step is specially tailored to the ghost fluid method that will be presented in Sec. 6.2.

5.1. Cell-center curvature

From Eq. (6), the curvature κ can be evaluated as

$$\kappa = -\frac{\phi_{yy}\phi_x^2 + \phi_{xx}\phi_y^2 - 2\phi_x\phi_y\phi_{xy}}{(\phi_x^2 + \phi_y^2)^{3/2}} \quad (26)$$

and as

$$\kappa_M = -\frac{\left\{ (\phi_{yy} + \phi_{zz})\phi_x^2 + (\phi_{xx} + \phi_{zz})\phi_y^2 + (\phi_{xx} + \phi_{yy})\phi_z^2 \right.}{\left. -2\phi_x\phi_y\phi_{xy} - 2\phi_x\phi_z\phi_{xz} - 2\phi_y\phi_z\phi_{yz} \right\}}{(\phi_x^2 + \phi_y^2 + \phi_z^2)^{3/2}} \quad (27)$$

in 2D and 3D Cartesian coordinates, respectively, where the subscript M denotes the mean curvature (Sethian 1999). The curvature can be determined from these expressions using simple central finite-differences. It has to be noted, however, that such evaluation of κ involves second derivatives of the level set field $\phi(\mathbf{x})$. As a consequence, if the calculation of ϕ is only second-order accurate, the resulting κ will be of order zero. To nonetheless retain a grid converging κ , one can use the compact least-squares scheme proposed by Marchandise *et al.* (2007). Their approach provides a second-order, grid converging evaluation of the cell-center curvature. It moreover smears out undesired high frequency oscillations possibly introduced by the velocity field. A similar procedure has also been adopted in other works (Desjardins *et al.* 2008; Luo *et al.* 2015b).

The principle of the least squares approach is to solve an over-determined linear system, $\mathbf{Ax} = \mathbf{b}$, where \mathbf{A} is a matrix built from the local coordinates, \mathbf{x} is a unknown array containing the reconstructed level set values and its spatial derivatives, and \mathbf{b} is the original level set field. The detailed descriptions can be found in Marchandise *et al.* (2007). Here, we only note that the level set function remains unmodified after this step. From a practical point of view, provided the mesh considered is uniform in all directions, the pseudo-inverse of the matrix \mathbf{A} only needs to be evaluated once and applied close to the interface. Therefore, the computational cost of this least-squares calculation is negligible.

5.2. Interface curvature

The least-squares approach described in the previous section only allows one to compute the nodal curvature κ of the level set field ϕ . For computations using the GFM (Sec. 6.2), one might however require an accurate evaluation of the curvature at the exact location of the interface. Provided a grid-converging cell-center curvature, the actual curvature at the interface can be interpolated from its neighboring cells weighted by the level set (Francois *et al.* 2006; Luo *et al.* 2015a). Here we present a slightly different but robust algorithm to estimate the interface curvature, with a straight-forward geometrical interpretation.

2D estimation. Suppose the interface Γ cuts through two adjacent cells, (i, j) and $(i+1, j)$, where the cell-center curvatures $\kappa_{i,j}$ and $\kappa_{i+1,j}$ are known.

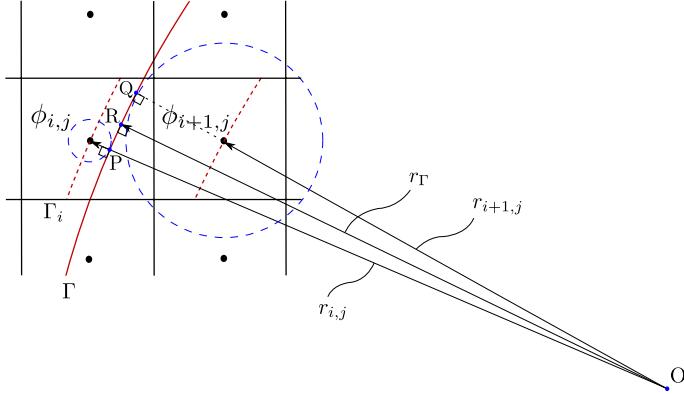


Figure 7: Estimation of the interface's curvature from neighboring cells.

In 2D, we can determine the radius of curvature at each cell directly from

$$\kappa_{i,j} = -\frac{1}{r_{i,j}}, \quad \kappa_{i+1,j} = -\frac{1}{r_{i+1,j}}, \quad (28)$$

as illustrated in Fig. 7. Since the level set is defined as the signed distance to the interface, Γ must be tangent to a circle of radius $|\phi_{i,j}|$ centered at (i, j) , and parallel to the contour line of $\Gamma_i = \{\mathbf{x} | \phi = \phi_{i,j}\}$ (otherwise they will not remain equidistant). We also know Γ lies between (i, j) and $(i + 1, j)$, then it must pass through P (see Fig. 7). Since Γ and Γ_i are parallel and there is only one line normal to both curves passing through P , $r_{i,j}$ and OP must originate from the same point, O . Then we get

$$|OP| = r_{i,j} - s_\Gamma \phi_{i,j}. \quad (29)$$

where s_Γ is a sign function equal to 1 if the interface wrapping the negative level set is convex, and equal to -1 if concave.

The same argument holds for cell $(i + 1, j)$, which yields $|OQ| = r_{i+1,j} - s_\Gamma \phi_{i+1,j}$. We can therefore write the radius of the interface curvature between (i, j) and $(i + 1, j)$ as

$$r_\Gamma = \frac{|OP| + |OQ|}{2}, \quad (30)$$

so that the interface curvature becomes

$$\kappa_\Gamma = \frac{2}{\kappa_{i,j}^{-1} + \kappa_{i+1,j}^{-1} + s_\Gamma(\phi_{i,j} + \phi_{i+1,j})}. \quad (31)$$

The above derivation provides a relation between the interface curvature and that at the adjacent cell-centers in the x direction. Similar results can be obtained in the y direction (e.g. between $\phi_{i,j}$ and $\phi_{i,j-1}$). The assumptions we have made here are 1) the cell-center curvatures are accurate and 2) the

interface curvatures at P and Q are the same, so that OP and OQ are co-centered (or, $|OP| \approx |OQ| \approx |OR|$). The second assumption is essentially a sub-cell approximation, and we expect it to be valid as long as the interface is well-resolved. One exception we have found is when two interfaces are closer than about $2\Delta x$, the local level set field will develop “corners”. In that case, the cell-center curvatures are erroneous and the underlying assumptions we require here are not fulfilled. We do not discuss that case in the present paper. However, we demonstrate in the next section that a second-order convergence is achieved when the interface is resolved.

3D estimation. In three dimensions, the mean curvature of a surface can be written as

$$\kappa_\Gamma = -\left(\frac{1}{r_{\Gamma 1}} + \frac{1}{r_{\Gamma 2}}\right), \quad (32)$$

where $r_{\Gamma 1}$ and $r_{\Gamma 2}$ are the two principal radii corresponding to the maximal and minimal planar radius of curvature. Note that we do not need to approximate the interface as a sphere since there is always a plane where the previous picture (Fig. 7) holds. Under the same assumption as for the 2D case, that the interface at P and Q have the same principal radii (hence the same curvature), one can again relate the nodal curvatures to their nearby interface as

$$\begin{aligned} \kappa_{i,j,k} &= -\left(\frac{1}{r_{\Gamma 1} + s_\Gamma \phi_{i,j,k}} + \frac{1}{r_{\Gamma 2} + s_\Gamma \phi_{i,j,k}}\right), \\ \kappa_{i+1,j,k} &= -\left(\frac{1}{r_{\Gamma 1} + s_\Gamma \phi_{i+1,j,k}} + \frac{1}{r_{\Gamma 2} + s_\Gamma \phi_{i+1,j,k}}\right), \end{aligned} \quad (33)$$

where s_Γ is the same sign function defined for the 2D case. Comparing equations (32) and (33), it is natural to expand Eq. (33) into a Taylor series and to approximate the interface curvature directly as

$$\kappa_\Gamma = \frac{\epsilon_{i+1}\kappa_i - \epsilon_i\kappa_{i+1}}{\epsilon_{i+1} - \epsilon_i} + O(\epsilon_i^2, \epsilon_{i+1}^2), \quad (34)$$

where

$$\epsilon_i = s_\Gamma \phi_{i,j,k}. \quad (35)$$

Since the level set must change sign across the interface, Eq. (34) is always defined and it reduces to the exact value if the cell center happens to be on the interface. Similarly, the whole procedure is repeated in the y and z directions.

Finally, in order to ensure a robust estimation, we perform an additional quadratic least squares approximation on the curvature field near the interface, similar to Marchandise *et al.* (2007). This procedure takes place before the 3D estimation (Eq. (34)), and essentially improves the accuracy of cell-center curvatures by removing possible high-frequency noise. We note that the second averaging is optional, and different methods can be found in literature to evaluate the cell-center curvatures (du Chéné *et al.* 2008). In the present paper, the least squares approach mentioned in Sec. 5.1 is used for all the cases.

To assess the accuracy of our interface curvature estimation, we calculate the L_∞ norm of a circle/sphere of radius 0.25 centered in a unit square/cube.

Table 1: Grid convergence of the current interface curvature calculation in both 2D and 3D.

Points per diameter	16	32	48	64
L_∞ 2D	1.144×10^{-2}	2.904×10^{-3}	1.285×10^{-3}	7.227×10^{-4}
L_∞ 3D	1.527×10^{-2}	3.888×10^{-3}	1.732×10^{-3}	9.753×10^{-4}

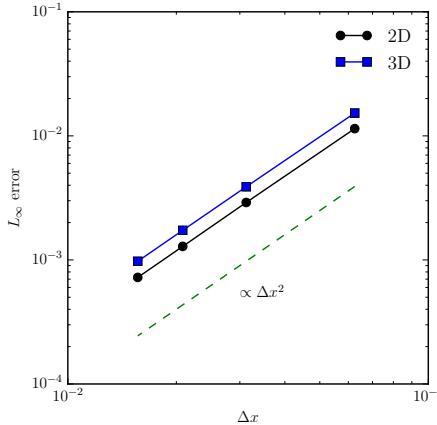


Figure 8: Second order convergence of the interface curvature computation in both 2D and 3D.

Table 1 summarizes the error after one step of the calculations on different resolutions, which are also plotted in Fig. 8. Clearly, second-order convergence is achieved in both 2D and 3D cases.

6. Solution of the Navier-Stokes equations

In this section, we outline the flow solver developed from that of Breugem (2012) for particle-laden flows. After advancing the level set from ϕ^n to ϕ^{n+1} , the density and viscosity fields are updated by

$$\rho^{n+1} = \rho_1 H_s(\phi^{n+1}) + \rho_2(1 - H_s(\phi^{n+1})), \quad (36a)$$

$$\mu^{n+1} = \mu_1 H_s(\phi^{n+1}) + \mu_2(1 - H_s(\phi^{n+1})), \quad (36b)$$

where

$$H_s(\phi) = \begin{cases} 1 & \text{if } \phi > 0 \\ 0 & \text{otherwise,} \end{cases} \quad (37)$$

is a simple step function.

Next, a prediction velocity \mathbf{u}^* is computed by defining \mathbf{RU}^n as

$$\mathbf{RU}^n = -\nabla \cdot (\mathbf{u}^n \mathbf{u}^n) + \frac{1}{Re} \left(\frac{1}{\rho^{n+1}} \nabla \cdot [\mu^{n+1} (\nabla \mathbf{u}^n + (\nabla \mathbf{u}^n)^T)] \right) + \frac{1}{Fr} \mathbf{g}, \quad (38)$$

which is the right-hand side of the momentum equation (1b) excluding the pressure gradient term. Integrating in time with the second-order Adams-Bashforth scheme (AB2) yields

$$\mathbf{u}^* = \mathbf{u}^n + \Delta t \left(\frac{3}{2} \mathbf{RU}^n - \frac{1}{2} \mathbf{RU}^{n-1} \right). \quad (39)$$

To enforce a divergence-free velocity field (Eq. (1a)), we proceed by solving the Poisson equation for the pressure as in the standard projection method (Chorin 1968), *i.e.*

$$\nabla \cdot \left(\frac{1}{\rho^{n+1}} \nabla p^{n+1} \right) = \frac{1}{\Delta t} \nabla \cdot \mathbf{u}^*. \quad (40)$$

The surface tension between two fluids is also computed during this step, using the ghost fluid method (Fedkiw *et al.* 1999) (Sec. 6.2). This allows for an accurate and sharp evaluation of the pressure jump even at large density contrasts (Desjardins *et al.* 2008). Finally, the velocity at the next time level is updated as

$$\mathbf{u}^{n+1} = \mathbf{u}^* - \frac{\Delta t}{\rho^{n+1}} \nabla p^{n+1}. \quad (41)$$

6.1. Fast pressure-correction method

In the above outline, a Poisson equation for the pressure (Eq. (40)) must be solved at each time step. This operation takes most of the computational time in the projection method, as it is usually solved iteratively. In addition, the operation count of iterative methods depends on the problem parameters (e.g. density ratio) and the convergence tolerance (Dodd & Ferrante 2014). On the other hand, Dong & Shen (2012) recently developed a velocity-correction method that transforms the variable-coefficient Poisson equation into a constant-coefficient one. The essential idea is to split the pressure gradient term in Eq. (40) in two parts, one with constant coefficients, the other with variable coefficients, *i.e.*

$$\frac{1}{\rho^{n+1}} \nabla p^{n+1} \rightarrow \frac{1}{\rho_0} \nabla p^{n+1} + \left(\frac{1}{\rho^{n+1}} - \frac{1}{\rho_0} \right) \nabla \hat{p}, \quad (42)$$

where $\rho_0 = \min(\rho_1, \rho_2)$ and \hat{p} is the approximate pressure at time level $n + 1$. This splitting reduces to the exact form of Eq. (40) within the lower-density phase, while its validity in the higher-density phase and at the interface depends on the choice of \hat{p} . Later, Dodd & Ferrante (2014) showed that by explicitly estimating \hat{p} from two previous time levels as

$$\hat{p} = 2p^n - p^{n-1}, \quad (43)$$

the resulting velocity field in Eq. (41) will be second-order accurate in both space and time, independent of the interface advection method. Furthermore, if the computational domain includes periodic boundaries or can be represented by

certain combination of homogeneous Dirichlet/Neumann conditions (Schumann & Sweet 1988), the constant-coefficient part of Eq. (42) can be solved directly using Gauss elimination in the Fourier space. Such a FFT-based solver can lead to a speed-up of 10 – 40 times, thus the name fast pressure-correction method (FastP*). Following this approach, Eqs. (40) and (41) are modified as

$$\nabla^2 p^{n+1} = \nabla \cdot \left[\left(1 - \frac{\rho_0}{\rho^{n+1}} \right) \nabla \hat{p} \right] + \frac{\rho_0}{\Delta t} \nabla \cdot \mathbf{u}^* \quad (44)$$

and

$$\mathbf{u}^{n+1} = \mathbf{u}^* - \Delta t \left[\frac{1}{\rho_0} \nabla p^{n+1} + \left(\frac{1}{\rho^{n+1}} - \frac{1}{\rho_0} \right) \nabla \hat{p} \right]. \quad (45)$$

6.2. Ghost fluid method

As discussed before, surface tension is commonly computed using the continuum surface force (CSF) model (Brackbill *et al.* 1992), in which the pressure jump across an interface is represented as a forcing term on the right-hand side of Eq. (1b). Despite its simplicity, CSF introduces an unfavorable smearing in the density and pressure profiles, resulting in an artificial spreading of the interface (typically over a thickness of $3\Delta x$). An alternative approach is the so-called ghost fluid method (GFM), originally developed by Fedkiw *et al.* (1999) to capture the boundary conditions in the inviscid compressible Euler equations. Unlike CSF, GFM enables a numerical discretization of the gradient operator while preserving the discontinuity of the differentiated quantity. It was extended to viscous flows by Kang *et al.* (2000) and has been successfully utilized in multiphase flow simulations, see e.g. Desjardins *et al.* (2008); Coyajee & Boersma (2009); Tanguy & Berlemont (2005).

Recall from Eq. (2) that the pressure jump has two components, one arising from the surface tension, the other from the viscosity difference of the two fluids. In Kang *et al.* (2000), a complete algorithm is provided to compute the two contributions, making the density, viscosity, and pressure all sharp. However, having a sharp viscosity profile requires an extra step to evaluate the divergence of the deformation tensor (see Eq. (38)). That is, for cells adjacent to the interface, the second derivatives of the velocity must be evaluated using the techniques developed in Liu *et al.* (2000); Kang *et al.* (2000). However, rewriting Eq. (2) as

$$[p]_\Gamma = \frac{1}{Re} \left(\frac{\kappa}{Ca} + 2[\mu]_\Gamma \mathbf{n}^T \cdot \nabla \mathbf{u} \cdot \mathbf{n} \right), \quad (46)$$

reveals that surface tension is the dominant term when the Capillary number, $Ca = We/Re$, is small. For the applications we are interested in, e.g. colloidal droplets in microfluidic channels, Ca is of the order of 10^{-5} . Therefore, in the present implementation, we regularize the viscosity profile (*i.e.* replacing $H_s(\phi)$ in Eq. (36b) with $H_\epsilon(\phi)$ in Eq. (18)) and use GFM only for the pressure jump.

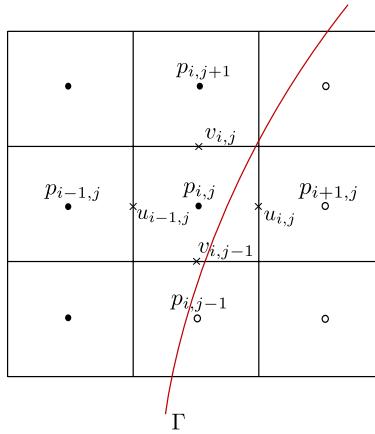


Figure 9: Schematic of the 2D staggered grid where pressure locates at cell centers and velocity components locate at cell faces. The curved line specifies the interface Γ ; filled and empty circles indicate discontinuous pressure (or density) values in phase 1 and 2, respectively.

6.2.1. Spatial discretization

Eqs. (38), (44), and (45) are discretized on a standard staggered grid using a second-order conservative finite volume method. It is equivalent to central differences in all three directions if the mesh is uniform. A detailed description of the discretization of the individual terms can be found in Dodd & Ferrante (2014), Sec. 2.2.1. For brevity, we show here only the 2D evaluations of ∇p and $\nabla^2 p$ due to GFM.

As sketched in Fig. 9, computing $\nabla^2 p$ at node (i, j) requires three entries of p in each direction. If CSF is used, all gradient terms can be evaluated with the straightforward central-difference, *i.e.*

$$(\nabla^2 p)_{i,j} = \frac{p_{i-1,j}^s - 2p_{i,j}^s + p_{i+1,j}^s}{\Delta x^2} + \frac{p_{i,j-1}^s - 2p_{i,j}^s + p_{i,j+1}^s}{\Delta y^2}. \quad (47)$$

However, the pressure at the cells adjacent to the interface will have to be smeared out; hence we denote them with p^s . In order for the pressure to be sharp, GFM creates an artificial fluid (the “ghost” fluid) and assumes that the discontinuity can be extended beyond the physical interface. That is, if we know the corresponding jumps of pressure, then its derivatives can be evaluated without smearing by removing such jumps. For the particular case depicted in Fig. 9, Eq. (47) can be re-written as (see Liu *et al.* (2000) for the intermediate

steps)

$$(\nabla^2 p)_{i,j} = \frac{p_{i-1,j} - 2p_{i,j} + p_{i+1,j}}{\Delta x^2} - \frac{[p]_{i,j}}{\Delta x^2} - \frac{1}{\Delta x} \left[\frac{\partial p}{\partial x} \right]_{i+1/2,j} + \frac{p_{i,j-1} - 2p_{i,j} + p_{i,j+1}}{\Delta y^2} - \frac{[p]_{i,j-1}}{\Delta y^2}, \quad (48)$$

where we recall $[\cdot]_{i,j}$ denotes the discontinuity from fluid 1 to fluid 2 at cell (i,j) (same for $[\cdot]_{i,j-1}$, etc.).

To determine the jump terms in Eq. (48), we first note that the velocity and its material derivatives across the interface of viscous flows are continuous Kang *et al.* (2000); Desjardins *et al.* (2008), resulting in

$$\left[\frac{1}{\rho^{n+1}} \nabla p^{n+1} \right]_\Gamma = \mathbf{0}. \quad (49)$$

Furthermore, owing to the splitting that allows us to solve only for a constant-coefficient Poisson equation (Eq. (44)), Eqs. (42) and (49) lead to

$$\left[\frac{1}{\rho_0} \nabla p^{n+1} \right]_\Gamma + \left[\left(\frac{1}{\rho^{n+1}} - \frac{1}{\rho_0} \right) \nabla \hat{p} \right]_\Gamma = \mathbf{0}, \quad (50)$$

which also implies that the pressure gradient terms are continuous everywhere (e.g. the subscript can be $(i+1/2, j)$), along any direction.

Denoting the right-hand side of Eq. (44) as RP , it is discretized as

$$RP_{i,j} = \left(\left(1 - \frac{\rho_0}{\rho_{i+1/2,j}^{n+1}} \right) \frac{\partial \hat{p}}{\partial x}_{i+1/2,j} - \left(1 - \frac{\rho_0}{\rho_{i-1/2,j}^{n+1}} \right) \frac{\partial \hat{p}}{\partial x}_{i-1/2,j} \right) / \Delta x + \left(\left(1 - \frac{\rho_0}{\rho_{i,j+1/2}^{n+1}} \right) \frac{\partial \hat{p}}{\partial y}_{i,j+1/2} - \left(1 - \frac{\rho_0}{\rho_{i,j-1/2}^{n+1}} \right) \frac{\partial \hat{p}}{\partial y}_{i,j-1/2} \right) / \Delta y - \frac{1}{\Delta x} \left[\left(1 - \frac{\rho_0}{\rho^{n+1}} \right) \frac{\partial \hat{p}}{\partial x} \right]_{i+1/2,j} + \frac{\rho_0}{\Delta t} \left(\frac{u_{i,j}^* - u_{i-1,j}^*}{\Delta x} + \frac{v_{i,j}^* - v_{i,j-1}^*}{\Delta y} \right), \quad (51)$$

again using GFM (Liu *et al.* 2000). Comparing Eqs. (48) and (51), we note that the jump of the first derivatives cancels out recognizing Eq. (50). With a modified right-hand side, RP^* , defined as

$$RP_{i,j}^* = RP_{i,j} + \frac{1}{\Delta x} \left[\left(1 - \frac{\rho_0}{\rho^{n+1}} \right) \frac{\partial \hat{p}}{\partial x} \right]_{i+1/2,j}, \quad (52)$$

the discrete form of Eq. (44) reduces to

$$\frac{p_{i-1,j}^{n+1} - 2p_{i,j}^{n+1} + p_{i+1,j}^{n+1}}{\Delta x^2} + \frac{p_{i,j-1}^{n+1} - 2p_{i,j}^{n+1} + p_{i,j+1}^{n+1}}{\Delta y^2} = \frac{[p]_{i,j}^{n+1}}{\Delta x^2} + \frac{[p]_{i,j-1}^{n+1}}{\Delta y^2} + RP_{i,j}^*. \quad (53)$$

Eq. (53) is still not ready to solve, since the pressure jumps for the first point away from the interface (e.g. $[p]_{i,j}^{n+1}$) are not known. Following Desjardins

et al. (2008), we perform a Taylor series expansion around Γ ,

$$[p]_{i,j}^{n+1} = [p]_{\Gamma}^{n+1} + (x_i - x_{\Gamma}) \left[\frac{\partial p}{\partial x} \right]_{\Gamma}^{n+1} + O((x_i - x_{\Gamma})^2), \quad (54)$$

where $[p]_{\Gamma}^{n+1} = \kappa_{\Gamma,x}/We$, and $\kappa_{\Gamma,x}$ is estimated from Eq. (31) in 2D and from Eq. (34) in 3D, along the x direction using $\phi_{i,j}^{n+1}$ and $\phi_{i+1,j}^{n+1}$. The jump of the pressure gradient at the interface can be similarly expanded at (i,j)

$$\left[\frac{\partial p}{\partial x} \right]_{\Gamma}^{n+1} = \left[\frac{\partial p}{\partial x} \right]_{i,j}^{n+1} + O(x_{\Gamma} - x_i), \quad (55)$$

resulting in

$$[p]_{i,j}^{n+1} = \frac{\kappa_{\Gamma,x}}{We} + (x_i - x_{\Gamma}) \left[\frac{\partial p}{\partial x} \right]_{i,j}^{n+1} + O((x_i - x_{\Gamma})^2). \quad (56)$$

Using Eq. (50), we can re-write Eq. (56) as

$$[p]_{i,j}^{n+1} = \frac{\kappa_{\Gamma,x}}{We} + (x_i - x_{\Gamma}) \left[\left(1 - \frac{\rho_0}{\rho^{n+1}}\right) \frac{\partial \hat{p}}{\partial x} \right]_{i,j} + O((x_i - x_{\Gamma})^2), \quad (57)$$

where the jump term on the right-hand side can be explicitly calculated using the family of identities of the form (Kang *et al.* 2000)

$$[AB] = [A]\tilde{B} + \tilde{A}[B], \quad \tilde{A} = aA_1 + bA_2, \quad a + b = 1. \quad (58)$$

Although Eqs. (57) and (58) lead to a second-order pressure jump, it is much simpler to keep only the leading-order term, *i.e.*

$$[p]_{i,j}^{n+1} = \frac{\kappa_{\Gamma,x}}{We} + O(x_i - x_{\Gamma}). \quad (59)$$

This way, the pressure jump varies only with the local curvature, remains invariant across the interface, and is second-order accurate when the density is uniform. For the test cases shown below, Eq. (59) is used. Thus, the complete discretization of Eq. (44) reads

$$\frac{p_{i-1,j}^{n+1} - 2p_{i,j}^{n+1} + p_{i+1,j}^{n+1}}{\Delta x^2} + \frac{p_{i,j-1}^{n+1} - 2p_{i,j}^{n+1} + p_{i,j+1}^{n+1}}{\Delta y^2} = \frac{1}{We} \left(\frac{\kappa_{\Gamma,x}}{\Delta x^2} + \frac{\kappa_{\Gamma,y}}{\Delta y^2} \right) + RP_{i,j}^*, \quad (60)$$

with $RP_{i,j}^*$ defined in Eq. (52) corresponding to Fig. 9.

Clearly, the resulting linear system (Eq. (60)) has a standard positive definite, symmetric coefficient matrix, and it can be solved directly using the FFT-based fast Poisson solver (Sec. 6.1). Care should be exercised when a nodal point crosses the interface in more than one direction. In those cases, the interface curvature of each crossing direction may be different and it shall not be averaged. Otherwise, the projection (Eq. (44)) and correction (Eq. (45)) steps can become inconsistent, making the velocity not divergence-free. Additionally,

when taking the gradient of the pressure-correction term; e.g. its derivative along the x direction, the correct discretization should be

$$\frac{\partial \hat{p}}{\partial x_{i,j}} = \frac{(\hat{p}_{i+1,j} - (2[p]_{i+1,j}^n - [p]_{i+1,j}^{n-1})) - \hat{p}_{i,j}}{\Delta x}. \quad (61)$$

After removing the jump, the divergence of the bracket term in Eq. (44) is evaluated in the same way as in Dodd & Ferrante (2014).

Finally, we can re-write Eqs. (44) and (45) compactly as

$$\nabla^2 p^{n+1} = \nabla_g^2 [p]_\Gamma + \nabla \cdot \left[\left(1 - \frac{\rho_0}{\rho^{n+1}} \right) \nabla_g \hat{p} \right] + \frac{\rho_0}{\Delta t} \nabla \cdot \mathbf{u}^*, \quad (62)$$

$$\mathbf{u}^{n+1} = \mathbf{u}^* - \Delta t \left[\frac{1}{\rho_0} \nabla_g p^{n+1} + \left(\frac{1}{\rho^{n+1}} - \frac{1}{\rho_0} \right) \nabla_g \hat{p} \right]. \quad (63)$$

where ∇_g and $\nabla_g^2 [p]_\Gamma$ denote, respectively, the gradient operator considering the jump and the extra jump terms from the laplacian operator due to GFM.

6.3. Time integration

In the current work, a second-order accurate Adams-Bashforth scheme is used for the time integration. The time step is restricted by convection, diffusion, surface tension, and gravity, due to our explicit treatment of these terms. As suggested in Kang *et al.* (2000), the overall time step restriction is

$$\Delta t \leq 1 / \left(C_{CFL} + V_{CFL} + \sqrt{(C_{CFL} + V_{CFL})^2 + 4G_{CFL}^2 + 4S_{CFL}^2} \right), \quad (64)$$

where C_{CFL} , V_{CFL} , G_{CFL} , and S_{CFL} are the “speeds” due to convection, viscosity, gravity, and surface tension, respectively. Specifically, they are given as

$$C_{CFL} = \frac{|u|_{max}}{\Delta x} + \frac{|v|_{max}}{\Delta y} + \frac{|w|_{max}}{\Delta z}, \quad (65)$$

$$V_{CFL} = \frac{1}{Re} \max \left(\frac{\mu_1}{\rho_1}, \frac{\mu_2}{\rho_2} \right) \left(\frac{2}{\Delta x^2} + \frac{2}{\Delta y^2} + \frac{2}{\Delta z^2} \right), \quad (66)$$

$$G_{CFL} = \sqrt{\frac{1}{Fr} \frac{|(1 - \frac{\rho_1 + \rho_2}{2\rho})g|_{max}}{\min(\Delta x, \Delta y, \Delta z)}}, \quad (67)$$

$$S_{CFL} = \sqrt{\frac{1}{We} \frac{|\kappa|_{max}}{\min(\rho_1, \rho_2) [\min(\Delta x, \Delta y, \Delta z)]^2}}. \quad (68)$$

where $|\kappa|_{max}$ in (68) can be approximated by $1/\Delta x$ in 2D and $2/\Delta x$ in 3D, assuming Δx is the smallest grid spacing.

The reasons we choose an explicit temporal scheme rather than an implicit one are twofold. First, for applications involving a large density and viscosity

contrast, the stability restriction imposed by surface tension is usually greater than that imposed by diffusion. Second, an implicit formulation of GFM has been admitted to be challenging to develop (Desjardins *et al.* 2008), and it was shown in a recent study (Denner & Wachem 2015) that a capillary time-step constraint exists, irrespective of the type of implementation, due to the temporal sampling of surface capillary waves. Fortunately, the fast pressure-correction method enables the use of FFT for the constant-coefficient Poisson equation and hence an accurate and fast solution of the two-fluid Navier-Stokes equation can be obtained.

6.4. Full solution procedure

We summarize the full solution procedure as follows:

1. Advance the interface explicitly from ϕ^n to ϕ^{n+1} using the ICLS, and update the density ρ^{n+1} and the viscosity μ^{n+1} .
2. Advance the velocity field explicitly from \mathbf{u}^n to \mathbf{u}^* with Eqs. (38) and (39).
3. Project the velocity field by solving the constant-coefficient Poisson Eq. (62) making use of the FastP* and the GFM.
4. Update the velocity from \mathbf{u}^* to \mathbf{u}^{n+1} explicitly with Eq. (63), again using the FastP* and the GFM.

6.5. Validations

In this section, we validate the coupled ICLS/NS solver using three benchmark examples with increasing complexities. Specifically, the first example verifies the discrete momentum balance for fluids of the same density and viscosity. This concerns the surface tension computed by the GFM using interface curvatures. Then, the density and viscosity ratios are significantly increased (up to 10^4) to test the combined FastP* and GFM. Using the same test, we also provide a convergence check of the complete flow solver. Finally, the overall accuracy is assessed by simulating a 3D bubble in comparison with experiments.

6.5.1. Spurious currents

A common problem in multiphase-flow simulations is the artificial velocity generated at the fluid interface due to errors in the curvature computation. To access the significance of such spurious currents, we test a stationary droplet of diameter $D = 0.4$ placed at the center of a unit box. The surface tension between the inner and outer fluid is $\sigma = 1$, the viscosity is uniformly $\mu = 0.1$, and the density ratio is 1. By changing the density ρ of both fluids, the Laplace number $La = \sigma\rho D/\mu^2$ can be varied. The spurious currents are thus determined from the resulting capillary number $Ca = |U_{max}|\mu/\sigma$ at a non-dimensional time $t\sigma/(\mu D) = 250$. Here, we compare the results on a 32×32 mesh with the GFM implementation by Desjardins *et al.* (2008). As listed in Table 2, the capillary numbers from both tests remain very small for all the Laplace numbers, with the present results being one-order smaller.

Table 2: Dependence of spurious current capillary number Ca on the Laplace number for a static droplet with surface tension on a 32×32 mesh in comparison with Desjardins *et al.* (2008).

La	12	120	1,200	12,000	120,000	1,200,000
Ca	2.85×10^{-6}	3.14×10^{-6}	3.63×10^{-6}	3.87×10^{-6}	3.41×10^{-6}	5.79×10^{-7}
Ca (ref)	4.54×10^{-5}	3.67×10^{-5}	3.62×10^{-5}	4.15×10^{-5}	3.75×10^{-5}	8.19×10^{-6}

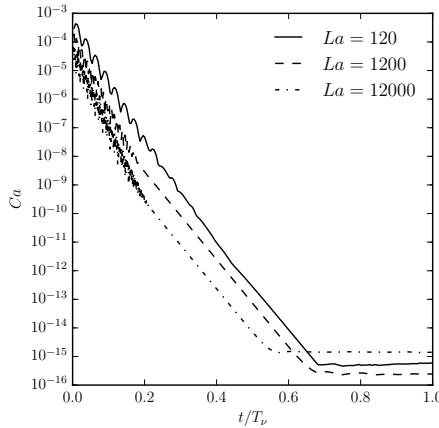


Figure 10: Temporal evolution of the spurious currents without performing level set reinitialization at three Laplace numbers as in Popinet (2009).

We also note that the spurious currents reported in Table 2 are obtained by performing the level set reinitialization at about every 100 time steps. However, if we turn off the reinitialization, such spurious velocity will eventually go to machine zero, as shown in Fig. 10, where time is non-dimensionalized with the viscous time scale, $T_\nu = \rho D^2 / \mu$. The nearly exponential decay of Ca and the collapsing of the three curves are the result of the viscous damping of the spurious velocity, as the shape of droplet relaxes to its numerical equilibrium. Similar results are obtained and explained in greater detail in Popinet (2009) using a balanced-force continuum-surface-force surface-tension formulation and the VOF. The result in Fig. 10 therefore validates the computation of the surface tension with the GFM.

6.5.2. Capillary wave

To verify the solver at large density and (dynamic) viscosity contrasts, we simulate a small-amplitude capillary wave for which there exists an analytical solution derived by Prosperetti (1981). Specifically, an initially sinusoidal

Table 3: Temporal and spatial convergence rates for the velocity component u and the pressure p .

	$L_2^{4\Delta t, 2\Delta t}$	$L_2^{2\Delta t, \Delta t}$	Rate	$L_2^{4\Delta x, 2\Delta x}$	$L_2^{2\Delta x, \Delta x}$	Rate
u	2.46×10^{-8}	1.03×10^{-8}	1.19	2.16×10^{-7}	5.95×10^{-8}	1.82
p	1.13×10^{-6}	3.85×10^{-7}	1.46	3.25×10^{-3}	6.11×10^{-4}	2.67

interface is imposed between two immiscible, viscous fluids of infinite depth and lateral extent. When the lower fluid is heavier, the balance between inertia, viscosity, and surface tension results in a decaying free-surface wave. By requiring matching kinematic viscosity $\nu_u = \nu_l$ (u for upper, l for lower), the solution of the wave amplitude in terms of Laplace transforms can be inverted analytically and compared with the simulation results.

We set up our simulation in the same way as suggested in Dodd & Ferrante (2014). Here, two fluids of equal depth are placed in a 1×3 (64×192 grid points) domain, where the streamwise direction ($L = 1$) is periodic and the vertical direction ($H = 3$) wall-bounded. The interface has an initial wavelength of $\lambda = 1$ and an amplitude of $a_0 = 0.01$. With varying density ratios ρ_l/ρ_u , the non-dimensional parameters for the test are

$$Re = 100, \quad We = 1, \quad Fr = \infty, \quad \rho_l/\rho_u = 10 - 10,000, \quad \nu_l = \nu_u. \quad (69)$$

The CFL number $\Delta t/\Delta x$ is 2.5×10^{-2} for $\rho_l/\rho_u = 10$ and 10^2 , and it is reduced to 2.5×10^{-3} for $\rho_l/\rho_u = 10^3$ and 2.5×10^{-4} for $\rho_l/\rho_u = 10^4$.

Fig. 11 shows the temporal evolution of the wave amplitude up to $t = 10$. The excellent agreement with Prosperetti's analytical solution (Prosperetti 1981) confirms the normal stress balance computed using the GFM. And accurate results at very large density contrasts are realized by combining the FastP* with GFM. Note that the dynamic viscosity ratio μ_l/μ_u also varies from 10 to 10^4 . However, neglecting its contribution to the pressure jump by regularizing the viscosity profile yields accurate results since the Capillary number is small ($Ca = We/Re = 0.01$), as discussed in conjunction with Eq. (46).

6.5.3. Convergence

We continue to check the temporal and spatial convergence rates of the coupled ICLS/NS solver. Here, the same test problem as in Sec. 6.5.2 is used, with the non-dimensional parameters given as

$$Re = 500, \quad We = 1, \quad Fr = \infty, \quad H_0 = 0.05, \quad \rho_l/\rho_u = 20, \quad \mu_l/\mu_u = 20, \quad (70)$$

again following Dodd & Ferrante (2014). Placing the fluids in a 1×1 box, the flow is simulated under different time steps or on different meshes so that the errors can be computed between successive solutions.

Table 3 shows the convergence rates for the velocity component u and the pressure p in the L_2 norm. Here, the temporal convergence is evaluated at

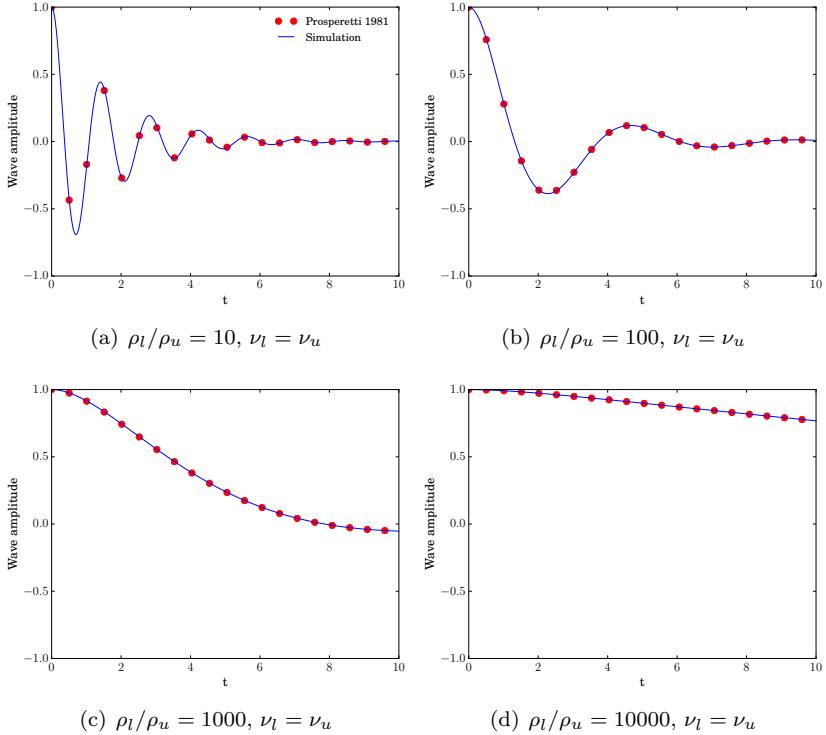


Figure 11: Time development of the capillary wave amplitude (normalized to a_0) for increasing density ratios and matching kinematic viscosity in comparison with Prosperetti's analytical solution (Prosperetti 1981).

$t = 6.25 \times 10^{-2}$ on a 256^2 grid, by increasing the time step from $\Delta t = 4.88 \times 10^{-5}$ to $2\Delta t$ and $4\Delta t$. Two iterations of reinitialization are performed every 25 – 100 time steps. The observed convergence rates for both velocity and pressure is between first and second order. Considering that we use RK3 for LS and AB2 for NS, the reduced convergence is probably due to the reinitialization that perturbs the interface. Changing the frequency of the reinitialization, we indeed observe different convergence rates (they can also exceed second order if the density ratio is 1, not shown). Next, the spatial convergence is obtained by successively refining the grid from 32^2 to 64^2 to 128^2 . Using the same time step $\Delta t = 4.88 \times 10^{-5}$ and interpolating the solution to the coarse grid after one solve, the results display nearly second order convergence for the velocity and a super-convergence for the pressure. We note that the GFM has been proven convergent (but without a rate) for variable-coefficient Poisson equations (Liu & Sideris 2003). Our results thus show improved accuracy in two fluid problems,

when a constant-coefficient Poisson equation is obtained by combining the GFM with the FastP*.

6.5.4. Rising bubble

Finally, we compute four cases of a rising bubble to access the overall accuracy of the current ICLS/NS solver in 3D in the presence of moderate deformations. Originally documented by Grace (1973), it was observed that a single gas bubble rising in quiescent liquid has four characteristic shapes: spherical, ellipsoidal, skirted, or dimpled. The governing non-dimensional numbers are the Morton number M , Eotvos number Eo (sometimes referred to as the Bond number), and the terminal Reynolds number Re_t , defined as

$$M = \frac{g\mu_l^4}{\rho_l\sigma^3}, \quad Eo = \frac{\Delta\rho gd^2}{\sigma}, \quad Re_t = \frac{\rho_l U_\infty d}{\mu_l}, \quad (71)$$

where d is the bubble diameter, $\Delta\rho$ is the density difference, U_∞ is the terminal velocity of the bubble, and the subscripts l and g denote, in order, the liquid and gas phase. The Morton and Eotvos number are defined purely by the material properties of the chosen fluids, while the terminal Reynolds number provides a measure of the steady-state bubble velocity.

Table 4 lists the four representative cases we select for the simulations. A spherical bubble of diameter $d = 1$ is centered in a domain of size $(L_x \times L_y \times L_z) = (3d \times 6d \times 3d)$. A grid of $96 \times 192 \times 96$ points is used, giving the bubble an initial resolution of 32 points per diameter. Periodic boundary conditions are imposed in the x (spanwise) and y (rising) directions whereas no friction, no penetration is enforced in the z direction. As suggested by Annaland *et al.* (2005), a ratio of 100 between the density and viscosity of liquid and gas is sufficiently high to approximate such gas-liquid systems, leading to $\Delta\rho \approx \rho_l$. Re and We in Eq. (3) can thus be obtained from M and Eo as

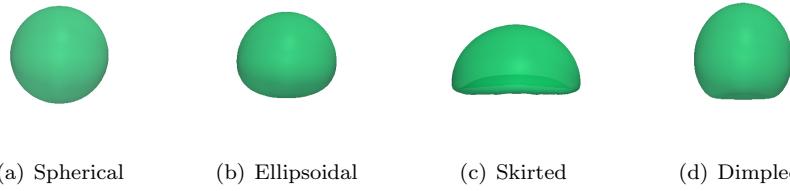
$$Re = \left(\frac{Eo^3}{M} \right)^{1/4}, \quad We = Eo. \quad (72)$$

The CFL number, $\Delta t / \Delta x$, is 1.6×10^{-4} for cases (a), (b), and (d), and 1.6×10^{-3} for case (c). The simulation is integrated in time up to $t = 10$ to ensure the bubble reaches nearly steady state.

The results of the bubble terminal velocities are presented in Table 4. The difference between the computed Reynolds, Re_C , and the terminal Reynolds, Re_G , measured by Grace (1973) remains small for all four cases. The bubble mass is conserved, with a maximal mass loss of about 0.02% found in the skirted case, where the bubble undergoes a large and rapid deformation. The corresponding bubble shapes are illustrated in Fig. 12, which clearly displays spherical, ellipsoidal, skirted, and dimpled shapes. We can therefore conclude that the dynamics of a single rising bubble is well-captured.

Table 4: Comparison of computed terminal Reynolds number (Re_C) and experimental terminal Reynolds number (Re_G) obtained from the Grace diagram (Grace 1973) under four different Morton (M) and Eotvos (Eo) numbers.

Case	Bubble regime	M	Eo	Re_G	Re_C	Mass loss (%)
(a)	Spherical	1×10^{-3}	1	1.7	1.73	9.86×10^{-5}
(b)	Ellipsoidal	0.1	10	4.6	4.57	3.32×10^{-4}
(c)	Skirted	1	100	20.0	19.21	1.64×10^{-2}
(d)	Dimpled	1000	100	1.5	1.71	3.28×10^{-3}



(a) Spherical (b) Ellipsoidal (c) Skirted (d) Dimpled

Figure 12: Bubble shapes resulting from different Morton (M) and Eotvos (Eo) numbers, as indicated in Table 4.

7. Droplet interactions

A unique feature of colloidal suspensions is the interaction between neighboring droplets, displaying fascinating behaviors such as self-assembly, self-replication, etc. . The reason for such interactions is rather complex; it often arises from a combination of fluid mechanical effects and physicochemical properties of the substance. To study the droplet interactions in the present ICLS/NS framework, we provide in this section a hydrodynamic model for the depletion forces. The method is a natural extension of the LS and GFM, and we demonstrate the clustering of droplets in various structures from a dumbbell to a face-centered cubic crystal.

7.1. Extension to multiple level set

The level set method discussed so far involves one marker function; we call it single level set (SLS) method. Thanks to its Eulerian nature, SLS can describe many droplets at the same time, provided that they do not need to be distinguished from each other. On the other hand, SLS can also be extended to multiple level set (MLS), so that each droplet has its own color function. This has several benefits including distinction and tracking of each droplet, independent curvature computation, and ability to prevent numerical

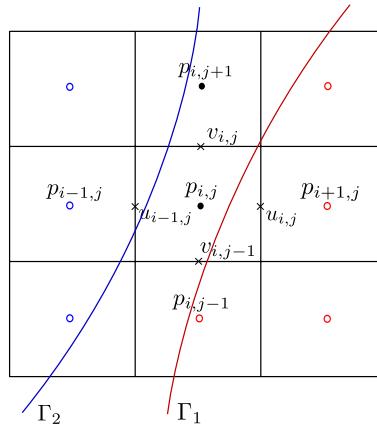


Figure 13: Pressure jump in the presence of multiple interfaces within two grid cells. Red and blue circles indicate nodal pressure in droplet 1 and 2, respectively. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

coalescence, etc. . Furthermore, with the narrow band approach (Adalsteinsson & Sethian 1995; Peng *et al.* 1999) and the various other techniques introduced in Sec. 1 (Nielsen & Museth 2006; Brun *et al.* 2012), the additional computational and memory cost as the number of the level set functions increases is limited.

The extension from SLS to MLS is straightforward. Assuming no droplets will overlap, each level set function is simply advected successively. When two droplets get close (typically within two grid cells, see Fig. 13), the pressure jump across each interface needs to be considered and superimposed. That is, Eq. (48) (corresponding to Fig. 9) should be modified as

$$\begin{aligned} (\nabla^2 p)_{i,j} = & \frac{p_{i-1,j} - 2p_{i,j} + p_{i+1,j}}{\Delta x^2} - 2 \frac{[p]_{i,j}}{\Delta x^2} - \frac{1}{\Delta x} \left[\frac{\partial p}{\partial x} \right]_{i+1/2,j} + \frac{1}{\Delta x} \left[\frac{\partial p}{\partial x} \right]_{i-1/2,j} \\ & + \frac{p_{i,j-1} - 2p_{i,j} + p_{i,j+1}}{\Delta y^2} - \frac{[p]_{i,j-1}}{\Delta y^2}, \end{aligned} \quad (73)$$

Similarly, all the jumps should be removed consistently when computing the pressure gradient in the subsequent step. The above modification applies to both SLS and MLS, as the compact formulas (Eqs. (62) and (63)) remain the same; although MLS is clearly more accurate in resolving the near field structure.

7.2. Near-field interactions

As introduced earlier, colloidal droplets transported in microfluidic devices are subject to various forces, a typical of which is the depletion force. The depletion force arises from the exclusion of the surfactant micelles in the

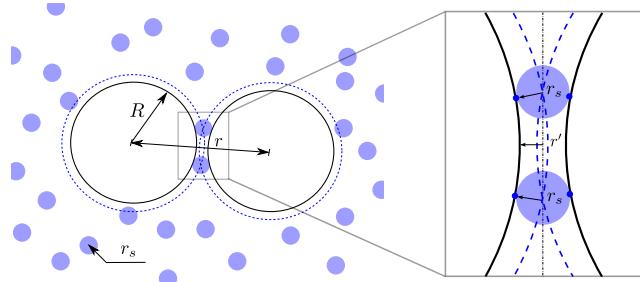


Figure 14: Depletion of surfactant micelles of radius r_s between larger colloidal droplets of radius R , separated by distance r . The dashed lines around larger spheres represent the region from which the centers of small spheres are excluded. They overlap when $r \leq 2R + 2r_s$. Inset: a zoom-in sketch of two droplets near contact.

colloidal suspension. It is often characterized as a near-field attracting potential (Asakura & Oosawa 1958; Mewis & Wagner 2012), and plays a key role in the droplet dynamics (Shen *et al.* 2016; Shen 2014). Below, we first provide a brief background on the colloidal theory of the depletion potential, then present a numerical model to enforce the depletion force using MLS and GFM.

7.2.1. The colloidal theory of the depletion potential

The original depletion potential model proposed by Asakura & Oosawa (1958) assumes the surfactant micelles as non-interacting hard-spheres. As sketched in Fig. 14, a suspension of such small spheres around the large colloidal droplets creates an osmotic pressure on the droplet surface. When the distance between two droplets is less than the diameter of the surfactant micelles, there will be a pressure defect due to the exclusion of the micelles, thus creating an attracting force. Integrating this force with respect to the inter-droplet distance r leads to a potential energy

$$U(r) = \begin{cases} \infty & \text{if } r \leq 2R \\ -p_{os}V_{ex} & \text{if } 2R < r \leq 2R + 2r_s \\ 0 & \text{otherwise,} \end{cases} \quad (74)$$

where V_{ex} is the excluded volume and p_{os} is the osmotic pressure. For spherical droplets, V_{ex} can be calculated analytically

$$V_{ex}(r) = \frac{4\pi(R + r_s)^3}{3} \left[1 - \frac{3r}{4(R + r_s)} + \frac{r^3}{16(R + r_s)^3} \right], \quad (75)$$

where R and r_s are, respectively, the radii of the big and small spheres. The osmotic pressure is given as

$$p_{os} = nkT, \quad (76)$$

where n is the number density of the small spheres, k is the Boltzmann constant, and T is the temperature. The negative sign in Eq. (74) corresponds to the tendency of the system to reduce its potential energy as the overlap increases. This is equivalent to increasing the total entropy of the small spheres (Melby *et al.* 2007), and it provides a physical description of the depletion force even when the droplets are deformable, or when p_{os} cannot be expressed by the van't Hoff's formula (Eq. (76)) (Asakura & Oosawa 1958).

7.2.2. A hydrodynamic model for the depletion force

Based on the above theory, the depletion force acting on a droplet is simply the derivative of the depletion potential, *i.e.* $F(r) = dU/dr = -p_{os}dV_{ex}/dr$. However, dV_{ex}/dr is not always straightforward to evaluate for non-spherical droplets; and unlike rigid-body dynamics, $F(r)$ cannot be applied directly to the motion of a liquid drop. In order to induce locally an aggregation, we take a closer look at the overlap region. As illustrated in Fig. 14, when the surface distance between two colloidal droplets is less than $2r_s$, there is a small area in which the osmotic pressure is subject to a jump. Assuming the concentration of the surfactant micelles changes abruptly, it resembles the jump of the Laplace pressure; however, it will not generate any flow if the pressure is uniform in the depleted region. On the contrary, if the osmotic pressure varies continuously within the overlap, *i.e.* $p' = p'(r')$, then we can write it as a Taylor-series expansion from $r' = r_s$

$$p'(r'/r_s) = p'(1) + \left(\frac{r'}{r_s} - 1 \right) \frac{\partial p'}{\partial r'/r_s}, \quad (77)$$

where the distance to the droplet surface r' is normalized by the surfactant micelle radius. An expansion of the osmotic pressure with the distance corresponds to a gradient of the micelle concentration near the gap. And if the micelle is much smaller than the droplet, as it is in many microfluidic devices (Shen *et al.* 2016), the gradient will be very sharp. Conversely, when the distance to the surface varies slowly, such as in the gap of a droplet and a flat wall, a uniform pressure will be recovered. Furthermore, a favorable pressure gradient from the overlap center will generate an outflow, pulling the droplets towards each other. Hence, Eq. (77) provides a hydrodynamic model for the depletion force.

In Eq. (77), the gradient of the osmotic pressure $\partial p'/\partial(r'/r_s)$ is not known *a priori*. It can be obtained by equating the depletion force acting on one droplet, *i.e.*

$$-p_{os}A_{ex} = \int_{\Omega} (p'(1) - p'(r'/r_s)) dS, \quad (78)$$

where A_{ex} is the effective area of the overlap Ω . Assuming a constant $\partial p'/\partial(r'/r_s)$, the above yields a linear dependence of the osmotic pressure on r' . Note that this is not the same as p' varying linearly with the distance to the overlap center (see Fig. 14). A description of the implementation and verification will be shown in the next section.

Algorithm 3: A pseudo code for computing the depletion force.

Enter the pressure solver. Compute the right-hand side of Eq. (62).

for $m = 1 : (N - 1)$ **do**

 Get the level set for droplet m , ϕ_m .

for $n = (m + 1) : N$ **do**

 Get the level set for droplet n , ϕ_n .

where $\phi_m < r_s$ and $\phi_n < r_s$ **do** $r' = (\phi_m + \phi_n)/2$, tag as
 overlap.

 Compute $[p']_\Omega$ from Eqs. (78) and (79) within *overlap*.

forall i, j, k **do**

if *entering overlap* **then**

 | Add the osmotic pressure jump $[p']_{i,j,k}$.

else

 | Remove the osmotic pressure jump $[p']_{i,j,k}$.

end

end

end

end

Solve for p^{n+1} regularly using the FastP* and GFM. Exit the pressure solver.

7.2.3. A MLS/GFM-based method for computing the depletion force

Provided a hydrodynamic model for the depletion force between two droplets, we can easily generalize it to multiple droplets using the MLS. Thanks to the distance information embedded in the level set functions, it is straightforward to identify the overlap region of arbitrary geometries. Furthermore, as the jump of the osmotic pressure occurs only across the overlap shell, we can define

$$[p']_\Omega = p'(r'/r_s) - p'(1), \quad (79)$$

similar to the Laplace pressure jump $[p]_\Gamma$ implemented by the GFM. Based on these observations, we propose a numerical method to compute the depletion force as laid out in Algorithm 3.

The overall idea of Algorithm 3 is to enforce the depletion attraction in the projection step through the use of MLS and GFM. Specifically, we first locate the overlap region of a pair of droplets with its own level set function, and define r' as the average of the two distances. Then, Eq. (78) can be integrated numerically to obtain $\partial p'/\partial(r'/r_s)$, which together with Eqs. (77) and (79) gives $[p']_\Omega$. This variable pressure jump manifests itself as a modification term on the right-hand side of Eq. (62), allowing us to use GFM to impose it across a sharp overlap shell. The resulting flow is divergence-free provided that all the jump terms are removed consistently in the correction step. Therefore, Eqs. (62) and

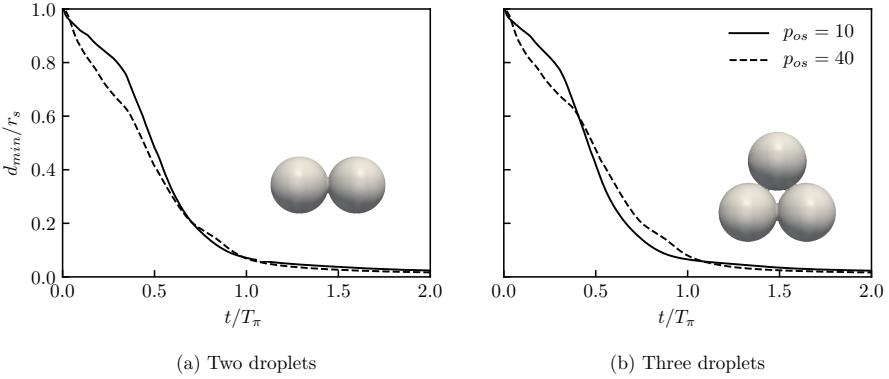


Figure 15: Minimal distance between the droplet surfaces as function of time in the presence of depletion forces proportional to $p_{os} = 10$ (solid line) and $p_{os} = 40$ (dashed line). Simulation of (a) two droplets and (b) three droplets suspended in an initially quiescent fluid. Due to symmetry, only the minimal distance is plotted.

(63) are re-formulated as ¹

$$\nabla^2 p^{n+1} = \nabla_g^2 ([p]_\Gamma + [p']_\Omega) + \nabla \cdot \left[\left(1 - \frac{\rho_0}{\rho^{n+1}} \right) \nabla_g \hat{p} \right] + \frac{\rho_0}{\Delta t} \nabla \cdot \mathbf{u}^*, \quad (80)$$

and

$$\mathbf{u}^{n+1} = \mathbf{u}^* - \Delta t \left[\frac{1}{\rho_0} \nabla_g p^{n+1} + \left(\frac{1}{\rho^{n+1}} - \frac{1}{\rho_0} \right) \nabla_g \hat{p} \right]. \quad (81)$$

Approaching drops. We verify the depletion force model and its numerical implementation by simulating 2 to 14 approaching droplets in a quiescent fluid environment. Specifically, we set the droplet radius $R = 0.5$, the computational domain $3 \times 3 \times 3$, and the resolution $\Delta x = 1/32$. The radius of the surfactant micelle is set to be $r_s = 1/16$, corresponding to $2\Delta x$. The viscosity and density ratios of the droplet to the ambient fluid are both 1. The non-dimensional parameters are $La = 2000$ and $Fr = \infty$, leading to a reference Laplace pressure jump $p_\sigma = 80$ and neglected gravity. The uniform osmotic pressure is either 10 or 40.

The temporal evolutions of the minimal surface distances in the case of two and three droplets are shown in Fig. 15. Here, time is scaled by a factor $T_\pi = (r_s/R)(p_\sigma/p_{os})$. The droplets, originally separated by a distance of r_s , get closer to the limit of the grid spacing at $t \approx T_\pi$. For the present study, we let the droplets aggregate without applying any repulsion models, except that

¹Eqs. (63) and (81) are identical in form; however, $[p']_\Omega$ has to be removed when evaluating $\nabla_g p^{n+1}$ and $\nabla_g \hat{p}$ in Eq. (81), as it is done in Eq. (61)

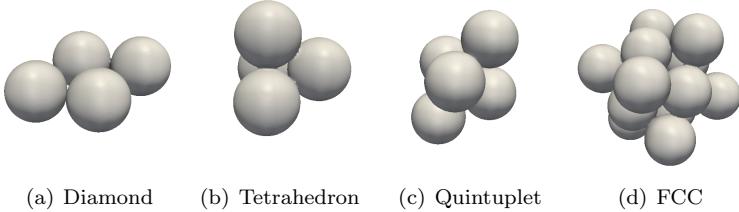


Figure 16: Examples of droplet clusters of different structures.

the magnitude of the osmotic pressure is reduced when $d_{min}/r_s < 0.1$. The smooth approaching in all cases and the collapse of the distance curve clearly evidence an attracting depletion force. To assess the robustness of the method, we further tested clustering of droplets into shapes from a 2D diamond to a face-centered cubic (FCC) composed of 14 drops, illustrated here in Fig. 16. FCC represents the unit structure of one of the most compact sphere packings. Therefore, we can conclude that the hydrodynamic model implemented by the MLS/GFM-based method is accurate and robust in computing the depletion forces.

8. Conclusion

A numerical method mainly intended for the hydrodynamic simulations of colloidal droplets in microfluidic devices has been developed and validated. The code is based on an efficient and sharp solver of the incompressible, two-fluid Navier-Stokes equations, and uses a mass-conserving level set method to capture the fluid interface. This combination provides a general framework for any multiphase flow problems (see e.g. our recent study on jet instabilities (Tammisola *et al.* 2017)), and allows us to develop specific methods for the simulations of droplets in saturated surfactant suspensions with depletion forces as in the recent experiment in Shen *et al.* (2016). Particularly, we have developed or extended four numerical techniques to improve the general accuracy:

1. A mass-conserving, interface-correction level set method (ICLS) is proposed. As a standalone level set module, it is efficient, accurate, guarantees global mass conservation, and is simple to implement. It also enables corrections that can depend on the local curvature or any other parameter of interest.
2. A geometric estimation of the interface curvature based on nodal curvatures is introduced. As an important ingredient both for the mass correction (ICLS) and the surface tension computation, we show that the calculation converges in second-order both in 2D and 3D, and can lead to machine-zero spurious currents for a stationary 2D droplet.

3. The ghost fluid method (GFM) for the computation of surface tension is combined with the FastP* method (Dodd & Ferrante 2014). This enables the use of FFT-based solvers for a direct pressure solve, and can accurately account for surface tension at large density ratios.
4. A ghost fluid/multiple level set (GFM/MLS-based) method is also proposed to compute the interaction force caused by depletion potentials between multiple droplets or between droplets and a nearby wall. The approach can possibly be extended to account for surfactant diffusion at the interface and in the liquid.

The last technique applies specifically to the simulation of colloidal droplets in microfluidic devices. This will enable us to further explore the effects of the near-field interactions as those observed experimentally in Shen *et al.* (2016), and potentially improve the design of microfluidic devices. In addition, the combination of the GFM for sharp interfaces and the FastP* method (Dodd & Ferrante 2014) can be exploited for the simulations of droplet in turbulent flows as in Dodd & Ferrante (2016), adding an accurate representation of evaporation thanks to the ICLS approach proposed here.

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Appendix A. Discretization error of $\int_{\Gamma} \mathbf{n} \cdot \mathbf{u}_c d\Gamma$

Similar to Engquist *et al.* (2005), we define the discretization error

$$E = \left| \left(\prod_{k=1}^d \Delta x_k \right) \sum_{j \in Z^d} \hat{\delta}_{\epsilon}(\Gamma, g, \mathbf{x}_j) - \int_{\Gamma} \mathbf{n} \cdot \mathbf{u}_c d\Gamma \right|, \quad (82)$$

where $\hat{\delta}_{\epsilon}$ is a Dirac delta function of variable strength g supported on the surface Γ , and $\mathbf{x} \in \mathbb{R}^d$. Following the derivations in Sec. 4, the extension of g to \mathbb{R}^d is provided by Eq. (22), allowing one to write

$$E = \left| \left(\prod_{k=1}^d \Delta x_k \right) \sum_{j \in Z^d} \frac{\delta V}{\delta t} \frac{f_s \delta_{\epsilon}(\phi(\mathbf{x}_j)) |\nabla \phi(\mathbf{x}_j)|}{A_f} - \int_{\Gamma} \mathbf{n} \cdot \mathbf{u}_c d\Gamma \right|. \quad (83)$$

Here, $\delta_{\epsilon}(\phi)$ is a one dimensional regularized delta function depending on the level set ϕ , and the expression is simplified noting that $\mathbf{n} \cdot \nabla \phi = |\nabla \phi|$ (it does

not have to be a distance function). By definition, $A_f = \int_{\Gamma} f_s \delta_{\epsilon}(\phi) |\nabla \phi| d\Gamma$, discretely reducing Eq. (83) to

$$E = \left| \frac{\delta V}{\delta t} - \int_{\Gamma} \mathbf{n} \cdot \mathbf{u}_c d\Gamma \right|. \quad (84)$$

Comparing with Eq. (13), it is obvious that $E = 0$. That is, the discretization error of $\int_{\Gamma} \mathbf{n} \cdot \mathbf{u}_c d\Gamma$ used in the mass correction is identically zero, independent of the choice of the regularized delta function.

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2

Paper 2

Flow-assisted droplet assembly in a 3D microfluidic channel

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Self-assembly of soft matter, such as droplets or colloids, has become a promising scheme to engineer novel materials, model living matter, and explore non-equilibrium statistical mechanics. In this article, we present detailed numerical simulations of few non-Brownian droplets in various flow conditions, specifically, focusing on their self-assembly within a short distance in a three-dimensional (*3D*) microfluidic channel, cf. Shen *et al.* (2016). Contrary to quasi two-dimensional (*q2D*) systems, where dipolar interaction is the key mechanism for droplet rearrangement, droplets in *3D* confinement produce much less disturbance to the underlying flow, thus experiencing weaker dipolar interactions. Using confined simple shear and Poiseuille flows as reference flows, we show that the droplet dynamics is mostly affected by the shear-induced cross-stream migration, which favors chain structures if the droplets are under an attractive depletion force. For more compact clusters, such as three droplets in a triangular shape, our results suggest that an inhomogeneous cross-sectional inflow profile is further required. Overall, the accelerated self-assembly of a small-size droplet cluster results from the combined effects of strong depletion forces, confinement-mediated shear alignments, and fine-tuned inflow conditions. The deterministic nature of the flow-assisted self-assembly implies the possibility of large throughputs, though calibration of all different effects to directly produce large droplet crystals is generally difficult.

1. Introduction

Recent advances in colloidal science have generated a growing interest in the fabrication of functional materials, especially those possessing photonic band gaps (Ho *et al.* 1990; Subramanian *et al.* 1999; Seelig *et al.* 2003; Wong *et al.* 2003; Shen *et al.* 2016b). Colloids – particles or droplets of (sub-)micron scale – are manipulated with high precision to self-organize into controlled patterns, which then form a library of basic building blocks for more complex structures (Sacanna & Pine 2011). Conceivably, such direct assembly is also the most efficient mechanism for material synthesis. Colloidal self-assembly has thus become a promising scheme to engineer novel materials, mimicking the machinery of nature (van Blaaderen 2003).

Currently, there are many strategies to synthesize colloidal building blocks, e.g. creating a short-range depletion potential in a microwell (Meng *et al.* 2010), exploiting shape/surface anisotropy of the colloids (Sacanna & Pine 2011; Evers *et al.* 2016), using patterned substrates as templates (Yin *et al.* 2001), or even jamming droplets with a micro-mixer (Ricouvier *et al.* 2017). Among these, *flow-assisted* self-assembly seems especially appealing, since the microfluidic droplets are driven by an external flow rather than Brownian motions, reducing the range of assembly time from days down to seconds (c.f. the experiment of (McMullen *et al.* 2018) as an example). With the potential of being optimized and applied in parallel, microfluidics appears to be the fastest pathway towards photonic material generation.

Following this approach, Shen *et al.* (2016b) recently demonstrated the generation and self-assembly of droplet strings into a rich variety of (non-)compact structures, including chains, triangles, diamonds, crosses, etc. . in a simple microfluidic device (see Fig. 1). Unlike most previous studies (Cui *et al.* 2004; Beatus *et al.* 2006; Janssen *et al.* 2012; Uspal *et al.* 2013; Desreumaux *et al.* 2013; Zhu & Gallaire 2016), where droplets under strong confinement interact via dipolar flows (Beatus *et al.* 2017; Diamant 2009), these droplets are smaller than the smallest dimension of the microfluidic channel. Specifically, the droplets are tens to hundreds microns in diameter, while the channel height is about three times larger. As a consequence, typical quasi two-dimensional ($q2D$) arguments do not apply, and the dynamics is fully three-dimensional ($3D$). In the latter case, the complete Navier-Stokes (or Stokes) equations have to be solved to obtain a correct physical understanding of the system,¹ wherein prior simulations mainly focus on the deformation and migration of a single drop, or an evenly spaced array of drops (Couliette & Pozrikidis 1998; Griggs *et al.* 2007; Janssen & Anderson 2007). Interestingly, assuming a tunable far-field dipolar interaction, Shen *et al.* (2016b) could however model the droplet motions up to a “semiquantitative” level, reproducing the sophisticated self-assembly observed in experiments. This apparent contradiction motivates us to pursue a detailed numerical study of the relatively fast droplet self-assembly in order to investigate the interactions in their microfluidic channel.

In the following, we first present a numerical methodology for the dynamics of two viscous fluids, including a hydrodynamic model for the near-field depletion force, a localized attractive force between suspending droplets. Applying this methodology, we then show results of extensive simulations of two to ten droplets in quiescent, shear-, and pressure-driven channel flows, each of which isolating an individual effect contributing to the self-assembly. Here, the focus is on the clustering and droplet interactions within a short distance from their initial release rather than the production of the droplets, which has been studied previously Chakraborty *et al.* (2017). Our aim is to elucidate the physical

¹A notable exception is the dynamics of two droplets in unbounded linear flows solved analytically by Batchelor & Green (1972a).

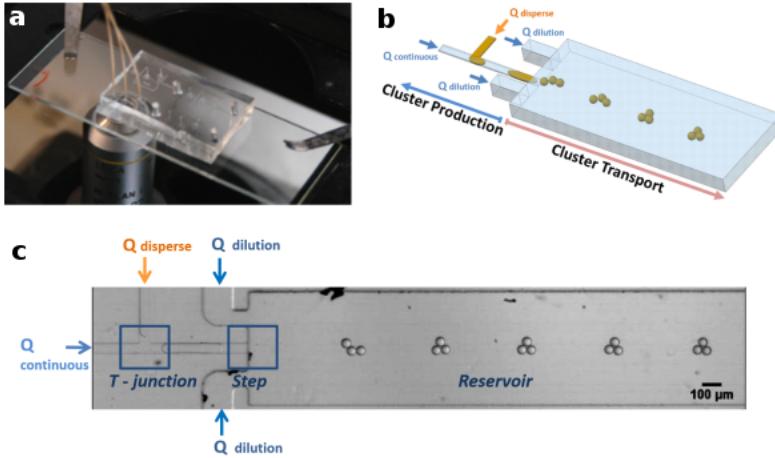


Figure 1: (a) Photo of a microfluidic channel placed above a desktop microscope. (b) Schematic of the channel geometry and generation of the droplet clusters. The main channel has a typical dimension of ($50 \mu\text{m} \times 600 \mu\text{m} \times 5000 \mu\text{m}$) in height, width, and length; hence, it can be considered as a Hele-Shaw cell. (c) Bottom view of the droplet self-assembly observed from the microscope. Pictures courtesy of Dr. Joshua Ricouvier. For more details, see Shen *et al.* (2016b).

picture of the hydrodynamic interactions, potentially improving the design of more efficient microfluidic systems.

2. Models and methods

2.1. Hydrodynamic model

The dynamics of two immiscible, Newtonian fluids in incompressible flows is governed by the Navier-Stokes equations

$$\nabla \cdot \mathbf{u} = 0, \quad (1a)$$

$$\frac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{u} = \frac{1}{\rho_i Re} \left(-\nabla p + \nabla \cdot [\mu_i (\nabla \mathbf{u} + \nabla \mathbf{u}^T)] \right) + \frac{\mathbf{g}}{Fr}, \quad (1b)$$

where \mathbf{u} , p , \mathbf{g} , ρ_i and μ_i denote the non-dimensional velocity, pressure, unit gravitational vector, density, and dynamic viscosity, respectively. Eq. (1b) is written for each fluid component, i ($= 1$ for the carrier fluid, 2 for the droplet), requiring a pressure boundary condition across the fluid interface

$$p_2 - p_1 = \frac{\kappa}{Ca}, \quad (2)$$

with κ being the mean curvature (the contribution due to the viscosity difference is neglected assuming matching viscosity (Batchelor 1967)).

So far, we have introduced three non-dimensional numbers: Re , Ca , and Fr , denoting the Reynolds, capillary, and Froude numbers, separately. Choosing fluid 1 as the reference phase, they are defined as

$$Re = \frac{\tilde{\rho}_1 \tilde{U} \tilde{L}}{\tilde{\mu}_1}, \quad Ca = \frac{\tilde{\mu}_1 \tilde{U}}{\tilde{\sigma}}, \quad Fr = \frac{\tilde{U}^2}{\tilde{g} \tilde{L}}, \quad (3)$$

where \tilde{U} , \tilde{L} , $\tilde{\rho}_1$, $\tilde{\mu}_1$, $\tilde{\sigma}$, and \tilde{g} denote the reference *dimensional* velocity, length, density, dynamic viscosity, surface tension, and gravitational acceleration. Following the experiments in Shen *et al.* (2016b), typical values of the reference velocity and length are $\tilde{U} \sim 100 \text{ } \mu\text{m/s}$ and $\tilde{L} \sim 100 \text{ } \mu\text{m}$, leading to $Re \sim 10^{-2}$, $Ca \sim 10^{-5}$, and $Fr \sim 10^{-5}$ for oil-water systems. Therefore, inertial (but not gravitational) effects are negligible and the droplets shall remain mostly spherical. In the simulations that we are going to present, however, these numbers are enlarged within reasonable physical limits also to reduce the computation time. Specifically, Re has been increased up to 10 in most cases except those in Sec. 3.2 (where $Re = 1$) and those in Sec. 3.3.2 (where $Re = 0.1$); Ca is in the order of $10^{-3} \sim 10^{-2}$ depending on the Re ; whereas $Fr = 0.025$ if gravity is enabled (otherwise ∞). In general, the Reynolds number cannot be arbitrarily increased as it is proportional to the fluid inertia. The reason we *can* increase it here is essentially due to the uniformity of the underlying flow. In this case, the mere effect of further reducing the Re is stretching the time scale, making the computations significantly longer. We have tested all cases at smaller Re 's to ensure that the differences are negligible in the case of fully developed Couette and Poiseuille flows. This is discussed in more details in Appendix A.

Finally, we note that the surface tension can also vary due to temperature or surfactant concentration gradients, leading to Marangoni stress along the surface. The detailed chemistry is rather complex Eastoe & Dalton (2000); in the present paper, we assume constant and uniform surface tension $\tilde{\sigma}$ to reduce the number of the governing parameters.

2.2. Depletion theory

Droplets suspended in an ambient fluid dissolved by surfactant molecules typically experience an attractive depletion force. The first model to describe such interaction was proposed by Asakura & Oosawa (1958), who assumed the surfactant micelles to be non-interacting hard spheres. As sketched in Fig. 2, a suspension of such small spheres around the large colloidal droplets creates an osmotic pressure on the droplet surface. When the distance between two droplets is less than the diameter of the surfactant micelles, the pressure defect due to the exclusion of the micelles pulls the drops even closer, corresponding to an attractive force. Integrating this force with respect to the inter-droplet distance r leads to a potential energy

$$U(r) = \begin{cases} \infty & \text{if } r \leqslant 2R \\ -p_{os} V_{ex} & \text{if } 2R < r \leqslant 2R + 2r_s \\ 0 & \text{otherwise,} \end{cases} \quad (4)$$

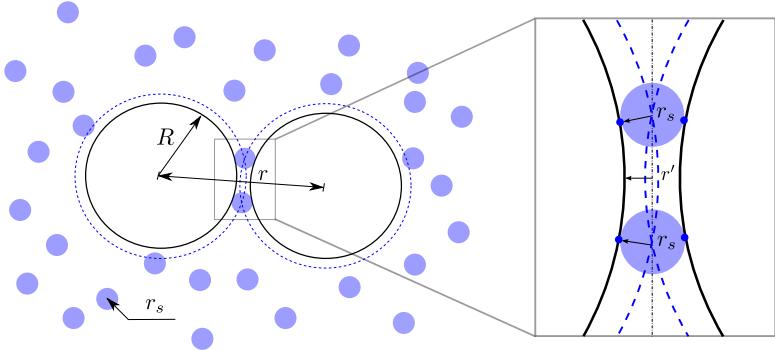


Figure 2: Depletion of surfactant micelles of radius r_s between larger colloidal droplets of radius R , separated by distance r . The dashed lines around larger spheres represent the region from which the centers of small spheres are excluded. They overlap when $r \leq 2R + 2r_s$. Inset: a zoom-in sketch of two droplets near contact.

where V_{ex} is the excluded volume and p_{os} is the osmotic pressure. For spherical droplets, V_{ex} can be calculated analytically

$$V_{ex}(r) = \frac{4\pi(R + r_s)^3}{3} \left[1 - \frac{3r}{4(R + r_s)} + \frac{r^3}{16(R + r_s)^3} \right], \quad (5)$$

where R and r_s are, respectively, the radii of the big and small spheres. The osmotic pressure is given as

$$p_{os} = nkT, \quad (6)$$

where n is the number density of the small spheres, k the Boltzmann constant, and T the temperature. The negative sign in Eq. (4) corresponds to the tendency of the system to reduce its potential energy as the overlap increases. This is equivalent to increasing the total entropy of the small spheres (Melby *et al.* 2007), and it provides a physical description of the depletion force even when the droplets are deformable, or when p_{os} cannot be expressed by the van't Hoff's formula (Eq. (6)) (Asakura & Oosawa 1958).

Based on the above theory, we consider the depletion potential in the hydrodynamic model by adding an osmotic pressure, $p'(r')$, near the overlap region (r' being the normalized distance to the droplet surface, see Fig. 2). Specifically, we write p' as a Taylor-series expansion from $r' = r_s$

$$p'(r'/r_s) = p'(1) + \left(\frac{r'}{r_s} - 1 \right) \frac{\partial p'}{\partial r'/r_s}, \quad (7)$$



Figure 3: (left) Two droplets approaching in a quiescent flow, and (right) close-up of the flows in the gap due to the depletion force.

with a constant $\partial p'/\partial(r'/r_s)$ satisfying the original depletion force acting on one droplet, i.e.

$$-p_{os}A_{ex} = \int_{\Omega} (p'(1) - p'(r'/r_s))dS, \quad (8)$$

where A_{ex} is the effective area of the overlap Ω . Note that, in this formulation, we do not require p_{os} to be a thermodynamic pressure; instead, its strength can be defined by a non-dimensional number

$$\Pi = \frac{p_{os}}{\kappa/Ca}, \quad (9)$$

which normalizes p_{os} by the Laplace pressure due to surface tension. Doing so, the osmotic pressure varies continuously within the overlap and depends linearly on r' . An expansion of the osmotic pressure with the distance corresponds to a gradient of the micelle concentration near the gap; and if the micelle is much smaller than the droplet, as it is in the microfluidic device of interest (Shen *et al.* 2016*b*), the gradient will be very sharp. Conversely, when the distance to the surface varies slowly, such as in the gap of a squeezed droplet and a flat wall, a uniform pressure will be recovered. In essence, what we propose here is a method to model the osmotic pressure as an equivalent mechanical pressure such that a favorable pressure gradient from the center of the overlap region generates an outflow, pulling the droplets towards each other. This is clearly illustrated in Fig. 3, where two droplets approach each other due to the locally induced depletion flow.

2.3. Numerical methods

The above governing equations are solved numerically using the interface-correction level set/ghost fluid method (ICLS/GFM) (Ge *et al.* 2018). The equations are discretized in space by the finite volume method, and integrated in time using the second-order Adam-Bashforth scheme. Combining several computational techniques, including the fast pressure correction method (Dodd & Ferrante 2014), the GFM (Fedkiw *et al.* 1999), and the fast Fourier transform (FFT) (Schumann & Sweet 1988), discontinuous quantities are treated sharply

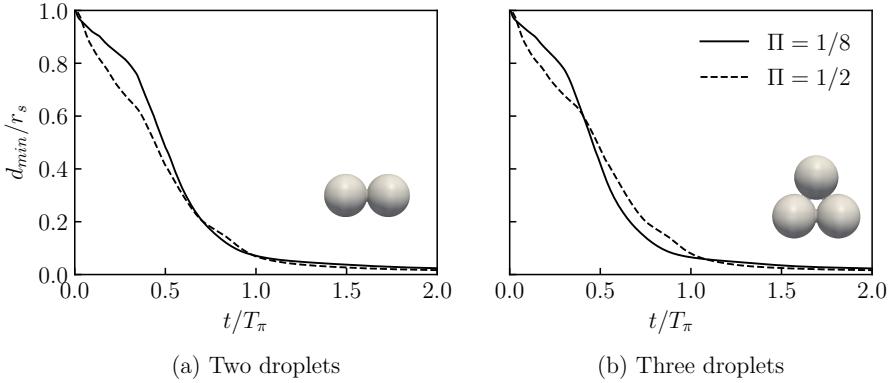


Figure 4: Minimal distance between the droplet surfaces as function of time in the presence of depletion forces proportional to $\Pi = 1/8$ (solid line) and $\Pi = 1/2$ (dashed line). Simulation of (a) two droplets and (b) three droplets suspended in an initially quiescent fluid. Due to symmetry, only the minimal distance is plotted.

at high efficiency. For the detailed algorithm and validations, we refer to Ge *et al.* (2018). The source code is also publicly available on GitHub (Ge 2018).

3. Results

In the following, we present simulation results of droplet motions in three types of flows: quiescent, shear-, and pressure-driven channel flows. In the last case, both Poiseuille flows and a non-uniform channel flow are considered. The purpose of using different flow types is to disentangle the effects of the depletion force, the droplet-droplet hydrodynamic interaction, and the droplet-flow interaction. The simpler cases can be seen as model problems towards understanding of the more complex motions of droplet ensembles in realistic microfluidic devices.

3.1. Approaching droplets in quiescent flows

3.1.1. 2 and 3 droplets

The simplest case of droplet self-assembly is identical drops approaching in quiescent flows, i.e. no external flow motions. In such a case, the remaining fluid parameters can be grouped into two non-dimensional numbers, i) the Laplace number $La = \tilde{\sigma} \tilde{\rho}_1 (2\tilde{R}) / \mu_1^2$, where \tilde{R} is the droplet radius; and ii) the osmotic-to-Laplace pressure ratio $\Pi = p_{os}/p$.³ La relates surface tension to

³The density and viscosity ratios between the drop and the carrier fluids are assumed to be unity. See Appendix A for the detailed numerical setup.

the viscous stress, which can also be expressed as $La = Re/Ca$. Π indicates the magnitude of the uniform osmotic pressure p_{os} due to depletion of the surfactant micelles, scaled by a reference Laplace pressure p due to surface tension (cf. Eq. (9)). In the following, we assume $La = 2000$ and $\Pi = 1/8$ or $1/2$, corresponding to the limit $Ca \ll Re \ll 1$ and conditions above the critical micelle concentrations (CMC, see dimensional analysis below) as in the experiments (Shen *et al.* 2016*b*).

The approach of two and three droplets is illustrated in Fig. 4, where the minimal distance between the droplet surfaces d_{min} , normalized by the surfactant micelle radius r_s , is shown as a function of time. Here, time is re-scaled by the factor $T_\pi = \tilde{r}_s/(\tilde{R}\Pi)$ to account for the size contrast of the droplet and the surfactant micelle, thus indicating an inverse scaling of the approaching time with the osmotic pressure for inertialess droplets, i.e. $T \propto \Pi^{-1}$. Indeed, for both $\Pi = 1/8$ and $1/2$, our results show that d_{min} approaches the limit of the grid spacing at $t \approx T_\pi$. The smooth approach in both cases and the collapse of the distance curves thus verify our modeling of the near field chemical interaction, consistent with an attracting depletion force.

We note that T_π is not a physical time scale (it is dimensionless). One possible definition for the depletion time scale is $\tilde{\tau}_\pi = \tilde{r}_s \tilde{\mu}_1 / (\tilde{R} \tilde{p}_{os})$, which can be rewritten as $T_\pi Ca \tilde{\tau}$, with a convection time scale $\tilde{\tau} = 2\tilde{R}/\tilde{U}$ in the range of 0.1 to 1 s typically. By substitution of usual values of colloidal systems, e.g. $\tilde{r}_s = 1$ nm, $\tilde{\mu}_1 = 10^{-3}$ kg/m-s, $\tilde{R} = 10$ μ m, and $\tilde{p}_{os} = 100$ Pa (corresponding to surfactant micelles concentration of 5 CMC, see Shen (2014), p112), the estimated time scale is 1 ns. Although $\tilde{\tau}_\pi$ can be amplified by increasing the viscosity of the suspending fluid or reducing the micelles concentration, its magnitude is so small that the approaching can be considered instantaneous. Therefore, in practical microfluidic devices such as those in Shen *et al.* (2016*b*), one cannot expect to detect the dynamical approaching process due to depletion forces. The droplets will appear either bound or separated, depending on the surfactant concentration and flow conditions.

3.1.2. 4 to 10 droplets

To further demonstrate the effect of the depletion force, we “virtually” assemble four to ten droplets under various initial configurations to form stable clusters as illustrated in Fig. 5. These clusters can be either 2D or 3D, exhibiting different levels/kinds of symmetry. In our simulations, the shape of the cluster is solely determined by the initial droplet arrangement, in the absence of any disturbance or other driving forces. Permitting disturbances, such as vibrations or thermal noises, would eventually lead to the formation of “rigid clusters”, i.e. clusters that cannot be reshaped by a small amount of inter-droplet displacement. Analytically, the number of possible rigid clusters grows rapidly with the number of droplets (N). For example, there is only one possible rigid cluster for $N = 4$, while there are 259 possibilities for $N = 10$, for packing of $3N - 6$ contacts

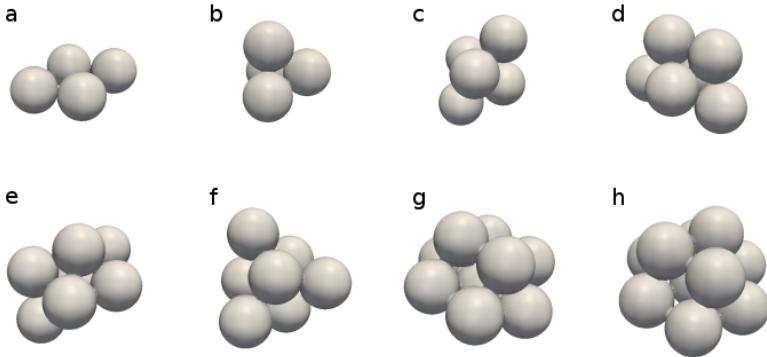


Figure 5: Packing of N droplets due to the near field depletion force. (a-b) $N = 4$, (c) $N = 5$, (d) $N = 6$, (e) $N = 7$, (f) $N = 8$, (g) $N = 9$, (h) $N = 10$.

(Holmes-Cerfon 2016). For brevity, we only illustrate two examples for $N = 4$ (including one planar cluster) and one example for $N = 5$ to 10 in Fig. 5.

We remind that the specific coordinates of the sphere packings bear no more significance than other possibilities in our simulations. They are arbitrarily chosen to illustrate the self-assembly due to the near field attraction. This is not the case for trapped equilibrium clusters, where less symmetric geometries are found to be favored by the entropic depletion force (Meng *et al.* 2010; Klein *et al.* 2018). Nor is it similar to colloidal particles interacting via short-range attractive, long-range repulsive potentials, where complex phase transitions emerge depending on the competition of the interactions (Mani *et al.* 2014; Das *et al.* 2018). Here, the self-assembly is microfluidic-based, driven by the hydrodynamics rather than the minimization of free energy over long periods. We examine the effect of the flow next.

3.2. Sticky droplets in shear-driven channel flows

When the droplets are carried by an external flow, their interactions are undoubtedly affected by the flow conditions, droplet-flow interactions and flow-induced droplet-droplet interactions (Fouxon *et al.* 2017). To study these additional effects, we consider an elementary flow field, the wall-bounded simple shear flow, defined as $(u, v, w) = (0, \dot{\gamma}z, 0)$ for $z \in [-L_z/2, L_z/2]$. Here, v is the only non-zero velocity component, its magnitude varies linearly with the z coordinate, and $\dot{\gamma}$ is the shear rate (see Fig. 6a). The presence of droplets will locally modify this flow field, which we sustain by enforcing opposite motions of two moving plates at $z = \pm L_z/2$.

Dating back to Taylor (1934), the deformation and motion of single or multiple droplet(s)/particle(s) have been studied extensively in simple shear flows (Lin *et al.* 1970; Batchelor & Green 1972a,b; Zinchenko 1983, 1984;

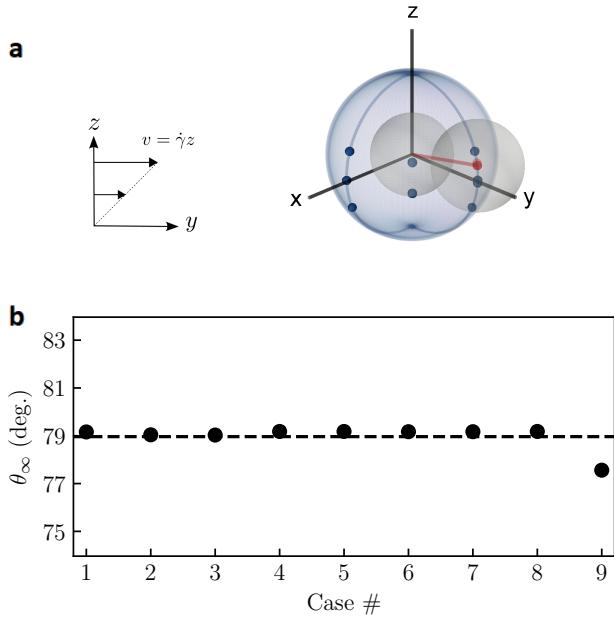


Figure 6: Stable configuration of a droplet pair in the simple shear flow. (a) Spherical diagram of the initial (blue dots) and final (red dots) positions of the second droplet in the reference frame of the first droplet. The blue arcs mark the border of the first quadrant where nine initial positions are considered. The undisturbed flow is a simple shear in the yz plane. (b) Steady-state polar angle, θ_∞ , corresponding to the cases in Table 1. The average value is 79 deg. (dashed line). The stable azimuthal angle is identically 0.

Zurita-Gotor *et al.* 2007). In the case of spherical particles/droplets, previously identified interaction modes include closed trajectories (particles rotate around each other in the vorticity plane), open-and-symmetric trajectories (particles return to their original z positions after passing each other), and swapping trajectories (particles exchange the z position after a binary encounter). The first two modes are generic features of a dilute suspension of particles or non-deforming/non-coalescing droplets, while the last mode arises when the particles are under relatively large geometric confinement. Conceivably, adding a near-field depletion force shall not alter these three modes as it is only activated at nearly touching, when the particles are already in a bound pair. What is yet to be explored, however, is when the gap between the two confining plates is smaller than the sum of the particle diameters (i.e. $L_z/D < 2$), thus disabling the occurrence of the Batchelor-Green type of closed orbit.

Table 1: Initial polar (θ) and azimuthal (ϕ) angles of the second droplet in the reference frame of the first droplet. θ and ϕ are measured from the z - and x -axes, respectively (see Fig. 6(a)).

Case #	1	2	3	4	5	6	7	8	9
θ (deg.)	70	70	70	89	89	89	91	110	110
ϕ (deg.)	90	45	0	90	45	0	90	90	0

Fig. 6(a) illustrates various initial conditions of two touching droplets, corresponding to the nine cases listed in Table 1, at $L_z/D = 1.5$. Specifically, the initial positions of the second droplet is given in the spherical coordinates centered at the first one, where all cases are located in one quadrant-sphere as we do not distinguish between the two droplets (the rest are equivalent due to symmetry). Contrary to the less confined conditions where a bound pair would rotate indefinitely relative to each other under shear (Batchelor & Green 1972a; Zinchenko 1984), Fig. 6 shows that the droplets tend to reside in the vorticity plane (i.e. the yz plane) with a stable polar angle $\theta_\infty \approx 79$ deg. This is true even if the two droplets are not initially in the same vorticity plane (case 2,3,5,6,9), or if the second droplet is in the lower hemisphere relative to the first one (case 7,8,9). Particularly, in cases 8 and 9, the droplet pair first rotates clockwise, then slides along the wall, before finally reaching the stable orientation (see ESI Video 1). The anomalous trajectory is a clear evidence of the influence of the walls, which, together with the attractive depletion force, break the symmetry of the droplet binary interactions.

The above results suggest that, for two droplets subject to an attractive depletion force in strongly confined simple shear flows, only one configuration is dynamically stable. It further implies that, for multiple droplets ($N > 2$) traveling in a pressure-driven channel with $H/D < 4$ (H being the channel height, see Sec. 3.3), a chain-like structure oriented in the flow direction is expected. We remark that the precise configuration of the droplet cluster may depend on the flow conditions and the level of confinement (see Appendix B for further discussion); however, the *qualitative* picture of the pairwise interaction shall remain unchanged, provided that the number of contact is $N - 1$. As this is often the case before a compact cluster is formed, we proceed to examine the droplet self-assembly in pressure-driven flows.

3.3. Droplet clusters in pressure-driven channel flows

In the following, clusters of three or four droplets are initialized to be in contact ($N - 1$ contacts for N droplets) and are released into different regions of a microfluidic channel to study their transport behavior. The production of these droplets is omitted, as the step-emulsifier is typically much smaller than the size of the channel, allowing for separation of the two processes (Shen 2014; Chakraborty *et al.* 2017). We note that, although the droplets are already in a

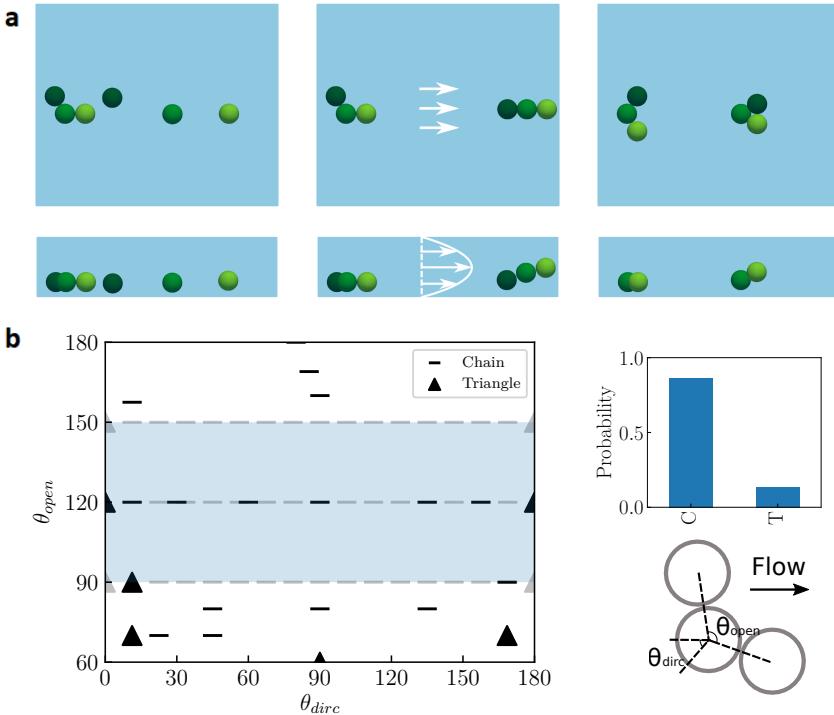


Figure 7: Three droplets in the Poiseuille flow. (a) Top (upper panels) and side (lower panels) views of the droplet positions. The droplets are colored differently only for visualization purposes. Left: without any depletion force (scattered droplets). Middle: with depletion force (forming a chain). Right: with depletion force (forming a triangle due to a different initial configuration). (b) Phase diagram showing the final configuration of three droplets under depletion force. θ_{open} and θ_{dirc} denote the initial opening and the direction angles (see inset for illustration). Interpolating the results for $\theta_{open} \in [90, 150]$ (shaded region), chain is clearly the predominant structure as shown in the probability distribution.

cluster initially, their relative rearrangement is still important as it determines the cluster morphology in the final state. The latter results primarily from the droplet-flow interaction and has direct consequence on the photonic properties of the droplet lattice, as we will discuss in details below.

3.3.1. Uniform region (the Poiseuille flow)

First, we consider droplet clusters in a Poiseuille flow; that is, we place the droplets in a channel whose undisturbed velocity is given as $(u, v, w) = (0, 6z/H(1 - z/H), 0)$, with the channel height $H = 3D$. As in the simple

shear flow, the droplets are neutrally buoyant, and x , y , z denote the spanwise, streamwise, and wall-normal directions, respectively. Enforcing periodic boundary conditions in both x and y directions, the flow can be computed efficiently using FFT, approximating the flow field far from the edges of the Hele-Shaw channel (see Fig. 1b).

As a control case, we simulate three droplets initially located near the bottom of the channel without any depletion forces. This is shown in the first column of Fig. 7a, where the top and bottom panels illustrate the top and side views of the channel (see Fig. 1). The two snapshots are separated by 13.5 convection time units (i.e. $\tilde{\tau}$). Clearly, the droplets quickly scatter as carried by the flow. In a previous work, we theoretically predicted the emergence of singlets and pairs of a dilute particle suspension due to weak particle-particle interactions (Fouxon *et al.* 2017). This example illustrates the separation of a droplet cluster, enhanced by their initial proximity already at $N = 3$, supporting our theoretical predictions.

In contrast, when the droplets are bound by a strong depletion force, the same initial condition can lead to a chain structure oriented in the flow direction, see the second column of Fig. 7a. Inspection of the side view reveals the apparent reason: the leading droplets migrate towards the middle of the channel due to the shear, thus experiencing faster flows; the attractive depletion force prevents the cluster from separating into singlets and pairs, yielding the eventual droplet string parallel to the stream. We propose that this shear-induced alignment mechanism is fundamentally due to the confinement-mediated pairwise interaction discussed in the previous section. The difference is that the Poiseuille flow has no simple analytical solution in the presence of droplets (hence the two cannot be compared exactly), and the confinement requirement is halved due to the symmetry of the parabolic velocity profile.

To further test the robustness of the shear-alignment mechanism, we consider multiple initial configurations of the triplet in the same flow. As sketched in Fig. 7b, three touching droplets whose centers are in the same xy plane can be completely described by two angles: θ_{open} , denoting the opening angle of the triplet, and θ_{dir_c} , denoting the angle between the bisector of the triplet and the direction of the undisturbed flow. For identical droplets, admissible angles are $\theta_{open} \in [60, 180]$ deg., and $\theta_{dir_c} \in [0, 180]$ deg. Extensive tests show that the chain structure is far more favorable than the closed triangular cluster in Poiseuille flows (see Fig. 7b). One case of the triangle cluster is visualized in the third column of Fig. 7a, where two droplets initially on the sides migrate towards the center, eventually leading to the closure of the open chain. In the vast majority of the cases, however, a straight droplet string aligned with the flow is observed, even if they are close to a triangle initially (note the small θ_{open} cases in Fig. 7b).

The above results confirm that the chain-like structure is indeed the predominant configuration of droplets bound by short-range depletion forces in the Poiseuille flow. Experimentally, this corresponds to strong diluting flows at the

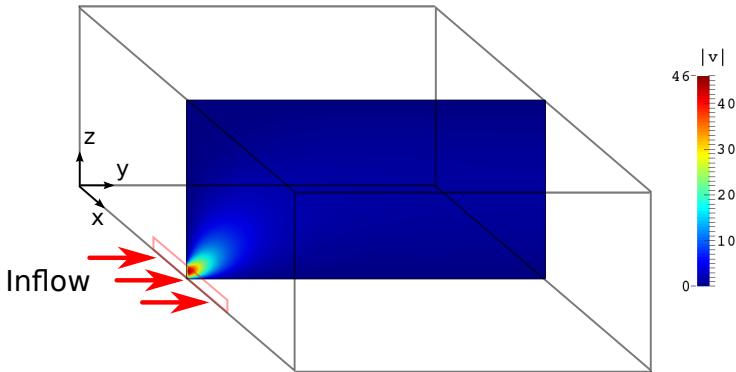


Figure 8: Cross-section of the channel inlet and magnitude of the initial velocity field at the central plane without droplets. The flow is injected from the step-emulsifier (red rectangle), where the droplets (not shown) are produced, into the channel (cf. Fig. 1b). The velocity is normalized such that $|v| = 1$ corresponds to the bulk flow velocity averaged over the entire channel.

channel inlet, where long droplet strings are also observed further downstream (see Shen (2014), p137). More importantly, our simulations suggest that aligning of the droplets is a 3D shear-induced effect mediated by the confinement. The cross-stream migration of the droplets happens within a much shorter time span than any tangential rearrangement due to the dipolar interactions (cf. Diamant (2009) and Fouxon *et al.* (2017)). This is one key difference between our 3D microfluidic channel and other *q2D* devices.

3.3.2. Entry region (with a non-uniform inflow)

So far, we have showed i) the self-assembly of two to ten droplets in quiescent flows, ii) the alignment of a droplet pair in confined simple shear flows, and iii) the chaining (or, sometimes clustering) of a triplet in the Poiseuille flow. Of these, i) is caused solely by the near-field depletion force, and provides the necessary condition for ii) and iii); ii) and iii) are closely related, and in principle can be generalized to clusters of $N > 3$. In addition, we have distinguished our 3D channel from typical *q2D* ones. The remaining question is what makes the droplets self-assemble into compact clusters within short distances (i.e. $\sim \mathcal{O}(10D)^4$) as seen in the experiment of Shen *et al.* (2016b)?

To answer this question, we perform series of simulations of a triplet/quadruplet cluster in a non-uniform channel, similar to the actual entry region of the microfluidic channel (see Fig. 1). Fig. 8 illustrates the cross-sectional design of the

⁴Dipolar interactions under similar confinement require at least $\mathcal{O}(100D)$ distance to see any clustering effect, see Fouxon *et al.* (2017).

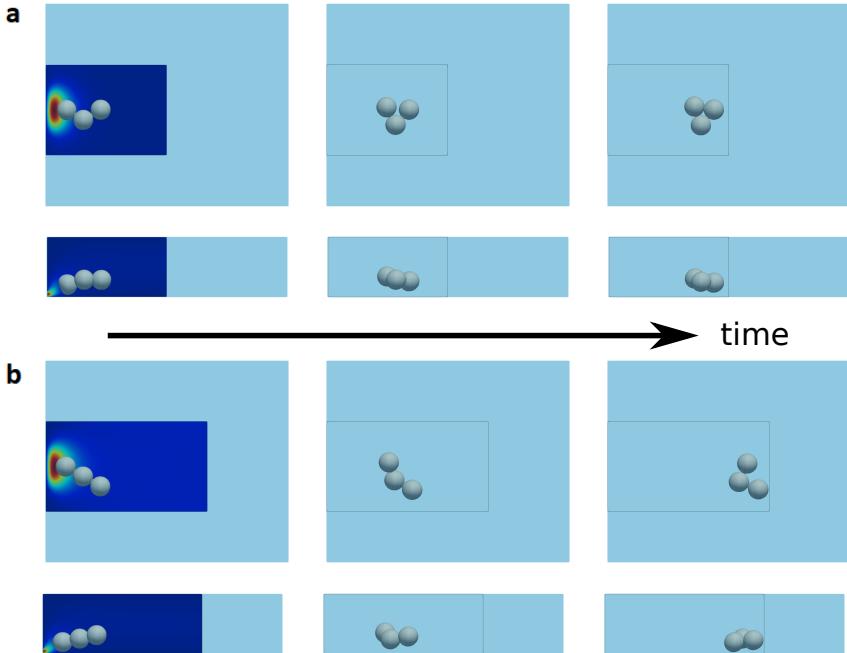


Figure 9: Clustering of three droplets in a channel with a non-uniform inflow. (a) and (b) show the top (upper panel) and side (lower panel) view of the droplet positions under two initial conditions at different times. The framed boxes depicts the actual computational domain (see Fig. 8), in comparison to the Poiseuille channel in Fig. 7. The color contours illustrate the velocity magnitude in two planes orthogonal to the viewing direction, where the color legend is the same as in Fig. 8.

channel inlet and the obtained velocity distribution in the central plane. Specifically, the computational domain has a size of $(L_x/D, L_y/D, L_z/D) = (4.5, 6, 3)$ or $(4.5, 8, 3)$ depending on the cases, and the ratio of the inflow area to the entire cross-section is $A_{in}/A_{tot} = 1/32$, resulting in a highly non-uniform velocity profile. Near the inlet, the peak velocity reaches 46 times the average bulk velocity, then quickly smoothens downstream. To utilise the efficient FFT solver, we again use periodic boundary condition in the spanwise (x) direction, mimicking the effect of diluting flows on the sides. The droplet-to-carrier-fluid density ratio is $\tilde{\rho}_2/\tilde{\rho}_1 = 1.8$, corresponding to silicone oil in water. The rest of the governing parameters are $Re = 0.1$, $Ca = 0.025$, $Fr = 0.0027$, and $\Pi = 1$.

Figs. 9 and 10 demonstrate four representative cases of the self-assembly of three and four droplets, respectively, within a distance of $\sim 10D$ from their initial release. Specifically, the droplets in Fig. 9(a) are initialized with

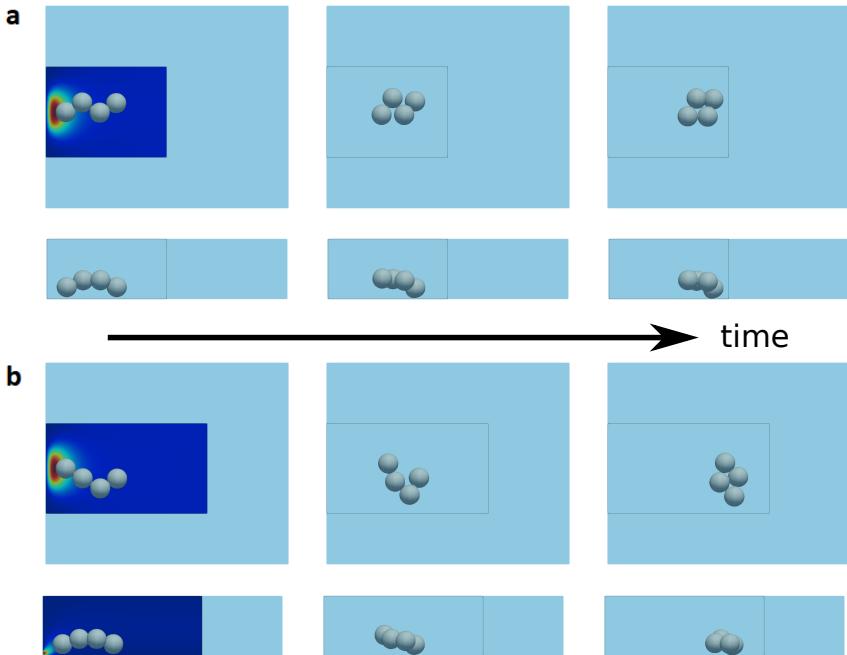


Figure 10: Clustering of four droplets in a channel with a non-uniform inflow. The organization of the plots is similar to Fig. 9.

$\theta_{open} = 120$ deg. and $\theta_{dirc} = 90$ deg. According to Fig. 7b, this triplet would become a chain in the Poiseuille flow. Here, due to the rapid expansion of the flow immediately after the inlet, the trailing droplet undergoes a upward motion to the high velocity region; and if the velocity gained during this sprint is large enough, as in Fig. 9(a), the droplets will soon form a triangle; otherwise, the cluster will at least form a V-shape pointing upstream, as shown in the example of Fig. 9(b). In the latter cases, the final shape of the cluster can be estimated by its orientation relative to the flow (i.e. θ_{open} and θ_{dirc}). Instead of simulating a full evolution of the clustering process (which may require a very large simulation domain and long time), one can simply read the last ($\theta_{open}, \theta_{dirc}$) in the phase diagram of Fig. 7(b). In the case discussed above (Fig. 9(b)), we verified that a triangular cluster is eventually obtained (see ESI Video 2).

Similar observations are made for four droplets, for which we show two examples of clustering into diamond shapes in Fig. 10. Here, the initial conditions are similar to those in Fig. 9, only a fourth droplet is appended to the droplet string at a slightly lower vertical position (due to gravity). Note that the vertical coordinates of the droplets as they move downstream are in opposite orders

(cf. the middle panels of Fig. 7a). Consequently, the trailing droplets travel faster than the frontal ones, leading to the rapid closure of the cluster into more compact shapes.

The above examples clearly illustrate the direct effect of the non-uniform inflow. If properly matched with the initial droplet configuration, the droplets can form a compact structure within a much shorter distance than by the long-range dipolar interaction. And if the near-field depletion force is strong enough, the obtained compact cluster will stay bound further downstream. On the other hand, if the initial non-uniform inflow fails to bring the droplets sufficiently close within its range of influence, i.e. before viscous diffusion smoothens the initial velocity gradients (typically $\sim 10D$), the shear-induced cross-stream migration can break the clustering of the droplets, eventually leading to chain-like structures. This inflow effect, often neglected in theoretical models (Shen *et al.* 2016b), is what we propose to be the key reason for the accelerated droplet assembly.

Finally, we remark that the simulated inlet configuration is only one simplified version of the experimental microfluidic channel. To fully reproduce the condition in the actual setup is unrealistic due to the size contrast of the different inlets; however, it is perhaps also unnecessary as the qualitative features of the clustering do not depend on the fine details at the device level. Depending on the governing parameters and the specific operating conditions, it is possible to optimize the geometry of the microfluidic device to achieve higher throughputs of compact droplet clusters at the outlet; however, in practice, tuning of the geometry and inflow conditions may still involve trial and error, since the final self-assembly results from the combination of all 3D effects with no simple parametric dependence. This is possibly the bottleneck of upscaling the current microfluidic strategy to directly create large photonic crystals.

4. Summary and outlook

Motivated by the recent experiment of flow-assisted droplet assembly (Shen *et al.* 2016b) and its potential application for photonic material synthesis, we present a numerical study of finite numbers of non-Brownian droplets in a 3D microfluidic channel. The newly developed numerical methodology (Ge *et al.* 2018) allows for direct simulations of the two-fluid Navier-Stokes equations, and can account for the short-range attractive depletion force between the drops in a sharp fashion.

Under this framework, we considered three types of flows with increasing complexity: quiescent, confined simple shear, and pressure-driven channel flows. The case of quiescent flows allows us to disentangle the effect of the depletion force from that of the flow. The simulation of two to three droplets shows that the approaching time is inversely proportional to the osmotic number Π , a ratio between the surfactant-induced osmotic pressure and the Laplace pressure. We further assembled four to ten droplets using an arbitrary enumeration of the corresponding sphere packing. Without any external driving motion or noise,

the obtained structure is purely determined by the closest neighbors in the initial state. This seemingly obvious result lays the basis for our subsequent reasoning.

As we place a droplet pair in the confined simple shear flow, the geometric obstruction combined with the depletion force results in a single steady configuration within the shear plane. The specific value of the alignment angle depends on the level of confinement and the shear rate; for nearly spherical drops between moving plates separated by $L_z/D = 1.5$, we find the stable polar angle of the pair to be $\theta_\infty \approx 79$ deg. This alignment arises from the bifurcation of the relative trajectories of two droplets constrained by short-range attractive depletion forces. We expect the phenomenon to persist also for more than two drops, at least in the initial state where pairwise interaction dominates.

The dynamics of droplet clusters in the channel flow depends strongly on the homogeneity of the velocity profile. Using the reference Poiseuille flow, we find that the chain-like structure is far more favorable than the triangular cluster despite the latter is mechanically more stable. This is in contrast to $q2D$ systems where dipolar interactions provide the tangential motion destabilizing the droplet string. When the channel height is larger than the droplet diameter, as it is the case here, the dipolar flow becomes insignificant and the shear-induced cross-stream migration is a genuine $3D$ effect.

To fully understand the fast self-assembly observed in the experiment (Shen *et al.* 2016*b*), we also simulated three and four droplets near a step-emulsifier that is much smaller than the bulk channel. Under suitable initial conditions, the triplet/quadruplet indeed forms a more compact cluster from a chain. The nearly reversed inter-droplet motions comparing to the Poiseuille case clearly highlight the effect of the inhomogeneous flow. For practical microfluidic devices aiming for large throughputs, geometric optimization and fine tuning of the flow condition appear to be the key.

The above depicts the complete physical picture of depletion/hydrodynamic interactions of few non-Brownian droplets in a $3D$ microfluidic channel. Correctly identifying these mechanisms may help experimentalists design microfluidic chips not only for the fabrication of photonic metamaterials, but also other functionalities in general. We note that, although it remains a challenge to directly produce large, defect-free photonic crystals (typically of diamond-like structures) using the current microfluidic setup, alternative strategies have been recently proposed to *indirectly* assemble droplet lattices composed of smaller clusters (Morozov & Leshansky 0), or creating hyperuniform droplet ensembles using a similar microfluidic device (Torquato & Stillinger 2003; Ricouvier *et al.* 2017). The latter is an active on-going research area, and we hope our findings provide additional guidelines to rationalize the design procedure of these miniature devices.

Acknowledgments

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Appendix

A. Numerical setup

The simulations are performed in rectangular Cartesian domains with periodic and/or inflow-outflow boundary conditions in two directions and wall boundary condition (i.e. no slip/no penetration) in the third direction. The streamwise and spanwise dimensions of the computational box are at least three times bigger than the initial diameter of the droplet to prevent possible long-range interactions caused by the image droplet. The droplets are resolved by 32 grid points per diameter (i.e. $\Delta x = 1/32$) to ensure that the interface curvature and the pressure jump are accurately computed, see Ge *et al.* (2018) for detailed verification.

As mentioned earlier, the numerical values of Re and Ca are artificially increased to facilitate faster simulations over a larger parameter space. Specifically, we set $Re = 10$, $Ca = 0.005$ in Secs. 3.1 and 3.3.1; $Re = 1$, $Ca = 0.0025$ in Sec. 3.2; and $Re = 0.1$, $Ca = 0.025$ in Sec. 3.3.2. Fr is effectively ∞ by setting the density ratio equal to unity in all cases except in Sec. 3.3.2, where it is 0.025. The viscosity ratio is always 1.

In Sec. 3.1, the suspending fluid has no underlying velocity, so the actual droplet Reynolds number should be rescaled by the ratio of the average approaching speed and the mean velocity of the channel². In the case of two droplets, this factor is 1/64 as one droplet moves the distance of Δx within $t = 2$, as comparing to a displacement of 2 within $t = 2$. The rescaled Reynolds is then $Re_r \approx 0.16$.

In Sec. 3.2, we reduce Re to 1 as the flow is shear-driven, thus the fluid inertia is expected to play a role. Testing various Re , as shown in Fig. 11, we observe that the stable polar angle approaches the same value for $Re \leq 1$ under the confinement of $L_z/D = 1.5$, justifying the use of $Re = 1$. For $L_z/D = 3$, however, $Re < 1$ must be used to obtain the true closed orbit of the two bounding pair (i.e. the obtained θ_∞ in that case is an artifact of the inertia).

In Sec. 3.3.1, we increase Re to 10 as the cross-stream migration in Poiseuille flow is a fairly robust phenomenon, only weakly dependent on the Reynolds number in the sense that a lower Re imposes a longer time scale. Since we are interested in the final shape of the droplet cluster - a qualitative result rather

²We keep the same definition of the Reynolds number due to bookkeeping reasons in the numerical code.

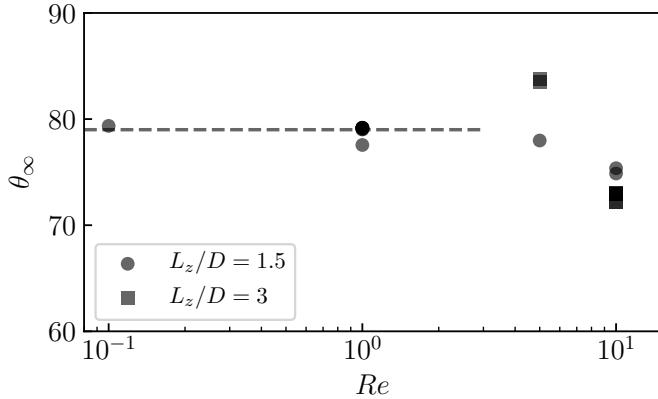


Figure 11: Stable polar angle of two droplets in a shear-driven channel at different Reynolds numbers and confinement, cf. Sec. 3.2. The dashed line corresponds to $\theta_\infty = 79$ deg.

than the detail dynamics - $Re = 10$ is used to speed up the simulations (the computational time step is roughly inversely proportional to Re).

Finally, in Sec. 3.3.2 where the entry region of the microfluidic channel is considered, we set the lowest Reynolds number to mimic the actual flow environment. Here, the capillary number is amplified to 0.025, larger than in the previous cases but still well within the low capillary limit. As a visual proof, the droplets shown in Fig. 9 all remain nearly spherical during the convection. Further reducing Ca shall have no effect but refine the sphericity of the drops.

B. Confinement-mediated interaction

For unbounded simple shear flows, we know that a pair of spherical particles/droplets can undergo either closed orbits or open-and-symmetric trajectories (Batchelor & Green 1972a; Zinchenko 1984). With moderate confinements, swapping trajectories are also possible (Zurita-Gotor *et al.* 2007). In Sec. 3.2, we show that strong geometric confinements combined with an attractive depletion force lead to pair alignments in the vorticity plane; particularly, the obtained stable polar angle is $\theta_\infty = 79$ deg. for $L_z/D = 1.5$. Below, we provide further evidence to support the symmetry-breaking argument and give a qualitative explanation of the observed θ_∞ .

In Fig. 11, we obtain non-converging values of θ_∞ at $L_z/D = 3$. Under this confinement, the binding droplets in the low-Reynolds-number limit exhibit cyclic motions as if they are unconfined (see ESI Video 3). Clearly, the center-to-center depletion force does not play any role since the hydrodynamic stresses already keep the droplet together; the droplets deform slightly, but are essentially spherical. As we double the confinement, i.e. reducing L_z/D by half, Fig. 12

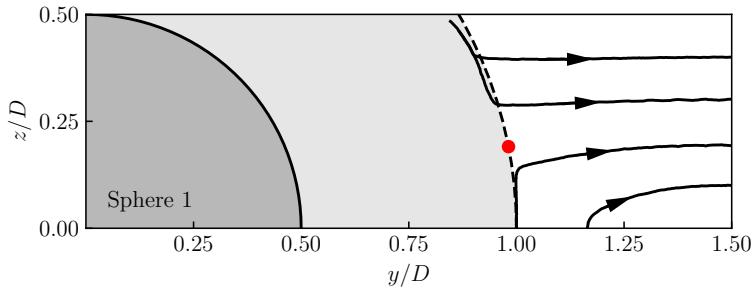


Figure 12: Relative trajectories of the second sphere in the vorticity plane of the simple-shear channel under confinement $L_z/D = 1.5$. The pair is not constrained by any depletion force. The shaded region denotes locations inaccessible to the second droplet if it is perfectly spherical. Overlap of the trajectories with the shades is a result of the small droplet deformation. The red dot corresponds to the polar angle of 79 deg, c.f. Fig. 6.

illustrates the relative trajectories in the absence of depletion forces (see also ESI Video 4,5). Here, only a quadrant of the plane in the vicinity of the first droplet is shown due to symmetry. Comparing to less confined conditions, the droplet pair displays only passing and swapping trajectories, while the Batchelor-Green type of closed orbit is completely suppressed (c.f. Fig. 3 in (Zurita-Gotor *et al.* 2007)). Arguably, such a result is obvious as the droplets cannot simply rotate in the same vorticity plane, whereas 3D rotations would violate either time-reversal or mirror symmetry.³ Regardless the reason, the results demonstrate that droplets cannot stay together indefinitely due solely to the hydrodynamic interactions. More importantly, plotting the position corresponding to the angle of 79 deg. (the red dot in Fig. 12) in the trajectory map clearly rationalizes the existence of a stable polar angle: the second droplet would travel in either direction above or below the saddle point; with a radial depletion force, only at $\theta_\infty = 79$ deg. can it stay dynamically stable.

³The symmetry argument, however, is only a necessary but not sufficient condition, since droplet sliding on the walls already invokes effects of the noise (otherwise two droplets stuck by the walls should be regarded as an admissible solution).

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Paper 3

3

Integral representation of channel flow with interacting particles

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We construct a boundary integral representation for the low-Reynolds-number flow in a channel in the presence of freely-suspended particles (or droplets) of arbitrary size and shape. We demonstrate that lubrication theory holds away from the particles at horizontal distances exceeding the channel height and derive a multipole expansion of the flow which is dipolar to the leading approximation. We show that the dipole moment of an arbitrary particle is a weighted integral of the stress and the flow at the particle surface, which can be determined numerically. We introduce the equation of motion that describes hydrodynamic interactions between arbitrary, possibly different, distant particles, with interactions determined by the product of the mobility matrix and the dipole moment. Further, the problem of three identical interacting spheres initially aligned in the streamwise direction is considered and the experimentally observed “pair exchange” phenomenon is derived analytically and confirmed numerically. For non-aligned particles, we demonstrate the formation of a configuration with one particle separating from a stable pair. Our results suggest that in a dilute initially homogenous particulate suspension flowing in a channel the particles will eventually separate into singlets and pairs.

1. Introduction

Hydrodynamic interactions among particles flowing in the fluid confined between two parallel walls at low Reynolds number have recently attracted a considerable attention (Carbajal-Tinoco *et al.* 1997; Acuña Campa *et al.* 1998; Pesché & Nägele 2000*a,b*; Lançon *et al.* 2001; Marcus *et al.* 1999; Santana-Solano & Arauz-Lara 2001, 2002; Cui *et al.* 2004; Cohen *et al.* 2004; Tlusty 2006; Beatus *et al.* 2006, 2012; Sarig *et al.* 2016; Swan & Brady 2010; Durlofsky & Brady 1989; Nott & Brady 1994; Morris & Brady 1998; SINGH & NOTT 2000; Morris 2001; Bhattacharya *et al.* 2006; Shani *et al.* 2014; Shen *et al.* 2016, 2014). The case of particles driven by thermal noise in the absence of a macroscopic flow

was studied in Carbajal-Tinoco *et al.* (1997); Acuña Campa *et al.* (1998); Pesché & Nägele (2000a,b); Lançon *et al.* (2001); Marcus *et al.* (1999); Santana-Solano & Arauz-Lara (2001, 2002); Cui *et al.* (2004); Cohen *et al.* (2004); Tlusty (2006). The hydrodynamic interactions cause long-range correlations in their diffusive motions that are measurable even at distances ten times larger than the particle size (Carbajal-Tinoco *et al.* 1997; Cui *et al.* 2004). In the case of pressure-driven Poiseuille or shear flow the particles are, in addition, dragged by the flow (Durlofsky & Brady 1989; Nott & Brady 1994; Morris & Brady 1998; SINGH & NOTT 2000; Morris 2001; Bhattacharya *et al.* 2006; Beatus *et al.* 2006, 2012; Shani *et al.* 2014; Shen *et al.* 2016, 2014; Sarig *et al.* 2016).

Identical particles at similar positions inside the channel move at the same velocity if not for hydrodynamic interactions. These interactions induce particle relative motions, which can result in considerable changes of their configuration inside the channel. In the case of a large number of particles the interactions cause also chaotic collisions among the particles (Shani *et al.* 2014).

Theoretical progress has mainly relied on the observation that the far flow caused by a particle confined in a channel is a dipolar flow decaying quadratically with the distance (Cui *et al.* 2004). For disk-like particles with thickness close to the channel width h , the dipolar flow and its moments were derived from lubrication theory in Tlusty (2006). The dipolar flow holds at distances much larger than the disk radius, where it gives also the leading order hydrodynamic interactions among particles (Tlusty 2006; Beatus *et al.* 2006, 2012; Shani *et al.* 2014).

It was observed in Beatus *et al.* (2012), however, that hydrodynamic interactions of pancake-like disks can also be described at much smaller distances between the disks where dipolar approximation breaks down, yet lubrication theory still holds (Batchelor 1967; Tavakol *et al.* 2017; Szeri 2005; Bruce 2012). This theory predicts that at distances from the particle boundary much larger than h the depth-averaged flow is an ideal two dimensional flow with potential obeying the Laplace equation. The boundary condition (b. c.), derived somewhat heuristically, is the usual ideal flow b. c. prescribing the velocity component normal to the particle surfaces (Landau & Lifshitz 1987), which allowed to find the hydrodynamic interactions of two close disks, see Beatus *et al.* (2012). Moreover, it was observed that the non-rigidity of the particles makes the lubrication theory valid up to distances from the particles smaller than h . The calculation of the hydrodynamic interactions for disks of different radii requires solving the Laplace equation with the help of bipolar coordinates, see Sarig *et al.* (2016).

Recently, a practical applications of hydrodynamic interactions among particles in a channel has been proposed. In particular, it is suggested that the combined action of adhesive (non-hydrodynamic) forces and hydrodynamic interactions between microdroplets can result in the formation of regular particle clusters and can thus be potentially used for the production of new materials (Shen *et al.* 2014, 2016). The hydrodynamic forces are believed to be a significant

factor in these structure formation. Though the particles forming the structure are in a close proximity in the experiments mentioned above, the hydrodynamic interactions are described phenomenologically by a dipolar flow, formally only valid at larger distances. Despite the use of the far-field dipolar flow beyond its domain of validity, the numerical simulations in Shen *et al.* (2014) showed very good agreement with the experimental results (Shen *et al.* 2016). The above motivates the need for the detailed theoretical study of hydrodynamic interactions among particles in narrow channels.

In this work, we introduce a boundary integral representation of the channel flow in the presence of freely suspended particles. The particles can be rigid or soft (droplets). The representation does not depend on the particle equation of motion, defined by inertia. Boundary integral representations are known to be useful in unconfined flows and can also be applied to confined geometries (Happel & Brenner 1983; Pozrikidis 1992). The flow is here expressed as the sum of the undisturbed Poiseuille flow and an integral over the surfaces of all particles, where the particles can have arbitrary shapes. The derivation is performed for a pressure-driven flow, but identical considerations can be applied to shear flows.

Our representation results in a formula for calculation of the dipole moment, which was previously available only for the case of disk-like particles. The moment is given in terms of a weighted integral of the stress tensor and the flow over the surface(s) of the particle(s). Once this integral is numerically tabulated, the result can be used to approximate the flow in different configurations. Here, we perform simulations for the case of neutrally buoyant rigid spherical particles and compute the integral for different positions of the particle center and different ratios of the particle radius to the channel height, i.e. different confinements.

We use this new integral representation to show that the lubrication theory holds at the particle near proximity, closer than what typically expected. As an example, we solve the problem of three aligned particles moving along the line defined by their centers and the case of three nonaligned particles. We conjecture that this solution is the attractor to which the long-time evolution of arbitrary initial condition converges. We conclude by proposing a mean field description of strong hydrodynamic interactions of close particles in a dense suspension.

2. Integral representation for channel flow with particles

In this Section we derive the boundary integral representation for channel flow in the presence of an arbitrary number of particles of arbitrary shape (see Fig. 1 where spherical particles are shown for illustration). It is assumed that the Reynolds number is low and the Stokes equations hold. The derivation uses the reciprocal theorem with the reciprocal flow given by the Stokeslet in a channel (Liron & Mochon 1976), similarly to the derivations in infinite space, see e. g. Pozrikidis (1992). In this Section we make no assumptions on the form of the

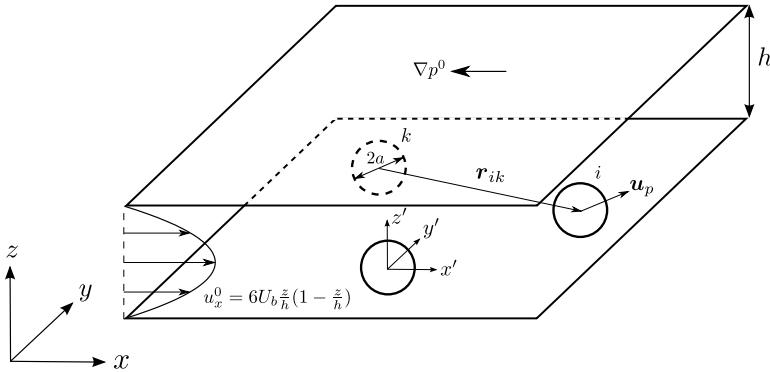


Figure 1: Schematic configuration of spherical particles flowing in the pressure-driven (Poiseuille) flow in a channel.

equation of motion of the particles which may change according to the relevance of inertia. The particles can be rigid, droplets or, e.g. viscoelastic.

The undisturbed flow \mathbf{u}^0 , in the absence of particles, is the Poiseuille flow driven by the constant pressure gradient ∇p^0 ,

$$u_x^0 = \frac{z(z-h)\nabla_x p^0}{2\eta}, \quad \nabla p^0 = \eta \nabla^2 \mathbf{u}^0. \quad (1)$$

where η is the fluid viscosity, z is the vertical coordinate and h is the channel width. The flow is in x -direction, $\nabla_x p^0 = -|\nabla_x p^0| \hat{\mathbf{x}}$. In the presence of a freely suspended particle we look for the solution of,

$$\begin{aligned} \nabla p &= \eta \nabla^2 \mathbf{u}, \quad \nabla \cdot \mathbf{u} = 0, \quad \mathbf{u}(z=0) = \mathbf{u}(z=h) = 0, \\ u_x(\infty) &= \frac{z(z-h)\nabla_x p^0}{2\eta}, \end{aligned} \quad (2)$$

which holds outside the particle. The flow is completely determined when solving for the particle motion, i.e. knowing the instantaneous particle position as it determines the boundary condition $\mathbf{u}_S(\mathbf{x})$ on the particle surface S . In the case of a rigid particle, $\mathbf{u}_S(\mathbf{x}) = \mathbf{v} + \boldsymbol{\omega} \times (\mathbf{x} - \mathbf{y})$ where \mathbf{v} and $\boldsymbol{\omega}$ are the particle translational and angular velocities and $\mathbf{y} = (x_p, y_p, z_p)$ is the coordinate of the particle center of mass. The velocities \mathbf{v} and $\boldsymbol{\omega}$ are determined by the solution of the equation for the particle motion coupled with the flow. These velocities could be time-independent as in the case of the steady motion of a neutrally buoyant rigid particle or the case of a non-neutrally buoyant particle after sedimentation when reaching the bottom wall (the theoretical determination of these velocities is impossible generally because of the interaction with the walls). These velocities can also be time-dependent as in the case of a transient flow or the gravitational settling of a non-neutrally buoyant particle. If several

particles are considered, a time-dependent configuration can be induced by their interactions. We assume here that the time variations are not fast so that the unsteady time-derivative term of the Navier-Stokes equations is negligible (for the steady motion of one particle the time-derivative is the spatial derivative of the flow along the streamline which is small because of the smallness of the Reynolds number). In the case of droplets the boundary condition on the surface is determined by matching with the inner flow. However there is no need for solving for this inner flow since the detailed form of $\mathbf{u}_S(\mathbf{x})$ is irrelevant for the derivation of the present representation. The generalization of the problem to the case of many particles is obvious.

Implications of lubrication theory.—Some conclusions on the flow at distances from the particle much larger than the channel width h can be obtained from lubrication theory (Batchelor 1967; Tavakol *et al.* 2017; Szeri 2005; Bruce 2012). The lubrication theory predicts that at these distances,

$$\mathbf{u} = \frac{z(z-h)\nabla p}{2\eta}, \quad (3)$$

with a certain z -independent p . Clearly, at large distances, $p \approx p^0$ at the leading order. The depth-averaged velocity \mathbf{u}_d is the ideal potential two-dimensional flow,

$$\mathbf{u}_d = \nabla\phi, \quad \phi = -\frac{h^2 p}{12\eta}, \quad \nabla^2\phi = 0. \quad (4)$$

In some cases this helps determining the flow completely.

Large disks.—Large non-wetting droplets squeezed between the walls of a Hele-Shaw cell have pancake-like shapes. These can be modeled as disks with radius $a \gg h$ and height close to h , i.e., they almost fill the entire channel height (Thulst 2006; Beatus *et al.* 2006, 2012; Shani *et al.* 2014). In this case, one can use Eqs. (3)-(4) at distances from the body that are much larger than h but much smaller than a . Thus the ideal flow holds outside the narrow boundary layer near the particle surface whose characteristic size l_0 is of the order h , much smaller than the particle horizontal size a . We call the layer containing the flow vorticity the viscous layer and assume that there is no flux of mass through the surface of the droplet, which keeps its shape and volume. Thus, in the frame of reference moving with the disk, the normal velocity component is zero at the outer boundary of the layer and the ideal flow outside the layer is determined uniquely by this boundary condition. However the geometry of the layer is not always known and the complete determination of the flow not possible. If we are only interested in the flow outside the immediate l_0 -vicinity of the viscous layer, we can set the boundary condition for the ideal flow on the disk itself, exploiting the fact that $l_0 \ll a$. The flow is then found as the dipole potential (Thulst 2006),

$$\phi = -\frac{h^2 p^0}{12\eta} - \frac{\mathbf{d} \cdot \hat{\mathbf{r}}}{r}, \quad \nabla^2\phi = 0, \quad \mathbf{d} = a^2 \mathbf{v}, \quad (5)$$

where \mathbf{d} is the dipole moment, $\mathbf{v} = -v\hat{\mathbf{x}}$ is the difference between the disk velocity and $-h^2\nabla p_0/(12\eta)$. It is readily seen that the normal, radial component of the velocity $\nabla\phi$ on the surface of the disk is $[\mathbf{v} - h^2\nabla p^0/(12\eta)] \cdot \hat{r}$. Note that the velocity $v > 0$ since the particle moves slower than the fluid.

The tangential velocity component on the outer boundary of the viscous layer obtained from Eq. (5) does not match the tangential velocity of the droplet surface. In contrast with the normal component, which can be considered almost constant through the viscous layer, the tangential component changes quickly through this layer to match the inner flow at the droplet surface. As example, in the limit of high droplet viscosity, the condition on the disk surface is that the flow is the appropriate superposition of translation and solid body rotation. Similar viscous layers occur for rigid bodies oscillating in the fluid (Landau & Lifshitz 1987). The ideal flow was obtained in Thusty (2006) and here we describe how this ideal flow fits the complete equations for the viscous flow.

Boxes.—Another case where the flow can be fixed without detailed calculations is the case of a box whose smallest dimension is close to h and the longer dimensions are much larger than h . If the box is located in the channel so that the flow is perpendicular to its longer axis with length $l \gg h$ then far from the ends of the box we find the ideal two-dimensional flow with constant velocity on the line. The solution for the ideal flow with constant velocity on an infinite linear boundary is the uniform flow. We thus conclude that in the frame moving with the box there is a region of stagnant flow behind the box, whose size is of order l .

Generally, the flow can be inferred from the lubrication theory in quite a detail for particles whose horizontal dimensions are much larger than h and whose vertical dimension is close to h . The flow outside the narrow viscous layer near the particle surface is $z(z-h)$ times the two-dimensional ideal flow determined by the boundary condition of zero normal velocity on the particle surface. The tangential velocity changes fast across the viscous layer. However, if the particle horizontal dimensions are not large or the dimensions are large but the vertical dimension is not close to h , a different approach is needed.

Boundary integral representation from the reciprocal theorem.—We use the reciprocal theorem (Happel & Brenner 1983; Pozrikidis 1992) using as the reciprocal flow the solution of Liron & Mochon (1976) for the point-force or Stokeslet between two parallel plates, i.e.

$$\begin{aligned} -\nabla p^S + \eta \nabla^2 \mathbf{u}^S + \mathbf{g}\delta(\mathbf{x} - \mathbf{x}_0) &= 0, \quad \nabla \cdot \mathbf{u}^S = 0, \\ \mathbf{u}^S(z=0) &= \mathbf{u}^S(z=h) = 0, \quad \mathbf{u}^S(x^2 + y^2 \rightarrow \infty) = 0. \end{aligned} \quad (6)$$

Analogously to the flow due to a point-force acting on a viscous fluid in infinite space (e.g. Pozrikidis (1992)), the solution depends linearly on the source forcing \mathbf{g} ,

$$\mathbf{u}^S(\mathbf{x}) = \frac{1}{8\pi\eta} S_{ik}(\mathbf{x}, \mathbf{x}_0) g_k, \quad (7)$$

where we introduced the tensor S_{ik} independent of \mathbf{g} . Similarly for the stress tensor of the Stokeslet solution we can write,

$$\begin{aligned}\sigma_{ik}^S(\mathbf{x}) &= -p^S \delta_{ik} + \eta (\nabla_k u_i^S + \nabla_i u_k^S) = \frac{T_{ilk}(\mathbf{x}, \mathbf{x}_0) g_l}{8\pi}, \\ \nabla_k T_{ilk} &= -8\pi \delta_{il} \delta(\mathbf{x} - \mathbf{x}_0), \quad T_{ilk} = -p_l \delta_{ik} + \nabla_k S_{il} + \nabla_i S_{kl},\end{aligned}\quad (8)$$

where T_{ilk} is a third-rank tensor independent of \mathbf{g} and we defined the \mathbf{g} -independent p_l by $p^S = \mathbf{p} \cdot \mathbf{g}/(8\pi)$. We use the Lorentz identity,

$$\nabla_k [u_i \sigma_{ik}^S - u_i^S \sigma_{ik}] + \mathbf{u} \cdot \mathbf{g} \delta(\mathbf{x} - \mathbf{x}_0) = 0, \quad (9)$$

readily inferred from the Stokes equations with σ_{ik} the stress tensor of the flow defined by Eqs. (2),

$$\sigma_{ik} = -p \delta_{ik} + \eta (\nabla_i u_k + \nabla_k u_i). \quad (10)$$

Substituting \mathbf{u}^S and $\boldsymbol{\sigma}^S$ from Eqs. (7)-(8) in Eq. (9) and using the above identity we find that,

$$\begin{aligned}8\pi \eta u_l(\mathbf{x}') \delta(\mathbf{x}' - \mathbf{x}) &= \frac{\partial}{\partial x'_k} [S_{il}(\mathbf{x}', \mathbf{x}) \sigma_{ik}(\mathbf{x}')] \\ &\quad - \eta u_i(\mathbf{x}') T_{ilk}(\mathbf{x}', \mathbf{x}).\end{aligned}\quad (11)$$

Integrating this equation over \mathbf{x}' outside the particles,

$$\begin{aligned}u_l &= f_l - \sum_n \int_{S_n} \frac{S_{il}(\mathbf{x}', \mathbf{x}) \sigma'_{ik} dS'_k}{8\pi \eta} \\ &\quad + \sum_n \int_{S_n} \frac{u_i(\mathbf{x}') T_{ilk}(\mathbf{x}', \mathbf{x}) dS'_k}{8\pi},\end{aligned}\quad (12)$$

where f is the integral over the far surface at infinity, S_n is the surface of the n -th particle and dS_k is aligned with the outward normal to the particle surface. There is no contribution from the channel boundaries $z = 0$ and $z = h$ since both flows vanish there.

We assume that the particles are confined in a finite region so that the flow far from the particles is the Poiseuille flow given by Eq. (1), see Eq. (2). Since the Stokeslet decays far from the source, the second and the third terms on the RHS of Eq. (12) decay to zero at large distances from the particles. Thus, the asymptotic approach of \mathbf{u} to the Poiseuille flow at large distances implies the asymptotic equality of f to the Poiseuille flow. It is readily seen using the asymptotic form of the Stokeslet at large horizontal distances, provided in the next Section, and the asymptotic, Poiseuille, form of the flow, that f is determined by terms independent of the particles, that is terms that would be the same for the case of no particles. Thus necessarily f is the Poiseuille flow given by Eq. (1). This is confirmed by the direct calculation in Appendix

Appendix A. We therefore conclude that,

$$\begin{aligned} u_l = & \frac{\delta_{lx} z(z-h) \nabla_x p^0}{2\eta} - \sum_n \int_{S_n} \frac{S_{il}(\mathbf{x}', \mathbf{x}) \sigma'_{ik} dS'_k}{8\pi\eta} \\ & + \sum_n \int_{S_n} \frac{u_i(\mathbf{x}') T_{ilk}(\mathbf{x}', \mathbf{x}) dS'_k}{8\pi}. \end{aligned} \quad (13)$$

This integral representation of the flow involves no approximations and holds for particles of arbitrary shape. The flow is determined by the values of \mathbf{u} and σ_{ik} at the particle surface. When the distance between the particles are much larger than their size the hydrodynamic interactions are negligible and \mathbf{u} and σ_{ik} are approximately those of an isolated particle. However, the current representation works also when the particles are close to each other so that the hydrodynamic interactions change significantly the values of \mathbf{u} and σ_{ik} at the particle surface.

Simplification for rigid surface.—The integral representation above simplifies in the case of rigid surfaces when the flow on the surface of the particles is the superposition of translation and solid-body rotation. This is not only the case of rigid particles described after Eq. (2), but often also the case of small bubbles where impurities present in the fluid accumulate at the surface making it effectively rigid. In this case, experiments demonstrate that the behavior of these bubbles is similar to that of rigid particles (Happel & Brenner 1983; Levich 1977). In these and similar cases the flow at the particle surface $u_i(\mathbf{x}')$ has the form $c_i + \epsilon_{irm} x'_m \tilde{c}_r$ where both \mathbf{c} and $\tilde{\mathbf{c}}$ are independent of \mathbf{x}' . Hence the last term in Eq. (13) drops out because for \mathbf{x} outside the particle interior V_p ,

$$\begin{aligned} \int_{S_p} dS'_k T_{ilk}(\mathbf{x}', \mathbf{x}) &= \int_{V_p} dV \nabla'_k T_{ilk} = 0, \\ \epsilon_{irm} \int_S dS'_k x'_m T_{ilk}(\mathbf{x}', \mathbf{x}) &= \epsilon_{irm} \int_{V_p} dV \delta_{mk} T_{ilk} = 0, \end{aligned} \quad (14)$$

cf. Pozrikidis (1992). Note that we made use of the fact that ϵ_{irm} is antisymmetric over indices i, m whereas T_{ilm} is symmetric with respect to those indices. We conclude that in the case of many particles with rigid surfaces,

$$u_l = \frac{\delta_{lx} z(z-h) \nabla_x p^0}{2\eta} - \sum_n \int_{S_n} \frac{S_{il}(\mathbf{x}', \mathbf{x}) \sigma'_{ik} dS'_k}{8\pi\eta}, \quad (15)$$

The representations derived here are thus useful to describe the flow.

3. Derivation of lubrication theory

In this Section we demonstrate that Eqs. (13) and (15) imply that the predictions of the lubrication theory hold at horizontal distances from the particles larger than h . This is less restrictive than the usual condition of applicability of the lubrication theory for distances much larger than h , cf. Sarig *et al.* (2016). This

property comes from the Stokeslet flow which obeys the lubrication theory at distances larger than h . We use the representation

$$S_{il}(\mathbf{x}', \mathbf{x}) = \frac{12z'(h-z')z(z-h)\nabla_i\nabla_l \ln \rho}{h^3} + \tilde{S}_{il}(\mathbf{x}', \mathbf{x}), \quad (16)$$

where ρ is the length of $\boldsymbol{\rho} = (x - x', y - y')$ (thus $\nabla_i\nabla_l \ln \rho = 0$ if one of the indices is z). It was observed in Liron & Mochon (1976) that \tilde{S}_{il} decays exponentially in ρ with exponent at least π/h , that is the smallness is at least $\exp(-\pi\rho/h)$. Thus already at $\rho \approx h$ we can discard the last, non-potential term in Eq. (16). The resulting approximation to the Stokeslet flow,

$$S_{il}(\mathbf{x}', \mathbf{x}) \approx S_{il}^0(\mathbf{x}', \mathbf{x}) = \frac{12z'(h-z')z(z-h)\nabla_i\nabla_l \ln \rho}{h^3}, \quad (17)$$

is the two-dimensional potential flow times $z'(h-z')$, in agreement with the predictions of the lubrication theory, see Eq. (3). Note that $S_{il}^0(\mathbf{x}', \mathbf{x})$ is a symmetric function of \mathbf{x} and \mathbf{x}' , whose dependence on the horizontal coordinates is via the difference $\boldsymbol{\rho}$ only. We consider the corresponding pressure p^0 that approximately solves the corresponding Stokes equation $\nabla'_i p_l^0(\mathbf{x}', \mathbf{x}) = \nabla'^2 S_{il}^0(\mathbf{x}', \mathbf{x})$, see the second of Eqs. (8). Here ∇'_i designates the derivative over x'_i and we do not write the $\delta(\mathbf{x}' - \mathbf{x})$ term on the RHS. We thus find ($l \neq z$),

$$p_l^0(\mathbf{x}', \mathbf{x}) = \frac{24z(z-h)\rho_l}{h^3\rho^2} = \frac{24z(z-h)}{h^3}\nabla_l \ln \rho. \quad (18)$$

Here, p^0 is the leading order approximation for the pressure of the Stokeslet at large distances (Liron & Mochon 1976), with an exponentially small correction. We can write ($l \neq z$ but i or k can be z),

$$T_{ilk}(\mathbf{x}', \mathbf{x}) = -p_l^0 \delta_{ik} + \nabla'_k S_{il}^0 + \nabla'_i S_{kl}^0 + \tilde{T}_{ilk}(\mathbf{x}', \mathbf{x}), \quad (19)$$

where $\tilde{T}_{ilk}(\mathbf{x}', \mathbf{x})$ decays exponentially in ρ with exponent at least π/h , cf. Eq. (8). The stress tensor $T_{ilk}(\mathbf{x}', \mathbf{x})$ is exponentially small when one of the indices is z . We find using the expressions for p_0 and S_{il}^0 ,

$$\begin{aligned} T_{ilk}(\mathbf{x}', \mathbf{x}) &\approx \frac{24z(h-z)}{h^3} \nabla_l [(\delta_{ik} + z'(h-z')\nabla_i\nabla_k) \ln \rho] \\ &+ \frac{12(h-2z')z(z-h)}{h^3} (\delta_{kz}\nabla_i + \delta_{iz}\nabla_k) \nabla_l \ln \rho, \end{aligned} \quad (20)$$

where we neglected exponentially small correction. We find from Eq. (13) that,

$$\mathbf{u}(\mathbf{x}) = \frac{z(z-h)\nabla p}{2\eta} + O\left(e^{-\pi \min[\rho_n]/h}\right); \quad \nabla^2 p = 0, \quad (21)$$

where $\mathbf{u} = (u_x, u_y)$ and $\min[\rho_n]$ is the distance from \mathbf{x} to the closest boundary of a particle. The pressure p in this formula is independent of z as predicted by

the lubrication theory used in Eq. (3) with,

$$\begin{aligned} p = p^0 + \sum_n \delta p_n, \quad \delta p_n = & \frac{3\nabla_i}{\pi h^3} \int_{S_n} z'(z' - h) \ln \rho \sigma'_{ik} dS'_k \\ & - \frac{6\eta}{\pi h^3} \int_{S_n} dS'_k (\delta_{ik} + z'(h - z') \nabla_i \nabla_k) u_i(\mathbf{x}') \ln \rho \\ & + \frac{3\eta}{\pi h^3} \int_{S_n} dS'_k (h - 2z') (\delta_{kz} \nabla_i + \delta_{iz} \nabla_k) u_i(\mathbf{x}') \ln \rho, \end{aligned} \quad (22)$$

where δp_n is the pressure perturbation due to the n -th particle and the summation over repeated indices is from 1 to 3. Eqs. (21)-(22) are one of the main results of our work. These provide a refinement of the lubrication theory demonstrating that Eq. (3) holds under the condition $\exp(-\pi \min[\rho_n]/h) \ll 1$, which is difficult to show using the classic lubrication theory as it demands the strong inequality $\min[\rho_n] \gg h$. For instance, at $\min[\rho_n] = h$ the exponential factor is ~ 0.04 . The result holds both for droplets and rigid particles where for rigid particles the last two lines of Eq. (22) become zero and the equation reduces to

$$p = p^0 + \frac{3\nabla_i}{\pi h^3} \sum_n \int_{S_n} z'(z' - h) \ln \rho \sigma'_{ik} dS'_k. \quad (23)$$

We have good control of the correction terms to Eqs. (21)-(23) from the series representation of $\tilde{S}_{il}(\mathbf{x}', \mathbf{x})$ provided in Liron & Mochon (1976).

The pressure p solves the two-dimensional Laplace equation in the domain between the particles since it is formed by integrals of the fundamental solution of the Laplace equation $\ln \rho$ over the particle boundaries. The formula for p matches the ideal flow that holds beyond the horizontal distance h from the particles with the fully viscous flow near the particles. The viscous layer is the neighborhood of the boundary of each particle where Eq. (22) breaks down. Though the solution for p is given in terms of the unknown velocities and stress tensors on the surfaces of the particles, it seems that this is as much as can be done generally: the matching problem is not solvable for any general particle shape. It does simplify for disk-like particles as described previously.

Hydrodynamic interactions of pancake-like droplets.—Eqs. (21)-(22) provide support for the observation that the width of the viscous layer around disk-like droplets is not larger than h . The formulae tell that, unless the distance between the droplet surfaces is smaller than h , the (horizontal) flow outside the viscous layers near the particles is an ideal potential flow. This flow can be determined using the boundary condition that the normal velocity at the outer boundary of the viscous layer coincides with the normal component of the translational velocity of the particle. Since the layer width is of the order of h , and as long as the distance between the droplets is larger than h (but possibly much smaller than a) we can impose the boundary condition on the particle surface, neglecting the finite width of this viscous layer as we did for the case of the single large disk, see Eq. (5). Similarly, in the presence of many particles whose

separation is larger than h , the flow outside the boundary layer is described by a pressure field p that obeys (Sarig *et al.* 2016; Beatus *et al.* 2012),

$$\nabla^2 p = 0, \quad \left(\mathbf{v}_n + \frac{h^2 \nabla p}{12\eta} \right) \cdot \hat{n}_n = 0, \quad (24)$$

where \mathbf{v}_n is the velocity of n -th particle, \hat{n}_n is the unit vector normal to the surface of the n -th particle. The pressure gradient is taken at the outer boundary of the viscous layer of the n -th particle. However, since the latter is narrow, one can consider ∇p on the surface of the n -th particle without affecting significantly the solution for the pressure outside the viscous layers. To find the pressure inside the layers would require a separate study. For close droplets the pressure determined by Eq. (24) is different from the superposition of the dipole solutions given by Eqs. (4)-(5) due to the near-field interactions.

Finally, we demonstrate that the force exerted on the particles, determined by the viscous stress tensor at the particle surface, can be obtained from the ideal flow description. The force \mathbf{F}^n on particle n is determined by the following integral over the particle surface,

$$\mathbf{F}_i^n = \int_{S_n} \sigma_{ik} dS_k = \int_{outer} \sigma_{ik} dS_k, \quad (25)$$

where the last integral is over the outer boundary of the viscous layer of the n -th particle and we used $\nabla_k \sigma_{ik} = 0$. We can neglect the viscous contribution to the stress tensor at the outer boundary and find

$$\mathbf{F}^n \approx - \int_{outer} p dS \approx - \int_{S_n} p dS, \quad (26)$$

where we must use the pressure p determined from Eq. (24) in the last term and not the true pressure on the surface of the particle. Thus, the force coincides with that in an ideal flow and, effectively, we can assume that the ideal flow holds everywhere disregarding the no-slip boundary condition. This provides a consistent basis for the study of hydrodynamic interactions between large droplets at small distances as performed in Sarig *et al.* (2016); Beatus *et al.* (2012).

4. Multipole expansion

The flow at large distances from the particles can be effectively studied using the multipole expansion. The distances must be larger than h and much larger than the particle size. We perform here this expansion in terms of δp_n in Eq. (22), solution of the two-dimensional Laplace equation. We write δp_n as,

$$\begin{aligned} \delta p_n = & \frac{3}{\pi h^3} \int_{S_n} dS'_k (z'(z' - h) \sigma'_{ik} \nabla_i - \eta u_i(\mathbf{x}') (2\delta_{ik} \\ & + 2z'(h - z') \nabla_i \nabla_k + (h - 2z') (\delta_{kz} \nabla_i + \delta_{iz} \nabla_k))) \ln \rho. \end{aligned} \quad (27)$$

We provide next the expansion in Cartesian and polar coordinates as in three-dimensional electrostatics (Jackson 1962).

We set the origin of the coordinate system inside the n -th particle. To determine the multipole expansion in Cartesian coordinates we consider the Taylor series (remind that $\rho = |\mathbf{r} - \mathbf{r}'|$),

$$\ln \rho = \ln r - r'_l \nabla_l \ln r + \frac{r'_l r'_p}{2} \nabla_l \nabla_p \ln r + \dots, \quad (28)$$

where dots stand for higher-order terms. Substituting into Eq. (27) one obtains the Cartesian form of the multipole expansion. The leading-order $\ln r$ term in the series,

$$\begin{aligned} \delta p_n = & \frac{3}{\pi h^3} \int_{S_n} dS'_k (z'(z' - h) \sigma'_{ik} \nabla_i - \eta u_i(\mathbf{x}') (2\delta_{ik} \\ & + 2z'(h - z') \nabla_i \nabla_k + (h - 2z') (\delta_{kz} \nabla_i + \delta_{iz} \nabla_k))) \ln r, \end{aligned} \quad (29)$$

has a contribution proportional to $\int \mathbf{u} \cdot d\mathbf{S}$, i.e. proportional to $\ln r$. Further assuming the droplet is incompressible $\int \mathbf{u} \cdot d\mathbf{S} = 0$. In this case, the leading order term at larger distances is given by the dipole term,

$$\begin{aligned} \delta p_n = & \frac{3}{\pi h^3} \int_{S_n} dS'_k (z'(z' - h) \sigma'_{ik} \nabla_i - \eta u_i(\mathbf{x}') (h - 2z') \\ & (\delta_{kz} \nabla_i + \delta_{iz} \nabla_k)) \ln r + \frac{6\eta \nabla_l \ln r}{\pi h^3} \int_{S_n} r'_l \mathbf{u} \cdot d\mathbf{S}, \end{aligned} \quad (30)$$

where the last term comes from the next-order term in the expansion of the logarithm. This term can be simplified for droplets that do not change their shape, such as the pancake-like droplets considered previously, since the slip and flow on the surface are irrelevant. For instance, for a spherical droplet whose center moves with velocity \mathbf{v} , one obtains $\int_{S_n} r'_l \mathbf{u} \cdot d\mathbf{S} = v_k \int_{S_n} r'_l dS_k = 4\pi a^3 v_l / 3$. The complete expansion becomes,

$$\begin{aligned} \delta p_n = & \frac{3}{\pi h^3} \int_{S_n} dS'_k (z'(z' - h) \sigma'_{ik} \nabla_i - \eta u_i(\mathbf{x}') (2\delta_{ik} \\ & + 2z'(h - z') \nabla_i \nabla_k + (h - 2z') (\delta_{kz} \nabla_i + \delta_{iz} \nabla_k))) (\ln r \\ & - r'_l \nabla_l \ln r + \frac{r'_l r'_p}{2} \nabla_l \nabla_p \ln r + \dots). \end{aligned} \quad (31)$$

The expansion in polar coordinates is found observing that for $r' < r$,

$$\begin{aligned} \ln |\mathbf{r} - \mathbf{r}'| = & \ln r \\ & - \sum_{n=1}^{\infty} \left(\frac{r'}{r} \right)^n \frac{\cos(n\theta) \cos(n\theta') + \sin(n\theta) \sin(n\theta')}{n}. \end{aligned} \quad (32)$$

This formula represents the fundamental solution $\ln |\mathbf{r} - \mathbf{r}'|$ in terms of the elementary solutions of Laplace equation, $r^{-k} \exp(ik\theta)$ and $r'^p \exp(ip\theta')$ with k and p positive integers. This is the counterpart of the expansion of $|\mathbf{r} - \mathbf{r}'|^{-1}$ in spherical harmonics adopted in three-dimensional multipole expansion in electrostatics (Jackson 1962) and, in fact, it can be derived from that expansion

by confining \mathbf{r} , \mathbf{r}' in a plane. We provide here a simpler derivation. We consider,

$$\ln |\mathbf{r} - \mathbf{r}'| = \ln r + \frac{\ln(1 - 2\epsilon \cos \gamma + \epsilon^2)}{2}, \quad \epsilon = \frac{r'}{r}, \quad (33)$$

where γ is the angle between \mathbf{r} and \mathbf{r}' and $\epsilon < 1$. We recall the Fourier series,

$$\ln(1 - 2\epsilon \cos \gamma + \epsilon^2) = - \sum_{n=1}^{\infty} \frac{2\epsilon^n \cos(n\gamma)}{n} \quad (34)$$

where the integrals for the Fourier coefficients can be obtained using the residue theorem (Gradshteyn & Ryzhik 2014). Finally, introducing the polar angles θ and θ' for \mathbf{r} and \mathbf{r}' , respectively, and using $\gamma = \theta' - \theta$ we obtain Eq. (32). The multipolar expansion in polar coordinates is finally

$$\begin{aligned} \delta p_n = & \frac{3}{\pi h^3} \int_{S_n} dS'_k (z'(z' - h)\sigma'_{ik} \nabla_i - \eta u_i(\mathbf{x}') (2\delta_{ik} \\ & + 2z'(h - z')\nabla_i \nabla_k + (h - 2z')(\delta_{kz}\nabla_i + \delta_{iz}\nabla_k))) (\ln r \\ & - \sum_{n=1}^{\infty} \left(\frac{r'}{r}\right)^n \frac{\cos(n\theta) \cos(n\theta') + \sin(n\theta) \sin(n\theta')}{n}), \end{aligned} \quad (35)$$

which gives the pressure as a superposition of elementary solutions $r^{-k} \exp(ik\theta)$. The formulae simplify for rigid particles to

$$\begin{aligned} \delta p_n = & \frac{3}{\pi h^3} \int_{S_n} dS'_k z'(z' - h)\sigma'_{ik} \nabla_i (\ln r \\ & - r'_l \nabla_l \ln r + \frac{r'_l r'_p}{2} \nabla_l \nabla_p \ln r + \dots) \end{aligned} \quad (36)$$

and

$$\begin{aligned} \delta p_n = & \frac{3}{\pi h^3} \int_{S_n} dS'_k z'(z' - h)\sigma'_{ik} \nabla_i (\ln r \\ & - \sum_{n=1}^{\infty} \left(\frac{r'}{r}\right)^n \frac{\cos(n\theta) \cos(n\theta') + \sin(n\theta) \sin(n\theta')}{n}). \end{aligned} \quad (37)$$

The multipole expansion of the flow is derived by taking the gradient of the pressure using Eq. (21). As an example, the perturbation of the Poiseuille flow due to a rigid particle, δu_k , is up to a cubically decaying term,

$$\begin{aligned} \delta u_k = & -\frac{3z(z-h)s_i}{2\pi h^3 \eta} \nabla_k \nabla_i \ln r - \frac{3z(z-h)}{2\pi h^3 \eta} \\ & \times \nabla_k \nabla_i \frac{1}{r} \int_S dS'_k z'(z' - h)\sigma'_{ik} r' \cos(\theta - \theta'), \end{aligned} \quad (38)$$

where we introduced,

$$s_i = \int_S z'(h - z')\sigma'_{ik} dS'_k. \quad (39)$$

The first term in Eq. (38) is a dipole and the second term a quadrupole. Similarly, we can write the corresponding and higher-order terms for droplets.

5. Leading-order behavior at large distances

In this Section we consider the leading order behavior of the flow at large horizontal distances from the particle(s). The distances must be larger than h (but not much larger) and much larger than the size of the particles. The far-field flow perturbation $\delta\mathbf{u}_n$ due to the n -th particle is given by the dipole flow,

$$\delta\mathbf{u}_n = \frac{z(z-h)}{2\eta} \nabla \delta p_n, \quad \delta p_n = -\frac{3}{\pi h^3} (\mathbf{s}_n \cdot \nabla) \ln r, \quad (40)$$

where

$$\begin{aligned} (\mathbf{s}_n)_i &= \int_{S_n} dS'_k z' (h - z') \sigma'_{ik} + \eta \int_{S_n} dS'_z (h - 2z') u_i(\mathbf{x}') \\ &\quad + \eta \int_{S_n} dS'_i (h - 2z') u_z(\mathbf{x}') - 2\eta \int_{S_n} r'_i \mathbf{u} \cdot dS, \end{aligned} \quad (41)$$

see Eqs. (21) and (30). For rigid particles this reduces to Eq. (39) which is why we use the same letter for the coefficient s_i . We find that the perturbation of the potential of the depth-averaged flow $\delta\phi_n = -h^2 \delta p_n / (12\eta)$ is,

$$\delta\phi_n = -\frac{\tilde{\mathbf{d}}_n \cdot \hat{r}}{r}, \quad \tilde{\mathbf{d}}_n = -\frac{\mathbf{s}_n}{4\pi\eta h}, \quad (42)$$

see Eq. (4). Thus, the flow perturbation at large distances is the dipolar flow with effective dipole moment $\tilde{\mathbf{d}}$. We can describe the far-field impact of a particle of arbitrary shape on the flow introducing the source in the potential equation, $\nabla^2 \delta\phi_n = -2\pi(\tilde{\mathbf{d}}_n \cdot \nabla)\delta(x)\delta(y)$ so that the full potential ϕ obeys,

$$\nabla^2 \phi = -2\pi \sum_n (\tilde{\mathbf{d}}_n \cdot \nabla) \delta(x - x_n) \delta(y - y_n) \quad (43)$$

where (x_n, y_n) are the horizontal coordinates of some point inside the n -th particle (observe that p^0 is a linear function and has zero laplacian).

The resulting correction to the Poiseuille flow is that of a particle that moves in direction of \mathbf{s} , see Eq. (5),

$$\delta u_k(\mathbf{x}) = \frac{3s_i z(h-z)}{\pi\eta h^3(x^2 + y^2)} \left[\frac{\delta_{ik}}{2} - \frac{x_i x_k}{x^2 + y^2} \right], \quad \rho \gg h. \quad (44)$$

The lateral, y and z , components of \mathbf{s} vanish for particles that have fore-and-aft symmetry. This can be shown in the same way as for the absence of lateral migration of spheres in a channel (Leal 2007; Bretherton 1962). The reversal of the sign of \mathbf{u} and p produces another solution of the system of Eqs. (2). This solution has opposite sign of the stress tensor and velocity and thus of \mathbf{s} . However, it describes the same physical situation and thus must have the same lateral components of \mathbf{s} , hence these components must vanish. Thus, for spheres or ellipsoids $\mathbf{s} = s\hat{\mathbf{x}}$. In contrast, for particles whose shape is an arc or similar one can have a non-zero s_y and s_z .

We consider a spherical particle as an example of a particle with fore-and-aft symmetry. We can introduce $s_i = s(z_p)\delta_{ix}$ where,

$$\begin{aligned} s(z_p) &= \int_S z'(h-z')\sigma'_{xk}dS'_k + \eta \int_{S_n} dS'_z(h-2z')u_x(\mathbf{x}') \\ &\quad + \eta \int_{S_n} dS'_x(h-2z')u_z(\mathbf{x}') - 2\eta \int_{S_n} r'_x \mathbf{u} \cdot d\mathbf{S}, \end{aligned} \quad (45)$$

with z_p the vertical position of the particle center. In this case, the flow is

$$\begin{aligned} \delta u_x(\mathbf{x}) &= \frac{3s(z_p)z(h-z)}{2\pi\eta h^3(x^2+y^2)} \frac{y^2-x^2}{x^2+y^2}, \\ \delta u_y(\mathbf{x}) &= -\frac{3s(z_p)z(h-z)}{2\pi\eta h^3(x^2+y^2)} \frac{2xy}{x^2+y^2}. \end{aligned} \quad (46)$$

It is plausible that $s(z_p) > 0$ because the particle is always lagging behind the local flow. This is confirmed by the direct numerical simulations reported below.

The formulas provided here give the possibility of tabulating the particle dipole moments from numerical simulations for future use. For a spherical particle of fixed radius the dipole moment depends on the vertical coordinate z_p . The solution of the flow equations in the presence of an isolated sphere would give the stress tensor and the surface velocity with which we can find $s(z_p)$. We illustrate this procedure for the case where the particle is a rigid sphere with same density as that of the fluid. The equation of motion is,

$$m \frac{dv_i}{dt} = \int_S \sigma_{ik} dS_k, \quad (47)$$

where m is the mass of the particle and gravity does not influence the motion since the particle is assumed to be neutrally buoyant. This equation is coupled to the time- dependent Navier-Stokes equations where the unsteady term is not negligible during the transients. The particle eventually reaches a constant velocity and the fluid flow is governed by the steady Stokes equations due to the small Reynolds number. Thus, our derivation of the far flow holds with the dipole coefficient for rigid particles,

$$s(z_p) = \int_S z'(h-z')\sigma'_{xk}dS'_k. \quad (48)$$

We computed here $s(z_p)$ using the numerically determined σ_{xk} for different z_p and different radii of the sphere. The results are summarized in Tables 1 - 3 (see Appendix B for details). Here, the particle relative velocity, defined as $\delta u_p = u_p - u_x^0$ where u_p is the particle center velocity, is non-dimensionalized by the bulk velocity of the undisturbed channel flow $U_b = -h^2 \nabla_x p^0 / (12\eta)$. The reduction of the translational velocity as the particle is placed closer to one wall or as the particle size increases is consistent with previous computations using the boundary integral method (Staben *et al.* 2003).

We also report the values of $\hat{s}(z_p) = 3s(z_p)/(2\pi\eta h^3)$, the common pre-factor in Eq. (46). We note that, though overall small, \hat{s} increases as the particle

Table 1: Relative particle velocity $\delta u_p/U_b$ and magnitude of \hat{s} as function of the particle centre position z_p/h for spherical particles of radius $a = h/6$ obtained from the numerical simulations.

z_p/h	0.50	0.55	0.60	0.65	0.70	0.75	0.80
$\delta u_p/U_b$	-0.06	-0.06	-0.06	-0.07	-0.07	-0.09	-0.17
$\hat{s} \times 10^{-3}$	1.8	2.2	3.7	6.3	10.6	17.8	30.0

Table 2: Relative particle velocity $\delta u_p/U_b$ and magnitude of \hat{s} as function of the particle centre position z_p/h for spherical particles of radius $a = h/3$ obtained from the numerical simulations.

z_p/h	0.50	0.55	0.60	0.65
$\delta u_p/U_b$	-0.24	-0.25	-0.30	-0.45
$\hat{s} \times 10^{-2}$	4.1	4.9	7.6	15.8

Table 3: Relative particle velocity $\delta u_p/U_b$ and magnitude of \hat{s} as function of the particle centre position z_p/h for spherical particles of radius $a = h/2.25$ obtained from the numerical simulations.

z_p/h	0.50
$\delta u_p/U_b$	-0.52
$\hat{s} \times 10^{-1}$	2.1

approaches one wall or as the confinement increases, as the relative velocity δu_p . The resulting \hat{s} , quantifying the local velocity disturbance generated by one particle, along with the spatial dependence in the horizontal plane, allows for predictions of the far-field interactions of spheres. These will be examined in Section 6.

Finally, we return to the lubrication theory by showing some typical depth-average velocity field in Fig. 2 and the velocity decay in Fig. 3. As mentioned earlier, the lubrication theory is valid at horizontal distances larger than the height of the channel. Fig. 2 depicts the flow field due to a sphere of diameter equal to $2/3$ of the channel height. The non-zero vertical vorticity outside the particle indicates the non-ideal structure of the depth-average flow, in contrast to the simple mass dipole of a disk (see Eq. 5). However, as the confinement increases, the disturbance velocity asymptotes the leading-order quadratic decay, as shown in Fig. 3.

6. Interactions

In this Section, we introduce equations that describe interactions of well-separated particles and solve them in some specific cases. We start by observing

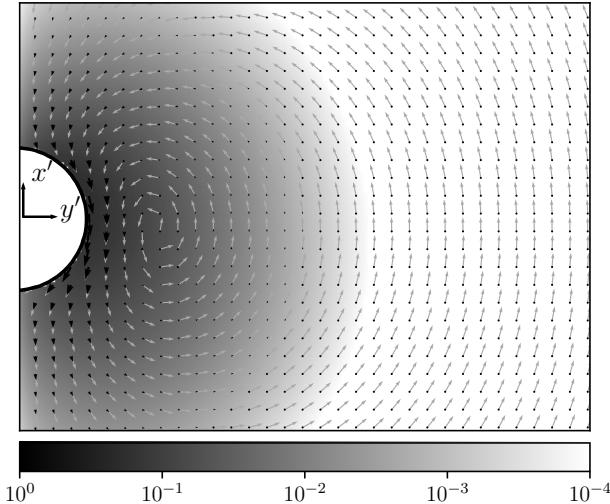


Figure 2: Depth-averaged disturbance flow around a sphere from the numerical simulations. The heavy arrows indicate the magnitude and light arrows the direction. The color in the background depicts the decay of the vorticity outside the sphere. The particle travels in the x' direction and is located at the mid-channel ($z_p/h = 0.5$), with $h/(2a) = 1.5$. Only half of the plane is shown due to symmetry.

that because of the linearity of the problem, the steady state (horizontal) velocity \mathbf{v}_0 of an isolated particle driven by the Poiseuille flow according to Eq. (2) is given by

$$\mathbf{v}_0 = -\hat{M}\nabla p^0, \quad (49)$$

where we assume \mathbf{v}_0 is a function of ∇p^0 that can be any constant vector in the plane. Indeed, \mathbf{v}_0 is a linear function of ∇p^0 that is zero when there is no driving flow. Since \hat{M} connects the velocity \mathbf{v}_0 with the force per unit volume of the fluid, we call \hat{M} the mobility matrix though it differs from the more commonly used coefficient between the velocity and the force on the particle (Happel & Brenner 1983). The two-by-two mobility matrix \hat{M} depends on the shape of the particle, whether the particle is rigid or droplet, and the particle position in the channel. The equation neglects gravitational settling, absent for neutrally buoyant particles or particles whose sedimentation is stopped by interactions with the walls, (as the pancake-like droplets) or because settling is negligible at relevant time scales. In cases with sedimentation velocity \mathbf{v}_s so low that the particle stays in quasi-steady state we have,

$$\mathbf{v}_0 = -\hat{M}(t)\nabla p^0 + \mathbf{v}_s, \quad (50)$$

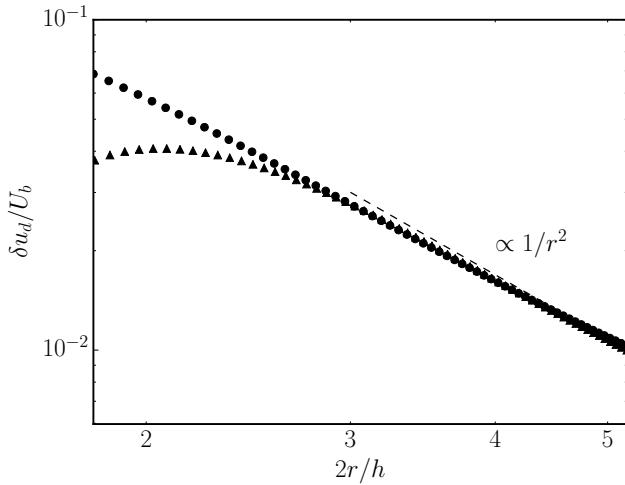


Figure 3: Spatial variation of the normalized streamwise depth-averaged disturbance-velocity, $\delta u_d/U_b$, along the streamwise ($y = 0$, circle) and spanwise ($x = 0$, triangle) directions away from the particle center. The particle is located at the mid-channel ($z_p/h = 0.5$), with $h/(2a) = 1.125$. The collapse of the disturbance-velocity away from the particle confirms the leading-order dipolar decay (dashed line).

where the matrix $\hat{M}(t)$ is determined by the instantaneous configuration in the channel, which may depend on time due to sedimentation.

We next consider interactions of many well-separated particles. The flow induced at the position of the i -th particle by the other particles is a quasi-Poiseuille flow,

$$\mathbf{u} = \frac{z(z-h)\nabla p}{2\eta}, \quad p = p^0 - \sum_{k \neq i} \frac{3}{\pi h^3} (\mathbf{s}_k \cdot \nabla) \ln |\mathbf{r} - \mathbf{r}_k|, \quad (51)$$

where \mathbf{r}_k is the horizontal position of the k -th particle, see Eqs. (21), (40). We observe that we can neglect variations of ∇p over the particle since the rest of the particles are well-separated. Thus, at the leading order in large distances between the particles the i -th particle assumes the horizontal velocity,

$$\frac{d\mathbf{r}_i}{dt} = -\hat{M}_i \nabla \left(p^0 - \sum_{k \neq i} \frac{3}{\pi h^3} (\mathbf{s}_k \cdot \nabla) \ln |\mathbf{r} - \mathbf{r}_k| \right)_{\mathbf{r}=\mathbf{r}_i}, \quad (52)$$

where \hat{M}_i describes the geometry of the i -th particle. The sedimentation velocity can be included in a straightforward way. This is the equation that describes the long-range interactions of the particles. The presented derivation avoids the problem with boundary conditions encountered in the derivation of

Shani *et al.* (2014) for the case of droplets. In that case, the derivation started with the flow induced by other particles at the position of the i -th particle and not the pressure. Since for particles of finite extent it becomes non-obvious where the three-dimensional flow must be considered, our derivation seems to be useful for a proper consideration of particles whose vertical size is smaller than h .

We consider the case of spherical particles or droplets of radius a smaller than $h/2$. In this case \hat{M}_i is $M(z_i)$ times the unit matrix where the scalar coefficient M depends on the vertical coordinate z_i of the i -th particle. Similarly $s_k = s(z_k)\hat{x}$ where $s(z)$ was introduced previously. We find,

$$\dot{\mathbf{r}}_i = -M(z_i)\nabla p^0 + \sum_{k \neq i} \frac{3M(z_i)s(z_k)}{\pi h^3 r_{ik}^2} \left[\hat{\mathbf{x}} - \frac{2(\mathbf{r}_{ik} \cdot \hat{\mathbf{x}}) \mathbf{r}_{ik}}{r_{ik}^2} \right],$$

where $\mathbf{r}_{ik} = \mathbf{r}_i - \mathbf{r}_k$. Thus for pair of particles,

$$\dot{\mathbf{r}} = (M(z_1) - M(z_2))\nabla p^0 + \frac{3\delta_{12}}{\pi h^3 r^2} \left[\hat{\mathbf{x}} - \frac{2x\mathbf{r}}{r^2} \right], \quad (53)$$

where $\mathbf{r} = \mathbf{r}_2 - \mathbf{r}_1 = (x, y, z)$ and we introduced,

$$\delta_{12} = M(z_2)s(z_1) - M(z_1)s(z_2). \quad (54)$$

Another case when Eqs. (52) simplify significantly is for pancake-like droplets that almost completely fill the channel in the vertical direction. In this case M and s are constant since no variation of the vertical position of the particles is possible. We see immediately that the configuration of two droplets is stable in the dipole approximation where $\dot{\mathbf{r}}$ in Eq. (53) is zero (in a higher-order quadrupole approximation proportional to r^{-3} the pair would not be stable). For many particles, the equations of motion in the frame that moves with the velocity of the isolated droplet $-M\nabla p^0$ become,

$$\frac{d\mathbf{r}_i}{dt} = \sum_{k \neq i} \frac{q}{r_{ik}^2} \left[\hat{\mathbf{x}} - \frac{2(\mathbf{r}_{ik} \cdot \hat{\mathbf{x}}) \mathbf{r}_{ik}}{r_{ik}^2} \right], \quad q = \frac{3Ms}{\pi h^3}. \quad (55)$$

These equations hold also for spherical particles located at the same distance from the mid-plane where we must use for M and s the values at the corresponding z . This is the case where the particles have identical vertical coordinate or their coordinates can be obtained by reflection with respect to the mid plane. Other cases of symmetric particles where Eq. (55) hold can be considered. If gravitational settling is relevant, \mathbf{r}_{ik} will change via time-dependent $s = s(z(t))$. It is assumed below that the change of $s = s(z(t))$ can be neglected over the time scales of interest.

It is often the case that we have two spherical particles at the same vertical distance from the walls. This can be the case of spherical droplets created at some fixed place in the channel and then transported down the flow Shen *et al.* (2014). In this case, an isolated pair is stable in the dipole approximation: we have $\dot{\mathbf{r}} = 0$ in Eq. (53) for $z_1 = z_2$.

This characterizes the basic property of the interaction given by Eq. (55), that the velocity induced by particle i at the position of the k -th particle is equal to the velocity induced by particle k at the position of the i -th particle. Thus the interparticle distances can change only if there are three or more particles. We can re-write the equation of motion as

$$\begin{aligned} \frac{d\mathbf{r}_{ik}}{dt} = & \sum_{l \neq i, l \neq k} \left(\frac{q}{r_{il}^2} \left[\hat{\mathbf{x}} - \frac{2(\mathbf{r}_{il} \cdot \hat{\mathbf{x}}) \mathbf{r}_{il}}{r_{il}^2} \right] \right. \\ & \left. - \frac{q}{r_{kl}^2} \left[\hat{\mathbf{x}} - \frac{2(\mathbf{r}_{kl} \cdot \hat{\mathbf{x}}) \mathbf{r}_{kl}}{r_{kl}^2} \right] \right). \end{aligned} \quad (56)$$

We start by considering in more detail the simplest case of two particles whose distance is constant in time. If the particles have the same y -coordinate then the x -coordinates obey,

$$\frac{dx_1}{dt} = \frac{dx_2}{dt} = -\frac{q}{(x_1 - x_2)^2}. \quad (57)$$

In this case the particles form a simple cluster with fixed distance that moves as a whole slower than the particles separately. We consider now two particles at different spanwise locations, $y_1 = y$, $y_2 = 0$,

$$\begin{aligned} \frac{dx_1}{dt} = \frac{dx_2}{dt} &= q \frac{(y_1 - y_2)^2 - (x_1 - x_2)^2}{[(x_1 - x_2)^2 + (y_1 - y_2)^2]^2}, \\ \frac{dy_1}{dt} = \frac{dy_2}{dt} &= -q \frac{2(x_1 - x_2)(y_1 - y_2)}{[(x_1 - x_2)^2 + (y_1 - y_2)^2]^2}. \end{aligned} \quad (58)$$

The RHSs are constant because inter-particle distances are but the velocity of the cluster of the two particles can change sign unlike the previous case. For two particles with the same x coordinate, the x - component of their velocity increases while the y -component is zero, see Shen *et al.* (2014) for experimental observations.

Next, we consider the simplest case with changing inter-particle distances: three particles at the same height. From the analysis of the two-particle dynamics, a possible solution is that particles form a cluster of two particles with the third farther away. The interactions of the single distant particle with the clustered particles decay quadratically with the distance and can be assumed negligible. Thus, the isolated particle moves with the velocity of one single sphere. The cluster keeps its configuration and moves at a constant velocity $(q/r^2)[\hat{\mathbf{x}} - 2(\mathbf{r} \cdot \hat{\mathbf{x}}) \mathbf{r}/r^2]$ with respect to the third particle. If this velocity is such to increase the separation between the cluster and the third particle, this solution will continue ad infinitum. It is thus plausible to assume that any arbitrary initial configuration of three particles will separate asymptotically in one cluster and one particle. We will prove this below for the practically important case of three particles aligned in the streamwise, x -, direction. This case can be observed when the particles are injected in the flow at the same location.

The distances between three particles are determined by two vectors \mathbf{r}_{12} and \mathbf{r}_{13} that obey,

$$\begin{aligned}\dot{\mathbf{r}}_{12} &= \frac{q}{r_{13}^2} \left[\hat{\mathbf{x}} - \frac{2(\mathbf{r}_{13} \cdot \hat{\mathbf{x}}) \mathbf{r}_{13}}{r_{13}^2} \right] - \frac{q}{r_{23}^2} \left[\hat{\mathbf{x}} - \frac{2(\mathbf{r}_{23} \cdot \hat{\mathbf{x}}) \mathbf{r}_{23}}{r_{23}^2} \right], \\ \dot{\mathbf{r}}_{13} &= \frac{q}{r_{12}^2} \left[\hat{\mathbf{x}} - \frac{2(\mathbf{r}_{12} \cdot \hat{\mathbf{x}}) \mathbf{r}_{12}}{r_{12}^2} \right] - \frac{q}{r_{23}^2} \left[\hat{\mathbf{x}} - \frac{2(\mathbf{r}_{23} \cdot \hat{\mathbf{x}}) \mathbf{r}_{23}}{r_{23}^2} \right].\end{aligned}\quad (59)$$

The solution described above pertaining the cluster of two particles (named here 2 and 3) and the faraway particle 1 corresponds to neglecting the first terms in the RHSs,

$$\dot{\mathbf{r}}_{12} \approx \dot{\mathbf{r}}_{13} \approx -\frac{q}{r_{23}^2} \left[\hat{\mathbf{x}} - \frac{2(\mathbf{r}_{23} \cdot \hat{\mathbf{x}}) \mathbf{r}_{23}}{r_{23}^2} \right] \approx \text{const}, \quad (60)$$

where \mathbf{r}_{23} is approximately constant. At large times the constant vector \mathbf{r}_{23} has become much smaller than the linearly growing \mathbf{r}_{12} and \mathbf{r}_{13} . We have that $\mathbf{r}_{23} = \mathbf{r}_{13} - \mathbf{r}_{12}$ obeys the equation,

$$\dot{\mathbf{r}}_{23} = \frac{q}{r_{12}^2} \left[\hat{\mathbf{x}} - \frac{2(\mathbf{r}_{12} \cdot \hat{\mathbf{x}}) \mathbf{r}_{12}}{r_{12}^2} \right] - \frac{q}{r_{13}^2} \left[\hat{\mathbf{x}} - \frac{2(\mathbf{r}_{13} \cdot \hat{\mathbf{x}}) \mathbf{r}_{13}}{r_{13}^2} \right],$$

where the RHS decays quadratically with time, in agreement with the assumption of constant \mathbf{r}_{23} .

We prove that the separation of 3 particles into one binary cluster and one isolated particle holds for arbitrary initial conditions when all three particles lie on the same line in the x -direction. It is clear that the separation can occur in two ways in this case: either particles 1 and 2 form a cluster or 2 and 3. Here we assume the ordering $x_1 > x_2 > x_3$. As the cluster moves slower than the isolated particle, the third particle would catch up with the binary cluster, made of 1 and 2. Hence the only stable configuration is a cluster of particles 2 and 3 whose distance from particle 1 increases linearly with time due to the cluster deceleration. Formally,

$$\begin{aligned}\dot{x}_{12} &= -q \left(\frac{1}{x_{13}^2} - \frac{1}{(x_{13} - x_{12})^2} \right), \\ \dot{x}_{13} &= -q \left(\frac{1}{x_{12}^2} - \frac{1}{(x_{13} - x_{12})^2} \right).\end{aligned}$$

Introducing $x = x_{12}$ and $r = x_{13}/x_{12}$ where $r > 1$, we can write

$$\begin{aligned}\dot{x} &= -\frac{q}{x^2} \left(\frac{1}{r^2} - \frac{1}{(r-1)^2} \right) = -\frac{q(1-2r)}{x^2 r^2 (r-1)^2}, \\ \dot{x} + \frac{x\dot{r}}{r} &= -\frac{q}{x^2} \left(\frac{1}{r} - \frac{1}{r(r-1)^2} \right).\end{aligned}\quad (61)$$

The distance r obeys,

$$\begin{aligned} x^3 \dot{r} &= -q \left(1 - \frac{1}{(r-1)^2} \right) + q \left(\frac{1}{r} - \frac{r}{(r-1)^2} \right) \\ &= -\frac{q(r-1)}{r} - \frac{q}{r-1} = -\frac{q(r^2-r+1)}{r(r-1)}. \end{aligned} \quad (62)$$

Hence $r(t)$ decreases in time monotonously and we can write

$$\begin{aligned} \frac{d \ln x}{ds} &= q \left(\frac{1}{(r-1)^2} - \frac{1}{r^2} \right), \\ \frac{dr}{ds} &= -\frac{q(r^2-r+1)}{r(r-1)}, \quad \frac{ds}{dt} = \frac{1}{x^3(t)}. \end{aligned} \quad (63)$$

We can solve for $s(r)$,

$$\begin{aligned} \frac{ds}{dr} &= -\frac{1}{q} + \frac{1}{q(r^2-r+1)}, \quad s(r) = \frac{r_0 - r}{q} \\ &+ \frac{2}{q\sqrt{3}} \arctan \left(\frac{2r-1}{\sqrt{3}} \right) - \frac{2}{q\sqrt{3}} \arctan \left(\frac{2r_0-1}{\sqrt{3}} \right), \end{aligned} \quad (64)$$

where $r_0 = r(s=0)$. The inversion of this formula, to find $r(s)$, gives a transcendental equation. However, the asymptotic properties of the solution can be derived without solving the equation. When s increases, r decreases reaching $r = 1$ at a finite value $s = s_*$ where,

$$s_* = \frac{r_0 - 1}{q} + \frac{\pi}{3q\sqrt{3}} - \frac{2}{q\sqrt{3}} \arctan \left(\frac{2r_0 - 1}{\sqrt{3}} \right). \quad (65)$$

The situation of r reaching 1 would correspond to coalescence of the second and the third particles. This happens only asymptotically, as $s = s_*$ corresponds to infinite physical time, $t(s_*) = \infty$. We have directly from Eq. (64) that $s'(r=1) = 0$ and $s'' = (1-2r)/[q(r^2-r+1)^2]$ which gives,

$$\begin{aligned} s(r) &\approx s_* + (r-1)s'(r=1) + \frac{(r-1)^2 s''(r=1)}{2} \\ &= s_* - \frac{(r-1)^2}{2q}, \quad (r-1)^2 = 2q(s_* - s). \end{aligned} \quad (66)$$

We can find x as a function of r observing that,

$$\frac{d \ln x}{dr} = \frac{d \ln x}{ds} \frac{ds}{dr} = - \left(\frac{1}{r-1} + \frac{1}{r} \right) \frac{1}{r^2-r+1}. \quad (67)$$

Integration of the above gives (with $x_0 = x(t=0)$),

$$\ln \left(\frac{x}{x_0} \right) = \ln \frac{r_0(r_0-1)}{r_0^2-r_0+1} - \ln \frac{r(r-1)}{r^2-r+1}, \quad (68)$$

where we used,

$$\int \left(\frac{1}{r-1} + \frac{1}{r} \right) \frac{1}{r^2-r+1} dr = \ln \frac{r(r-1)}{r^2-r+1}. \quad (69)$$

We find from Eq. (68) that,

$$x = x_0 \frac{r_0(r_0 - 1)(r^2 - r + 1)}{r(r - 1)(r_0^2 - r_0 + 1)}. \quad (70)$$

In the limit of large times where r approaches 1 from above we have,

$$x \approx \frac{r_0(r_0 - 1)x_0}{(r - 1)(r_0^2 - r_0 + 1)} \approx \frac{r_0(r_0 - 1)x_0}{(r_0^2 - r_0 + 1)\sqrt{2q(s_* - s)}}, \quad (71)$$

where we used Eq. (66). Finally we restore the physical time using,

$$\frac{dt}{ds} = x^3(s) \approx \frac{r_0^3(r_0 - 1)^3 x_0^3}{(r_0^2 - r_0 + 1)^3 (2q)^{3/2} (s_* - s)^{3/2}}, \quad (72)$$

and obtain,

$$t(s) \approx \frac{r_0^3(r_0 - 1)^3 x_0^3}{q(r_0^2 - r_0 + 1)^3 \sqrt{2q(s_* - s)}}. \quad (73)$$

We conclude from Eqs. (66), (71) that the long-time asymptotic form of the solution is,

$$x(t) = \frac{qt(r_0^2 - r_0 + 1)^2}{r_0^2(r_0 - 1)^2 x_0^2}, \quad r(t) = 1 + \frac{x_0^3 r_0^3 (r_0 - 1)^3}{qt(r_0^2 - r_0 + 1)^3}. \quad (74)$$

This implies that the distance between the second and the third particles reaches a constant value at large times,

$$\begin{aligned} x_{23}(t) &= x(t)(r(t) - 1) \approx \frac{x_0 r_0 (r_0 - 1)}{r_0^2 - r_0 + 1} \\ &= \frac{x_{13}(0)x_{23}(0)x_{12}(0)}{x_{13}(0)x_{23}(0) + x_{12}^2(0)}. \end{aligned} \quad (75)$$

To conclude, we can write,

$$x(t) = \frac{qt}{x_{23}^2}, \quad (76)$$

in agreement with the form given by Eq. (60). This relation proves the separation in cluster and faraway particle and provides the distance between the particles in the cluster as function of the initial conditions.

The obtained formulas provide a theoretical explanation for the pair exchange observed in the experiments by Shen *et al.* (2014), and confirmed by numerical simulations as illustrated in Fig. 4. We consider initial conditions for which particles 1 and 2 are close and the third particle is trailing behind. In this case $x_{12}^2(0) \ll x_{13}(0), x_{23}(0)$ and Eq. (75) becomes,

$$x_{23}(t) = x_{12}(0). \quad (77)$$

Thus, for long times the distance between the third and the second particle becomes equal to the initial distance between the first and the second particle, that is an exchange takes place.

We can also prove the separation in one cluster and one faraway particle for initial conditions where the particles are "almost" aligned, i.e. y_{12} and y_{13}

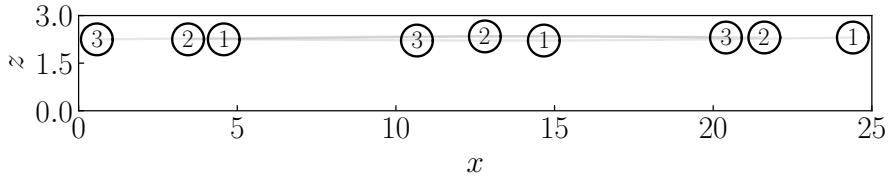


Figure 4: Pair exchange phenomenon as obtained from numerical simulations. Initially particles 1 are 2 are close and particle 3 is trailing behind the pair. As a result of hydrodynamic interactions the trailing particle is catching up with the pair, while the leading particle breaks away from the newly formed pair, whereas the trailing particles 2 and 3 are separated by the same distance as 1 and 2 were initially.

are much smaller than the smallest of x_{12} and x_{13} . If the y -components of the particle positions are linearly ordered, the equations for x_{ik} do not change and the evolution of x_{ik} is as above. The y -components obey,

$$\begin{aligned}\dot{y}_{12} &= -\frac{2qy_{13}}{x_{13}^3} + \frac{2qy_{23}}{(x_{13} - x_{12})^3}, \quad \dot{y}_{23} = \frac{2qy_{13}}{x_{13}^3} - \frac{2qy_{12}}{x_{12}^3} \\ \dot{y}_{13} &= -\frac{2qy_{12}}{x_{12}^3} + \frac{2qy_{23}}{(x_{13} - x_{12})^3},\end{aligned}\tag{78}$$

where $x_{ik}(t)$ are determined from the previous solution. Since x_{12} , x_{13} grow linearly with time at large times then the asymptotic form of the solution is,

$$y_{12} = y_{13} = \frac{2qct}{x_0^3}, \quad y_{23} = c = \text{const.}$$

This solution indicates that the first particle separates from the binary cluster at constant small but finite angle ϕ with respect to the x -direction given by,

$$\phi = \frac{y_{12}}{x_{12}} \approx \frac{y_{13}}{x_{13}} \approx \frac{c}{x_0 \sqrt{2}},\tag{79}$$

Self-consistency with the assumption of smallness of y_{ik} demands that $c \ll x_0$. This assumption can be guaranteed by the smallness of the initial conditions on y_{ik} because of the linearity of the equations in y_{ik} .

Thus we demonstrated the for initial conditions where the particles are aligned in the x -direction, or almost aligned, the solution at large times takes the form of a cluster of particles 2 and 3, those two initially upstream, with constant distance r_{23} and the first particle separating from the cluster according to Eq. (60).

We formulate the hypothesis that any arbitrary initial configuration of three particles will lead at large times to a binary cluster and the third particle linearly separating from it. It seems that the evolution from any arbitrary initial conditions cannot be solved analytically, but only numerically: a reduction from

four to three degrees of freedom can be obtained, but the resulting equations could not be solved.

To conclude, we describe the properties of cluster solutions assuming constant \mathbf{r}_{23} , of magnitude r_{23} , and angle ϕ with respect to the x -axis. Using Eq. (60), we write

$$\dot{x}_{12} \approx \dot{x}_{13} \approx \frac{q \cos(2\phi)}{r_{23}^2}, \quad \dot{y}_{12} \approx \dot{y}_{13} \approx \frac{q \sin(2\phi)}{r_{23}^2}.$$

We can assume with no loss of generality that $x_2 \geq x_3$ so that ϕ is in the range $-\pi/2 \leq \phi \leq \pi/2$. Four different solutions can therefore be identified, assuming the cluster at large time can be denoted as a point at the origin. In the range $0 \leq \phi \leq \pi/4$ the first particle leaves the cluster behind when going to infinity inside the first quadrant. In the range $-\pi/4 \leq \phi \leq 0$, the first particle leaves the cluster behind when going to infinity inside the second quadrant. In the case of $\pi/4 \leq \phi \leq \pi/2$ the cluster leaves the first particle behind and to the right. Finally in the case of $-\pi/2 \leq \phi \leq -\pi/4$ the cluster leaves the first particle behind and to the left.

It is clear from the above that the configuration with distant pairs or singlets of particles is stable: the singlets separate ballistically from the stable pairs which maintain the pair distance constant. In contrast, clusters of three and probably more particles are unstable. Thus, we conjecture that under arbitrary initial conditions an arbitrary number of distant particles will separate at large times into a collection of singlets and pairs if the solution is dilute. For suspensions of many particles this implies that hydrodynamic interactions increase the probability of particles to be isolated or in pairs, rather than forming clusters composed of many particles. In dense suspensions, the interactions discussed above do not have time to occur and we rather expect chaotic collisions of particles (Shani *et al.* 2014).

7. Concluding remarks

In the present paper, we provided a boundary integral representation for the flow due to particles (rigid ones or droplets) freely-suspended in a channel flow. The particle number, size and shape and the inter-particle distances are arbitrary. We have thus demonstrated the utility of this representation.

For an isolated particle, the proposed representation is useful for the study of the far-field flow. At the leading order, the far flow is dipolar flow with the dipole moment given by a weighted integral of the stress tensor and the flow over the particle surface. This defines the far flow completely. The flow was previously available only for strongly confined pancake-like droplets that almost block the channel in the vertical direction. We also determine the dipole moment integral numerically for neutrally buoyant rigid sphere. Further, we provide the multipole expansion from which the far flow can be found with any desired accuracy.

For close particles, the representation is helpful for the study of hydrodynamic interactions. It demonstrates clearly that the range of validity of the lubrication theory is larger than expected from the usual approach (Batchelor 1967; Tavakol *et al.* 2017; Szeri 2005; Bruce 2012). Our representation also solves the problem of matching the ideal flow holding far from the particles with the fully viscous flow near the particle surface. The solution is expressed in terms of the unknown surface velocity and stress tensor. This sheds light on the use of the ideal flow approximation in previous works on disk-like particles (Shani *et al.* 2014; Sarig *et al.* 2016) and helps to consider particles of other shapes.

We introduced the equation of motion of particles interacting at long distances, refining previous derivation for droplets (Shani *et al.* 2014) and extending it to the case of arbitrary, possibly different, interacting particles. We solved the three-body problem of hydrodynamic interactions for the case of identical symmetric particles aligned in the stream-wise direction. This solution provides theoretical support for the pair exchange phenomenon observed previously in experiments (Shen *et al.* 2014). We provide special solutions for the three-body problem and demonstrate that it is plausible that these solutions describe the long-time asymptotic evolution for arbitrary initial conditions. We further demonstrate the application of the theory to the many body problem.

We did not consider potential lateral migration induced by hydrodynamic interactions. It can be readily seen that a pair of distant spherical particles with different vertical coordinates will separate laterally because the coordinate-dependent mobility matrices and dipole moments will differ for these particles. The study of this instability will be the object of future work.

The representation proposed here is a good starting point for a mean-field description of strong hydrodynamic interactions of close particles. We notice after Eq. (15) that the effect of the interactions can be described by a change of the stress-tensors and velocities at the particle surfaces. Thus the model description of the interaction boils down to the model description of surface stress tensors and velocity. This can be done by introducing the mean field ∇p whose direction can differ from the direction of the undisturbed flow. We assume that the stress tensor and flow on the surface of each particle is that for an isolated particle in the Poiseuille flow with pressure gradient ∇p (e.g., the particle velocity is equal to minus the mobility matrix times ∇p). Using this in Eq. (15) one can find the flow in terms of ∇p . A closed integral equation for ∇p can then be obtained from the Stokes flow equations. The study of this equation is planned as future work.

This study was majorly motivated by recent experiments on the formation of droplet clusters in a microfluidic channel (Shen *et al.* 2014, 2016). The theoretical modeling in Shen *et al.* (2016) assumed that flow-assisted clustering of weakly confined spherical droplets in close proximity is driven by the combination of non-hydrodynamic (adhesive, e.g., depletion forces) and hydrodynamic interactions of dipolar nature similar to interactions of strongly confined (pancake-like)

droplets in Hele-Shaw cells. The qualitative agreement between the results of the numerical simulations and experimental results in Shen *et al.* (2016) suggested that *ad hoc* modeling of hydrodynamic interactions by dipolar flow is admissible. The present study shows that far-field interactions of weakly confined droplets are indeed of dipolar nature, however their magnitude is too weak to lead to relative motion between freely suspended particles on the time scale of the experiment. Moreover, the present study, as well as the calculations of the interactions at small distances in Beatus *et al.* (2012); Sarig *et al.* (2016), suggest that hydrodynamic interactions at close proximity cannot be described by dipolar flows. We thus believe that the reason for the qualitative agreement between the numerical results and the experiment (using unknown magnitude of the adhesive forces as an adjustable parameter) is that the adhesive force diverges at contact dominating the particle dynamics. The dipolar hydrodynamic interactions provided the source of sliding (tangential) motions of the particles necessary for the particle rearrangement and not provided by the adhesive (radial) forces. However, the particular functional form (e.g., dipolar or other) of these interactions seems to be of minor importance as long as these provide some tangential mobility. An accurate *quantitative* predictive theory of flow-assisted clustering requires the knowledge of the near-field hydrodynamic interactions, including an accurate treatment of the non-uniform flow near the inlet. This will be the object of a future work.

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Appendix A. Calculation of the far-field term $f_l(\mathbf{x}_0)$

In this appendix, we present a direct calculation of the far-field term $f_l(\mathbf{x}_0)$ in Eq. (12). We find from volume integration of Eq. (11) the boundary term,

$$\begin{aligned}
f_l(\mathbf{x}_0) = & \frac{1}{8\pi\eta} \int_{-L}^L dy \int_0^h dz [S_{il}(L - x_0, y - y_0, z, z_0) \\
& \sigma_{ix}(L, y, z) - S_{il}(-L - x_0, y - y_0, z, z_0)\sigma_{ix}(-L, y, z)] \\
& + \frac{1}{8\pi\eta} \int_{-L}^L dx \int_0^h dz [S_{il}(x - x_0, L - y_0, z, z_0) \\
& \sigma_{iy}(x, L, z) - S_{il}(x - x_0, -L - y_0, z, z_0)\sigma_{iy}(x, -L, z)] \\
& - \frac{1}{8\pi} \int_{-L}^L dy \int_0^h dz [T_{ilx}(L - x_0, y - y_0, z, z_0)u_i(L, y, z) \\
& - T_{ilx}(-L - x_0, y - y_0, z, z_0)u_i(-L, y, z)] - \frac{1}{8\pi} \\
& \times \int_{-L}^L dx \int_0^h dz [T_{ily}(x - x_0, L - y_0, z, z_0)u_i(x, L, z) \\
& - T_{ily}(x - x_0, -L - y_0, z, z_0)u_i(x, -L, z)]; \quad L \rightarrow \infty.
\end{aligned}$$

The Stokeslet decays exponentially in the z -direction so $f_z = 0$. To find the remaining components we use

$$\sigma_{ix}(L, y, z) = -L\nabla_x p^0 \delta_{ix} + \frac{(2z - h)\nabla_x p^0 \delta_{iz}}{2}. \quad (80)$$

We find that f_x is determined by the asymptotic solution for channel flow and is not affected by the presence of the spherical particle,

$$\begin{aligned}
f_x(\mathbf{x}_0) = & -\frac{L\nabla_x p^0}{8\pi\eta} \int_{-L}^L dy \int_0^h dz [S_{xx}(L, y - y_0, z, z_0) \\
& + S_{xx}(-L, y - y_0, z, z_0)] \\
& - \frac{\nabla_x p^0}{8\pi\eta} \int_{-L}^L dx \int_0^h dz [S_{yx}(x - x_0, L, z, z_0) \\
& - S_{yx}(x - x_0, -L, z, z_0)] \\
& - \frac{1}{8\pi} \int_{-L}^L dy \int_0^h dz \frac{z(z - h)\nabla_x p^0}{2\eta} [T_{xxx}(L, y - y_0, z, z_0) \\
& - T_{xxx}(-L, y - y_0, z - z_0)] \\
& - \frac{1}{8\pi} \int_{-L}^L dx \int_0^h dz \frac{z(z - h)\nabla_x p^0}{2\eta} [T_{xxy}(x - x_0, L, z, z_0) \\
& - T_{xxy}(x - x_0, -L, z - z_0)]; \quad L \rightarrow \infty. \quad (81)
\end{aligned}$$

Thus, this must be the unperturbed channel flow as readily verified. Rescaling the integration variable by L and keeping leading order terms we have,

$$\begin{aligned}
 f_x(\mathbf{x}_0) = & -\frac{L^2 \nabla_x p^0}{8\pi\eta} \int_{-1}^1 dy \int_0^h dz [S_{xx}(L, Ly, z, z_0) \\
 & + S_{xx}(-L, Ly, z, z_0)] \\
 & -\frac{L^2 \nabla_x p^0}{8\pi\eta} \int_{-1}^1 dx \int_0^h dzx [S_{yx}(Lx, L, z, z_0) \\
 & - S_{yx}(Lx, -L, z, z_0)] \\
 & -\frac{L}{8\pi} \int_{-1}^1 dy \int_0^h dz \frac{z(z-h)\nabla_x p}{2\eta} [T_{xxx}(L, Ly, z, z_0) \\
 & - T_{xxx}(-L, Ly, z - z_0)] \\
 & -\frac{L}{8\pi} \int_{-1}^1 dx \int_0^h dz \frac{z(z-h)\nabla_x p}{2\eta} [T_{xxy}(Lx, L, z, z_0) \\
 & - T_{xxy}(Lx, -L, z - z_0)]. \tag{82}
 \end{aligned}$$

So far the calculation involved the complete Stokeslet solution. To determine f_l we can use the asymptotic form of the Stokeslet at large distances, which is for the stress tensor,

$$T_{ilk} = -24 \frac{r_l}{\rho^2} (z_0/h^2) (1 - z_0/h) \delta_{ik} + O\left(\frac{1}{\rho^2}\right). \tag{83}$$

Using these formulas for S_{ik} and T_{ilk} , one can write

$$\begin{aligned}
 f_x = & \frac{\nabla_x p^0}{8\pi\eta} \int_{-1}^1 dy \int_0^h dz \frac{24z(z-h)z_0(z_0-h)(y^2-1)}{(1+y^2)^2 h^3} \\
 & -\frac{\nabla_x p}{4\pi\eta} \int_{-1}^1 dx \int_0^h dzx^2 \left[\frac{24z(z-h)z_0(z_0-h)}{(1+x^2)^2 h^3} \right] \\
 & -\frac{1}{4\pi} \int_{-1}^1 dy \int_0^h dz \frac{z(z-h)\nabla_x p}{2\eta} \left[24 \frac{z_0(z_0-h)}{(1+y^2)h^3} \right].
 \end{aligned}$$

Integrating over z , this can be written as,

$$\begin{aligned}
 f_x = & \frac{\nabla_x p z_0 (z_0-h)}{2\eta} \left(\int_{-1}^1 \frac{dy}{\pi(1+y^2)} \left[\frac{2}{1+y^2} - 1 \right] \right. \\
 & \left. + \frac{1}{\pi} \int_{-1}^1 dx \frac{2x^2}{(1+x^2)^2} + \frac{1}{\pi} \int_{-1}^1 dy \left[\frac{1}{(1+y^2)} \right] \right). \tag{84}
 \end{aligned}$$

Performing the integrals we confirm that indeed f_x is the flow given by Eq. (1).

Appendix B. Numerical integration of s

We compute the weighted dipole moment \mathbf{s} in Eq. (39) by directly simulating a rigid spherical particle of radius a transported in a doubly-periodic channel using the immersed boundary method (IBM), see Breugem (2012); Lambert *et al.* (2013); Picano *et al.* (2015) for more details and validations.

In the IBM, there are two meshes; one Eulerian mesh for the flow and one Lagrangian mesh for the moving particle. The two meshes are coupled through a multidirect forcing scheme that ensures the approximate no-slip/no-penetration condition on the particle surface.

The motion of the particle is described by the Newton-Euler equations, given for the translational velocity by Eq. (47)). The equation does not contain the gravitational force which is assumed to be balanced either by the particle interactions with the bottom wall (the case of particle near the wall) or by buoyancy (the case of density-matched particle). We also assume that particle-wall collisions are absent (*cf.* Eq. (3a) in Breugem (2012)).

The flow outside the particle is governed by the incompressible Navier-Stokes (NS) equations with the no-slip boundary conditions on the surface of the rigid particle described in connection with Eq. (2). Although the steady state flow obeys the Stokes equations, computation of the transients demands inclusion of the time derivative in the NS equations. In our simulations, the full NS equations are computed at a small Reynolds number ($\sim 10^{-1}$), *viz.*

$$Re \left(\frac{\partial \mathbf{u}}{\partial t} + \nabla \cdot (\mathbf{u}\mathbf{u}) \right) = \nabla \cdot \boldsymbol{\sigma} + \mathbf{f}, \quad (85)$$

where $Re = \rho_f U_b (2a)/\eta$ is the Reynolds number, U_b the channel bulk velocity, ρ_f ($= \rho_p$) the fluid density and \mathbf{f} the IBM force enforcing that the no-slip boundary condition (in this formulation pressure is rescaled by Re .) We discretize these equations using a second-order finite volume scheme. Finding the flow at given translational and rotational particle velocities we obtain the viscous stress which is used for updating these velocities as in Eq. (47). Numerically, the LHS of Eq. (47) is computed at each time step by summing the forces exerted on all the Lagrangian points, in addition to the volumetric forces inside the particle (see Eq. (8a) in Breugem (2012) for the full expression). In our case, this is simply

$$m \frac{d\mathbf{v}}{dt} \approx - \sum_{l=1}^{N_l} \mathbf{F}_l \Delta V_l + \rho_f \frac{d}{dt} \left(\int_{V_p} \mathbf{u} dV \right), \quad (86)$$

where $-\mathbf{F}_l$ the force acting on the l Lagrangian point centred at a shell element of volume ΔV_l , and N_l the total number of Lagrangian points.

At the steady state,

$$\int_S \boldsymbol{\sigma} \cdot d\mathbf{S} \approx - \sum_{l=1}^{N_l} \mathbf{F}_l \Delta V_l, \quad (87)$$

corresponding to the solution of the steady state the Stokes equations, Eq. (2).

Provided that the interpolation and spreading between $\mathbf{f}_{i,j,k}$ and $-\mathbf{F}_l$ preserves the local stress, we obtain the dimensionless weighted dipole moment $\tilde{\mathbf{s}}$ needed to compute particle interactions as

$$\tilde{\mathbf{s}} = - \sum_{l=1}^{N_l} z_l (h - z_l) \mathbf{F}_l \Delta V_l. \quad (88)$$

The dimensional s is thus $\eta U_b(2a)^3 \tilde{s}$.

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Paper 4

4

The theory of hydrodynamic interaction of two spheres in wall-bounded shear flow

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The seminal Batchelor-Green's (BG) theory on the hydrodynamic interaction of two spherical particles of radii a suspended in a viscous shear flow assumes unbounded fluid. In the present paper we study how a rigid plane wall modifies this interaction. Using an integral equation for the surface traction we derive the expression for the particles' relative velocity as a sum of the BG's velocity and the term due to the presence of a wall at finite distance, z_0 . Our calculation is not the perturbation theory of the BG solution, so the contribution due to the wall is not necessarily small. We indeed demonstrate that the presence of the wall is a singular perturbation, i.e., its effect cannot be neglected even at large distances. The distance at which the wall significantly alters the particles interaction scales as $z_0^{3/5}$. The phase portrait of the particles' relative motion is different from the BG theory, where there are two singly-connected regions of open and closed trajectories both of infinite volume. For finite z_0 , besides the BG's domains of open and closed trajectories, there is a domain of closed (dancing) and open (swapping) trajectories that do not materialize in an unbounded shear flow. The width of this region grows as $1/z_0$ at smaller separations from the wall. Along the swapping trajectories, that have been previously observed numerically, the incoming particle is turning back after the encounter with the reference particle, rather than passing it by, as the BG theory anticipates. The region of dancing trajectories has infinite volume and is separated from a BG-type domain of closed trajectories that becomes compact due to presence of the wall. We found a one-parameter family of equilibrium states that were previously overlooked, whereas the pair of spheres flows as a whole without changing its configuration. These states are marginally stable and their perturbation yields a two-parameter family of the dancing trajectories, whereas the test particle is orbiting around a fixed point in a frame co-moving with the reference particle. We suggest that the phase portrait obtained at $z_0 \gg a$ is topologically stable and can be extended down to rather small z_0 .

of several particle diameters. We confirm this hypothesis by direct numerical simulations of the Navier-Stokes equations with $z_0 = 5a$. Qualitatively the distant wall is the third body that changes the global topology of the phase portrait of two-particle interaction.

1. Introduction

Small particles, droplets and bubbles are ubiquitously present in flowing fluids. When a suspended particle is transported by a viscous fluid, it modifies the flow around it. If another particle happens to be in the region of the modified flow, mutual hydrodynamic interactions between the particles will take place. The interactions are given implicitly by imposing boundary conditions on the flow that must hold simultaneously on the surfaces of all interacting particles (Happel & Brenner 1983; Kim & Karrila 2013). This setting is inconvenient for analyses, both theoretical and numerical. Thus there is no answer to even simplest questions, for instance whether there can be a non-trivial stationary configuration of particles that would flow as a whole due to the hydrodynamic interaction. Although these interactions somewhat resemble electrostatic interactions, there is no a hydrodynamic counterpart of Earnshaw's theorem (Purcell & Morin 2013) stating that such simple configurations are impossible. Here we provide an example of this possibility in the presence of a boundary and demonstrate that boundaries can have surprising and non-trivial effects on hydrodynamic interactions.

The only well-studied case of hydrodynamic interactions of particles transported by non-uniform flow is the case of two particles in a time-independent low Reynolds number linear flow. This was studied in the seminal Batchelor & Green (1972a) paper, see also Arp & Mason (1977) for more details and an account of various contributions to the problem. If the particles' (and fluid) inertia can be entirely neglected, the vector between the particle centers obeys an autonomous first-order evolution equation, which gives its rate of change as a function of the instantaneous value. The use of the symmetries makes it possible to quantify the interaction by the two scalar functions of the distance, which have been tabulated (Batchelor & Green 1972a), see also Kim & Karrila (2013). This case presents no stationary configurations for the two particles. One of the main applications is the Poiseuille flow in the channel shown in Fig. 1. If both particles are far from the walls, $z_0 \gg a, r$, they can seemingly be considered as flowing in an unbounded shear flow and the analysis of Batchelor & Green (1972a); Arp & Mason (1977); Lin *et al.* (1970) applies. The BG theory thus predicts that there are no possible stationary configurations of the particle pair. Here we demonstrate that the approximation of an unbounded flow overlooks such configurations and also other phenomena, which hold independently of how large z_0 is, cf. Zurita-Gotor *et al.* (2007). Thus, the presence of the wall exhibits a singular perturbation of the BG theory.

Recently, stationary configurations of particles transported in microfluidic channels attracted attention due to the possibility of flow-assisted microfabrication by using a combination of hydrodynamic and non-hydrodynamic (*i.e.* adhesive) interactions (Shen *et al.* 2014, 2016). Under certain conditions, the particles can self-assemble into clusters of different morphology that flow with no change of inter-particle distances, see Ge *et al.* (2019) for detailed discussions. These micron-scale clusters can then be solidified and collected from the flow, and be potentially used for fabrication of functional metamaterials. To theoretically explain and subsequently predict the structure formation of the suspended particles observed in experiments, Shen *et al.* (2016) proposed a model based on a dipolar asymptotic form of hydrodynamic interaction. Notice that the dipolar form only holds at large particle separations (it was derived in detail using the fundamental solution for the channel flow (Liron & Mochon 1976), see Fouxon *et al.* (2017) and also references therein). One reason for the apparent applicability of this description, despite the particles were *close to* each other, is likely the dominance of the adhesive radial forces between particles at close proximity, such that any hydrodynamic interaction producing a non-zero tangential velocity component would yield a similar cluster formation dynamics (see Ge *et al.* (2019) for further evidence). In contrast, a consistent predictive theory of hydrodynamic interactions should hold irrespective of the presence of adhesive forces, and allow for analysis of interaction of flowing particles at close proximity and near the wall, as in experimental setup (Shen *et al.* 2016). The present paper is a step toward this theory.

There are two differences between the channel flow and the unbounded shear flow considered in the BG theory (Lin *et al.* 1970; Batchelor & Green 1972a). The velocity profile of the channel flow is quadratic (Haber & Brenner 1999) in the coordinate rather than linear. This difference is often irrelevant when the interacting particles are located much closer to one of the channel walls, so that the flow can be closely approximated by the linear shear flow. This is the case we consider in the present paper. Another difference, is that the no-slip rigid wall is always at a finite distance and it interacts with the flowing particles.

We first consider the evolution of the inter-particle distance when the effect of the wall is neglected and the BG theory applies. It is useful to consider the three-dimensional phase space spanned by all possible distances \mathbf{r} between the spheres' centers where one of them is at the origin. The distance $\mathbf{r}(t)$ between the spheres' centers obeys an autonomous evolution equation which means that there is a well-defined phase space flow $\mathbf{V}^0(\mathbf{r})$ such that $\dot{\mathbf{r}} = \mathbf{V}^0(\mathbf{r}(t))$ and a unique trajectory passes through each point. This is the consequence of neglecting particles' and fluid inertia and the translational invariance due to which the shear resistance matrix depends on \mathbf{r} only, cf. Brenner & O'Neill (1972) (translation in a linear flow changes the flow by a constant vector, irrelevant by Galilean invariance). The flow $\mathbf{V}^0(\mathbf{r})$ does not vanish anywhere so that there are no steady configurations. The absence of critical points with

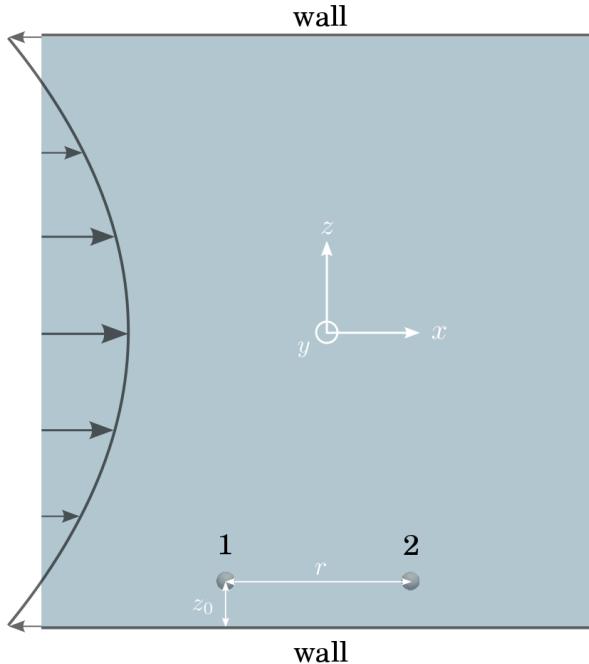


Figure 1: Setup of a particle pair in the Poiseuille flow (depicted in a comoving reference frame). In this work we study the case $z_0 \gg a$, where a is the particle radius, however does not necessarily require $z_0 \gg r$. The upper wall is assumed to be much further away from the particle pair than z_0 .

$\mathbf{V}^0(\mathbf{r}) = 0$ implies a simple structure of the phase space. This can be most readily observed in the symmetry plane formed by the horizontal flow direction x and the vertical velocity gradient direction z (see Fig. 2). The trajectories that belong to the plane never leave it, $V_y(y=0) = 0$, and can be considered separately. There is a simple dichotomy of the trajectories: closed trajectories crossing the x -axis and open trajectories that do not cross the x -axis. The open trajectories describe the faster particle overtaking the slower one. The particles return to their original vertical positions following the hydrodynamic encounter and there is fore-and-aft symmetry of the phase portrait. In contrast, the trajectories that cross the x -axis are closed, corresponding to a bound pair of spheres orbiting around each other. Open and closed trajectories are separated by the separatrix that touches the x -axis asymptotically at large distances (Lin *et al.* 1970). Rotation of this separatrix around the z -axis creates an axisymmetric surface that separates the regions of open and closed trajectories in space (it is not readily evident how this axial symmetry could be guessed *a priori* without writing down the equations). The region of closed trajectories has an infinite volume, which presents difficulties in, e.g., calculation of the

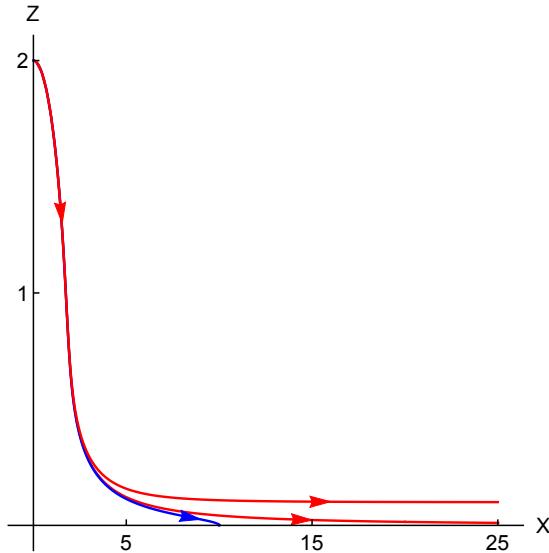


Figure 2: Phase portrait of the trajectories in the symmetry xz -plane, within the BG approximation of $z_0 = \infty$. All lengths are scaled with the particle radius a . Throughout the paper the reference sphere is at the origin and the trajectories of the second sphere are shown. Due to the fore-and-aft and top-down symmetries only one quadrant is depicted. The two types of the trajectories – closed (blue) and open (red), are separated by the separatrix, the open trajectory that asymptotically approaches the x -axis (Lin *et al.* 1970; Batchelor & Green 1972a). For two spheres at the same vertical line, the maximal separation for closed trajectories is of order of 10^{-5} , see Arp & Mason (1977). As a result, at this resolution, the trajectories are indistinguishable when approaching the z -axis. This includes the shown open trajectory that crosses the z -axis slightly above the closed trajectories. The time-period of revolution along the shown closed trajectory is more than 700 (here and thereafter the time units of inverse shear rate $\dot{\gamma}^{-1}$ are used).

effective viscosity of a dilute hard-sphere suspension at the quadratic order in concentration (Batchelor & Green 1972b).

We demonstrate here that when a distant wall is considered, the evolution of \mathbf{r} remains autonomous in the leading approximation, $\dot{\mathbf{r}} = \mathbf{V}(\mathbf{r}(t))$. Thus at any finite $z_0 \gg a$ we can still examine the phase portrait, which is however qualitatively different from that in Fig. 2. Our calculation is not a perturbation theory of the BG solution as we do not assume $\mathbf{V} \approx \mathbf{V}^0$, so the disturbed phase space flow $\mathbf{V}(\mathbf{r})$ is significantly different from \mathbf{V}^0 . The change in topology occurs because at finite z_0 there exist critical points at which $\mathbf{V}(\mathbf{r})$ vanishes, see the phase portrait in the symmetry plane in Fig. 3. The saddle (hyperbolic)

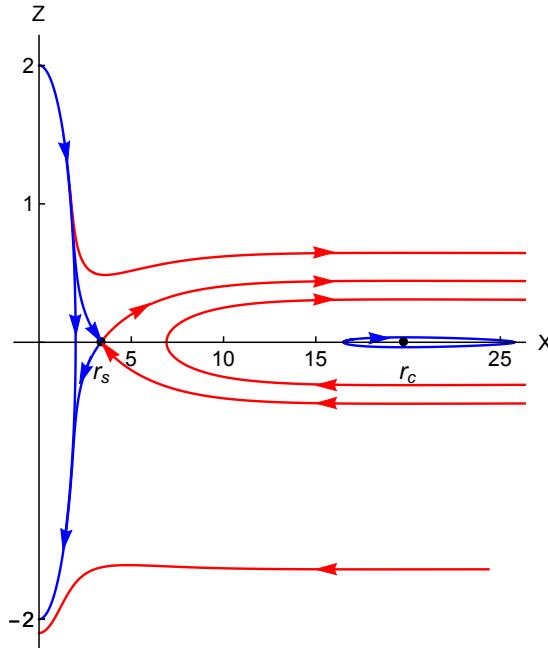


Figure 3: The phase portrait in the symmetry xz -plane at finite $z_0 = 5$. The fore-and-aft and top-down symmetries survive the wall perturbation in the leading order. The phase portrait exhibits two critical (equilibrium) points: the saddle (hyperbolic) point r_s and the neutral equilibrium (elliptic) point r_c , representing a completely different topology from the BG theory in Fig. 2. As $z_0 \rightarrow \infty$, the topology of the phase portrait is preserved, while the critical points are being shifted to infinity.

point r_s , the closer of the two critical points to the origin, is unstable. The other neutral equilibrium (elliptic) point r_c corresponds to a marginally stable configuration, where the pair flows without changing its inter-particle distance and orientation, see Fig. 4(a). Not too large deviations from this state result in the sphere orbiting around this elliptic point. These dancing trajectories would have rather unusual appearance when considered in the laboratory frame: while one sphere travels downstream, the other sphere revolves around a point co-moving in space with the first sphere, see Fig. 4(b). The phase plane at $x > 0$ is characterized by two disconnected regions of closed trajectories shown in Fig. 3 by the blue curves. The region to the left of the separatrix (blue) that crosses $x = r_s$ resembles the BG's closed trajectories. The trajectories around the elliptic point $(r_c, 0)$ are solely due to the presence of the wall. The lowest red curve is an open trajectory similar to the BG's: after the encounter the vertical separation is restored to its initial value. In contrast to that, along

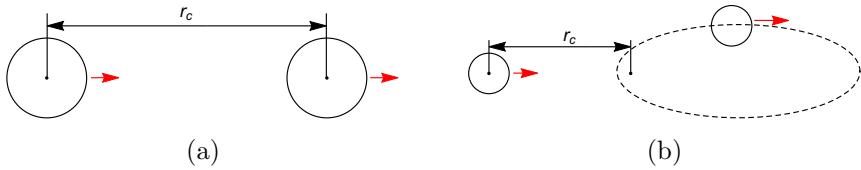


Figure 4: (a) There is a unique value of the stream-wise separation distance between two particles, $x = r_c$, flowing along the same streamline of the Poiseuille flow, for which they flow steadily without changing their configuration. The stability of such motion is marginal, cf. (b). The value $r_c = 4z_0$ is confirmed in the numerical simulations of the Navier-Stokes equations for $z_0 = 5$. (b) Trajectories that pass through points around the stationary point $x = r_c$, $z = 0$, crossing x -axis at distance larger than $2\sqrt{2}z_0$, exhibit a peculiar dancing dynamics. In the coordinate frame co-moving with the trailing (left) particle, the leading (right) sphere follows an elongated closed orbit around $(r_c, 0)$. Similar trajectories hold outside the symmetry plane.

the (red) trajectories circumventing the elliptic point, the vertical separation of the particles reverses sign after the encounter. We call these “swapping trajectories”, since they seem to correspond to the numerical findings of Zurita-Gotor *et al.* (2007) at $z_0 \sim a$, where trajectories with particles swapping their vertical positions after the encounter in a channel flow were reported (careful consideration of the Figure presented in Zurita-Gotor *et al.* (2007) reveals slight changes of the vertical coordinates which seem to be a higher order effect than that considered here). The phase portrait for the evolution of inter-particle distance along the swapping trajectory obtained in Zurita-Gotor *et al.* (2007) numerically, agrees with that predicted here theoretically. To prove that the sign-reversal of the vertical separation predicted here implies swapping, it has to be shown that the center of mass of the pair is not displaced vertically as a result of the encounter. We leave the rigorous proof for future work, focusing here on the evolution of inter-particle distance only. Thus, the use of the term “swapping trajectories” here, strictly speaking, refers to open trajectories with sign-reversal of the vertical separation following the encounter.

The three-dimensional trajectories are more complex. The circle of radius r_c around the z -axis provides the critical curve with $\mathbf{V}(\mathbf{r}) = 0$. The configurations with \mathbf{r} on that circle are stationary, so that for instance there is a stationary pair where only the y -coordinates of the particles are different. Displacements from these stationary configurations result in closed trajectories that loop around the critical circle, see Fig. 5. In contrast with the symmetry plane, where the BG trajectories display no behavior similar to dancing, some of the three-dimensional BG trajectories do look rather similar (notice that it was not stressed in the original work or in Arp & Mason (1977)), see Fig. 6 for comparison.

We emphasize the topological difference between the phase portrait in Fig. 3 and that of the BG theory in Fig. 2. There are two disconnected regions of closed trajectories. In one region the particles orbit each other, similarly to $z_0 = \infty$ approximation, however the volume of this region is finite. The other region contains dancing closed trajectories and at large $x^2 + y^2$ it is bounded by the surface of revolution $|z| \propto z_0(x^2 + y^2)^{-3/2}$. This is similar to the BG bounding surface, $|z| \propto (x^2 + y^2)^{-3/2}$, however boosted by the large z_0 factor. In both cases the volume of the phase space domain containing closed trajectories diverges, so the divergences in the second order in particle concentration stress calculations of Batchelor & Green (1972b) are not regularized by the wall. At finite z_0 the two regions of closed trajectories are separated by a region of a new type of open swapping trajectories that, in contrast to the BG theory, cross the x -axis. Then the top-down symmetry, which holds remarkably in the presence of the wall, implies that for open trajectories that cross the x -axis, the vertical component of the inter-particle distance reverses its sign, as in numerically observed swapping trajectories Zurita-Gotor *et al.* (2007). At least some features of the presented topology, derived theoretically at $z_0 \gg a$, work accurately down to $z_0 = 5a$, as demonstrated by our in-house numerical simulations of the Navier-Stokes equations.

In the next Section we present the detailed derivation of the evolution equation for the inter-particle distance in the wall-bounded shear flow. Sec. 3 demonstrates why the wall presents a singular perturbation of the BG theory. In Sec. 4 we review the BG trajectories that serve as the reference point of our study. We present the results of the numerical solutions of the derived evolution equation in Sec. 5. Sec. 6 presents full solution of equation of motion in the dancing-swapping region. Section 7 presents the confirmation of the theory by direct numerical simulations of the motion of a pair of spheres in the Poiseuille flow. In the last Section we conclude our results, discuss the applicability of the BG theory and formulate some open questions.

2. Evolution of the distance between two particles transported by a shear flow near wall

In this Section we derive the autonomous evolution equation for the distance $\mathbf{r}(t)$ between two spheres transported by the Poiseuille flow. We make the simplifying assumption that both spheres are much closer to one of the bounding walls than the other. Thus the particles are effectively transported by the shear flow and not the parabolic velocity profile. The hydrodynamic interaction of particles transported by an unbounded shear flow are well-studied and their velocities \mathbf{V}_α^0 , $\alpha = 1, 2$ were considered in Batchelor & Green (1972a). This analysis serves as a starting point of our study. We also assume that the distance to the wall is much larger than the particles' radii. We derive the particles' relative velocity as a sum of \mathbf{V}_α^0 and the correction velocity $\delta\mathbf{V}_\alpha$. The correction velocity is not necessarily smaller than \mathbf{V}^0 , as our solution is not a perturbation around the solution for an unbounded shear flow.

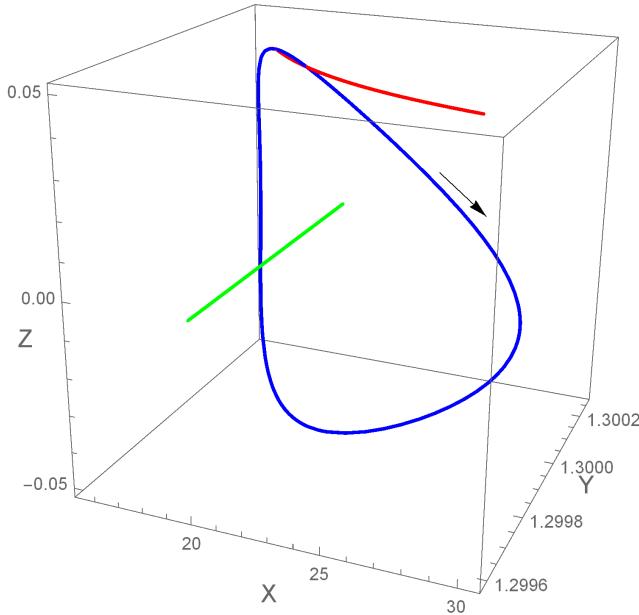


Figure 5: The blue line depicts the closed trajectory that forms a loop around the critical circle of radius r_c (whose segment is shown by the green line) for $z_0 = 5$. The trajectory can be shrunk to a (necessarily critical) point by continuously changing the initial conditions. The red line shows the BG trajectory that starts from the same initial condition as the blue line. The period of revolution along the closed trajectory is 1165.

2.1. Direct approach

We set the problem and consider its formulation using the flow for infinitely separated walls as a reference. The problem of two spheres driven by the Poiseuille flow is described by,

$$\begin{aligned} \nabla p &= \eta \nabla^2 \mathbf{u}, \quad \nabla \cdot \mathbf{u} = 0, \quad \mathbf{u}(z=0) = \mathbf{u}(z=h) = 0, \\ u_x(\infty) &= \frac{z(z-h)\nabla_x p^0}{2\eta}, \quad \mathbf{u}(S_\alpha) = \mathbf{V}_\alpha + \boldsymbol{\Omega}_\alpha \times (\mathbf{x} - \mathbf{x}_\alpha), \end{aligned} \quad (1)$$

where $\alpha = 1, 2$ are the indices of the spheres, x_α are the coordinates of the centers and S_α is the surface of the α -th sphere. We designate the flow by \mathbf{u} , and the translational and rotational velocities of the spheres by \mathbf{V}_α and $\boldsymbol{\Omega}_\alpha$. The constant pressure gradient $\nabla p^0 = -|\nabla_x p^0| \hat{\mathbf{x}}$ drives the flow in the positive x -direction, η is the fluid viscosity, z is the vertical coordinate and h the channel height. We assume that the spheres have equal radii a , although

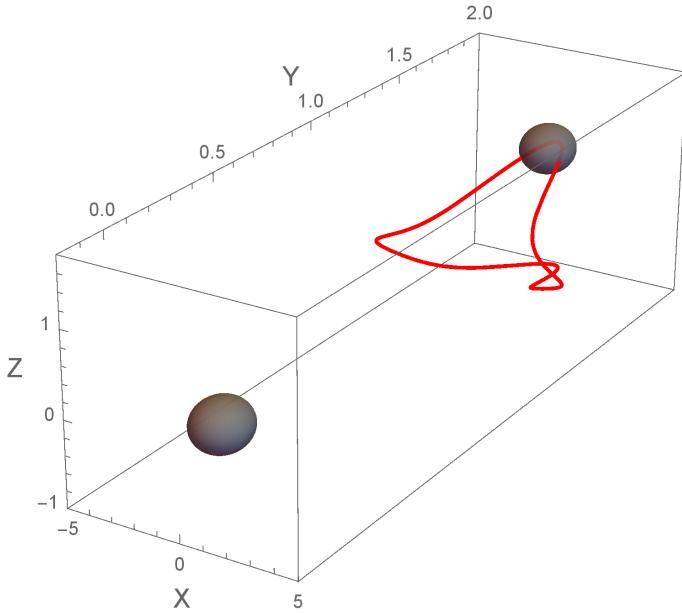


Figure 6: Three-dimensional BG trajectory (red curve) of the sphere in the frame co-moving with the reference sphere. The closed trajectory has a geometrical center on the y -axis. In contrast to the trajectory in Fig. 5, this curve cannot be shrunk to a point by a continuous change of the initial conditions. That point would have to be critical and the BG phase space does not admit those. For this trajectory, a distant wall is only a regular small perturbation. The period of revolution is 120.

most of the calculations below can be done without this assumption. We will use below a as the unit of length so that the radii are 1. We assume that the particle inertia is negligible so that the values of \mathbf{V}_α and $\boldsymbol{\Omega}_\alpha$ are determined from the conditions that the total force and torque from the fluid on either particle is zero,

$$\int_{S_\alpha} \mathbf{t} dS = 0, \quad \int_{S_\alpha} (\mathbf{x} - \mathbf{x}_\alpha) \times \mathbf{t} dS = 0, \quad (2)$$

where we have introduced the surface traction \mathbf{t} , which can be written via the stress tensor σ_{ik} as,

$$t_i(\mathbf{x}) = \frac{\sigma_{ik}(\mathbf{x} - \mathbf{x}_\alpha)_k}{a}, \quad \sigma_{ik} = -p\delta_{ik} + \eta(\nabla_i u_k + \nabla_k u_i), \quad (3)$$

where \mathbf{x} belongs to S_α . We observe that if the spheres are much closer to the wall at $z = 0$ than at $z = h$, then we can use different boundary conditions in

Eq. (1),

$$\mathbf{u}(z=0) = 0, \quad \mathbf{u}(\infty) = \dot{\gamma} z \hat{\mathbf{x}}, \quad \dot{\gamma} = \frac{h |\nabla_x p^0|}{2\eta}, \quad (4)$$

where we introduced the effective shear rate $\dot{\gamma}$ in terms of the parameters defining the Poiseuille flow. The boundary conditions at S_α are unchanged. Without the boundary condition at $z = 0$ we reduce to the problem of motion of two spheres in an unbounded shear flow considered in Batchelor & Green (1972a). We designate all quantities of this problem by the superscript zero. Thus \mathbf{u}^0 is the unbounded shear flow for the two spheres with translational and rotational velocities \mathbf{V}_α^0 and $\boldsymbol{\Omega}_\alpha^0$, which obey Eq. (2) with $\mathbf{t} = \mathbf{t}^0$. We look for the solution as superposition of \mathbf{u}^0 and the flow perturbation $\delta\mathbf{u}$ (where the use of δ does not imply smallness of $\delta\mathbf{u}$). We thus have,

$$\begin{aligned} \nabla \delta p &= \eta \nabla^2 \delta \mathbf{u}, \quad \nabla \cdot \delta \mathbf{u} = 0, \quad \delta \mathbf{u}(z=0) = -\mathbf{u}^0(z=0), \\ \delta \mathbf{u}(\infty) &= 0, \quad \delta \mathbf{u}(S_\alpha) = \delta \mathbf{V}_\alpha + \delta \boldsymbol{\Omega}_\alpha \times (\mathbf{x} - \mathbf{x}_\alpha), \end{aligned} \quad (5)$$

where we introduced deviations of the velocities and of the surface traction from their values in an infinite domain,

$$\delta \mathbf{V}_\alpha = \mathbf{V}_\alpha - \mathbf{V}_\alpha^0, \quad \delta \boldsymbol{\Omega}_\alpha = \boldsymbol{\Omega}_\alpha - \boldsymbol{\Omega}_\alpha^0, \quad \delta \mathbf{t} = \mathbf{t} - \mathbf{t}^0. \quad (6)$$

The deviations of the velocities are fixed by the condition that the deviation from the surface traction obeys Eqs. (2) with $\delta \mathbf{t}$ instead of \mathbf{t} . We notice that the flow $\mathbf{u}^0(z=0)$ in Eq. (55) is induced by the spheres, since the unperturbed flow vanishes at $z = 0$. Thus $\mathbf{u}^0(z=0)$ vanishes at infinity as necessary for consistency of the boundary conditions at the plane and at infinity. For the distant wall the flow $\mathbf{u}^0(z=0)$ can be simplified. This flow obeys the integral representation (see the derivation in Appendix A),

$$\begin{aligned} u_i^0(\mathbf{x}) &= \dot{\gamma} \delta_{i1} z - \sum_\alpha \int_{S_\alpha} \frac{Y_{il}(\mathbf{x} - \mathbf{x}') t_l^0(\mathbf{x}') dS}{8\pi\eta}; \\ Y_{il}(\mathbf{r}) &= \frac{\delta_{il}}{r} + \frac{r_i r_l}{r^3}, \quad \mathbf{r} = \mathbf{x} - \mathbf{x}', \end{aligned} \quad (7)$$

where Y_{il} is the Oseen tensor or the Green's function of the Stokes flow in an unbounded fluid Kim & Karrila (2013). If the vertical positions z_α of the centers of both spheres are much larger than their radii, a , then the asymptotic expansion of $\mathbf{u}^0(z=0)$ in a/z_α is obtained by Taylor expansion of $Y_{il}(\mathbf{x} - \mathbf{x}')$ in Eq. (7) near $\mathbf{x}' = \mathbf{x}_\alpha$. Using the condition of zero force we find that, at the leading order,

$$\begin{aligned} u_i^0(z=0) &\approx \frac{1}{8\pi\eta} \frac{\partial}{\partial x_m} \sum_\alpha Y_{il}(\mathbf{x} - \mathbf{x}_\alpha) S_{lm}^\alpha |_{z=0}; \\ S_{lm}^\alpha &\equiv \int_{S_\alpha} \left((\mathbf{x} - \mathbf{x}_\alpha)_m t_l^0(\mathbf{x}) - \frac{\delta_{ml}(\mathbf{x} - \mathbf{x}_\alpha)_p t_p^0(\mathbf{x})}{3} \right) dS, \end{aligned} \quad (8)$$

where the traceless tensor S_{lm}^α is Batchelor & Green (1972a) the force dipole strength of sphere α . The δ_{lm} term can be added since $\nabla_l Y_{il} = 0$. The force

dipole strengths obey a general form derived in Batchelor & Green (1972a). We have $S_{lm}^1 = S_{lm}^2 = S_{lm}$ with

$$\frac{3S_{lm}(\mathbf{r})}{10\pi\eta a^3\dot{\gamma}} = (\delta_{lx}\delta_{mz} + \delta_{mx}\delta_{lz})(1+K) + \left(\frac{r_l(x\delta_{mz} + z\delta_{mx}) + r_m(x\delta_{lz} + z\delta_{lx})}{r^2} - \frac{4xz\delta_{lm}}{3r^2} \right) L + \frac{2xz}{r^2} \left(\frac{r_l r_m}{r^2} - \frac{\delta_{lm}}{3} \right) M, \quad (9)$$

where the scalar functions K , L and M depend on the inter-particle distance r/a only (we omitted the prime in the notation of Batchelor & Green (1972a), as the spheres have identical radii in our case). These functions can be completely found only numerically and are considered below as given. We can use Eq. (8) instead of the boundary condition at $z = 0$ in Eq. (55). The first reflection Happel & Brenner (1983) gives the leading order approximation for $\delta\mathbf{V}_\alpha$ as in the Lorentz solution for a sphere in the presence of a distant wall Happel & Brenner (1983). The compact expansion can be found below from integral representations.

2.2. Integral equation for velocities

Here we derive the integral equation that determines the particle velocities. For future generalization to the case where the distances from the spheres to both walls are comparable we perform the derivation starting from the full formulation given by Eq. (1). We use the integral representation of the flow derived in Fouxon *et al.* (2017),

$$\mathbf{u}_i(\mathbf{x}) = \frac{\delta_{ix}z(z-h)\nabla_x p^0}{2\eta} - \sum_\alpha \int_{S_\alpha} \frac{S_{il}(\mathbf{x}, \mathbf{x}') t_l(\mathbf{x}') dS'}{8\pi\eta}, \quad (10)$$

where we exploited the symmetry (Pozrikidis 1992) of Green's function $S_{il}(\mathbf{x}, \mathbf{x}') = S_{li}(\mathbf{x}', \mathbf{x})$. This function is defined by,

$$\mathbf{u}^S(\mathbf{x}) = \frac{1}{8\pi\eta} S_{ik}(\mathbf{x}, \mathbf{x}_0) g_k, \quad (11)$$

where \mathbf{u}^S is the Stokeslet flow caused by a point force acting between two parallel plates,

$$\begin{aligned} -\nabla p^S + \eta\nabla^2 \mathbf{u}^S + \mathbf{g}\delta(\mathbf{x} - \mathbf{x}_0) &= 0, \quad \nabla \cdot \mathbf{u}^S = 0, \\ \mathbf{u}^S(z=0) &= \mathbf{u}^S(z=h) = 0, \quad \mathbf{u}^S(x^2 + y^2 \rightarrow \infty) = 0. \end{aligned} \quad (12)$$

The function S_{ik} is independent of \mathbf{g} and it was derived in Liron & Mochon (1976). We study the velocities \mathbf{V}_α using the integral equation for the surface traction $\mathbf{t}(\mathbf{x})$ obtained by taking \mathbf{x} in Eq. (10) to the surface of one of the spheres; this gives

$$(\mathbf{V}_\alpha)_i + (\boldsymbol{\Omega}_\alpha \times (\mathbf{x} - \mathbf{x}_\alpha))_i = \frac{\delta_{ix}z(z-h)\nabla_x p^0}{2\eta} - \sum_{\alpha'} \int_{S_{\alpha'}} \frac{S_{il}(\mathbf{x}, \mathbf{x}') t_l(\mathbf{x}') dS'}{8\pi\eta}, \quad (13)$$

cf. Pozrikidis (1992). This equation holds for all \mathbf{x} on S_α with $\alpha = 1, 2$. Together with the conditions of zero forces and torques it determines \mathbf{V}_α , $\boldsymbol{\Omega}_\alpha$ and the surface traction uniquely (Kim & Karrila 2013). We use the assumption $h \gg z_\alpha$,

meaning that the wall at $z = h$ is much further from the spheres than the one at $z = 0$. We can therefore approximately assume

$$S_{il}(\mathbf{x}, \mathbf{x}') \approx G_{il}(\mathbf{x}, \mathbf{x}'), \quad (14)$$

where $G_{il}(\mathbf{x}, \mathbf{x}')$ is the Stokeslet near a plane wall defined by

$$\begin{aligned} -\nabla p' + \eta \nabla^2 \mathbf{u}' + \mathbf{g} \delta(\mathbf{x} - \mathbf{x}') &= 0, \quad \nabla \cdot \mathbf{u}' = 0, \\ \mathbf{u}'(z = 0) &= \mathbf{u}'(x \rightarrow \infty) = 0, \quad \mathbf{u}'(\mathbf{x}) = \frac{G_{il}(\mathbf{x}, \mathbf{x}') g_l}{8\pi\eta}. \end{aligned} \quad (15)$$

The requirement that Eq. (14) holds when both \mathbf{x} and \mathbf{x}' belong to the spheres quantifies the assumption that one of the walls is much further than the other. In practice the difference between distances to the upper and lower walls does not have to be too large for the equation to hold. With $z_\alpha \ll h$ and this assumption, Eq. (13) becomes

$$(V_\alpha)_i + (\boldsymbol{\Omega}_\alpha \times (\mathbf{x} - \mathbf{x}_\alpha))_i = \dot{\gamma} \delta_{ix} z - \sum_{\alpha'} \int_{S_{\alpha'}} \frac{G_{il}(\mathbf{x}, \mathbf{x}') t_l(\mathbf{x}') dS'}{8\pi\eta}. \quad (16)$$

We next introduce the decomposition of $G_{il}(\mathbf{x}, \mathbf{x}')$ into the Stokeslet in an infinite space and the correction due to the wall \tilde{G}_{il} ,

$$G_{il}(\mathbf{x}, \mathbf{x}') = Y_{il}(\mathbf{r}) + \tilde{G}_{il}(\mathbf{x}, \mathbf{x}'), \quad (17)$$

with $\mathbf{r} = \mathbf{x} - \mathbf{x}'$, as above. The contribution \tilde{G}_{il} is induced by the images located at the reflection $(\mathbf{x}')^* = (x', y', -z')$ of the source position $\mathbf{x}' = (x', y', z')$ with respect to the plane $z = 0$. It was found in Blake (1971) that the image singularities are a point force of the same magnitude as the source, but with an opposite sign, a stokes-doublet and a source-doublet, see definitions in the paper. We can write the formula in Blake (1971) as follows

$$\begin{aligned} \tilde{G}_{il}(\mathbf{x}, \mathbf{x}') &= -Y_{il}(\mathbf{R}) + 2z' G_{il}^{(1)}(\mathbf{R}) + 2z'^2 G_{il}^{(2)il}(\mathbf{R}), \\ G_{il}^{(1)} &= (2\delta_{3l} - 1) \partial_l Y_{i3}, \quad G_{il}^{(2)} = \frac{(1 - 2\delta_{3l})(R^2 \delta_{il} - 3R_i R_l)}{R^5}, \end{aligned} \quad (18)$$

where $\mathbf{R} = \mathbf{x} - (\mathbf{x}')^*$ is the distance from the images and there is no summation over repeated indices. The symmetries of the Green's functions $G_{il}(\mathbf{x}, \mathbf{x}') = G_{li}(\mathbf{x}', \mathbf{x})$ and $Y_{il}(\mathbf{x}, \mathbf{x}') = Y_{li}(\mathbf{x}', \mathbf{x})$ imply the symmetry $\tilde{G}_{il}(\mathbf{x}, \mathbf{x}') = \tilde{G}_{il}(\mathbf{x}', \mathbf{x})$, which can be confirmed directly.

We compare Eq. (16) with the similar equation for two spheres driven by the unbounded shear flow that was considered above. The equation can be obtained by dropping \tilde{G}_{il} above, see Eq. (7), which yields

$$(V_\alpha^0)_i + (\boldsymbol{\Omega}_\alpha^0 \times (\mathbf{x} - \mathbf{x}_\alpha))_i = \dot{\gamma} \delta_{ix} z - \sum_{\alpha'} \int_{S_{\alpha'}} \frac{Y_{il}(\mathbf{x} - \mathbf{x}') t_l^0(\mathbf{x}') dS'}{8\pi\eta}. \quad (19)$$

Subtracting Eq. (19) from Eq. (16) we find

$$(\delta V_\alpha)_i + (\delta \Omega_\alpha \times (\mathbf{x} - \mathbf{x}_\alpha))_i + \sum_{\alpha'} \int_{S_{\alpha'}} \frac{\tilde{G}_{il}(\mathbf{x}, \mathbf{x}') t_l^0(\mathbf{x}') dS'}{8\pi\eta} = - \sum_{\alpha'} \int_{S_{\alpha'}} \frac{G_{il}(\mathbf{x}, \mathbf{x}') \delta t_l(\mathbf{x}') dS'}{8\pi\eta}. \quad (20)$$

Provided that $\mathbf{t}^0(\mathbf{x})$ is known, this is an integral equation on $\delta \mathbf{t}(\mathbf{x})$ which also obeys the conditions of zero forces and torques given by Eqs. (2) with $\delta \mathbf{t}$ replacing \mathbf{t} . So far we have not made any approximations besides that the spheres are much closer to one of the two walls of the channel.

2.3. Asymptotic solution for a distant wall

We consider the solution of Eq. (20) in the limit of a distant wall. The last (source) term in the left hand side of this equation, in contrast with the rest of the terms, does not involve properties of $\delta \mathbf{u}$. If it is dropped, then we find Eq. (16) with $\dot{\gamma} = 0$, that is the equation for two inertialess spheres moving near the wall in the fluid at rest, which unique solution is trivial – zero translational and angular velocities.

In fact, Eq. (20) coincides with the equation for the velocities of an inertialess swimmer, composed of two spheres, that swims near a plane wall at $z = 0$. In this case, the propulsion is powered by the swimming stroke prescribed by the velocity distribution at the spheres' surface as given by that last term.

When both spheres are separated from the wall by a distance much larger than a the asymptotic series solution can be obtained via the Taylor expansion of $\tilde{G}_{il}(\mathbf{x}, \mathbf{x}')$ near the centers of the spheres, cf. Sec. 2.1. Indeed, both arguments, \mathbf{x} and \mathbf{x}' , of $\tilde{G}_{il}(\mathbf{x}, \mathbf{x}')$ are confined in Eq. (20) to one of the spheres (possibly different ones). In this range $\tilde{G}_{il}(\mathbf{x}, \mathbf{x}')$ is a slowly varying function of its arguments because $z_\alpha \gg a$ and the image of \mathbf{x}' under this condition is separated from each sphere by a distance much larger than the radius, cf. with the Lorentz solution (Happel & Brenner 1983) and also Appendix of Fouxon & Leshansky (2018). This observation does not depend on the separation between the spheres that can be nonetheless arbitrary. Thus we write Eq. (20) as,

$$\begin{aligned} (\delta V_\alpha)_i + (\delta \Omega_\alpha \times (\mathbf{x} - \mathbf{x}_\alpha))_i + \sum_{\alpha'} \int_{S_{\alpha'}} \frac{\tilde{G}_{il}(\mathbf{x}_\alpha, \mathbf{x}') t_l^0(\mathbf{x}') dS'}{8\pi\eta} + \\ (\mathbf{x} - \mathbf{x}_\alpha)_k \frac{\partial}{\partial x_k} \sum_{\alpha'} \int_{S_{\alpha'}} \frac{\tilde{G}_{il}(\mathbf{x}, \mathbf{x}') t_l^0(\mathbf{x}') dS'}{8\pi\eta} |_{\mathbf{x}=\mathbf{x}_\alpha} + \dots \quad (21) \\ = - \sum_{\alpha'} \int_{S_{\alpha'}} \frac{G_{il}(\mathbf{x}, \mathbf{x}') \delta t_l(\mathbf{x}') dS'}{8\pi\eta}, \end{aligned}$$

where dots stand for higher order terms in the Taylor expansion. The asymptotic solution can be obtained by requiring that the equation holds at every order in $\max[a/z_1, a/z_2]$ (the case of disparate z_α seems to be of little interest so

$z_1 \sim z_2$ can be assumed below though this is not necessary for the analysis). The zero-order term determines the particle velocities,

$$(\delta V_\alpha)_i = -\sum_{\alpha'} \int_{S_{\alpha'}} \frac{\tilde{G}_{il}(\mathbf{x}_\alpha, \mathbf{x}') t_l^0(\mathbf{x}') dS'}{8\pi\eta}, \quad (22)$$

where δt_l is zero at this order. This formula can be simplified by noting that $\tilde{G}_{il}(\mathbf{x}_\alpha, \mathbf{x}')$ is a smooth function of \mathbf{x}' on each of the spheres for the same reasons as before due to the symmetry $\tilde{G}_{il}(\mathbf{x}, \mathbf{x}') = \tilde{G}_{li}(\mathbf{x}', \mathbf{x})$. The zero-order term in the expansion vanishes by the condition of zero force. We thus find that

$$\delta V_{\alpha i} = -\frac{S_{lm}}{8\pi\eta} \sum_{\alpha'} \frac{\partial \tilde{G}_{il}(\mathbf{x}_\alpha, \mathbf{x}_{\alpha'})}{\partial (x_{\alpha'})_m} + o\left(\frac{a}{z_\alpha}\right), \quad (23)$$

which is a more rigorous derivation of the result that might also be obtained using reflections as described in the beginning of the Section. The use of an integral representation allows us to precisely formulate the validity conditions and to provide a transparent structure of the asymptotic series. It is important for the further analysis that the derivation does not assume any *a priori* relation between \mathbf{V}_α^0 and $\delta \mathbf{V}_\alpha$. Actually, the absolute value of the velocities $\delta \mathbf{V}_\alpha$ would be smaller than \mathbf{V}_α^0 , however, this need not to be true for the relative velocities which are of main interest here.

2.4. Evolution equation of inter-particle distance

The velocity of the relative motion of the spheres is described by $\mathbf{V} = \mathbf{V}_2 - \mathbf{V}_1$ that, at the leading order, obeys

$$V_i = V_i^0 + \frac{S_{lm}}{8\pi\eta} \sum_{\alpha'} \left(\frac{\partial \tilde{G}_{il}(\mathbf{x}_1, \mathbf{x}_{\alpha'})}{\partial (x_{\alpha'})_m} - \frac{\partial \tilde{G}_{il}(\mathbf{x}_2, \mathbf{x}_{\alpha'})}{\partial (x_{\alpha'})_m} \right). \quad (24)$$

The relative velocity in an unbounded shear flow \mathbf{V}^0 can be written as (Batchelor & Green 1972a)

$$V_i^0(\mathbf{r}) = \dot{\gamma} z \delta_{i1} - \frac{\dot{\gamma} B z \delta_{i1}}{2} - \frac{\dot{\gamma} B x \delta_{i3}}{2} - \frac{\dot{\gamma} (A - B) x z r_i}{r^2}, \quad (25)$$

where $\mathbf{r} = \mathbf{x}_2 - \mathbf{x}_1$. The first term in the RHS is the driving shear flow. The remaining terms, due to hydrodynamic interactions, are described by the functions A and B , which depend on $|\mathbf{r}| = r$ only. These functions are considered, similarly to K , L and M above, as given in Batchelor & Green (1972a). We observe that \mathbf{V}^0 is determined uniquely by the distance between the particles and is independent of the particles' center of mass. Thus the evolution of $\mathbf{r}(t)$ without the wall is autonomous, i.e., the time derivative of $\mathbf{r}(t)$ is determined uniquely by the instantaneous value of $\mathbf{r}(t)$. We demonstrate that the evolution of $\mathbf{r}(t)$, described by Eq. (24), remains autonomous. This means that we can neglect in \mathbf{V} , which is a function of \mathbf{x}_i , the dependence on the center-of-mass coordinate $(\mathbf{x}_1 + \mathbf{x}_2)/2$. Since the horizontal coordinates of the center of mass are irrelevant by translational invariance in the horizontal directions, we need to consider only the dependence on $z_0 = (z_1 + z_2)/2$. This coordinate would not

change at all without the hydrodynamic interactions and the particles would move in straight lines parallel to the wall. The interactions cause temporal variations of z_0 ; however these occur only over the scale of these interactions which is the radius a . Moreover, this change is small by the assumption that $\max[a/z_1, a/z_2] \ll 1$. This allows to consider $z_0 = (z_1 + z_2)/2$ as constant during the whole time of the interactions giving $\mathbf{V} = \mathbf{V}(\mathbf{r}(t), z_0(t)) \approx \mathbf{V}(\mathbf{r}(t), z_0(t_0))$ where t_0 is arbitrary. For $\delta V_i \equiv V_i - V_i^0$ we have

$$\begin{aligned} \delta V_i &= \frac{S_{lm}}{8\pi\eta} \sum_{\alpha'} \left(\frac{\partial \tilde{G}_{li}(\mathbf{x}_{\alpha'}, \mathbf{x}_1)}{\partial (x_{\alpha'})_m} - \frac{\partial \tilde{G}_{li}(\mathbf{x}_{\alpha'}, \mathbf{x}_2)}{\partial (x_{\alpha'})_m} \right) = \\ &\frac{S_{lm}}{8\pi\eta} \sum_{\alpha'} \frac{\partial}{\partial (x_{\alpha'})_m} (Y_{li}(\mathbf{x}_{\alpha'} - \mathbf{x}_2^*) - Y_{li}(\mathbf{x}_{\alpha'} - \mathbf{x}_1^*) + 2z_1 G_{li}^1(\mathbf{x}_{\alpha'} - \mathbf{x}_1^*) + \\ &2z_1^2 G_{li}^2(\mathbf{x}_{\alpha'} - \mathbf{x}_1^*) - 2z_2 G_{li}^1(\mathbf{x}_{\alpha'} - \mathbf{x}_2^*) - 2z_2^2 G_{li}^2(\mathbf{x}_{\alpha'} - \mathbf{x}_2^*)), \end{aligned} \quad (26)$$

where we used Eq. (18). The derivatives in the above equation can be written via the tensors

$$\begin{aligned} T_{lim}^{(1)}(\mathbf{r}) &\equiv -\frac{\partial Y_{li}(\mathbf{r})}{\partial r_m} = \frac{r^2 (r_m \delta_{il} - r_i \delta_{lm} - r_l \delta_{im}) + 3r_i r_l r_m}{r^5}; \\ T_{lim}^{(2)}(\mathbf{r}) &\equiv \frac{\partial G_{li}^1}{\partial r_m} = (1 - 2\delta_{3i}) \left(\frac{\delta_{im} \delta_{3l} - \delta_{i3} \delta_{lm} - \delta_{il} \delta_{3m}}{r^3} \right. \\ &\left. - \frac{3r_i (r_m \delta_{3l} - r_3 \delta_{lm} - r_l \delta_{3m})}{r^5} + \frac{3(\delta_{i3} r_l r_m + \delta_{il} r_3 r_m + \delta_{im} r_3 r_l)}{r^5} - \frac{15r_i r_3 r_l r_m}{r^7} \right); \\ T_{lim}^{(3)}(\mathbf{r}) &\equiv \frac{\partial G_{li}^2}{\partial r_m} = -3(1 - 2\delta_{3i}) \left(\frac{r_m \delta_{il} + \delta_{im} r_l + \delta_{lm} r_i}{r^5} - \frac{5r_m r_i r_l}{r^7} \right), \end{aligned} \quad (27)$$

where we used Eqs. (17) and (18). We also observe that,

$$\mathbf{x}_{\alpha'} - \mathbf{x}_{\alpha}^* = (x_{\alpha'} - x_{\alpha}, y_{\alpha'} - y_{\alpha}, z_{\alpha'} + z_{\alpha}). \quad (28)$$

Thus we find,

$$\begin{aligned} \delta V_i &= \frac{S_{lm}}{8\pi\eta} [K_{lim}^{11} - K_{lim}^{12} + K_{lim}^{21} - K_{lim}^{22}]; \\ K_{lim}^{kn} &= T_{lim}^{(1)}(\mathbf{x}_k - \mathbf{x}_n^*) + 2z_j T_{lim}^{(2)}(\mathbf{x}_k - \mathbf{x}_n^*) + 2z_j^2 T_{lim}^{(3)}(\mathbf{x}_k - \mathbf{x}_n^*). \end{aligned} \quad (29)$$

We write above $z_2 = z_0 + r_3/2$ and $z_1 = z_0 - r_3/2$ where $r_3 = z_2 - z_1$ is the vertical component of the distance \mathbf{r} . We use $\mathbf{x}_{\alpha} - \mathbf{x}_{\alpha}^* = 2z_{\alpha}$ so that,

$$T_{lim}^{(k)}(\mathbf{x}_1 - \mathbf{x}_1^*) = T_{lim}^{(k)}(0, 0, 2z_0 - r_3); T_{lim}^{(k)}(\mathbf{x}_2 - \mathbf{x}_2^*) = T_{lim}^{(k)}(0, 0, 2z_0 + r_3). \quad (30)$$

Similarly, using $\mathbf{x}_1 - \mathbf{x}_2^* = (-r_1, -r_2, z_1 + z_2) = (-r_1, -r_2, 2z_0)$ and $\mathbf{x}_2 - \mathbf{x}_1^* = (r_1, r_2, 2z_0)$ we find,

$$T_{lim}^{(k)}(\mathbf{x}_1 - \mathbf{x}_2^*) = T_{lim}^{(k)}(-r_1, -r_2, 2z_0); T_{lim}^{(k)}(\mathbf{x}_2 - \mathbf{x}_1^*) = T_{lim}^{(k)}(r_1, r_2, 2z_0). \quad (31)$$

The last equations provide the velocity in Eq. (29) in terms of \mathbf{r} and z_0 . We consider z_0 as a constant given by the initial configuration, see the discussion

after Eq. (25). The remaining terms in Eq. (26) depend only on \mathbf{r} , providing an autonomous equation for \mathbf{r} .

The detailed form of the evolution equation for \mathbf{r} in Cartesian coordinates is given by Eqs. (74)-(77) in Appendix B. The more compact form is found by employing the cylindrical coordinate system with $x = \rho \cos \phi$, $y = \rho \sin \phi$, $z = z$. We find using the identities $x\dot{x} + y\dot{y} = \rho\dot{\rho}$ and $-y\dot{x} + x\dot{y} = \rho^2\dot{\phi}$ and the definitions $s^2 \equiv \rho^2 + 4z_0^2$ and $\sigma \equiv r^2 - s^2$ that (here and below we set $\dot{\gamma} = 1$ by passing in the equation of motion for \mathbf{r} to dimensionless time $\dot{\gamma}t$),

$$\begin{aligned}\dot{\rho} = & zc_\phi \left[1 - \frac{B}{2} - \frac{\rho^2(A - B)}{r^2} + \frac{5\rho^2P}{3r^4s^5} + \frac{10z_0R}{r^4\sigma^2} \right. \\ & \left. + \frac{5\rho^2\sigma}{2r^4s^7}(P + 2(\rho^4 - s^2\rho^2 + 4z^2z_0^2)M) \right].\end{aligned}\quad (32)$$

Here, we have introduced $c_\phi = \cos \phi$, $P = r^2s^2(L - M) + 3(\rho^4 + 4z^2z_0^2)M$ and $R = r^4(1 + K + L) + 2z^2\rho^2M$, see definitions in Eqs. (9) and (25). The dynamics of c_ϕ is

$$\dot{c}_\phi = \frac{z}{\rho} (c_\phi^2 - 1) \left[\frac{B}{2} - 1 + \frac{5\rho^2\sigma L}{2r^2s^5} - \frac{10z_0}{r^2\sigma^2}(r^2(1 + K) + z^2L) \right]. \quad (33)$$

Finally, the dynamics of z reads

$$\begin{aligned}\dot{z} = & \rho c_\phi \left[-\frac{B}{2} - \frac{z^2(A - B)}{r^2} + \frac{5(\rho^2 - 16z_0^2)\sigma R}{2r^4s^7} + \right. \\ & \left. \frac{5z^2P}{3r^4s^5} + \frac{10z^2z_0}{r^4\sigma^2}(r^2L + (2z^2 - \rho^2)M) \right].\end{aligned}\quad (34)$$

Further noting that $\rho^4 - s^2\rho^2 + 4z^2z_0^2 = \rho^2(\rho^2 - s^2) + 4z^2z_0^2 = -4z_0^2\rho^2 + 4z^2z_0^2 = 4z_0^2(z^2 - \rho^2)$ the evolution equation for ρ can be rewritten as

$$\begin{aligned}\dot{\rho} = & zc_\phi \left[1 - \frac{B}{2} - \frac{\rho^2(A - B)}{r^2} + \frac{5\rho^2P}{3r^4s^5} + \right. \\ & \left. \frac{10z_0R}{r^4\sigma^2} + \frac{5\rho^2\sigma P}{2r^4s^7} + \frac{20\rho^2z_0^2\sigma}{r^4s^7}(z^2 - \rho^2)M \right].\end{aligned}\quad (35)$$

It can be readily seen using $|s| \sim z_0$ and $|\sigma| \sim z_0^2$, that at fixed \mathbf{r} we have $\delta V_i \sim z_0^{-3}$ upon varying z_0 . Similarly if we fix z_0 then $\delta V_i \sim r^{-2}$ upon varying r . The inverse cubic dependence on z_0 is non-trivial. Derivatives of $\tilde{G}_{il}(\mathbf{r})$ contain terms of order r^{-2} which would give z_0^{-2} behavior in Eq. (23), cf. the dependence of T_{lim}^k on r in Eq. (27). Following rules for tensorial transformations upon the sign reversal of the argument, see, *e. g.*, Eqs. (30)-(31), the leading order z_0^{-2} terms cancel.

We remark that finding the next order correction to \mathbf{V} in the inverse distance to the wall would involve the quadratic surface moments originating from \mathbf{t}^0 . These were not considered previously and would be quite demanding to compute, see Eq. (22). It would also require considering the contributions in the second line of Eq. (21). The corresponding exceedingly complex calculations are beyond the scope of the present paper. We take here the practical approach

of trying to push our leading order calculation to smaller z_0 and compare the analytical prediction with the results of the direct numerical simulations.

The equations of motion have symmetries that can be described as the properties of the velocity components,

$$\begin{aligned} V_\rho(\rho, -\phi, z) &= V_\rho(\rho, \phi, z), \quad V_\rho(\rho, \phi, -z) = -V_\rho(\rho, \phi, z), \\ V_\phi(\rho, -\phi, z) &= V_\phi(\rho, \phi, z), \quad V_\phi(\rho, \phi, -z) = -V_\phi(\rho, \phi, z), \\ V_z(\rho, -\phi, z) &= V_z(\rho, \phi, z), \quad V_z(\rho, \phi, -z) = V_z(\rho, \phi, z). \end{aligned} \quad (36)$$

These properties allow to confine the study of the trajectories $\mathbf{r}(t)$ to $z \geq 0$, $0 \leq \phi \leq \pi/2$, besides the constraint $r \geq 2$.

The main result of this Section is the evolution equation for the distance between two spheres freely suspended in a shear flow near the wall,

$$\dot{\mathbf{r}} = \mathbf{V}(\mathbf{r}) = \mathbf{V}^0(\mathbf{r}) + \delta\mathbf{V}(\mathbf{r}). \quad (37)$$

Here $\mathbf{V}^0(\mathbf{r})$, given by Eq. (25), describes hydrodynamic interactions due to shear in unbounded flow and $\delta\mathbf{V}$ describes the effects of the wall, given at the leading order by Eqs. (74)-(77). Despite that the wall is assumed to be distant, its effect is not small even for large channels.

3. Singular effect of the wall at far distances

In this Section we demonstrate that the wall is a singular perturbation of the relative motion between the two spheres. Regardless of how large z_0 is, its influence cannot be entirely neglected. For any fixed \mathbf{r} we have $\mathbf{V}(\mathbf{r}) = \mathbf{V}^0(\mathbf{r})$ for $z_0 \rightarrow \infty$. However for any fixed $z_0 \gg 1$ there are large \mathbf{r} for which some velocity components satisfy $|\delta V_i| \gg V_i^0$. There is a competition between the different parameters: the hydrodynamic interactions are small by a/r whereas the interaction with the wall is small by a/z_0 . As a result at r given by a power of z_0 , whose exponent is determined by the details of the power laws of the particle-particle and particle-wall interactions, the interactions with the wall may dominate the evolution of \mathbf{r} . The resulting topology of the trajectories of the relative motion is hence different, as we will describe in the next Sections. First, we illustrate the differences numerically.

Trajectories that pass through points with $y = 0$ belong to the xz -plane by symmetry, as $V_y(y = 0) = 0$. We consider the remaining components V_x and V_z as functions of x and z in the xz -plane. We can restrict the analysis to positive x and z due to the symmetries described in the previous Section. For the streamwise component of the velocity the wall is a regular perturbation: the ratio V_x/V_x^0 is everywhere close to 1. Thus for $z_0 = 20$ the maximal deviation of V_x/V_x^0 from 1 is seen numerically to be less than 1%. Consider for instance the ratio at $x = y = 0$ where the only nonzero components are $V_x^0(0, 0, z) = (1 - B(z)/2)z$ and $\delta V_x(0, 0, z) = 10zz_0(1 + K(z) + L(z))/(z^2 - 4z_0^2)^2$, see Eqs. (25) and (74). When $z \sim 2$ all $B(z), K(z), L(z)$ are finite and less than unity Batchelor & Green (1972a). By taking the ratio we find that $\delta V_x/V_x^0 \sim 10/z_0^3 \ll 1$ at $z_0 \gg 1$. At large distances V_x^0 is dominated by the

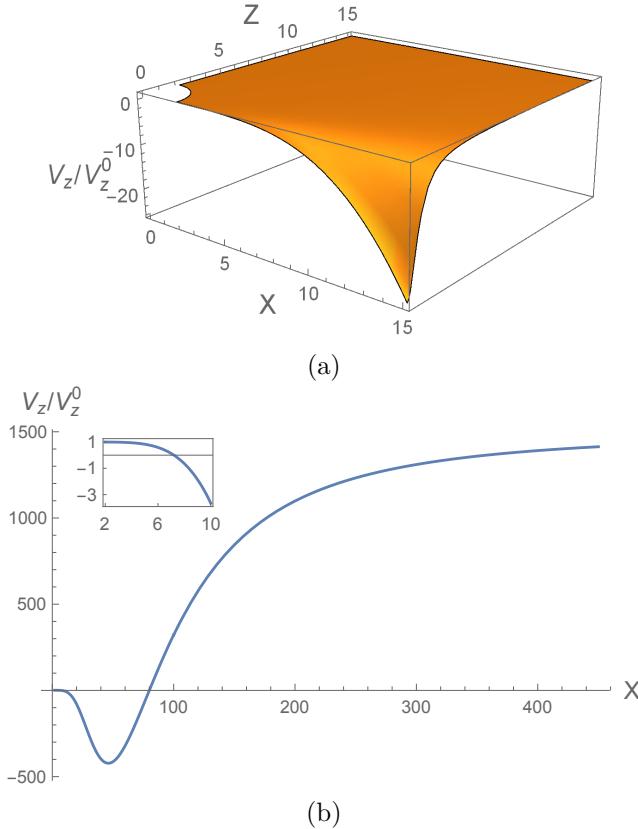


Figure 7: The ratio V_z/V_z^0 for $z_0 = 20$ (a) in the xz -plane and (b) along the x -axis. The presence of the wall increases the velocity at $x \gg z_0$ by a constant large factor of order z_0^2 . The two critical points are the neutral equilibrium point $r_c = 4z_0 = 80$ and the saddle point $r_s = (32z_0^3/15)^{1/5}$ (see the inset in (b)).

driving shear flow, see Eq. (25), and it is much larger than δV_x because the symmetry imposes proportionality of δV_x and z , see Eq. (74). Similarly, in other cases, $V_x/V_x^0 \approx 1$. Thus for practical purposes we can set

$$V_x(\mathbf{r}) \approx V_x^0(\mathbf{r}) = \left(1 - \frac{B(r)}{2}\right) z - \frac{(A(r) - B(r))x^2 z}{r^2}, \quad (38)$$

which at large distances reduces to the carrying shear flow difference given by z . The situation is quite different for V_z/V_z^0 in Fig. 7. We see that when the spheres are close, the difference is negligible and $V_z \approx V_z^0$. However the situation is quite different at large separations. From Eq. (25) we obtain that

for a wall at infinite distance,

$$V_z^0(\mathbf{r}) = -\frac{Bx}{2} - \frac{(A-B)xz^2}{r^2}, \quad (39)$$

which can be further simplified at large r using

$$A(r) = \frac{5}{r^3} + O\left(\frac{1}{r^5}\right), \quad B(r) = \frac{16}{3r^5} + o\left(\frac{1}{r^6}\right), \quad (40)$$

see Batchelor & Green (1972a). We thus find that

$$V_z^0(\mathbf{r}) \approx -\frac{8x}{3r^5} - \frac{5xz^2}{r^5}, \quad (41)$$

which is negative at $x > 0$. We observe from Eqs. (40) that the order of corrections is quite high so this formula might hold already at $r \simeq 3 - 4$. For δV_z we have from the equations in Appendix B that

$$\delta V_z(x, 0, 0) = \frac{10xz_0^2(16z_0^2 - x^2)}{(x^2 + 4z_0^2)^{7/2}} (1 + K(x) + L(x)). \quad (42)$$

This can be simplified at large x following Ref. Batchelor & Green (1972a),

$$K(r) \approx -\frac{2}{r^5}, \quad L(r) \approx -\frac{5}{2r^3}, \quad M(r) \approx \frac{25}{2r^3}, \quad r \gg 1, \quad (43)$$

by neglecting K and L compared to unity in Eq. (42). These functions decay fast with r implying that

$$\delta V_z(x, 0, 0) \approx \frac{10xz_0^2(16z_0^2 - x^2)}{(x^2 + 4z_0^2)^{7/2}}, \quad (44)$$

must hold already at $x = 3$ where the spheres are rather close. We also find for the ratio of velocities

$$\frac{\delta V_z(x, 0, 0)}{V_z^0(x, 0, 0)} = \frac{15x^5 z_0^2 (x^2 - 16z_0^2)}{4(x^2 + 4z_0^2)^{7/2}}; \quad x \gg 1. \quad (45)$$

The corrections are of order higher than $1/x$ so in practice this formula works at rather small x .

The ratio on the LHS of Eq. (45) equals -1 at x obeying the condition,

$$15x^5 z_0^2 (x^2 - 16z_0^2) + 4(x^2 + 4z_0^2)^{7/2} = 0. \quad (46)$$

This equation has two solutions. The first one is obtained when $x \ll 2z_0$ and hence $15x^5 = 32z_0^3$ leading to $r_s = (32z_0^3/15)^{1/5}$. This expression for the critical point, obtained from the $x \gg 1$ approximation given by Eq. (45), is indistinguishable from the numerical solution of $V_z(x, 0, 0) = 0$ with the full velocity given by Eqs. (74)-(77), at least down to $z_0 = 5$ which is the smallest z_0 considered in this work (we have $r_s \approx 3$ at $z_0 = 5$). This is reasonable in view of the remarks after Eqs. (41), (44) and (45). To find the other solution we notice that $x^2 = 16z_0^2 + \delta$ with $\delta \ll 16z_0^2$ solves the equation giving $r_c \approx 4z_0$. Both r_s and r_c are much larger than unity at $z_0 \gg 1$ confirming the consistency of the approach and can be used for $z_0 \geq 5$.

The obtained points obey $V_z(r_c, 0, 0) = V_z(r_s, 0, 0) = 0$. Moreover $V_x(x, 0, 0) = V_y(x, 0, 0) = 0$ since both $V_x(x, y, z)$ and $V_y(x, y, z)$ are odd functions of z , see Eq. (78). Thus, the points on the x -axis with $x = r_s$ and $x = r_c$ are the critical points with $\mathbf{V} = 0$. We demonstrate below that these are a saddle point and a stationary point, respectively.

Finally we would like to emphasise the singular nature of the perturbation due to the long-range interaction at finite z_0 . For motions in the plane $y = 0$, at large but finite z_0 , there are locations \mathbf{r} for which the wall-normal component of the velocity V_z is much larger than the BG velocity, see the $z_0 = 20$ case in Fig. 7. In fact, for $x \gg z_0$, the ratio $\delta V_z(x, 0, 0)/V_z^0(x, 0, 0)$ becomes an x -independent constant of order $z_0^2 \gg 1$. The wall contribution to the velocity is opposite in sign to the BG velocity. If we consider two particles on the same streamline of the unperturbed flow with $y = z = 0$, then the only non-vanishing, z -component of the velocity $V_z(x, 0, 0)$ is,

$$V_z = \frac{10xz_0^2(16z_0^2 - x^2)}{(x^2 + 4z_0^2)^{7/2}}(1 + K(x) + L(x)) - \frac{x B(x)}{2}. \quad (47)$$

In the BG limit of $z_0 \rightarrow \infty$, taken at fixed x , the first term drops, reducing the velocity to $V_z^0(x, 0, 0) = xB(x)/2$, see Eq. (39), and at large distances $xB(x)/2 \approx 8/(3x^4)$, see Eq. (40). In contrast, at any finite z_0 , for $x \gg z_0$, the range not considered in the BG approximation, the contribution due to the wall, described by the first term in Eq. (47) behaves as $z_0^2 x^{-4}$. We find, using that the functions $K(x)$ and $L(x)$ vanish at large distances by Eq. (43),

$$\lim_{x \rightarrow \infty} \frac{\delta V_z(x, 0, 0)}{V_z^0(x, 0, 0)} = \lim_{x \rightarrow \infty} \frac{20z_0^2(x^2 - 16z_0^2)}{B(x)(x^2 + 4z_0^2)^{7/2}} = \frac{15z_0^2}{4}. \quad (48)$$

Thus the interaction between particles flowing along the same streamline is dominated by the wall term at $x \gtrsim z_0$. This sets in non-uniformly. We see from Fig. 7(b) that for $z_0 = 20$ the absolute value $|V_z(x, 0, 0)/V_z^0(x, 0, 0)|$ grows fast with x . It crosses zero (which corresponds to $|\delta V_z(x, 0, 0)/V_z^0(x, 0, 0)| = 1$) at the critical saddle point $(32z_0^3/15)^{1/5} \approx 7$, a value smaller than the half of z_0 . One might have expected that $r \lesssim z_0$ guarantees at least a qualitative validity of the BG theory, however it does not. The ratio $|\delta V_z(x, 0, 0)/V_z^0(x, 0, 0)|$ rapidly grows with x , becoming of order one hundred already at $x \approx 30$. However, after reaching the maximum, it decreases to the value 1 at the critical point at $x = 4z_0$. Only at $x \gg 4z_0$ the asymptotic law $|V_z(x, 0, 0)/V_z^0(x, 0, 0)| \sim z_0^2$ starts to apply. We find numerically that the curve $|V_z(x, 0, 0)/V_z^0(x, 0, 0)|$ starts flattening at $x \sim 200$ when its value is about one thousand. The approach to the limiting value of 1500, imposed by Eq. (48), is quite slow: e.g. at $x \simeq 450$ the ratio is about 1400. We conclude that, at the considered value of z_0 , the wall dominates the interactions at all $x \gtrsim z_0/2$, excluding a small neighborhood of the neutral equilibrium critical point r_s .

The strong changes of \mathbf{V} induced by the presence of a wall described in this Section imply that the phase portrait is very different from that obtained

in the limit $z_0 \rightarrow \infty$. In the next section, we therefore start from reviewing the reference $z_0 = \infty$ case.

4. Trajectories for infinitely distant walls

We describe briefly the seminal results in Batchelor & Green (1972a) pertaining the relative motion of two spheres in unbounded shear flow, as determined by the equation of motion $\dot{\mathbf{r}} = \mathbf{V}^0(\mathbf{r})$. The trajectories can be obtained from the two integrals R_2 and R_3 (notice a different labelling of the axes compared to Batchelor & Green (1972a). We have y and z , and correspondingly R_2 and R_3 , switched)

$$\begin{aligned} R_2 &= y \exp \left(\int_r^\infty \frac{B(r') - A(r')}{1 - A(r')} \frac{dr'}{r'} \right); \\ R_3^2 &= z^2 \exp \left(2 \int_r^\infty \frac{B(r') - A(r')}{1 - A(r')} \frac{dr'}{r'} \right) - \\ &\quad \int_r^\infty \frac{B(r') r' dr'}{1 - A(r')} \exp \left(2 \int_{r'}^\infty \frac{B(r'') - A(r'')}{1 - A(r'')} \frac{dr''}{r''} \right). \end{aligned} \quad (49)$$

We consider trajectories in the symmetry xz -plane ($y = 0$) where $R_2 = 0$. The trajectories are given in the form $z = z(r)$ where ($r^2 = x^2 + z^2$),

$$\begin{aligned} z^2(r) &= R_3^2 \exp \left(2 \int_r^\infty \frac{A(r') - B(r')}{1 - A(r')} \frac{dr'}{r'} \right) + \\ &\quad \int_r^\infty \frac{B(r') r' dr'}{1 - A(r')} \exp \left(2 \int_r^{r'} \frac{A(r'') - B(r'')}{1 - A(r'')} \frac{dr''}{r''} \right). \end{aligned} \quad (50)$$

There are two types of trajectories: open and closed trajectories corresponding to $R_3^2 > 0$ and $R_3^2 < 0$, respectively. The regions in phase space occupied by open and closed trajectories are separated by the separatrix $z^s(r)$ whose equation is found by setting $R_3 = 0$,

$$(z^s)^2 = \int_r^\infty \frac{B(r') r' dr'}{1 - A(r')} \exp \left(2 \int_r^{r'} \frac{A(r'') - B(r'')}{1 - A(r'')} \frac{dr''}{r''} \right). \quad (51)$$

We can obtain $z^s(r)$ at large r using Eq. (40),

$$\int_r^{r'} \frac{A(r'') - B(r'')}{1 - A(r'')} \frac{dr''}{r''} \approx \int_r^{r'} \frac{5 dr''}{r''^4} = \frac{5}{3} \left(\frac{1}{r^3} - \frac{1}{r'^3} \right).$$

The separatrix equation becomes (this asymptotic form was not presented in Batchelor & Green (1972a)),

$$\begin{aligned} (z^s)^2 &\approx \exp \left(\frac{10}{3r^3} \right) \int_r^\infty \frac{16 dr'}{3r'^4} \exp \left(-\frac{10}{3r'^3} \right) = \\ &\quad \frac{8}{15} \left(\exp \left(\frac{10}{3r^3} \right) - 1 \right) \approx \frac{16}{9r^3} \approx \frac{16}{9x^3}, \end{aligned} \quad (52)$$

which shows that the separatrix asymptotically approaches the x -axis (Lin *et al.* 1970). The surface obtained by rotation of this curve around the z -axis separates closed and open three-dimensional trajectories. The volume of closed trajectories is infinite due to divergence of two dimensional integral of $(x^2 + y^2)^{-3/2}$.

We could not obtain a description of the particle-pair motion by integrals similar to R_2 and R_3 in the presence of the walls. For some trajectories, however, the wall is a small perturbation so that $\mathbf{V}(\mathbf{r}) \approx \mathbf{V}^0(\mathbf{r})$ holds everywhere along the trajectory. The trajectory equation is then $\dot{\mathbf{x}}(t) = \mathbf{x}^0(t) + \delta\mathbf{x}(t)$ where $\mathbf{x}^0(t)$ is a BG trajectory and $\delta\mathbf{x}(t)$ represents just a small modification. An example of these trajectories is the trajectory a in Fig. 11. These trajectories can be described with integrals of motion $R_i = R_i^0 + \delta R_i$ where δR_i is a small perturbation of the functional form of the R_i due to the wall. This perturbation can be found from perturbation theory. However this is of limited use since we are interested in trajectories for which the wall contribution is not small.

5. Trajectories for a wall at finite distance

Here, we present the results of numerical simulations of the evolution equation of the inter-particle distance obtained in Sec. 2.4. We apply the algorithm proposed in Filippov (2000), which allows us to compute the hydrodynamic interactions in a system of N spheres in a creeping flow. The algorithm is based on the multipole expansion of the Lamb solution for the fluid velocity field. We applied it to describe the motion in a system of two force- and torque-free solid spheres of unit radius in a shear flow for different distances r between the centers. Namely, for given components of the shear flow field and the vector \mathbf{r} connecting the sphere centers, we compute the velocity \mathbf{V} in Eq. (24). Thus we determined the functions A , B , K , L and M for $r \geq 2.01$ using the formulas in Batchelor & Green (1972a). When r approaches the value $r = 2$ the algorithm requires a very large number of spherical harmonics into the solution expansion, which leads to a very large system of linear equations for the coefficients of the harmonics.

The functions A and B were therefore smoothly continued to $r = 2$ using the asymptotic forms for almost touching spheres given by

$$A(r) = 1 - 4.077(r - 2) + O((r - 2)^{3/2}), \quad B \approx 0.406 - \frac{0.78}{\ln [(r - 2)^{-1}]} \quad (53)$$

The derivative of B diverges at $r = 2$, while the functions A , K , L and M are finite in the limit of touching spheres, $r \rightarrow 2$ and can be continued from $r \geq 2.01$ to $r < 2.01$ using a linear Taylor series approximation. A similar approach was used in Arp & Mason (1977), where, however, continuation was used only below 2.0002. Our main interest is in the behavior at larger r so we did not undertake the detailed solution for the small values of $r - 2$. A higher resolution is needed for the precise evaluation of the impact of the wall on the nearly touching BG trajectories and is left for future work.

Here, the equations of motion are generated employing the velocities given by the contributions Eqs. (25,29). These equations are solved numerically using the custom code in *Mathematica*, which reduces the integration step when the trajectory approaches the vicinity of $r = 2$. In this region, the different trajectories are very close to each other and one has to resolve them accurately. This necessity is obvious already from the BG trajectories in the symmetry plane. All trajectories when the spheres pass in close vicinity to $r = 2$ are closed. In other words, the trajectories that cross the z -axis at z obeying $2 \leq z \leq 2 + \Delta$ are closed; however, those crossing at $z > 2 + \Delta$ are open where Δ is a small number. The quantity Δ obeys the equation

$$(2 + \Delta)^2 = \int_{2+\Delta}^{\infty} \frac{B(r')r'dr'}{1 - A(r')} \exp \left(2 \int_{2+\Delta}^{r'} \frac{A(r'') - B(r'')}{1 - A(r'')} \frac{dr''}{r''} \right), \quad (54)$$

as readily seen from Eq. (51). The evaluation of Δ from this equation (not done in Batchelor & Green (1972a)) is beyond our scope here. Note however that Arp & Mason (1977) provide $\Delta \sim 10^{-5}$.

The smallness of Δ implies that small perturbations can readily turn closed trajectory into an open one, which is indeed what the distant wall does as shown in Fig. 8. The resolution of these small-scale effects demands high numerical precision.

To construct the separatrices (defined here as curves separating regions of qualitatively different behavior) in the xz -plane for given value of z_0 we first find the critical point on the x -axis ($r_s, 0, 0$) where the approximate value of r_s is given in Section III. One separatrix (red curve in Fig. 8) is stable, see Fig. 3 and thus is computed using integration of the original equations. The other separatrix (blue curve in Fig. 8) is unstable as seen from Fig. 3. Thus, it is found by backward integration in time, for which it is stable, until the trajectory reaches the z -axis. All the trajectories below the blue curve (region I) are closed, while those between the red and blue curves (region II) are open – they correspond to non-swapping trajectories. The trajectories between the red curve and the x -axis (region III) can be divided into two classes – open swapping trajectories (brown, black curves in the inset of Fig. 12) and closed trajectories characterized by a very large separation between the spheres (green, blue curves), see the captions of the Figures and detailed theory in the next Section.

We next consider three-dimensional trajectories. The axial symmetry of the governing equations (32)-(34) implies that the saddle points reside in the xy -plane on a circle with radius r_s . For each point on this curve one can construct the corresponding separatrices in 3D (see Fig. 9, where the third neutral direction is given by the circle $r = r_c$, not shown). All the separatrices belong to some surface of rotation (Fig. 10) which is obtained by the rotation of the curves in Fig. 12 around the z -axis.

At this point, it is instructive to compare the evolutions of the same representative initial conditions for $z_0 = \infty$ and finite z_0 . The evolution of

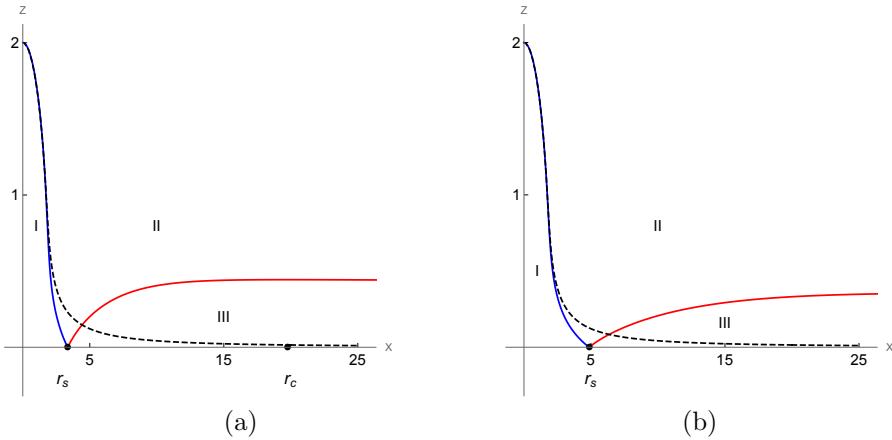


Figure 8: The phase portrait in the xz -plane for $z_0 = 5$ (a) and, $z_0 = 10$ (b). The BG separatrix obeying $z^2 = 16/(9x^3)$ at large x is depicted by the dashed (grey) line. For any finite z_0 , the phase portrait contains two disconnected regions of closed trajectories, in contrast to one region at $z_0 = \infty$. Region I, where all trajectories are closed and the spheres are close to each other, is similar to that at $z_0 = \infty$. Region II is also similar to the $z_0 = \infty$ case: all the trajectories are open and the vertical separation after the interaction returns to its original value. Region III has no counterpart at $z_0 = \infty$. This region contains both closed and open trajectories (see Fig. 12 for a more detailed description). The trajectories passing not far from the stationary point r_c are closed, orbiting around this point. The swapping open trajectories instead are characterized by a sign reversal of the vertical component of the separation vector after the encounter. The region of swapping trajectories is bounded from one side by the closed trajectories around r_c and from the other side by open non-swapping trajectories.

conditions that produce closed BG trajectories with small $x^2 + z^2$ in the limit $z_0 = \infty$ is only weakly influenced by far wall (unless passing near the BG separatrix where small perturbations are relevant), as in Fig. 6, see the caption. In contrast, the trajectories with large $x^2 + z^2$ may be very different as shown in Fig. 5 where the wall changes the evolution from an open trajectory to a closed one. The evolution of initial conditions leading to open trajectories for $z_0 = \infty$ may be only slightly changed by the wall, as in Fig. 11(a), or result in swapping as for the case in Fig. 11(b).

6. Theory of dancing-swapping region

In this Section we analyze the trajectories in the dancing-swapping region III, as shown in Figs. 8. We restrict the consideration to the symmetry plane $y = 0$. All trajectories in this region cross the x -axis. These trajectories are of two types,

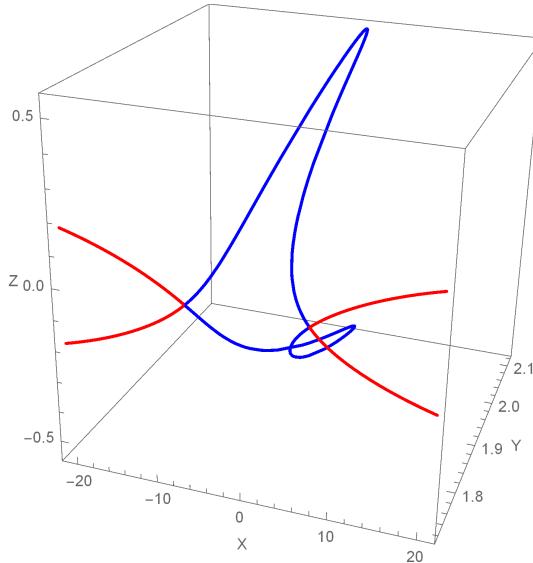


Figure 9: The separatrices for $z_0 = 20$ corresponding to the initial point $(r_s \cos \phi, r_s \sin \phi, 0)$ with $\phi = 7\pi/90$.

both are qualitatively different from the BG theory. The swapping trajectories are open, each crosses the x -axis at a single point x obeying $r_s < x < x_s$ where $r_s = (32z_0^3/15)^{1/5}$ and $x_s = 2\sqrt{2}z_0$ is determined below. For these trajectories the difference of the z coordinates of the particles changes sign as a result of the hydrodynamic encounter (as for black curve in Fig. 12). This sign-reversal corresponds to swapping of the vertical coordinates, see the Introduction. The larger crossing coordinate is, starting from $x = r_s$, the closer the trajectory is to the x -axis at large x . For the unique trajectory passing through $x = x_s$ the trajectory asymptotically approaches the x -axis indefinitely similarly to the BG's separatrix, dividing regions of open and closed trajectories. Finally, the trajectories that pass through a point $(x > x_s, 0, 0)$ are closed, each crossing the x -axis at two locations.

First we observe that the evolution of trajectories in the dancing-swapping region III admits $r \gg 1$ and thus can be simplified. It is readily seen numerically that, at least for $z_0 \geq 5$ that are of interest here, we have $V_x^0(\mathbf{r}) \approx z$ within less than 15% accuracy, meaning that the BG velocity difference is fully determined by the undisturbed shear flow. This is because the hydrodynamic interactions' correction to $V_x^0(\mathbf{r})$ decays quickly with the spheres' separation, see Eqs. (38), (40). We find from Eq. (38) that we can use $V_x(\mathbf{r}) \approx z$ everywhere in region III. Moreover, we observe that $\delta V_z(x, 0, z) - \delta V_z(x, 0, 0)$ grows quadratically with z , see Appendix B. It is then found that since small- z approximation holds (see below) then we can then use $\delta V_z(x, 0, z) \approx \delta V_z(x, 0, 0)$ in the whole

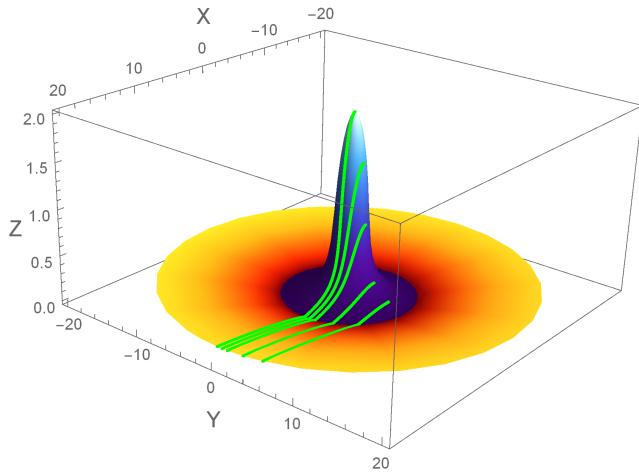


Figure 10: Surface of rotation formed by the separatrices that pass through $(r_s \cos \phi, \sin \phi, 0)$ with $0 \leq \phi \leq 2\pi$ at $z_0 = 20$. The green curves represent separatrices corresponding to $\phi = 0, \pi/36, \pi/18, 5\pi/36, \pi/4$. The trajectories inside the region formed by the blue surface are closed BG-type orbits, whereas the trajectories inside the orange surface are either open swapping or closed dancing trajectories, as in region III in Fig. 12, cf. Fig. 9.

region III. Finally, we can use the reduced Eq. (44). We find that the evolution of the trajectories in region III can be accurately described by the reduced system of equations,

$$\dot{x} = z, \quad \dot{z} = -\frac{8x}{3r^5} - \frac{5xz^2}{r^5} + \frac{10xz_0^2(16z_0^2 - x^2)}{(x^2 + 4z_0^2)^{7/2}}, \quad (55)$$

where we assumed $z_0 \geq 5$ and used Eq. (41). Furthermore, since region III is characterized by small z then it is seen that $r \approx x$ and the second term in the RHS of the equation on z can be dropped. Indeed, the ratio $15z^2/8$ of this term to the first term in the RHS is small at moderate x and not so small at larger x . However, at larger x the time-derivative \dot{z} is determined by the third term. Thus the second term is uniformly small everywhere in III as we verified numerically, and Eq. (55) is rewritten as

$$\dot{x} = z, \quad \dot{z} = -\frac{8}{3x^4} + \frac{10xz_0^2(16z_0^2 - x^2)}{(x^2 + 4z_0^2)^{7/2}}. \quad (56)$$

The trajectories produced by this system in region III are indistinguishable from those produced by the full $\mathbf{V}(\mathbf{r})$. The critical points of this approximate

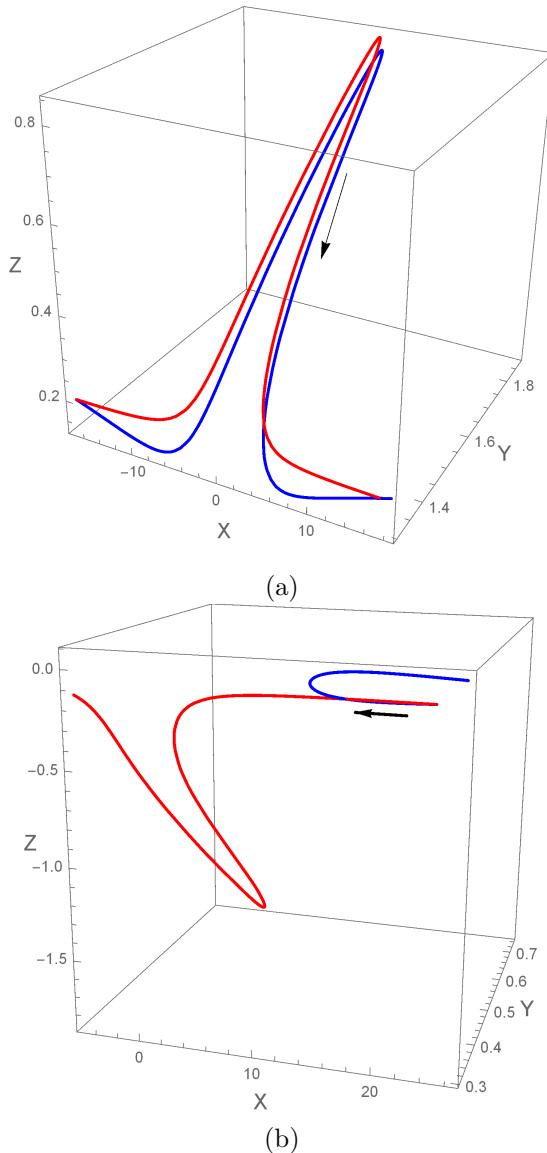


Figure 11: Comparison of representative open trajectories in the BG case (red) and in the case of a wall at a finite distance z_0 (blue): (a) typical open trajectories are qualitatively similar in both cases ($z_0 = 20$); (b) for some initial conditions the presence of the wall results in the appearance of a swapping trajectory ($z_0 = 5$). The black arrows indicate the direction of motion.

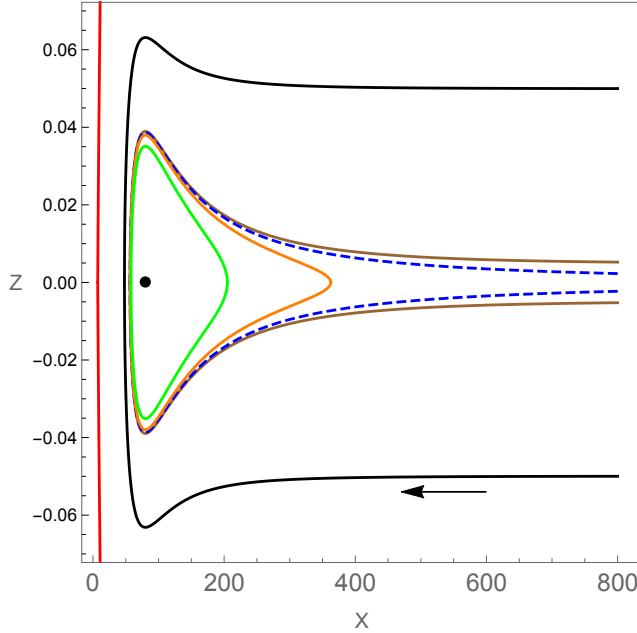


Figure 12: The dancing-swapping region III of Figs. 8 for $z_0 = 20$. The red line is the region's boundary that crosses the x -axis at $(32z_0^3/15)^{1/5}$. The dashed blue line separates open swapping and closed dancing trajectories and crosses the x -axis at $x_s = 2\sqrt{2}z_0$. The black dot is the equilibrium point ($r_c = 4z_0, 0$).

evolution obviously coincide with those obtained in Eq. (46), so that, e. g. $10r_s z_0^2(16z_0^2 - r_s^2)/(r_s^2 + 4z_0^2)^{7/2} = 8/(3r_s^4)$ with $r_s = (32z_0^3/15)^{1/5}$. Eliminating time variable we arrive at

$$\frac{d}{dx}z^2 = -\frac{16}{3x^4} + \frac{20xz_0^2(16z_0^2 - x^2)}{(x^2 + 4z_0^2)^{7/2}}. \quad (57)$$

The trajectory that crosses the x -axis at $x = x_i$ is given by the solution of the above equation and it reads

$$z^2 = \frac{16}{27x^3} + \frac{20z_0^2(x^2 - 8z_0^2)}{3(x^2 + 4z_0^2)^{5/2}} - \frac{16}{27x_i^3} - \frac{20z_0^2(x_i^2 - 8z_0^2)}{3(x_i^2 + 4z_0^2)^{5/2}}. \quad (58)$$

Setting here $x_i = r_s$ and using the condition on r_s provided after Eq. (56), we arrive at the equation of the separatrix bounding the dancing-swapping region and separating it from region II (red line in Fig. 8)

$$z_{II,III}^2 = \frac{16}{27x^3} + \frac{20z_0^2(x^2 - 8z_0^2)}{3(x^2 + 4z_0^2)^{5/2}} + \frac{40z_0^2(48z_0^4 - 2r_s^2z_0^2 - r_s^4)}{9(r_s^2 + 4z_0^2)^{7/2}}, \quad r_s = \left(\frac{32z_0^3}{15}\right)^{1/5}. \quad (59)$$

We further find the following asymptotic behavior

$$z_{II,III}^2(x = \infty) = \frac{40z_0^2(48z_0^4 - 2r_s^2z_0^2 - r_s^4)}{9(r_s^2 + 4z_0^2)^{7/2}} \approx \frac{5}{3z_0}, \quad (60)$$

where the first equality holds down to $z_0 = 5$ and the last equality assumes $z_0 \gg 1$. The equations confirm that region III has a finite width in z -direction, the fact underlying the validity of $\delta V_z(x, 0, z) \approx \delta V_z(x, 0, 0)$. The last equality provides the scaling law of growth of region III as the proximity to the wall decreases from $z_0 = \infty$ to some finite value.

There is a unique value of $x_i = x_s$ for which the last two terms in Eq. (58) vanish and the trajectory asymptotes the x -axis at large x . This value is determined by the condition $x_s^3 = 4(x_s^2 + 4z_0^2)^{5/2}/(45z_0^2(8z_0^2 - x_s^2))$. The solution is $x_s^2 = 8z_0^2 - \epsilon$ with $\epsilon \approx 8\sqrt{3}/(5\sqrt{2})$. The corresponding trajectory z_{sw} is the separatrix of swapping and dancing trajectories,

$$z_{sw}^2 = \frac{16}{27x^3} + \frac{20z_0^2(x^2 - 8z_0^2)}{3(x^2 + 4z_0^2)^{5/2}}; \quad z_{sw}^2(x \approx 2\sqrt{2}z_0) = 0. \quad (61)$$

This asymptotic behavior of this separatrix at $x \gg z_0$ is $z_{sw}^2 \sim 20z_0^2/(3x^3)$. Remarkably this is the same behavior as the BG asymptote given by Eq. (52), however with a much larger coefficient. Since the three-dimensional separatrix is obtained by revolution around the z -axis, we conclude that the volume of closed dancing trajectories is infinite. Thus the wall does not regularize the divergences in the stress calculation at the second order in concentration of Batchelor & Green (1972b). The volume of swapping trajectories is also infinite. Finally we remark that long-distance behavior of trajectories in regions I and II can also be described using the approach of this Section, however the global behavior in those regions involves close positions of the spheres and demands the full formulas.

7. Direct numerical simulation of a particle pair in Poiseuille flows

In this Section, we provide supporting evidence of the existence of the neutral equilibrium point $(r_c, 0, 0)$ from direct numerical simulations of the motion of a pair of particles in the Poiseuille flow. We simulate the Navier-Stokes equations with appropriate boundary conditions at a Reynolds number of 0.1 to approximate solutions of Stokes flows, as verified in previous work (Breugem 2012; Fouxon *et al.* 2017). A moderate distance from the wall ($z_0 = 5$) is considered to examine the accuracy of the theory in conditions typical of microfluidic channels, cf. Shen *et al.* (2016); Ge *et al.* (2019). In this way, we provide confirmation of the theory and demonstrate that it holds down to rather small z_0 .

We use interface-resolved, direct numerical simulations to obtain motions of the particles and full solutions of the ambient flow field. The particles are simulated either as solid spheres using an immersed boundary method or as

liquid droplets using the interface-correction level set/ghost fluid method; see Breugem (2012); Ge *et al.* (2018, 2019) and the Appendix B of Fouxon *et al.* (2017) for detailed descriptions of the governing equations and their numerical treatments. Note that, our solvers does not neglect any details (up to the numerical precision) of the fluid motion throughout a suspension, hence it is better suited for detailed studies of few hydrodynamically interacting particles despite being computationally more expansive, cf. Swan & Brady (2007).

Fig. 1 illustrates the schematic of the simulation setup. Here, two neutrally buoyant particles are transported inside a rectangular channel of dimensions L_x , L_y , and L_z , that are at least an order-of-magnitude larger than the particle radius a . The undisturbed flow is the Poiseuille flow shifted backwards by a constant velocity so that the position of the first particle remains roughly unchanged throughout the simulation.⁴ The particle pair is initially placed adjacent to the bottom wall, with $z_0 = 5$ and $L_z = 64$. L_x and L_y are chosen to be large enough so that the imposed boundary conditions (periodic or inflow/outflow) do not qualitatively affect the particle motion, which we verified by checking that changes in L_x and L_y do not affect the results appreciably. Thus we used $L_x = 12$ and 24 , and increased L_y from 60 up to 80 .

Fig. 13 depicts the vertical component of the relative velocity V_z of two solid particles at various initial separations r/z_0 , obtained asymptotically upon their release. That is, we extract V_z from the simulations when both particles are still approximately at the same vertical position z_0 within the accuracy of 10^{-4} . The theoretical values are computed according to Eqs. (41) and (45), which apply since the minimal considered distance is 15 . Remarkably, we observe a close agreement between the theoretical prediction and the numerical simulation, from the smallest studied distance of $r = 3z_0$. This is despite that the simulations are performed in a pressure-driven channel flow in the presence of two walls and z_0 is not so large. The deviation of the numerical results from the theoretical values at $r/z_0 \geq 5$ is probably due to numerical confinement; as the particles are further separated, larger computational boxes would be necessary to accurately isolate the interaction due solely to the neighbouring particle.

8. Conclusions

We presented here the theory of the hydrodynamic interactions of two spheres in a shear flow in the presence of a plane rigid wall. This theory provides a reference for consistent direct numerical or experimental studies of the particles' trajectories. Some of the predictions of the theory have been confirmed by direct numerical simulations in Poiseuille flow, demonstrating that neglecting

⁴Here, we exploit the fact that the particles travel nearly at a constant velocity relative to the wall despite the relative influence. This allows us to subtract the velocity of particle 1, taken as the undisturbed velocity evaluated at its initial center location, from the underlying flow to obtain a moving reference frame. Doing so automatically satisfies the Galilean invariance and significantly reduces the computational cost by limiting the size of the simulation box.

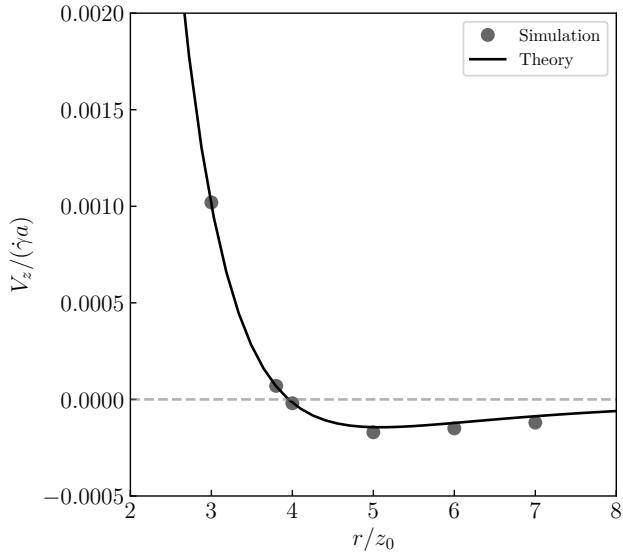


Figure 13: Asymptotic vertical velocity of particle 2 relative to particle 1 as a function of the horizontal separation, at $z_0 = 5a$ (cf. Fig. 1). We focus on the range where the theory predicts change of sign of the velocity and the associated critical point. The theory is seen to hold accurate predictions even in geometrically confined Poiseuille flows.

the farthest wall is a valid assumption and the theory holds at least down to distances from the wall of 5 particle radii, $z_0 \approx 5a$.

The immediate use of our work is the determination of the limitations of the BG theory (Batchelor & Green 1972a), see also Kim & Karrila (2013). Our theory indicates that for inter-particle distances r/a much smaller than $(z_0/a)^{3/5}$ variations of the inter-particle velocity with respect to the BG velocity is small. This condition, $r \ll z_0^{3/5} a^{2/5}$, is stricter than the naive estimate $r \ll z_0$. If this condition is not met then the wall correction to the BG inter-particle velocity is not small. The trajectories are then significantly altered in comparison with the BG predictions, both quantitatively and qualitatively.

Generally speaking our approach to the problem relies on finding the image of the full BG flow that describes particles' interactions in shear flow. It might be anticipated that the above critical distance $z_0^{3/5} a^{2/5}$ follows from the comparison of the scalings of the BG velocity with that of the image. In fact, the distance $\propto z_0^{3/5}$ arises by comparing the BG relative velocity $\propto 1/x^4$ with that $\propto x/z_0^3$ due to the image flow, see Sec. 3. However we did not find a simple explanation for $\propto 1/x^4$ and $\propto x/z_0^3$ scalings.

Corrections due to the wall can also be relevant at $r \ll z_0^{3/5} a^{2/5}$ despite their smallness. The reason is that the global behavior of the BG trajectories passing near the separatrix is sensitive to small perturbations. Thus, perturbations originating from the wall presence, Brownian noise, gravity, finite roughness of the particles' surface or any other source, may easily change the global portrait of the interactions. All the closed trajectories of the classic BG solution (Batchelor & Green 1972a) can be altered quite significantly by small perturbations, since they all pass near the separatrix. Indeed when particles, orbiting around each other in the symmetry plane, reach the vertical (side-by-side) orientation, the maximal distance between them is of order of $10^{-5}a$, see Arp & Mason (1977). The wall, even a distant one, can produce a small upward displacement which would shrink the region of closed trajectories. This is in fact what we see in the simulations where the separatrix in the presence of the wall crosses the z -axis at shorter distance from $z = 2a$ than without the wall. However, our simulations are not built for resolving distances as small as $10^{-5}a$ so this initial observation demands further, more accurate studies, which can take advantage of the evolution equation for the inter-particle separation derived here.

We notice that the problem considered here seemingly has a hidden symmetry. The presence of the wall makes the top-down symmetry, which is displayed by the trajectories, non-evident. Moreover, it is not so evident why the separatrices form a surface of rotation in both the BG and our cases.

Strictly speaking our analysis is not complete. The leading order correction that we found has naive order of smallness z_0^{-2} , and not the actually holding z_0^{-3} . We found this from the detailed calculation that revealed the vanishing of the z_0^{-2} contribution due to symmetry. It is possible that the symmetry would be irrelevant for the next order term which has the naive order of magnitude of z_0^{-3} and it cannot be neglected. We consider this scenario implausible and make the conjecture that the next order term is actually $\mathcal{O}(z_0^{-4})$ and can be consistently neglected. Proving this conjecture theoretically is a formidable task which was not undertaken here. It seemed more practical to test the predictions that we made by direct numerical simulations of the motion of two spheres in a shear flow in the presence of a wall. The performed numerical simulations of the Poiseuille flow closely confirmed the predictions of our theory.

Another confirmation of our theory comes from the previous, unguided by the theory, simulations of Zurita-Gotor *et al.* (2007). This work considered the shear flow between two parallel planes induced by the motion of the upper plane. This problem, with both walls included, could be considered as in Sec. 2 by using the Green's function for the Stokes flow between two infinite planes (Liron & Mochon 1976), which however is beyond the scope of the present paper. The interacting spheres in Zurita-Gotor *et al.* (2007), however, were located closer to the immobile lower wall which makes our theory applicable at least qualitatively. The phase portrait of Zurita-Gotor *et al.* (2007) for the evolution of the inter-particle distance in the symmetry plane agrees remarkably well with that provided here, though it lacks the neutral equilibrium point and the

closed trajectories revolving around it. The authors observed the saddle point at $z_0 = 4.8$ with distance 10 between the walls. In this case, our theory applies only qualitatively. However, when we use our formula $(32z_0^3/15)^{1/5}$ for the position of the saddle point, we find that our prediction agrees very well with the numerical findings of Zurita-Gotor *et al.* (2007). All these provide strong evidence for validity of our theory.

The complete proof showing that our open trajectories with sign-reversal of the vertical separation describe swapping of the vertical positions requires the computation of the vertical coordinate of the center of mass after the interaction. Although as we argued above, this seems inevitable, a proof demands the study of the motion of the center of mass, which was not undertaken here (the formulae of Sec. 2 can be used for this aim). For an unbounded shear flow, the motion of the center of mass could be obtained using the shear resistance matrix, function of the instantaneous distance between the spheres. This matrix can be written in terms of scalar coefficient functions, similar to A and B , with the asymptotic form of this matrix obtained at large separations in Brenner & O'Neill (1972) (see also Arp & Mason (1977)). Considering this matrix and the solution for the inter-particle distance as a function of time as given, one can readily find the center of mass velocity as a function of time. In our case the calculations are even more involved due to the presence of the wall. This is therefore left for future work.

The numerical and experimental tests of our predictions may focus on the emergence of the neutrally stable bound state, when the particle pair flows as a whole at some fixed distance from the wall z_0 . The horizontal component of the inter-particle distance in this state belongs to the circle of radius $4z_0$, although at small z_0 some deviations from $4z_0$ must occur.

The fluid inertia may have non-trivial effect on the relative motion of two spheres in shear flow. It was observed in Kulkarni & Morris (2008) that the BG trajectories are altered by finite but yet small Reynolds number $Re = 0.1$. Besides the open and closed BG-type trajectories, the authors found reversing trajectories that are similar to the swapping trajectories of Zurita-Gotor *et al.* (2007), considered here, and recirculating trajectories of Subramanian & Koch (2006). The authors also observed spiralling trajectories, that do not have a counterpart at zero Re , and did not observe the dancing trajectories discovered here. The authors ascribed the observations to the effect of finite Re of unbounded shear flow, despite being aware that reversing trajectories could be caused by the boundaries. At the same time they found that variation of the computational domain in the streamwise direction does alter the trajectories, so that the simulation results are influenced by the boundaries. The numerical simulations performed here demonstrate that the effects of the boundaries become negligible for the flow domain which is twice longer in the streamwise direction than that used in Kulkarni & Morris (2008). Future work on the effect of the finite fluid inertia must carefully and fully separate the effects by independent variation of Re and z_0 in wide intervals. Notice that even though our numerical

simulations used finite $\text{Re} = 0.1$, there is an excellent agreement with theoretical predictions corresponding to $\text{Re} = 0$, indicating that the described phenomena should be attributed to the wall-bounded flow and not the weak fluid inertia.

Since the experiments of Shen *et al.* (2014, 2016); Ge *et al.* (2019) used droplets and not rigid particles, we shall briefly address how the results obtained for the rigid particles here would change for droplets. Close interactions of rigid particles and droplets are quite different, both qualitatively and quantitatively, see e.g. Kim & Karrila (2013). However at large separations, when the effects of the wall are most relevant, the differences seem to be less significant. We have confirmed this again using direct numerical simulations for two liquid droplets in the same setup as for the solid particles in Sec. 7. We verified that $V_z > 0$ at $r/z_0 = 3$ while $V_z \lesssim 0$ at $r/z_0 = 5$. Thus, there is a point within this range where the velocity vanishes, as in the case of rigid particles. Therefore at least the prediction of the stationary point holds also in the case of liquid droplets. This suggests that the existence of states of marginal equilibrium is a robust phenomenon for pair of particles flowing next to a wall.

The theory presented here has direct generalisations to other distant boundaries. The developed approach can also be used to study the hydrodynamic interactions between suspended particles in other confined shearing flows, such as, e.g., Couette flow. The case of a third particle at a finite distance from the pair of spheres in an unbounded shear flow is also of interest. When the driving flow is enclosed between two parallel planes (*i.e.* a slit geometry), as in Zurita-Gotor *et al.* (2007), the inclusion of the second plane is required for a theoretical analysis, as suggested above.

The present finding of stable configurations of pairs of particles due to hydrodynamic interactions is probably due to the fact that the position of one of the three bodies in interaction – the wall – is fixed. The question whether such configurations can exist for three, or a larger number of flowing particles is left for future work.

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Appendix A. Integral representation of shear flow round spheres

We derive here the integral representation of the flow round spheres driven by shear in an unbounded fluid. The flow obeys,

$$\nabla p = \eta \nabla^2 \mathbf{u}, \quad \mathbf{u}(\infty) \sim \dot{\gamma} z \hat{x}, \quad \nabla \cdot \mathbf{u} = 0, \quad \mathbf{u}(S_\alpha) = \mathbf{V}_\alpha + \boldsymbol{\Omega}_\alpha \times (\mathbf{x} - \mathbf{x}_\alpha), \quad (62)$$

where $\dot{\gamma}$ is the shear rate and as in the main text $\alpha = 1, 2$ are the indices of the spheres and x_α are the coordinates of the centers. Translational and angular velocities are determined from the conditions that the fluid applies to each particle zero net force and torque,

$$\int \sigma_{ik} dS_{\alpha k} = 0, \quad \int (\mathbf{x} - \mathbf{x}_\alpha) \times \sigma_{ik} dS_{\alpha k} = 0, \quad (63)$$

$$\sigma_{ik} \equiv -p\delta_{ik} + \eta(\nabla_i u_k + \nabla_k u_i)$$

where σ_{ik} is the stress tensor. We use the Lorentz-type identity for \mathbf{x}' outside the volume of the spheres,

$$u_i(\mathbf{x}')\delta(\mathbf{x}' - \mathbf{x}) = \frac{\partial}{\partial x'_k} \left[\frac{Y_{il}(\mathbf{x} - \mathbf{x}')\sigma_{lk}(\mathbf{x}')}{8\pi\eta} + u_l(\mathbf{x}')\Sigma_{ilk}(\mathbf{x} - \mathbf{x}') \right]. \quad (64)$$

where Y_{il} is defined in Eq. (7) and Σ_{ilk} defines the stress tensor of the Stokeslet. We have (our definition differs from Kim & Karrila (2013) by insignificant permutation of indices of the symmetric tensor Σ_{ilk}),

$$Y_{il} = \frac{\delta_{il}}{r} + \frac{r_i r_l}{r^3}, \quad \Sigma_{ilk} = \frac{1}{8\pi} \left(\frac{\partial Y_{il}}{\partial r_k} + \frac{\partial Y_{ik}}{\partial r_l} \right) - \frac{r_i \delta_{lk}}{4\pi r^3} = -\frac{3}{4\pi} \frac{r_i r_l r_k}{r^5}, \quad (65)$$

$$\frac{\partial}{\partial x'_k} \Sigma_{ilk}(\mathbf{x} - \mathbf{x}') = \delta_{il}\delta(\mathbf{x} - \mathbf{x}').$$

Integrating Eq. (64) over \mathbf{x}' outside the particles, we find

$$u_i(\mathbf{x}) = \int_{S_\infty} \left(\frac{Y_{il}(\mathbf{x} - \mathbf{x}')\sigma_{lk}(\mathbf{x}')}{8\pi\eta} + u_l(\mathbf{x}')\Sigma_{ilk}(\mathbf{x} - \mathbf{x}') \right) dS'_k - \sum_\alpha \int_{S_\alpha} \frac{Y_{il}(\mathbf{x} - \mathbf{x}')\sigma_{lk}(\mathbf{x}')dS'_k}{8\pi\eta}, \quad (66)$$

where the direction of the normals is outward from the surfaces and S_∞ is the spherical surface with radius R taken to infinity. The prime designates that the integrals are over the \mathbf{x}' variable and the term with an integral of Σ over the particle surfaces vanishes by the rigid body boundary condition (Kim & Karrila 2013; Pozrikidis 1992; Fouxon *et al.* 2017). We observe that the disturbance of the flow caused by the spheres vanishes at infinity so that,

$$\mathbf{u} \sim \dot{\gamma} z \hat{x} + o(const), \quad \sigma_{lk} \sim \eta \dot{\gamma} (\delta_{lx}\delta_{kz} + \delta_{kx}\delta_{lz}) + o(r^{-1}).$$

We use these asymptotic forms for obtaining the integrals over S_∞ . Keeping the lowest order non-vanishing term in the Taylor series of $Y_{il}(\mathbf{x} - \mathbf{x}')$ in \mathbf{x} ,

$$\int_{S_\infty} \frac{Y_{il}(\mathbf{x} - \mathbf{x}')\sigma_{lr} dS'}{8\pi\eta\dot{\gamma}} = -x_m \int_{S_\infty} (\delta_{lx}z' + x'\delta_{lz}) \frac{dS'}{8\pi R} \frac{\partial Y_{il}(\mathbf{x}')}{\partial x'_m}.$$

We find using the form of Y_{ik} in Eq. (65),

$$\frac{\partial Y_{il}}{\partial r_m} = \frac{r^2(r_l\delta_{im} + r_i\delta_{lm} - r_m\delta_{il}) - 3r_i r_l r_m}{r^5}, \quad (67)$$

and obtain,

$$\int_{S_\infty} \frac{Y_{il}(\mathbf{x} - \mathbf{x}')\sigma_{lr}dS'}{8\pi\eta\dot{\gamma}} = -\frac{x_m}{2} \langle (\delta_{lx}z + x\delta_{lz})(x_l\delta_{im} + x_i\delta_{lm} - x_m\delta_{il} - 3x_ix_lx_m) \rangle, \quad (68)$$

where angular brackets stand for averages over the unit sphere,

$$\langle x_i x_k \rangle = \int_{S=1} \frac{x_i x_k dS}{4\pi} = \frac{\delta_{ik}}{3}, \quad \langle x_i x_k x_l x_m \rangle = \frac{\delta_{ik}\delta_{lm} + \delta_{il}\delta_{km} + \delta_{im}\delta_{kl}}{15}. \quad (69)$$

Further, by collecting the different terms,

$$\int_{S_\infty} \frac{Y_{il}(\mathbf{x} - \mathbf{x}')\sigma_{lr}dS'}{8\pi\eta\dot{\gamma}} = \frac{\delta_{iz}x + \delta_{ix}z}{5}. \quad (70)$$

We consider similarly the remaining integral over S_∞ ,

$$\int_{S_\infty} u_l(\mathbf{x}')\Sigma_{lik}(\mathbf{x} - \mathbf{x}')dS'_k = \dot{\gamma} \int_{S_\infty} z'\Sigma_{xik}(\mathbf{x} - \mathbf{x}')dS'_k = x_m \dot{\gamma} \int_{S_\infty} z' \frac{\partial \Sigma_{xik}(\mathbf{x}')}{\partial x'_m} dS'_k. \quad (71)$$

We have from Eq. (65) that,

$$\frac{\partial \Sigma_{xik}}{\partial r_m} = \frac{3}{4\pi} \left(\frac{5xr_ir_kr_m}{r^7} - \frac{r_ir_k\delta_{mx} + r_ix\delta_{mk} + r_kx\delta_{mi}}{r^5} \right),$$

and

$$\int_{S_\infty} u_l(\mathbf{x}')\Sigma_{lik}(\mathbf{x} - \mathbf{x}')dS'_k = 3x_m \dot{\gamma} \langle (4x_z r_i r_m - z r_i \delta_{mx} - x z \delta_{mi}) \rangle. \quad (72)$$

We obtain using Eq. (69),

$$\int_{S_\infty} u_l(\mathbf{x}')\Sigma_{lik}(\mathbf{x} - \mathbf{x}')dS'_k = \dot{\gamma} \left(\frac{4\delta_{ix}z - \delta_{iz}x}{5} \right). \quad (73)$$

Collecting the terms in Eq. (66), we obtain the integral representation given by Eq. (7) in the main text. This representation leads to Eq. (19) in the main text when taking \mathbf{x} on the surface of one of the spheres and using the proper boundary condition. The representation could also be derived by considering the standard integral representation for the correction flow $\mathbf{u} - \dot{\gamma}z\hat{x}$, which also obeys the Stokes equation. In that approach one would need to evaluate integrals on the particles' surfaces instead of S_∞ to find Eq. (7).

The integral representation in Eq. (7) gives readily the multipole expansion of the flow at large distances $x \gg x'$, see Pozrikidis (1992); Kim & Karrila (2013). The leading order term is provided in Eq. (8) of the main text where we use the condition of zero force. This approximation holds at $|\mathbf{x} - \mathbf{x}_\alpha|$ much larger than the radii of the spheres.

Appendix B. Dynamical equations in Cartesian coordinates

We can derive an explicit expression for δV_i using Eqs. (9) and (27). To this end, we introduce $s^2 = x^2 + y^2 + 4z_0^2$, $\sigma = z^2 - 4z_0^2$ and $g^2 = x^4 + y^4 + 4z^2 z_0^2$. We

find after simplifications that δV_i can be written as sums over four components c_i^k (we use dimensionless time $\dot{\gamma}t$),

$$\delta V_x = \frac{5z}{r^4} \sum_{k=1}^4 c_x^k, \quad \delta V_y = \frac{5xyz}{r^4} \sum_{k=1}^4 c_y^k, \quad \delta V_z = \frac{5x}{r^4} \sum_{k=1}^4 c_z^k. \quad (74)$$

The components of δV_x are given by,

$$\begin{aligned} c_x^1 &= \frac{y^2(2s^2x^2 - (s^2 - 5x^2)\sigma)(r^2L + 2x^2M)}{2s^7}, \quad c_x^3 = x^2 c_y^3, \\ c_x^2 &= \frac{2z_0}{\sigma^2} ((1+K)r^4 + (x^2 + z^2)r^2L(r) + 2x^2z^2M), \\ c_x^4 &= -\frac{x^2\sigma}{2s^7} [r^2(5y^2 - s^2)L + (s^2(r^2 + 2x^2) - 5g^2)M], \end{aligned} \quad (75)$$

where the components of δV_y read,

$$\begin{aligned} c_y^1 &= \frac{(2s^2y^2 - (s^2 - 5y^2)\sigma)(r^2L + 2x^2M)}{2s^7}, \\ c_y^2 &= \frac{2z_0}{\sigma^2} (r^2L + 2z^2M), \\ c_y^3 &= \frac{(s^2 - 3y^2)r^2L + (3g^2 - r^2s^2)M}{3s^5}, \\ c_y^4 &= -\frac{\sigma}{2s^7} [r^2(5y^2 - 3s^2)L + (s^2(r^2 + 2y^2) - 5g^2)M]. \end{aligned} \quad (76)$$

Finally the components of δV_z are,

$$\begin{aligned} c_z^1 &= \frac{\sigma}{2s^7} (s^2 - 20z_0^2)[r^4(1 + K + L) + 2z^2(r^2 - z^2)M], \\ c_z^2 &= \frac{2z^2z_0}{\sigma^2} (r^2L + (3z^2 - r^2)M), \\ c_z^3 &= z^2 c_y^3, \quad c_z^4 = \frac{y^2z^2}{s^5} (r^2L + 2x^2M). \end{aligned} \quad (77)$$

We observe from Eqs. (25) and (74)-(77) that $V_i = V_i^0 + \delta V_i$ obeys the symmetries,

$$\begin{aligned} V_x(-x, y, z) &= V_x(x, y, z), \quad V_x(x, y, -z) = -V_x(x, y, z), \\ V_x(x, -y, z) &= V_x(x, y, z), \quad V_y(-x, y, z) = -V_y(x, y, z), \\ V_y(x, -y, z) &= -V_y(x, y, z), \quad V_y(x, y, -z) = -V_y(x, y, z), \\ V_z(-x, y, z) &= -V_z(x, y, z), \quad V_z(x, -y, z) = V_z(x, y, z), \end{aligned} \quad (78)$$

These symmetries, which are rather simple in the case of the infinitely separated wall (Batchelor & Green 1972a), are not destroyed by the corrections due to the finiteness of the separation. They allow us to confine the study of the trajectories to the octant $x > 0$, $y > 0$ and $z > 0$.

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Paper 5

5

Droplets in homogeneous shear turbulence

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We simulate the flow of two immiscible and incompressible fluids separated by an interface in a homogeneous turbulent shear flow at a shear Reynolds number equal to 15 200. The viscosity and density of the two fluids are equal, and various surface tensions and initial droplet diameters are considered in the present study. We show that the two-phase flow reaches a statistically stationary turbulent state sustained by a non-zero mean turbulent production rate due to the presence of the mean shear. Compared to single-phase flow, we find that the resulting steady-state conditions exhibit reduced Taylor-microscale Reynolds numbers owing to the presence of the dispersed phase, which acts as a sink of turbulent kinetic energy for the carrier fluid. At steady state, the mean power of surface tension is zero and the turbulent productionrate is in balance with the turbulent dissipation rate, with their values being larger than in the reference single-phase case. The interface modifies the energy spectrum by introducing energy at small scales, with the difference from the single-phase case reducing as the Weber number increases. This is caused by both the number of droplets in the domain and the total surface area increasing monotonically with the Weber number. This reflects also in the droplet size distribution, which changes with the Weber number, with the peak of the distribution moving to smaller sizes as the Weber number increases. We show that the Hinze estimate for the maximum dropletsize, obtained considering break-up in homogeneous isotropic turbulence, provides an excellent estimate notwithstanding the action of significant coalescence and the presence of a mean shear.

1. Introduction

The understanding of liquid-liquid emulsions is important in many industrial processes e.g. hydrocarbon separation, suspension crystallization, and emulsion polymerization. These flows are characterized by density and viscosity ratios on the order of unity (e.g. water and oil mixtures) and a source of agitation (e.g. an impeller) that creates a turbulent two-phase mixture consisting of a dispersed phase of droplets and a continuous phase. The resulting turbulence in the

carrier phase is altered directly by the droplet feedback on the surrounding fluid and indirectly by droplet-droplet interactions. Many aspects of the complex interaction of the dispersed phase with the continuous phase are not well understood. In particular, there are questions related to the topological changes and to the role of the surface tension of the dispersed phase, the stationarity of the turbulent statistics, and the kinetic energy budget.

Liquid-liquid emulsions have been the subject of numerous experimental (Berkman & Calabrese 1988; Pacek *et al.* 1998; Lovick *et al.* 2005) and computational studies (Perlekar *et al.* 2012; Skartlien *et al.* 2013; Komrakova *et al.* 2015; Scarbolo *et al.* 2015; Dodd & Ferrante 2016). The computational studies can be broadly categorized as forced homogeneous isotropic turbulence (Perlekar *et al.* 2012; Skartlien *et al.* 2013; Komrakova *et al.* 2015), decaying homogeneous isotropic turbulence (Dodd & Ferrante 2016) and turbulent wall flows (Scarbolo *et al.* 2015). Forced homogeneous isotropic turbulence has the advantage of producing a statistically homogeneous and isotropic flow field that, in time, can reach a statistically stationary state. However, in forced homogeneous isotropic turbulence, the turbulent kinetic energy must be induced artificially via a forcing term in the Navier–Stokes equations. This is in contrast to a natural forcing mechanism that produces turbulent kinetic energy from finite Reynolds stresses interacting with a mean velocity gradient. While forcing homogeneous isotropic turbulence may be appropriate for studying the droplet size distributions, it has been argued that artificial forcing is inappropriate for studying two-way coupling effects (Elghobashi 2019). Therefore, for studying the turbulent kinetic energy budget, either decaying isotropic turbulence or turbulent shear flow might be preferable.

In decaying isotropic turbulence, it was shown that the presence of finite-size droplets always enhances the decay rate of the turbulent kinetic energy (Dodd & Ferrante 2016). Also, the deformation, breakup, or coalescence of the droplets introduces an additional term to the turbulent kinetic energy equation - the power of the surface tension - termed Ψ_σ by Dodd & Ferrante (2016), which describes the rate of change of the interfacial energy, balancing the kinetic energy transfer between the external fluid and the flow inside the droplets. Correct identification of these pathways for the turbulent kinetic energy exchange is fundamental to understand the turbulence modulation by the droplets and then to model it.

Building upon previous studies, we consider finite-size bubbles/droplets of Taylor length scale in homogeneous shear turbulence (Tavoularis & Corrsin 1981*a,b*; Pumir 1996; Mashayek 1998; Sekimoto *et al.* 2016). Homogeneous shear turbulence flow is conceivably the simplest case in which the flow remains statistically homogeneous in all spatial directions. Moreover, compared to forced isotropic turbulence, it has a natural energy production mechanism via a mean velocity gradient. We note that ideal homogeneous shear turbulence is self-similar, implying an unbounded energy growth within infinite domains (Sukheswalla *et al.* 2013). This condition limits any numerical simulations to

relatively short times, concerning only the initial shearing of isotropic turbulence (Rogers & Moin 1987; Lee *et al.* 1990; Sukheswala *et al.* 2013). However, as demonstrated by Pumir (1996) and Sekimoto *et al.* (2016) in single-phase flow, the finite computational box introduces a large-scale confinement effect similar to that enforced by a wall; thus, a meaningful statistically stationary state can be reached over long periods, termed statistically stationary homogeneous shear turbulence (SS–HST). In particular, Sekimoto *et al.* (2016) showed that long-term simulations of HST are “minimal” in the sense of containing on average only a few large-scale structures: all the one-point statistics agree well with those of the logarithmic layer in turbulent channel flows, particularly when scaled with the friction velocity derived from the measured Reynolds stresses. The same holds for the wall-parallel spectra of the wall-normal velocity. The authors concluded that the similarities between the steady state homogeneous shear turbulence and other shear flows, particularly with the logarithmic layer of wall turbulence, make it a promising system to study shear turbulence in general. These observations, combined with the insights recently gained in the droplet-turbulence interaction in decaying homogeneous isotropic turbulence, motivate us to further investigate turbulence modulation due to droplets/bubbles in steady state homogeneous shear turbulence.

In this paper, we present DNS of an emulsion created by droplets dispersed in homogeneous shear turbulence. By changing the initial size of the dispersed phase and the Weber number, we aim to answer the following questions:

- (a) Can a statistically stationary state be reached when the dispersed phase actively undergoes breakup and coalescence in homogeneous shear turbulence?
- (b) If so, what determines the steady-state size distribution of the dispersed phase?
- (c) How does the dispersed phase change the turbulent kinetic energy budget?

Homogeneous shear turbulence shares many similarities with other shear flows, including turbulent wall flows (Sekimoto *et al.* 2016); therefore, by answering these questions, we expect to improve our understanding of the droplet-turbulence interaction and, hopefully, help future modelers gain intuition about more complex conditions.

To capture the complex phenomena accurately in a direct numerical simulation of turbulent two-phase flow, we need a numerical method that is reliable and possess the following properties: (i) discrete mass, momentum and kinetic energy conservation, (ii) ability to handle large jumps in density, (iii) ability to handle complex topologies and separation of scales, and (iv) accurate surface tension implementation (Mirjalili *et al.* 2017). In the present work, we choose to use an algebraic volume of fluid method known as THINC (tangent of hyperbola for interface capturing) method, which is a sharp-interface method. This method is relatively new and has been demonstrated to be as accurate and also cost effective compared to the well known geometric volume of fluid methods

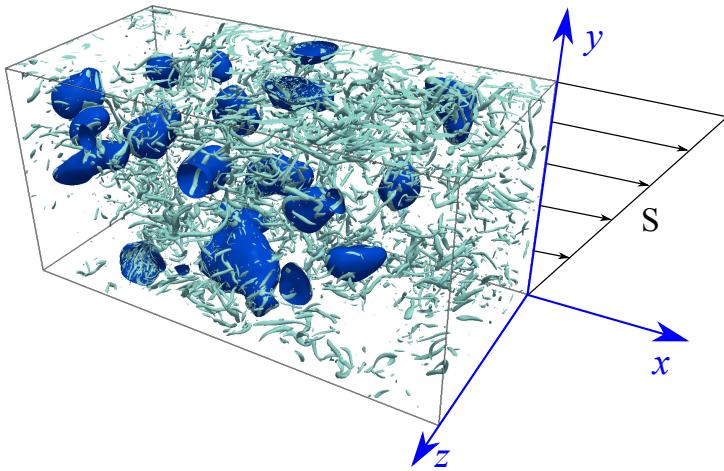


Figure 1: Sketch of the computational domain and of the Cartesian coordinate system. The visualization pertains the flow at $Re_z \approx 15000$ with 5% volume fraction of the dispersed phase at $We_\lambda \approx 0.75$. The blue color is used to depict the surface of the droplets.

in canonical test cases (Xie *et al.* 2014), which makes it a good alternative. However, Mirjalili *et al.* (2017) indicate that large-scale realistic simulations of turbulent two-phase flows using THINC methods are still lacking in the literature and are crucial to fully evaluate the capabilities of these methods (see Rosti *et al.* 2019, for the use of the THINC method for low Reynolds number flows). Hence we choose to use this method in the current study, which will serve as an evaluation of the robustness of THINC methods for complex realistic simulations.

This paper is organized as follows. In section 2, we first discuss the flow configuration and the governing equations and then present the numerical methodology used. The results on the fully developed two-phase homogeneous shear turbulent flow are presented in section 3, where we answer the questions discussed above based on our observations. In particular, we first show how the turbulent flow is modified by the droplets and how the droplets evolve in the turbulent flow, and then explain how these modifications occur by studying the turbulent kinetic energy balance in the two-phase flow. Finally, all the main findings and conclusions are summarized in section 4.

2. Methodology

2.1. Governing equations and numerical methods

We consider the flow of two immiscible incompressible fluids in a periodic box subject to a uniform mean shear S . Figure 1 shows a sketch of the geometry

and the Cartesian coordinate system, where x , y , and z (x_1 , x_2 , and x_3) denote the streamwise, shear, and spanwise coordinates, and u , v , and w (u_1 , u_2 , and u_3) denote the respective components of the velocity field. Standard periodic conditions are applied in x and z , and a shear-periodic boundary condition is enforced in y , *i.e.*

$$u_i(x_1 + L_x, x_2, x_3) = u_i(x_1, x_2, x_3), \quad (1)$$

$$u_i(x_1, x_2 + L_y, x_3) = u_i(x_1 - \mathcal{S}tL_z, x_2, x_3), \quad (2)$$

$$u_i(x_1, x_2, x_3 + L_z) = u_i(x_1, x_2, x_3). \quad (3)$$

The total velocity field u_i can be decomposed for convenience into the sum of a mean component $\langle u_i \rangle_{xz}$ generated by the imposed shear \mathcal{S} , *i.e.* $\langle u_i \rangle_{xz} = \mathcal{S}x_2\delta_{1i}$ where δ_{ij} is the Kronecker delta, and a fluctuating part u'_i ($u'_i = u_i - \langle u_i \rangle_{xz}$). In this article we indicate the spatial average in the x and z directions with $\langle \cdot \rangle_{xz}$, fluctuations with the prime symbol ('), and the average in the full volume with $\langle \cdot \rangle$. The time evolution of the fluctuating velocity u'_i is described by

$$\rho \left(\frac{\partial u'_i}{\partial t} + \frac{\partial u'_i u'_j}{\partial x_j} + \mathcal{S}x_2 \frac{\partial u'_i}{\partial x_1} + \mathcal{S}u'_2 \delta_{1i} \right) = - \frac{\partial p}{\partial x_i} + \frac{\partial \tau_{ij}}{\partial x_j} + f_i, \quad (4)$$

$$\frac{\partial u'_i}{\partial x_i} = 0, \quad (5)$$

where ρ is the fluid density, p is the pressure, $\tau_{ij} = 2\mu D_{ij}$ with μ the dynamic viscosity and D_{ij} the strain rate tensor ($D_{ij} = (\partial u_i / \partial x_j + \partial u_j / \partial x_i) / 2$), and f_i is the surface tension force defined as $f_i = \sigma \kappa n_i \delta$, where δ is the Dirac delta function at the interface, σ the interfacial surface tension, κ the interface curvature and n_i the normal to the interface. This equation is written in the so-called one-fluid formulation (Tryggvason *et al.* 2007) so that only one set of equations is solved in both phases. The problem is solved by introducing an indicator function H to identify each fluid phase so that $H = 1$ in the region occupied by the suspended dispersed fluid (fluid 1) and $H = 0$ in the carrier phase (fluid 2). Considering that both fluids are transported by the flow velocity, we update H in the Eulerian framework by the following advection equation written in divergence form,

$$\frac{\partial \phi}{\partial t} + \frac{\partial u_i H}{\partial x_i} = \phi \frac{\partial u_i}{\partial x_i}, \quad (6)$$

where ϕ is the cell-averaged value of the indicator function.

The above governing equations are solved numerically. First, the transport equation for ϕ is updated following the methodology described by Ii *et al.* (2012) and Rosti *et al.* (2019) in order to obtain ϕ^{n+1} which is used to update the density and viscosity of the fluids. In particular, the indicator function H is approximated as

$$H(X, Y, Z) \approx \hat{H}(X, Y, Z) = \frac{1}{2} \left(1 + \tanh(\beta(P(X, Y, Z) + d)) \right), \quad (7)$$

where $X, Y, Z \in [0, 1]$ is a centered local coordinate system defined in each cell, P is a three dimensional quadratic curved surface function determined algebraically by imposing the correct value of the three normal components and the six components of the Cartesian curvature tensor in each cell, d is a normalization parameter used to enforce that the integral of the indicator function in each cell equals ϕ and β is a sharpness parameter. β is set equal to 1 in the present work, the smallest value allowed by the method which ensures the sharpest possible interface for a given mesh size. Second, the momentum equation and the incompressibility constraint are solved following the method proposed by Gerz *et al.* (1989) and recently adopted by Tanaka (2017), in which the third term on the left-hand side of the momentum equation (equation (4)), *i.e.* the advection due to the mean shear flow, is solved separately using a Fourier approximation. In particular, the second-order Adams–Bashforth method is applied for the convection and viscous terms in equation (4) to obtain an intermediate velocity

$$u_i'^* = u_i'^n + \Delta t \left(\frac{3}{2} \text{rhs}_i^n - \frac{1}{2} \text{rhs}_i^{n-1} \right), \quad (8)$$

where Δt is the time step from time t^n to t^{n+1} and

$$\text{rhs}_i = -\mathcal{S}u_2'\delta_{i1} - \frac{\partial u_i' u_j'}{\partial x_j} + \frac{1}{\rho} \frac{\partial \tau_{ij}}{\partial x_j}. \quad (9)$$

The time step Δt is chosen such that the Courant–Friedrichs–Lewy (CFL) number $U_{max}\Delta t/\Delta x$ is smaller than unity, where $U_{max} = \mathcal{S}L_y$ is the maximum of the mean shear flow velocity inside the computational domain. The advection due to the mean shear flow is then solved separately using a Fourier approximation as

$$u_i'^{**} (x_1, x_2, x_3) = u_i'^* (x_1 - \Delta t \mathcal{S}x_2, x_2, x_3). \quad (10)$$

Note that Tanaka (2017) modified the approach of Gerz *et al.* (1989) by performing a similar additional step for the pressure. Our tests suggest that the original form by Gerz *et al.* (1989) is numerically more stable and physically consistent with the incompressibility constraint because the pressure is not a transported quantity. The surface tension term f_i is then taken into account by updating the velocity field: we use the continuum surface force model by Brackbill *et al.* (1992) to compute the surface tension force where the normals are obtained with the well known Youngs approach (Youngs 1982), *i.e.* $f_i = \sigma\kappa\partial\phi/\partial x_i$, thus obtaining

$$u_i'^{***} = u_i'^{**} + \Delta t \frac{f_i^{n+1}}{\rho}. \quad (11)$$

Then, we enforce the zero divergence of the velocity field by solving the following Poisson equation

$$\frac{\partial^2 p^{n+1}}{\partial x_j \partial x_j} = \frac{\rho}{\Delta t} \frac{\partial u_i'^{***}}{\partial x_i}, \quad (12)$$

Case	Symbol	D_0/L_z	N_0	We_{S_0}	We_{rms_0}	We_0	We_λ	Re_λ
1	-	—	—	—	—	—	—	145
2	●	0.36	4	0.2	0.04	0.5330	0.0220	83
3	●	0.16	51	0.2	0.2	0.8000	0.0776	101
4	●	0.08	564	0.2	1	2.0943	0.9339	111
5	■	0.36	4	1	0.2	2.0944	0.6773	113
6	■	0.16	51	1	1	4.0156	0.7536	117
7	■	0.08	564	1	5	10.4717	4.9313	132
8	▲	0.36	4	5	1	4.1890	2.0103	122
9	▲	0.16	51	5	5	7.9999	4.0868	131
10	▲	0.08	564	5	25	20.9432	13.3057	142

Table 1: Summary of the direct numerical simulations performed with different initial droplet sizes D_0 , numbers of droplets N_0 and surface tension σ , all at a fixed Reynolds number $Re_z = 15200$ and volume fraction $\Phi = 5\%$.

which is solved with a standard FFT-based solver by exploiting the periodic and shear-periodic boundary conditions as detailed in Tanaka (2017). Finally, we correct the velocity with p^{n+1} to enforce the incompressibility constraint

$$u_i'^{n+1} = u_i'^{***} - \Delta t \frac{1}{\rho} \frac{\partial p^{n+1}}{\partial x_i}. \quad (13)$$

Note that, our numerical scheme discretely conserves both momentum and kinetic energy (in absence of viscosity and surface tension) since we use second order centered finite difference on a staggered mesh and the divergence form of the convective terms (Morinishi *et al.* 1998).

2.2. Setup

The problem is governed by several dimensionless parameters, which define the problem under consideration. First, the computational box is defined by two aspect ratios $\mathcal{R}_{xz} = L_x/L_z$ and $\mathcal{R}_{yz} = L_y/L_z$ which are fixed equal to 2.05 and 1.025 respectively. These values have been chosen accordingly to what proposed by Sekimoto *et al.* (2016) as “acceptable” in the sense that they fall within the range of parameters in which the flow is as free as possible from box effects and can thus be used as a model of shear-driven turbulence in general. Indeed, homogeneous shear turbulence in an infinite domain evolves towards larger and larger length scales while simulations in a finite box are necessarily constrained to some degree by the box geometry. These authors noticed that the effect of the geometry can be reduced by ensuring that L_z is the main constraint, thus resulting in the flow being “minimal” in the spanwise direction. Next, once the size of the numerical box is fixed, to fully characterize the problem we define the shear Reynolds number based on the box width

$$Re_z = \frac{\mathcal{S}L_z^2}{\nu}, \quad (14)$$

the Weber number based on the initial droplet diameter D_0

$$We_{S_0} = \frac{\rho S^2 D_0^3}{\sigma}, \quad (15)$$

and the ratio of the initial droplet diameter to the box size $\mathcal{R}_{Dz} = D_0/L_z$. In the following, we consider one case of single-phase flow as reference and nine cases of two-phase flows, all at the same Reynolds number equal to 15200; in the multiphase cases, we vary the ratio \mathcal{R}_{Dz} and We_{S_0} , as summarized in table 1. Note that, the Weber number here is mainly determined by the interfacial surface tension σ . Two other nondimensional parameters are the density and viscosity ratios, which are fixed to unity to study the individual effect of the Weber number (interfacial surface tension).

Besides the parameters just defined and based on the geometrical dimensions and initial and boundary conditions alone, in the following discussion we will use other nondimensional numbers because they turned out to be more relevant to understand the problem at hand; in particular, the two non-dimensional parameters which characterize the single-phase homogeneous shear turbulent flows, the Taylor-microscale Reynolds number Re_λ and the shear-rate parameter S^* , defined as

$$Re_\lambda = \left(\frac{2\mathcal{K}}{3} \right)^{1/2} \frac{\lambda}{\nu} = \left(\frac{5}{3\nu\varepsilon} \right)^{1/2} 2\mathcal{K}, \quad (16)$$

and

$$S^* = \frac{2S\mathcal{K}}{\varepsilon}, \quad (17)$$

where $\lambda = \sqrt{10\nu\mathcal{K}/\varepsilon}$ is the Taylor microscale (Sekimoto *et al.* 2016), $\mathcal{K} = \langle \rho u'_i u'_i \rangle / 2$ is the turbulent kinetic energy per unit volume, and $\varepsilon = \mu \langle \partial u'_i / \partial x_j : \partial u'_i / \partial x_j \rangle$ is the dissipation rate of the fluctuating energy. These two non-dimensional numbers can be interpreted as the ratio of the eddy-turnover time $\tau_0 = (2\mathcal{K})^{1/2} / \varepsilon$ and the Kolmogorov time scale $\tau_K = (\nu/\varepsilon)^{1/2}$ and the mean shear time scale $\tau_S = 1/S$, respectively.

Weber numbers can be defined in several ways. In equation (15) we defined the Weber number based on the mean shear, but it can also be defined based on the velocity fluctuations, thus obtaining

$$We_{rms_0} = \frac{2\rho\mathcal{K}D_0}{\sigma}. \quad (18)$$

Note that, the latter definition is the one usually used in homogeneous isotropic turbulent flows in the absence of a mean flow (Dodd & Ferrante 2016). Both the Weber numbers We_{S_0} and We_{rms_0} are of interest since they are based on two different mechanisms that may affect the droplets dynamics: on large scales (large droplets) the effect of the mean shear is dominant, while on small scales (small droplets) the flow is mainly dominated by the isotropic turbulent fluctuations. Our set of parameters is chosen such that the ratio of these two Weber numbers We_{S_0}/We_{rms_0} equals 1/5, 1 and 5, as reported in figure 2. In

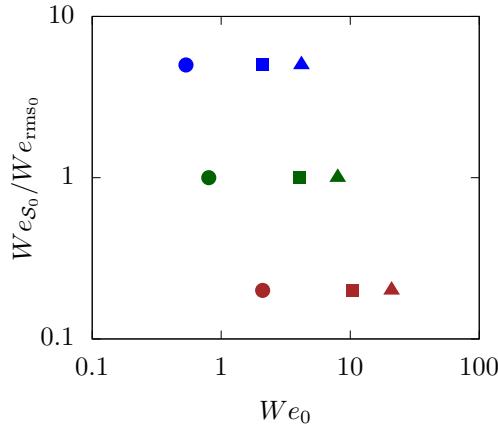


Figure 2: The ratio of the two Weber numbers introduced here, one based on the mean shear We_{S_0} and one on the velocity fluctuations We_{rms_0} , as a function of the Weber number based on the initial droplet size, We_0 . The circle, square and triangle symbols are used to distinguish cases with different surface tension but same ratio We_{S_0}/We_{rms_0} , while the brown, green and blue colors represent cases with the ratio We_{S_0}/We_{rms_0} equal to $1/5$, 1 and 5 , respectively. These symbols and color scheme will be used throughout the rest of the paper.

general both the mechanisms are present together and hence we can define a Weber number which incorporates both the effects as

$$We_0 = \frac{\rho (\sqrt{2\mathcal{K}} + \mathcal{S}D_0)^2 D_0}{\sigma}. \quad (19)$$

Finally, we can define a Weber number based on λ as

$$We_\lambda = \frac{\rho (\sqrt{2\mathcal{K}} + \mathcal{S}\lambda)^2 \lambda}{\sigma}. \quad (20)$$

The choice of using λ in the definition of the Weber number instead of a dimension associated to the suspended phase is due to the fact that the interface is not only deforming, thus losing its original spherical shape, but also actively undergoing merging and break-up processes, which makes the definition of a unique dimension difficult. Therefore, we propose to rely on a fluid length scale, which, as shown below in the results, yields a good collapse of our data. In the following discussion, we use We_λ to discuss the results; the value of We_0 is reported in order to fully characterize the initial conditions of the present simulations.

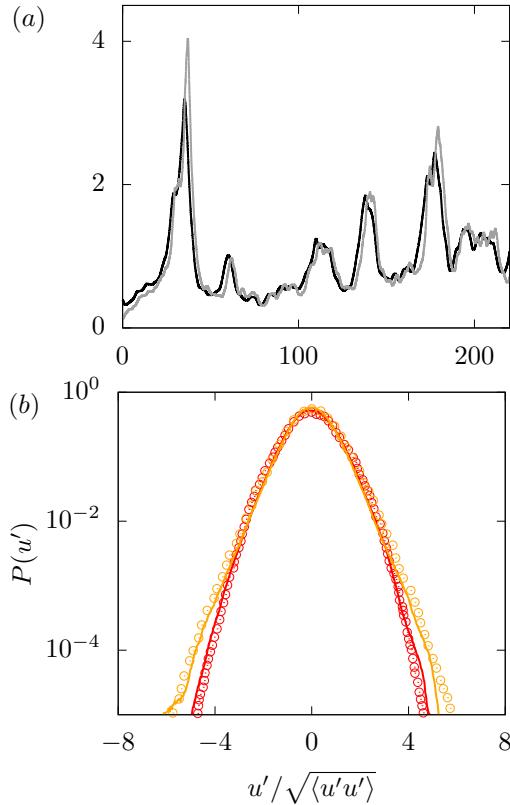


Figure 3: (a) Time history of the turbulent kinetic energy $\mathcal{K} = \langle u'_i u'_i \rangle / 2$ (black line) and enstrophy Ω (grey line), normalized by their mean values. (b) Normalized histogram of the streamwise (red) and shear (orange) components of the velocity fluctuations, u' and v' . The lines and symbols are used to distinguish our results (lines) from those by Pumir (1996).

2.3. Code validation

The numerical code used in this work has been extensively validated in the past for multiphase turbulent flows simulations (Rosti & Brandt 2017; Rosti *et al.* 2018a,b). Here, we provide one more comparison with literature results for the specific case of HST. The single-phase homogeneous shear turbulence has been validated by reproducing one of the cases investigated by Pumir (1996); in particular, we simulated the Run No. 2 in that paper. The initial condition at $t = 0$ is a homogeneous isotropic turbulent field at $Re_\lambda = 50.8$, obtained in a square computational box of size 2π discretised with 256 grid points in each direction. From the time history of the turbulent kinetic energy \mathcal{K} and of the enstrophy $\Omega = \langle \omega_i \omega_i \rangle$, shown in figure 3(a), we observe a first transient

phase for $0 \leq t\mathcal{S} \leq 30$, where the kinetic energy and enstrophy grow rapidly, followed by a statistically stationary state characterized by a cyclic succession of turbulent kinetic energy peaks rapidly followed by a peak in enstrophy with a time lag of approximately $5\mathcal{S}$. This behavior is well captured in our simulation. A quantitative validation is performed first by comparing the mean components of the velocity anisotropy tensor, $b_{ij} = \langle u_i u_j / u'_k u'_k - \delta_{ij}/3 \rangle$ computed in our simulations ($b_{11} = 0.231$, $b_{22} = -0.129$, $b_{33} = -0.101$, $b_{12} = -0.147$) with the data reported by Pumir (1996), and we found that the differences are below 5%. A further comparison is shown in figure 3(b) where the normalized histograms of the streamwise and shear components of the velocity obtained with the present simulations are compared with the results reported in the literature (Pumir 1996); again we observe a very good agreement.

3. Results

3.1. Statistically stationary state

We start our analysis by considering the single-phase flow at $Re_z = 15200$. The problem is solved numerically on a computational mesh of $1312 \times 640 \times 624$ grid points and the simulation is run for approximately $250\mathcal{S}$ time units. Note that, the grid spacing is chosen sufficiently small for good resolution of the smallest turbulent scales as indicated by $\Delta x/\eta \approx 0.7$, where η is the Kolmogorov scale defined as $\eta = (\mu/\rho)^{3/4}/\varepsilon^{1/4}$. The initial flow field is fully developed single-phase homogeneous isotropic turbulence, and the mean shear \mathcal{S} is applied from $t = 0$. As shown in figure 4(a), once the shear is applied, the flow undergoes an initial transient characterized by a strong increase in the production of turbulent kinetic energy, which is not in balance with the dissipation rate. After some time, however, the turbulent kinetic energy \mathcal{K} decreases owing to an increase in the dissipation, reaching a new statistically steady state where, on average, the production balances the dissipation ($\mathcal{P} \approx \varepsilon$). This state, called steady-state shear turbulence, was first found and characterized by Pumir (1996) and later investigated by others (e.g. Sekimoto *et al.* 2016). The resulting Taylor microscale Reynolds number at the steady state is equal to $Re_\lambda \approx 145$ with the averaged spectrum of the TKE reported in figure 4(b). Owing to the high Reynolds number, a clear $k^{-5/3}$ regime develops at intermediate scales. We also observe that the spectra of each individual component of the velocity are different at small wave numbers because of the large-scale anisotropy, while all spectra coincide at higher wave numbers, consistently with what observed by Pumir (1996).

We now consider the multiphase problem. After around $100\mathcal{S}$, when the single-phase flow has already reached a statistically steady state, we inject spherical droplets into the domain at random locations, globally enclosing a volume fraction of the carrier phase of 5%. The initial droplet diameter D_0 is in the inertial range, as shown in figure 4(b) with the vertical dashed lines. In particular, three different initial diameters are chosen, $D_0/L_z \approx 0.08$ (brown), 0.16 (green), and 0.32 (blue), corresponding to approximately 1.1, 2.5,

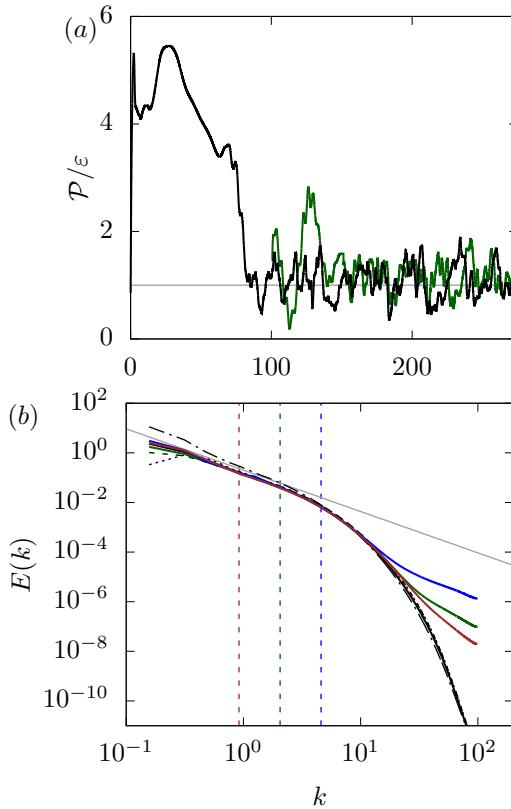


Figure 4: (a) Time history of the ratio between the turbulent production $\mathcal{P} = -\langle u'v' \rangle d\langle u \rangle / dy$ and the turbulent dissipation rate $\varepsilon = \mu \langle \partial u'_i / \partial x_j \partial u'_i / \partial x_j \rangle$. The black and green lines represent the single and multiphase flows ($D_0 = 0.16L_z$ and $We_\lambda \approx 0.75$), respectively. (b) Spectra of the mean turbulent kinetic energy (black solid line) and its three spatial components (black dashed, dotted, and dashed-dotted lines) for the single-phase flow. The other three colored solid lines (blue, green and brown) are used for the spectra of the two-phase flows with $We_\lambda = 0.02, 0.75$ and 5 . The grey line is $\propto k^{-5/3}$, and the three vertical dashed lines represent the initial size of the droplets. The spectra are normalized by multiplying by $\varepsilon^{-2/3}$.

and 5.6 times the single-phase Taylor microscale λ . After the introduction of the dispersed phase, a new short transient arises lasting approximately $50\mathcal{S}$, eventually leading to a new statistically steady state, as depicted in figure 4(a). Also, in the multiphase case, we observe that, at regime, the turbulent production balances on average the dissipation rate ($\mathcal{P} \approx \varepsilon$).

The presence of the droplets modifies the flow profoundly. The averaged spectrum of the turbulent kinetic energy in both phases in the two-phase case is reported in figure 4(b), where we observe that the interface mostly affects the large wave numbers (small scales) for which higher levels of energy are evident, while slightly lower energy is present at the large scales. Note that, the result is analogous to what was observed in decaying homogeneous isotropic turbulence for solid particles (Lucci *et al.* 2010) and bubbles (Dodd & Ferrante 2016); the increased energy at high wave numbers has been explained by the breakup of large eddies due to the presence of the suspended phase and the consequent creation of new eddies of smaller scale. In the same figure we can also observe that the effect of the droplets decreases as the Weber number increases; in other words, the spectra of the multiphase cases approach the single phase one as We increases, while for low We the spectra depart from the single phase case at smaller and smaller wavenumbers.

As already discussed above, We_0 is the Weber number based on the initial droplet size, but since the droplets break up or coalesce, this measure is not fully representative of the final state of the multiphase problem; because of that, in the following sections we prefer to use the Weber number based on a flow length scale, We_λ , reported in figure 5(a) as a function of We_0 . We can observe that the two Weber numbers are well correlated, with We_λ scaling approximately as the square of We_0 , *i.e.* $We_\lambda \propto We_0^2$. The good level of correlation between the two definitions is a further demonstration that for the parameter range considered here the Weber number variations are mainly due to the changes of the interfacial surface tension rather than the chosen length scale.

We quantify the turbulence modulation by examining the resulting Re_λ , shown for all our simulations in figure 5(b) as a function of the Weber number based on the Taylor microscale We_λ , and also reported in table 1. We can observe that the Reynolds number grows with We_λ and that all the two-phase flow cases exhibit lower Taylor microscale Reynolds numbers than the single-phase case. Moreover, we observe that the difference decreases as the Weber number increases, with the two-phase flow cases approaching the single phase one as We_λ increases, consistently with what was already observed in figure 4(b). Indeed, the Reynolds number for the case with the most rigid droplets ($We_\lambda \approx 0.02$) is approximately half the single phase value (-41%), while the difference with the single phase flow is only 2% in the most deformable case ($We_\lambda \approx 13$). Note that, in the context of unbounded forced turbulent flows, such as homogeneous isotropic turbulence and homogeneous shear turbulence, a reduction of the Reynolds number can be interpreted as a drag increase, contrary to what is usually found in wall-bounded flows with constant flow rates where a reduction in the friction Reynolds number leads to drag decrease.

As first noteworthy result, the above data demonstrate that a statistical stationarity is not unique to single-phase homogeneous shear turbulent flows, but it is also realizable in the presence of a second, dispersed phase. Here, we have defined the stationary state in terms of the statistical properties of the

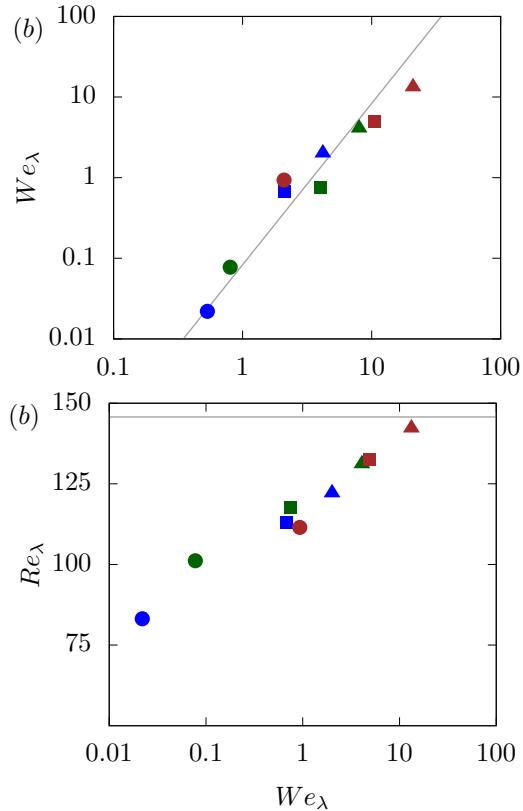


Figure 5: (a) Weber numbers based on the Taylor microscale, We_λ , as a function of the initial Weber number We_0 and (b) Reynolds numbers based on the Taylor microscale, Re_λ , as a function of We_λ . The grey solid line in the left panel is a fit to our data in the form of $We_\lambda \propto We_0^2$, while the grey solid line in the right panel represents the Taylor microscale Reynolds number Re_λ of the single phase flow.

flow averaged over both phases, but since the droplets can also break up or coalesce, it is natural to ask what the steady-state size distributions are and how that relates to the turbulence features. These questions are answered in the following sections.

3.2. Size distribution

We now study the transient and steady state property of the interface separating the two fluids. Figure 6 shows instantaneous snapshots of the two-phase flow at the statistically steady state, which is characterized by droplets with different sizes and shapes: in general we can observe that small droplets are approximately

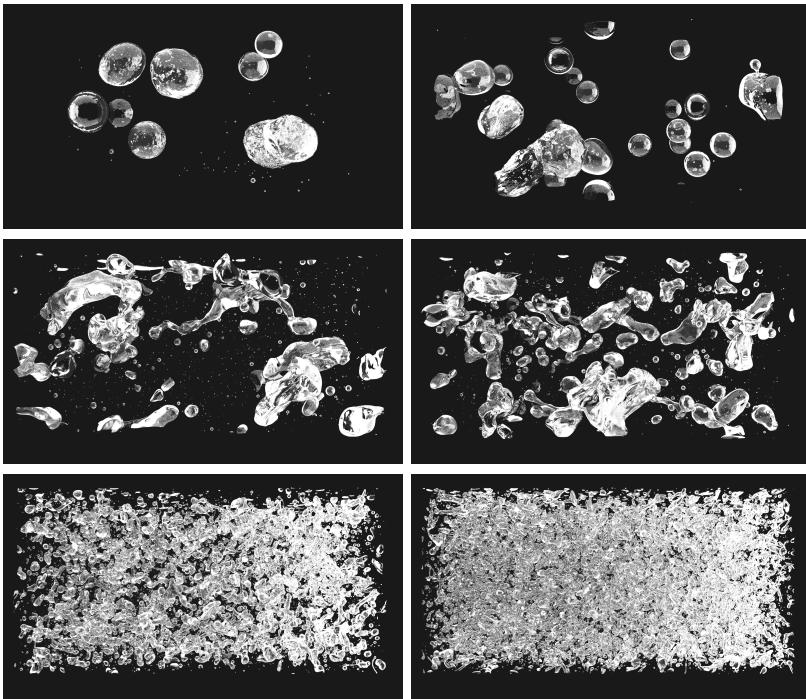


Figure 6: Visualisation in the $x - y$ plane of the interface in the homogeneous shear turbulent flow for different We_λ : (top left) $We_\lambda \approx 0.02$, (top right) 0.08, (middle left) 0.8, (middle right) 4, (bottom left) 5 and (bottom right) 13. In the figures the flow is from left to right.

spherical, while the largest ones have very anisotropic shapes and show a preferential alignment with the direction of the mean shear. Also, as the Weber number decreases, the droplets size increases and larger droplets can sustain the spherical shape.

Figure 7(a) shows the temporal evolution of the number of droplets (\mathcal{N}) under various We_λ and initial sizes D_0 . The counting of the droplets is conducted automatically by checking the connectivity of the local VOF field (ϕ) using a n -dimensional image processing library⁵. We observe that \mathcal{N} has an initial transient phase of same duration as the fluid transient phase observed previously in figure 4(a) ($t\mathcal{S} \lesssim 50$), before the droplets count approaches a statistically steady value for all the cases considered, consistently with the statistically stationarity of the averaged global flow quantities. Note that, the final state is a statistically steady state since the number of droplets \mathcal{N} is not constant but continuously varies and oscillates around a mean value, denoted later on

⁵Scipy.ndimage, <https://docs.scipy.org/doc/scipy/reference/ndimage.html>.

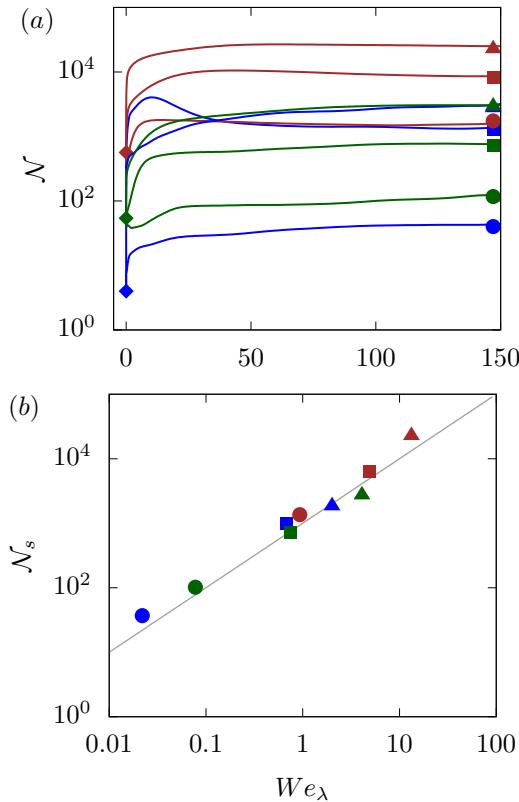


Figure 7: (a) Time history of the number of droplets \mathcal{N} in the domain for different Weber numbers. The rhombus symbols at $t = 0$ represent the initial number of droplets. (b) The mean number of droplets \mathcal{N}_s at the statistically steady state as a function of the Weber number We_λ . The grey solid line in the right panel is a fit to our data in the form of $\mathcal{N}_s \propto We_\lambda$.

as \mathcal{N}_s . From the figure we observe also that the initial transient phase differs among the cases, with three distinct behaviors evident: *i*) in most of the cases, \mathcal{N} increases rapidly after the injection (within $t\mathcal{S} \approx 10$); however, the growth slows down and \mathcal{N} reaches its final steady state value almost monotonically; *ii*) cases 4 and 5 exhibit a significant overshoot of the number of droplets \mathcal{N} for short times before \mathcal{N} reduces to the final regime values due to the coalescence; *iii*) case 3 shows an initial decrease of the number of droplets followed by an increase. Notwithstanding the different behaviors, in all the cases the final number of droplets is always larger than the initial one.

The steady-state value of the number of droplets \mathcal{N}_s as a function of We_λ is reported in figure 7(b); we observe that \mathcal{N}_s grows monotonically with We_λ

(see also the visualisations in figure 6) and that the growth is nearly linear over the three decades spanned in the present study, *i.e.* a fit to our data produces $\mathcal{N}_s \propto We_\lambda$ with an exponent of 1. Since a high Weber number corresponds to a low surface energy, we conjecture that \mathcal{N}_s grows indefinitely with We_λ . Note also that, cases 5 and 6 which have different initial droplet diameters, have almost the same final count of droplets \mathcal{N}_s as well as We_λ . This provides additional evidence that the droplet statistics are better defined by the Weber number We_λ based on the flow quantities rather than by that based on the initial droplet size We_0 . These results suggest that the relative strength between the breakup and coalescence reflects the history of the flow features, and at equilibrium measurable quantities depend only on the global physical parameters.

Next, we aim to characterize the steady state size distribution of the emulsion. Thus, we first examine the cumulative volume, \mathcal{V} , as a function of the equivalent spherical diameter D defined as the diameter of the sphere occupying the same volume, see figure 8(a). Specifically, figure 8(a) shows the steady-state distributions of all cases, where each point on the curves represents the total volume of the droplets with equivalent diameter lower than D . In the figure, both \mathcal{V} and D are normalized by the global maximal values so that the curves are bounded uniformly from above by 1. The figure shows that the cumulative volume distribution only has one inflection point ($d^2\mathcal{V}/dD^2 = 0$), thus indicating that the probability density plot ($d\mathcal{V}/dD$) is single peaked. In figure 8(a) the Weber number We_λ grows from right to left, as indicated by the list of symbols, suggesting that small droplets tend to be more common at high Weber numbers. Additionally, the range of the droplet diameters also narrows with increasing We_λ , since the cumulative volume grows faster to unity, as visually confirmed in figure 6. Case 2, blue line with circle, is the only simulation exhibiting a double peak (*i.e.* $d\mathcal{V}/dD$ has two local maxima): this is due to the presence of very small droplets together with few large ones as can be seen in figure 6(left). Nevertheless, the overall trend of decreasing size for increasing Weber number is still consistent with the linear scaling between \mathcal{N}_s and We_λ , as already observed in figure 7(b). The two bottom panels in figure 8, are contours of $\mathcal{V}/\mathcal{V}_{\text{tot}}$ as a function of the equivalent diameter D and time, and can thus be interpreted as a cumulative spectrogram with most of the droplets centered in the region where the gradient of the color is the largest. In particular, we selected two specific cases, with same initial Weber number $We_0 \approx 2$, but different initial droplet size and surface tension, thus leading to different We_λ . The two figures show the transient behavior for cases 4 and 5, respectively: in figure 8(b) the mean size distribution remains relatively unchanged over time but it is subject to strong fluctuations, while figure 8(c) shows a clear shift of the population from large droplets to small ones, with a statistically steady state characterized by small fluctuations.

Another important parameter related to the size distribution is the largest droplet size, D_{\max} . Assuming breakup of droplets due to the dynamic pressure

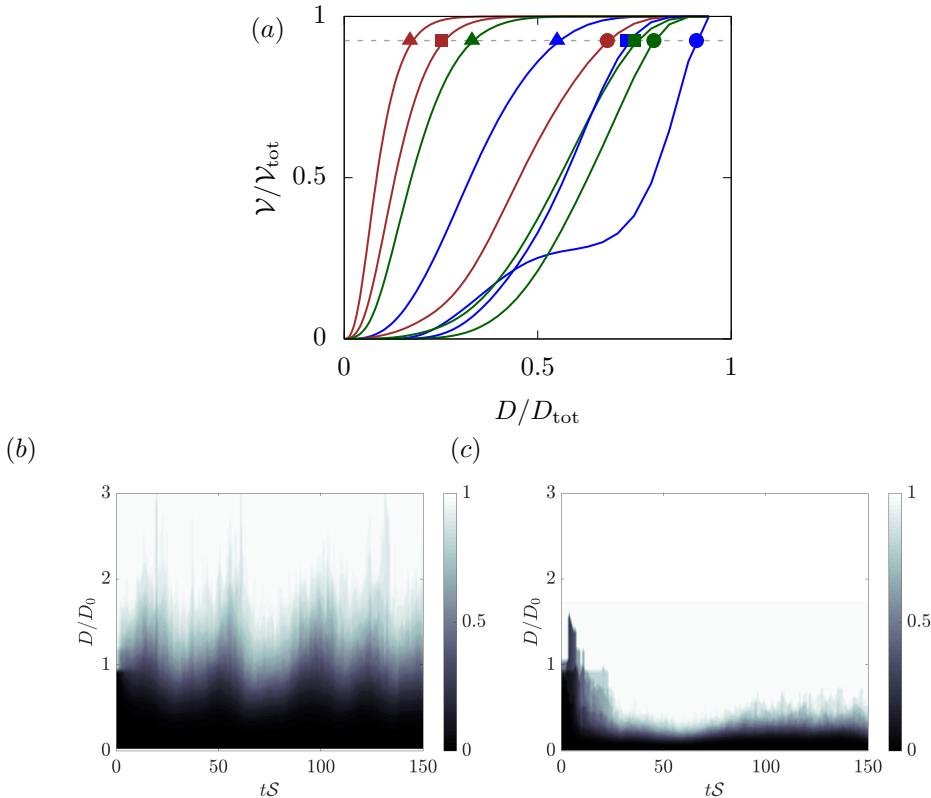


Figure 8: (a) Normalized cumulative volume distributions $\mathcal{V}/\mathcal{V}_{\text{tot}}$ of the dispersed phase at the steady state as a function of the equivalent spherical droplet diameters D . The horizontal grey line correspond to the level $\mathcal{V} = 0.95\mathcal{V}_{\text{tot}}$. (b-c) Contour of the temporal evolution of the normalized cumulative volume distributions of the dispersed phase as a function of the equivalent spherical droplet diameter for cases 4 (b) and 5 (c).

($\sim \rho U^2$), Hinze (1955) proposed that the largest possible droplet in a turbulent emulsifier is determined by the velocity fluctuation across D_{max} , *i.e.* one can define a critical Weber number $We_{\text{crit}} = \rho u'^2 D_{\text{max}}/\sigma$, above which the droplet breaks up. Hinze (1955) showed that simple dimensional analysis leads to $D_{\text{max}} \propto \varepsilon^{-2/5}$, if isotropy prevails and the scaling by Kolmogorov (1941) is assumed valid. D_{max} can be in general approximated by the diameter of the equivalent droplet occupying 95% of the total dispersed volume, *i.e.* $D_{\text{max}} \approx D_{95}$, which is represented in figure 8(top) with the dashed grey line. The symbols in the same figure provide the values of D_{95} for our data. Figure 9(a) shows the normalised D_{95} as a function of the scaled energy input, and indeed we can observe that our data scales with an approximately $-2/5$ slope. We remark

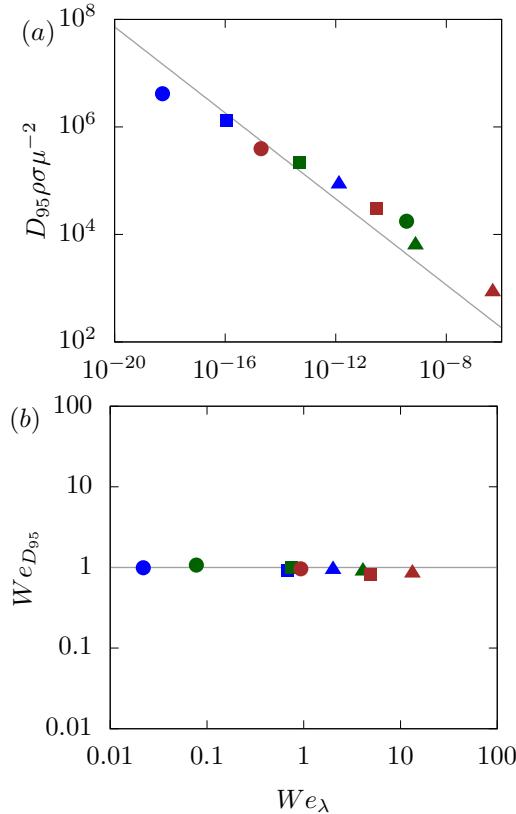


Figure 9: (a) Normalised maximum droplet size D_{95} as a function of the energy input ε . The grey solid line is the relation $\rho\sigma D_{95}/\mu^2 = 0.725 (\mu^5 \epsilon / \rho\sigma^4)^{-2/5}$ proposed by Hinze (1955). (b) Critical Weber number $We_{D_{95}}$ based on the maximum droplet size D_{95} for all the cases considered.

that, although Hinze developed his theory considering only isotropic turbulent flows dominated by the breakup process and neglecting the coalescence, he hypothesized that the same scaling law might still hold for non-isotropic flows provided that the droplet sizes fall within the inertial range, such as in all our cases. More importantly, the success of the Hinze theory relies on the central assumption that breakup results from the dynamic pressure force, corresponding to a fixed critical Weber number. This is clearly shown in figure 9(b), which shows the Weber number based on D_{95} as a function of We_λ . For all our cases, we obtain that $We_{crit} \approx 1$. Our results thus confirm that the $-2/5$ scaling between the maximum droplet diameter and the turbulence dissipation applies not only to isotropic turbulence, but also to the homogeneous shear turbulence that we have analyzed.

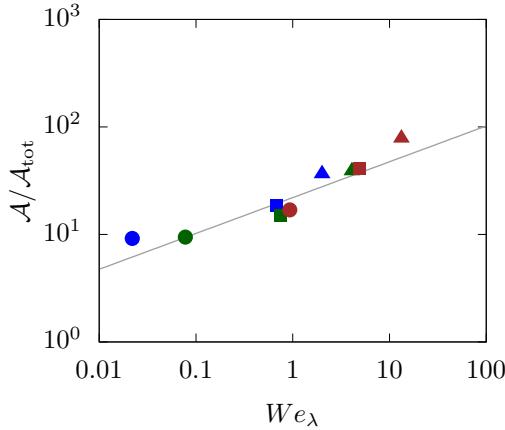


Figure 10: Total interfacial area \mathcal{A} as a function of the Weber number We_λ . The grey solid line is a fit to our data in the form of $\mathcal{A} \propto We_\lambda^{1/3}$.

Finally, we can further characterize the size distribution of the emulsion by inspecting the total surface area \mathcal{A} of the dispersed phase. This quantity is very important when studying multiphase flows with interfaces, since the rate of work due to the surface tension is equal to the product of the surface tension coefficient and the rate of change in interfacial surface area (Dodd & Ferrante 2016); also, for many industrial applications, the total surface area is often the most important parameter as surfactants tend to reside on the interface or it determines the chemical reaction rate. Figure 10 reports the steady state surface area \mathcal{A} as a function of the Weber number We_λ and clearly shows that the surface area increases monotonically with the Weber number. As we have shown above that $\mathcal{N} \propto We_\lambda$, combining with mass conservation, *i.e.* $\mathcal{N}D^3 \propto 1$, leads to the following relation for the total area: $\mathcal{A} \propto \mathcal{N}D^2 \propto We_\lambda^{1/3}$. In other words, the surface area of the droplets shall also increase with the Weber number defined by the Taylor length of the flow, with a slope of 1/3. Figure 10 verifies this scaling. We remark that in the derivation above, we have assumed that the droplets are spherical, which is not always true in our cases. However, provided the linear scaling between \mathcal{N} and We_λ remains valid, we expect the 1/3 scaling law to hold for a wide range of emulsions.

3.3. Turbulent kinetic energy budget

We now study how the multiphase nature of the problem affects the turbulent kinetic energy. To do so, we derive the turbulent kinetic energy evolution equation by first multiplying the momentum conservation equation equation (4)

by the velocity fluctuation u'_i ,

$$\rho \left(\frac{\partial u'_i u'_i / 2}{\partial t} + \frac{\partial u'_i u'_i u'_j / 2}{\partial x_j} + \mathcal{S} x_2 \frac{\partial u'_i u'_i / 2}{\partial x_1} + \mathcal{S} u'_1 u'_2 \right) = -\frac{\partial u'_i p}{\partial x_i} + u'_i \frac{\partial \tau_{ij}}{\partial x_j} + u'_i f_i. \quad (21)$$

We make use of

$$u'_i \frac{\partial \tau_{ij}}{\partial x_j} = \frac{\partial u'_i \tau_{ij}}{\partial x_j} - \tau_{ij} \frac{\partial u'_i}{\partial x_j} = \frac{\partial u'_i \tau_{ij}}{\partial x_j} - \tau_{ij} \mathcal{D}_{ij}, \quad (22)$$

to obtain

$$\begin{aligned} \rho \left(\frac{\partial u'_i u'_i / 2}{\partial t} + \frac{\partial u'_i u'_i u'_j / 2}{\partial x_j} + \mathcal{S} x_2 \frac{\partial u'_i u'_i / 2}{\partial x_1} + \mathcal{S} u'_1 u'_2 \right) &= -\frac{\partial u'_i p}{\partial x_i} + \frac{\partial u'_i \tau_{ij}}{\partial x_j} - \\ &\quad \tau_{ij} \mathcal{D}_{ij} + u'_i f_i. \end{aligned} \quad (23)$$

Equation (23) can then be either volume averaged over both phases to obtain the total kinetic energy equation, or phase averaged over the phase m (e.g. carrier or dispersed phase) to obtain the turbulent kinetic energy evolution equation for one phase only.

The equation for the two-fluid mixture is obtained by applying the volume averaging operator

$$\langle \cdot \rangle = \frac{1}{\mathcal{V}} \int_{\mathcal{V}} \cdot \, d\mathcal{V}, \quad (24)$$

leading to

$$\frac{d\mathcal{K}}{dt} = \mathcal{P} - \varepsilon + \Psi_{\sigma}, \quad (25)$$

where the different terms indicate the rate of change of turbulent kinetic energy \mathcal{K} , the turbulent production rate \mathcal{P} , the dissipation rate ε and the power of the surface tension Ψ_{σ} , defined as

$$\mathcal{K} = \langle \rho u'_i u'_i \rangle / 2, \quad \mathcal{P} = -\mathcal{S} \langle \rho u'_1 u'_2 \rangle, \quad \varepsilon = \langle \tau_{ij} \mathcal{D}_{ij} \rangle, \quad \Psi_{\sigma} = \langle u'_i f_i \rangle. \quad (26)$$

Ψ_{σ} is the rate of work performed by the surface tension force on the surrounding fluid. It represents exchange of turbulent kinetic energy and interfacial surface energy and can be either positive or negative and thus a source or sink of turbulent kinetic energy. In particular, Ψ_{σ} is proportional to the rate at which surface area is decreasing, *i.e.* $\Psi_{\sigma} \propto -dA/dt$ (Dodd & Ferrante 2016), and therefore decreasing (increasing) interfacial area through droplet restoration (deformation) or coalescence (breakup) is associated with Ψ_{σ} being a source (sink) of turbulent kinetic energy. Note that all the transport terms in equation (23) vanish due to the homogeneity of the domain. On the other hand, if we apply the phase average operator

$$\langle \cdot \rangle_m = \frac{1}{\mathcal{V}_m} \int_{\mathcal{V}_m} \cdot \, d\mathcal{V}, \quad (27)$$

we obtain

$$\frac{d\mathcal{K}_m}{dt} = \mathcal{P}_m - \varepsilon_m + \mathcal{T}_m^{\nu} + \mathcal{T}_m^p, \quad (28)$$

where the different terms now indicate the rate of change of turbulent kinetic energy \mathcal{K}_m , the turbulent production rate \mathcal{P}_m , the dissipation rate ε_m and the viscous \mathcal{T}_m^ν and pressure \mathcal{T}_m^p powers of the phase m , defined as

$$\begin{aligned} \mathcal{K} &= \langle \rho u'_j u'_j \rangle_m / 2, \quad \mathcal{P}_m = -\mathcal{S} \langle \rho u'_1 u'_2 \rangle_m, \quad \varepsilon = \langle \tau_{ij} \mathcal{D}_{ij} \rangle_m, \\ \mathcal{T}_m^\nu &= \left\langle \frac{\partial u'_i \tau_{ij}}{\partial x_j} \right\rangle_m, \quad \mathcal{T}_m^p = -\left\langle \frac{\partial u'_i p}{\partial x_i} \right\rangle_m. \end{aligned} \quad (29)$$

In this case, the viscous and pressure transport terms are retained to account for a net flux of turbulent kinetic energy from one phase to the other caused by the coupling between the droplets and the carrier fluid (this physical interpretation can be seen more clearly by applying the Gauss's theorem to rewrite the terms as surface integrals, thus resulting in surface integration over the droplet surface). Note finally that the convective transport terms are zero because the two fluids are immiscible and therefore turbulent eddies can not transport turbulent kinetic energy across the interface.

First, we focus on the equation for \mathcal{K} obtained by averaging over the whole volume and over both phases (equation (25)). At steady state, the rate of change of \mathcal{K} is obviously zero and the remaining terms are the production and dissipation rates and the power of surface tension. Figure 11 shows the production \mathcal{P} and dissipation ε rates, normalized by their single-phase values \mathcal{P}_0 and ε_0 , for all the simulations performed in the present study as a function of the Weber number We_λ . We observe that both the normalized production and dissipation rates are greater than unity and decrease monotonically as the We_λ increases, indicating that the presence of the droplets leads to turbulence augmentation. As We_λ decreases, the droplets become increasingly rigid, and therefore they exert a blocking effect on the surrounding turbulent flow. This effect abruptly re-orients the turbulent eddies leading to an increase in the magnitude of the Reynolds stress, $\langle u'_1 u'_2 \rangle$, causing an increase in \mathcal{P} , which also leads to an increase in the magnitude of the velocity gradients \mathcal{D}_{ij} , associated with an increase in ε relative to the single-phase flow, as shown in figure 11. Moreover, the two quantities have approximately the same value (the difference is less than 3%), thus indicating that at steady state the production balances the dissipation and that the power of surface tension is on average zero (*i.e.* $\mathcal{P} \approx \varepsilon$ and $\Psi_\sigma \approx 0$). These results are consistent with what was previously observed in figure 4(a) and indirectly confirm the relation $\Psi_\sigma = -\sigma / \mathcal{V}_m d\mathcal{A}/dt$ derived by Dodd & Ferrante (2016). Indeed, this relation implies that at steady state Ψ_σ is zero since the rate of change of \mathcal{A} is null.

Next, we focus on the equation obtained by phase averaging in one of the two fluids (equation (28)). Again, at steady state the time derivative on the left-hand side is zero and the relation states that the production and dissipation are balanced by the two transport terms \mathcal{T}_m^ν and \mathcal{T}_m^p . Figure 12 shows histograms of the production \mathcal{P}_m and dissipation ε_m rates in the two phases for three selected Weber numbers We_λ (cases 2, 6 and 10). We observe that the production rate is lower in the dispersed phase than in the carrier phase, while the dissipation rate

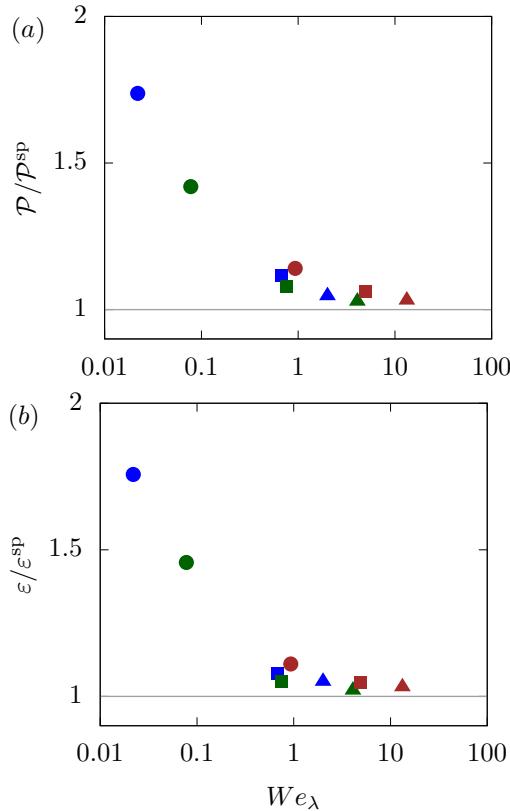


Figure 11: (a) Turbulent kinetic energy production \mathcal{P} and (b) dissipation ε rates averaged over both phases as a function of the Weber number We_λ , normalized by their value in the single-phase flow (\mathcal{P}_0 and ε_0).

is higher in the dispersed fluid than in the carrier fluid. These results indicate that the total transport term $\mathcal{T}_m = \mathcal{T}_m^\nu + \mathcal{T}_m^p$ is positive in the dispersed fluid and negative in the carrier, corresponding to a turbulent kinetic energy transfer from the carrier to the dispersed phase. In other words, the presence of the droplets is overall a sink for the turbulent kinetic energy of the bulk fluid \mathcal{K}_c . In addition, we observe that the difference in \mathcal{P}_m and ε_m decreases with We_λ .

Finally, figure 13 shows the decomposition of the total transport term \mathcal{T}_m into its pressure and viscous contributions, \mathcal{T}_m^p and \mathcal{T}_m^ν . In the dispersed phase shown in the left panel, the pressure transport term is very small and almost negligible, with most of the transport of turbulent kinetic energy (90–95%) due to the viscous contribution \mathcal{T}_d^ν . On the other hand, an opposite behavior is evident in the carrier phase shown in the right panel: the pressure transport term \mathcal{T}_c^p is dominant one and accounting for most of the transport of turbulent

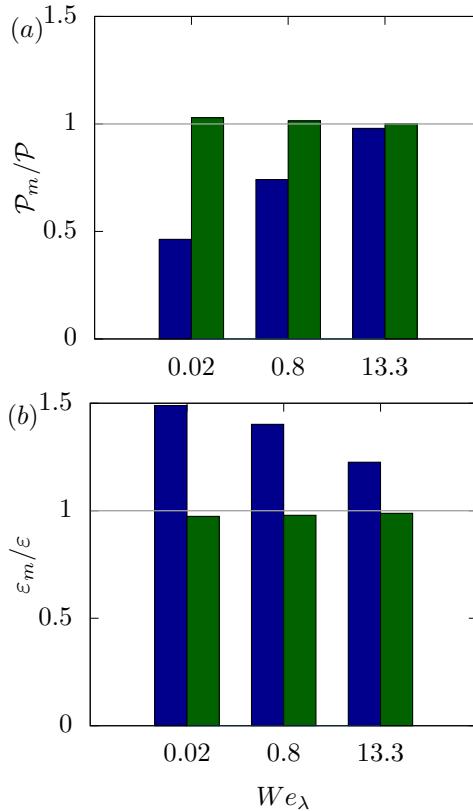


Figure 12: (a) Turbulent kinetic energy production \mathcal{P}_m and (b) dissipation ε_m rates averaged over the two phases separately as a function of the Weber number We_λ for cases 2, 6 and 10. The left and right columns are used to distinguish the dispersed and carrier phases, respectively.

kinetic energy (65–80%), while the pressure contribution is small. Moreover, we can observe that all the transport terms reduce for increasing Weber number, consistently with the discussion concerning figure 12.

The different mechanism of transport of turbulent kinetic energy between the carrier and dispersed phase is due to the different kind of flow experienced by the two fluids. This is discussed in figure 14 where the so-called flow topology parameter \mathcal{Q} (see e.g. De Vita *et al.* 2018) is presented. The flow topology parameter is defined as

$$\mathcal{Q} = \frac{\mathcal{D}^2 - \Omega^2}{\mathcal{D}^2 + \Omega^2}, \quad (30)$$

where $\mathcal{D}^2 = \mathcal{D}_{ij}\mathcal{D}_{ji}$ and $\Omega^2 = \Omega_{ij}\Omega_{ji}$, being Ω_{ij} the rate of rotation tensor, $\Omega_{ij} = (\partial u_i / \partial x_j - \partial u_j / \partial x_i)/2$. When $\mathcal{Q} = -1$ the flow is purely rotational,

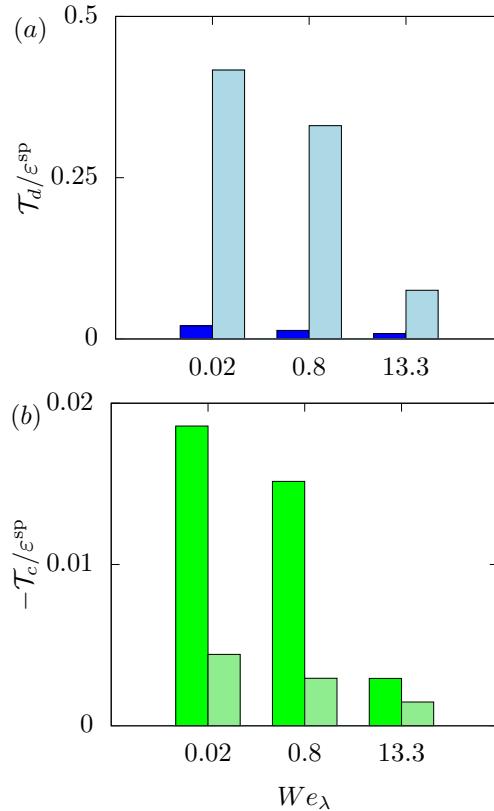


Figure 13: (a) Dispersed and (b) carrier transport terms \mathcal{T}_m , averaged over the two phases separately as a function of the Weber number We_λ for cases 2, 6 and 10. The left and right columns are used to distinguish the pressure and viscous contributions, respectively.

regions with $Q = 0$ represent pure shear flow and those with $Q = 1$ elongational flow. The distribution of the flow topology parameter for three selected cases is reported in figure 14. Note that, in the figure we show the probability density function (pdf) of Q in the two liquid phases separately. We observe that in the carrier fluid (dashed lines) the flow is mostly a shear flow as demonstrated by a single broad peak at $Q = 0$, and that little changes when changing the Weber number. On the other hand, the flow of the dispersed fluid (solid lines) still shows a broad single peak, now shifted towards negative values of Q , meaning that the flow is more rotational. Also, the relevance of the rotational flow is more and more evident as the Weber number increases. This is caused by the increased number of droplets and their consequent reduction in size: indeed, as the droplets size reduces the effect of the shear reduces as well.

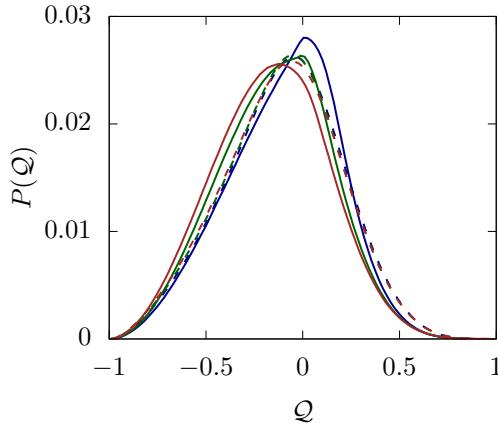


Figure 14: Probability density function of the flow topology parameter Q for three different Weber numbers: cases 2 (blue line), 6 (green line) and 10 (brown line), same as figure 13. The solid and dashed lines are used for the dispersed and carrier phase, respectively.

4. Conclusions

We perform direct numerical simulations of two-phase homogeneous shear turbulent flows at $Re_z = 15200$, where the two-phase nature of problem is tackled numerically using the MTHINC volume of fluid method recently developed. The droplets are initially spheres providing 5% volume fraction of the suspended phase and various Weber numbers and droplet initial diameters are investigated.

We show that the two-phase flow is able to reach a statistically steady state as indicated by a balance of turbulent kinetic energy production and dissipation. The results show that the presence of the droplets leads to turbulence augmentation by increasing the dissipation and production rates of the turbulence relative to the droplet-free flow. In particular, we find that as the Weber number decreases (higher droplet surface tension), the dissipation rate increases, causing the Taylor-microscale Reynolds number to decrease. This is explained by the surface tension force exerting a blocking effect on the surrounding turbulent flow. The turbulent production and dissipation rates are on average equal and in balance, with values larger than their single phase counterparts. Also, the surface tension power is on average zero. The flow modifications are caused by the presence of the dispersed phase, which acts as a sink of turbulent kinetic energy for the carrier phase, with a net flux going from the bulk of the fluid to the dispersed phase where it is dissipated. Moreover, the transport of turbulent kinetic energy in the carrier fluid is mainly due to the pressure transport, while the one inside the dispersed phase is dominated by the viscous contribution. This difference is explained by the different nature of the flow in the two phases: the carrier fluid is mainly a shear flow, while the

dispersed fluid is more rotational owing to its smaller length scales where the effect of the mean shear is reduced.

In addition to the flow properties, the droplet distribution eventually reaches a statistically stationary condition. Indeed, we show that the flow reaches a condition where the number of droplets remains almost constant, due to a balance between the break up and coalescence mechanisms, and that the number of droplets grows approximately linearly with the Weber number. A similar trend is found for the averaged surface area which also grows monotonically with the Weber number, but the growth rate is less than linear (the surface area grows with the Weber number to the power of 1/3, at least for moderately large Weber numbers). With the exception of one case, the droplet size distribution is single peaked, with the mean droplet size reducing with the Weber number. Based on the size distribution data, we show that the maximum droplets size scales well with the energy input as proposed by Hinze (1955), although the possibility of coalescence mechanism and the presence of a mean shear which were not considered in the original formulation by Hinze (1955).

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Paper 6

6

Effective slip over partially filled microcavities and its possible failure

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Motivated by the emerging applications of liquid-infused surfaces (LIS), we study the drag reduction and robustness of transverse flows over two-dimensional microcavities partially filled with an oily lubricant. Using separate simulations at different scales, characteristic contact line velocities at the fluid-solid intersection are first extracted from nano-scale phase field simulations and then applied to micron-scale two-phase flows, thus introducing a multiscale numerical framework to model the interface displacement and deformation within the cavities. As we explore the various effects of the lubricant-to-outer-fluid viscosity ratio $\tilde{\mu}_2/\tilde{\mu}_1$, the capillary number Ca, the static contact angle θ_s , and the filling fraction of the cavity δ , we find that the effective slip is most sensitive to the parameter δ . The effects of $\tilde{\mu}_2/\tilde{\mu}_1$ and θ_s are generally intertwined, but weakened if $\delta < 1$. Moreover, for an initial filling fraction $\delta = 0.94$, our results show that the effective slip is nearly independent of the capillary number, when it is small. Further increasing Ca to about $0.01\tilde{\mu}_1/\tilde{\mu}_2$, we identify a possible failure mode, associated with lubricants draining from the LIS, for $\tilde{\mu}_2/\tilde{\mu}_1 \lesssim 0.1$. Very viscous lubricants (e.g. $\tilde{\mu}_2/\tilde{\mu}_1 > 1$), on the other hand, are immune to such failure due to their generally larger contact line velocity.

1. Introduction

Advances in microfluidics and nanotechnology have boosted a rapid development of surface engineering in the last two decades. Among the different effects of micro-/nano-patterned surfaces, often inspired by observations in nature, one remarkable finding is that the introduction of micro-/nano-scale roughness on an otherwise smooth hydrophobic surface can sometimes significantly reduce the resistance to an external liquid flow. This slippery effect, due to entrapment of gas or vapor pockets under the surface asperities (superhydrophobic Cassie state), was first observed in the experiment of a water flow through a water-repellent pipe (Watanabe *et al.* 1999). Subsequently, a number of studies

have demonstrated various levels of drag reduction (Ou *et al.* 2004; Choi & Kim 2006; Schäffel *et al.* 2016; Lee *et al.* 2016), but also in some cases drag enhancement (Steinberger *et al.* 2007; Karatay *et al.* 2013). Despite the discrepancies in the literature, a common technological challenge for the application of superhydrophobic materials is their fragility (Bocquet & Lauga 2011). Under high pressures or external forces, such as turbulent fluctuation or phase change, the surface texture can be partially or fully impregnated by the outer fluid (Cassie-to-Wenzel transition), causing the system to lose the features it was designed for (Gentili *et al.* 2014; Giacomello *et al.* 2012; Seo *et al.* 2018).

Liquid-infused surfaces (LIS) are an alternative when aiming for drag reduction. They are more robust against pressure-induced failure, while displaying the same useful properties as conventional gas-cushioned superhydrophobic surfaces (Wexler *et al.* 2015). Two recent experiments have demonstrated, using microfabricated oil-impregnated pillars and grooves separately, up to 16% drag reduction in laminar flows (Solomon *et al.* 2014) and up to 14% drag reduction in turbulent flows (Rosenberg *et al.* 2016). In the case of the turbulent flow, the authors also tested superhydrophobic surfaces and measured approximately 10% drag reduction (Rosenberg *et al.* 2016). The values cited above, obtained at small lubricant-to-external-fluid viscosity ratios, can eventually decrease to nearly zero as the lubricant becomes more viscous. However, hybrid designs have been devised to maintain the performance, see e.g. a recent proof-of-concept study (Hemedha & Tafreshi 2016).

Analytically, the slippage over a superhydrophobic or liquid-infused surface can be characterized by an effective slip length. Analogous to the definition of the Navier slip, the effective slip length is an *averaged* quantity equal to the distance below the surface at which the velocity would extrapolate to zero (to be distinguished from the *intrinsic* slip of molecular nature (Gentili *et al.* 2014)). Extensive studies have been devoted to obtaining theoretical expressions of the effective slip for two-dimensional longitudinal or transverse grooves (Lauga & Stone 2003; Sbragaglia & Prosperetti 2007; Davis & Lauga 2009; Ng & Wang 2009; Schönecker *et al.* 2014; Nizkaya *et al.* 2014; Crowdy 2017*b,a*). Among these, Lauga & Stone (2003); Sbragaglia & Prosperetti (2007); Davis & Lauga (2009); Ng & Wang (2009); Crowdy (2017*a*) assume perfect slip along the liquid-gas interface, Ng & Wang (2009); Schönecker *et al.* (2014); Nizkaya *et al.* (2014) assume flat menisci, while the meniscus deformation, if considered, is either small (Crowdy 2017*b,a*) or in the dilute limit (*i.e.* the surface is mostly solid) (Lauga & Stone 2003). Furthermore, for purpose of calculation, the shape of the interface is always assumed symmetric (*i.e.* flat or circular) even under shear. This practically limits the application of the analytical results to the zero capillary limit, being the upper/lower bound of the drag reduction depending on the specific conditions.

Understanding the dependence of the slip length on the imposed shear and the lubricant viscosity in more realistic conditions may require a numerical

approach. There are, as yet, surprisingly few fully resolved hydrodynamic simulations able to solve the details of the flow reducing the underlying assumptions. Most prior numerical studies still consider flat/circular menisci with zero sub-phase viscosity (Davies *et al.* 2006; Martell *et al.* 2009; Cheng *et al.* 2009; Wang *et al.* 2014; Teo & Khoo 2014; Seo *et al.* 2018); however, they extend analytical solutions to more complex surface patterns or the finite-Reynolds-number regime. Flexible bubble shapes were first considered in Hyväloma & Harting (2008) for a uniform gas mattress, and later for a non-uniform distribution (Hyväloma *et al.* 2011). Using a two-phase Lattice Boltzmann method, Hyväloma & Harting (2008); Hyväloma *et al.* (2011) show that increasing the capillary number reduces the effective slip, even below zero (*i.e.* more friction than a solid plate). Specifically, their nanobubbles protrude strongly into the flow and remain trapped in the pores. Indeed, for very large protrusion angles, negative slip is both observed experimentally (Steinberger *et al.* 2007) and verified analytically (Davis & Lauga 2009). On the other hand, when the protrusion angle is smaller and the bubbles are allowed to slide on the substrate, the phase field simulation of Gao & Feng (2009) shows the opposite behavior: the effective slip is nearly shear-independent for relatively low capillary numbers, while it can increase dramatically if the capillary number is beyond some threshold. This threshold is not a single, universal value but depends on the spacing of the grooves and the initial filling of the gas; however, the enhancement of the slip is clearly due to depinning of the liquid-gas-solid contact line. We note that the depinning process considered in Gao & Feng (2009) might be an idealization, since realistic solid surfaces may not be smooth/chemically-homogeneous near the edge. Furthermore, both studies consider gas bubbles submerged in water under unrealistically large shear rates ($10^6 \sim 10^7 s^{-1}$)⁶. Whether this is stable or can be physically realized without generating significant heat remains an open question.

Here, we explore a slightly different flow configuration: planar shear flows over a micro-rough wall *partially* impregnated by a lubricant fluid. Using the newly developed multiscale numerical framework in Holmgren & Kreiss (2017), simulations at separate scales are performed to obtain the steady drag reduction, while capturing the dynamic wetting behavior in details. As we investigate the various effects of the viscosity ratio, the capillary number, and the static contact angle, we find that the filling fraction has the largest impact for drag reduction. It weakens the effects of other parameters, which are generally intertwined in a number of non-trivial ways. Moreover, for a given initial filling fraction (94%), our results show that the viscosity of the lubricant can not only influence the effective slip length, but also the robustness of the substrate under external shear. Shear-driven failure of LIS has recently been reported in Wexler *et al.* (2015); Jacobi *et al.* (2015); Liu *et al.* (2016) in the longitudinal case. Our study predicts that a similar drainage, though the viscosity dependence differs,

⁶In Hyväloma & Harting (2008), the shear rates were reported as $10^{-6} \sim 10^{-7} s^{-1}$. This must be a typo.

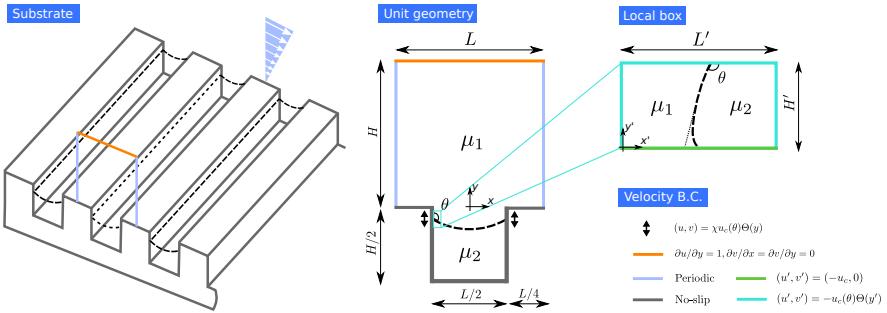


Figure 1: (color online) Schematic of the problem definition and setup for the two separate simulations. The substrate is patterned with an array of square cavities. The unit geometry shows the cross-section of the partially filled microcavity. The boundary condition at $y = H$ is equivalent to unit tangential stress and zero normal stress, while the arrows near the contact lines represent the slip boundary. The local box depicts the computational domain of the moving contact line model (the variables are denoted by a prime). Its velocity boundary conditions correspond to a moving wall in the bending interface reference frame.

may also occur in the transverse case. Understanding of this drainage failure is instructive for improved robustness of the surface design.

2. Microcavities partially filled with lubricants

2.1. Problem setup

We consider the transverse flow over an array of regularly spaced square cavities illustrated in Fig. 1. The outer fluid of viscosity μ_1 is driven by a constant shear $\dot{\gamma}$ in the x direction, imposed at distance H above the floor. The cavities of length $L/2$ and depth $H/2$ are partially filled with a lubricant fluid of viscosity μ_2 . When the number of the microcavities is large, the system is equivalent to a single cavity with periodic boundary conditions in the front and back. The solution at the (quasi-)steady state is determined by the incompressible Stokes equations, written in the non-dimensional form

$$\nabla \cdot \mathbf{u} = 0, \quad -\nabla p + \nabla \cdot [\mu_i(\nabla \mathbf{u} + \nabla \mathbf{u}^T)] = 0, \quad (1)$$

where $\mathbf{u} = (u, v)$ is the velocity, $p = p(x, y)$ the pressure, and $\mu_i = \tilde{\mu}_i/\tilde{\mu}_1$ ($i = 1$ or 2) the dimensionless viscosity, using \tilde{H} and $\tilde{\dot{\gamma}}\tilde{H}$ as the reference length and velocity respectively⁷. For viscous flows, the velocity and its tangential derivatives are continuous along the fluid interface (Batchelor 1967). The normal

⁷Dimensional values are denoted with a tilde throughout the manuscript.

stress is discontinuous due to the surface tension $\tilde{\sigma}$ and the viscosity difference, giving the pressure jump (denoted as $[A]_\Gamma = A_2 - A_1$)

$$[p]_\Gamma = \frac{\kappa}{\text{Ca}} + 2[\mu]_\Gamma \mathbf{n}^T \cdot \nabla \mathbf{u} \cdot \mathbf{n} \quad \text{on } \Gamma, \quad (2)$$

where \mathbf{n} is the outward-pointing normal at the interface Γ , κ its curvature, and $\text{Ca} = \tilde{\mu}_1 \dot{\gamma} \tilde{H} / \tilde{\sigma}$ the capillary number.

As the lubricant only partially fills the cavity initially, it may become distorted or splatter under the external shear. The associated contact line motion can be described by a second capillary number, $\text{Ca}_c = \tilde{\mu}_2 \tilde{U}_c / \tilde{\sigma}$, where \tilde{U}_c is the characteristic contact line velocity related to the liquid and solid surface energies. The ratio between this velocity and the shear, $\chi = \tilde{U}_c / (\dot{\gamma} \tilde{H})$, measures the magnitude of the local slip in the sheared dynamical system. It also scales the slip velocity near the contact line,

$$\mathbf{u} = \chi u_c(\theta) \Theta(y) \quad \text{on } \partial\Omega_\Gamma, \quad (3)$$

where $\chi u_c(\theta)$ is the renormalized nanoscale contact line velocity depending on the apparent contact angle θ , and $\Theta(y)$ provides the self-similar slip velocity function of the wall-parallel coordinate y that is imposed in the vicinity of the contact line on the boundary $\partial\Omega_\Gamma$ (see Fig. 1). Further details of $\chi u_c(\theta)$ and $\Theta(y)$ will be provided in Sec. 2.2.

In summary, Eqs. (1–3) are determined, neglecting the fluid inertia and fixing the substrate geometry, by the following non-dimensional parameters: (i) the viscosity ratio, $\tilde{\mu}_2/\tilde{\mu}_1$, (ii) the static contact angle, θ_s , (iii) the initial filling fraction of the cavity $\delta = 2d_0/H$ (where d_0 is the initial depth of the lubricant measured from the contact point to the bottom of the cavity), (iv) the capillary number based on the imposed shear Ca , and (v) the ratio between the characteristic contact line velocity and the shear, χ . The effect of the presence of the lubricating cavity and the corresponding apparent slip can be readily quantified by an effective slip length λ_e , defined as

$$\lambda_e = \frac{\bar{u}(H)}{\dot{\gamma} H} - 1, \quad (4)$$

with $\bar{u}(H)$ being the average streamwise velocity at distance H above the floor (averaged over the x direction).

In the following, we will consider various combinations of the governing parameters (i–v) and evaluate λ_e for each configuration. As the result will clearly depend on the motion of the impregnated lubricant, the multiscale modelling approach that we adopt is described next. The objective here is to provide an overall description of our methodology, rather than deriving the full mathematical/numerical details. For the latter, including validations, we refer to our previous work (Kronbichler & Kreiss 2017; Holmgren & Kreiss 2017).

2.2. Modelling of the moving contact lines

We model the contact line dynamics in two steps. First, we solve the Cahn-Hilliard equations within a Stokes system

$$\frac{\partial c}{\partial t} + \tilde{\mathbf{u}} \cdot \nabla c - \tilde{m} \nabla^2 \tilde{\psi} = 0, \quad \tilde{\psi} - \frac{3\tilde{\sigma}\tilde{\epsilon}}{4} \left(\frac{2}{\tilde{\epsilon}^2} (c^3 - c) - \nabla^2 c \right) = 0, \quad (5)$$

$$\nabla \cdot \tilde{\mathbf{u}} = 0, \quad -\nabla \tilde{p} + \nabla \cdot [\tilde{\mu}(\nabla \tilde{\mathbf{u}} + \nabla \tilde{\mathbf{u}}^T)] + \tilde{\psi} \nabla c = 0. \quad (6)$$

In the above, c is a non-dimensional phase parameter smoothly varying from $+1$ in one fluid to -1 in the other within a thickness of $\tilde{\epsilon}$, $\tilde{\psi}$ is the fluid chemical potential, \tilde{m} is the mobility, and $\tilde{\sigma}$, again, is the surface tension. The chemical potential $\tilde{\psi}$ measures the variation of the system free energy with respect to c . Its gradient determines the interfacial diffusion flux $-\tilde{m} \nabla \tilde{\psi}$, which together with the convective flux $\tilde{\mathbf{u}}c$, models the creation, movement, and dissolution of phase interfaces (Jacqmin 2000).

Technically, Eqs. (5–6) are solved in a rectangular box in the vicinity of a contact line using methods presented in Kronbichler & Kreiss (2017) (see Fig. 1, the local box and its velocity boundary condition). They are determined solely by the viscosity ratio, the surface tension, and the static contact angle (the rest are fixed choosing the proper non-dimensionalization); hence, the moving contact line can be simulated separately from the cavity flow. Inherently, we assume the length and time scales of the local box are much smaller than the cavity, *i.e.* $\tilde{H}'/\tilde{H} \ll 1$ and $\tilde{\tau}'/\tilde{\tau} \ll 1$ respectively. The first condition holds by definition and is enforced by providing enough resolution. The second condition is automatically satisfied realizing $\tilde{\tau}'/\tilde{\tau} = \tilde{H}'/\tilde{U}_c/\dot{\gamma}^{-1} = \tilde{H}'/\tilde{H}/\chi$. We will show in Sec. 3 that χ in our case is indeed much bigger than 1.

The steady-state solutions of Eqs. (5–6) give the contact line velocity, $\chi u_c(\theta)$, function of the apparent contact angle only. It is typically nonlinear, and is valid down to the nanometer scale. To impose this slip velocity in the micrometer cavity flow, as the second step, we modify the velocity boundary condition near the contact line using asymptotic matching (Holmgren & Kreiss 2017). Here, the self-similarity of the local velocity field is invoked and the singularity of the viscous stress is avoided (Huh & Scriven 1971). The end result is an algebraic operator, $\Theta(y)$, applied to $\chi u_c(\theta)$ on the boundary $\partial\Omega_\Gamma$.

We comment that our multiscale modelling approach is not limited to the phase-field model for the nanoscale; in principle, any model able to describe the contact line dynamics, e.g. the molecular dynamics (MD) (Johansson *et al.* 2015) or the Lattice-Boltzmann (LB) (Sbragaglia *et al.* 2006), can be used. We also note that, by solving Eqs. (5–6) in a square domain, we implicitly assume the solid surface is nanosmooth. Consequently, any deviation from the static contact angle will result in an interface displacement, bringing the phase field back to its local equilibrium. In practice, a real surface may have random roughness or defects smaller than the scale of the printed patterns, causing the interface to be pinned (*i.e.* contact angle hysteresis). Such effects can be included by modifying the geometry of the computational domain, or simply by

modifying the relation $u_c = u_c(\theta)$ so that $u_c = 0$ for a range of θ 's. In Sec. 3, we will take this second approach to account for a small contact angle hysteresis.

2.3. Numerical methods

The governing equations, together with the boundary conditions, Eqs. (1–3), are solved numerically using the two-phase flow solver described in Kronbichler *et al.* (2018), with suitable modifications for moving contact lines. The equations are discretized in space using the finite element method and the solver is implemented in the C++ based finite element open source library **deal.II** (Bangerth *et al.* 2007, 2016). The interface between the two fluids is evolved using the conservative level set method (Olsson & Kreiss 2005), so that only one fixed set of mesh is required. Specifically, we use uniformly distributed quadrilaterals (*i.e.* squares) with grid spacing $\Delta x = 1/160$, and time steps restricted by the stability condition $\Delta t_{max} = c_0 \text{Ca} \Delta x$ (c_0 is a constant) (Kronbichler *et al.* 2018). This leads to $\Delta t = 10^{-4} \sim 10^{-3}$ depending on the capillary number Ca .

The moving contact-line velocities are pre-computed by solving Eqs. (5–6) and used as tabulated inputs. In the simulations, additional numerical parameters include the frequency of the reinitialization (a technical procedure in the level set method, see Olsson & Kreiss (2005)), the size of the local box in the contact line model (*i.e.* L' and H' , see Fig. 1), and the size of a so-called bump function (related to $\partial\Omega_\Gamma$, see Holmgren & Kreiss (2017)). These are chosen to yield numerically-independent results as in Kronbichler & Kreiss (2017); Holmgren & Kreiss (2017) where validations are presented.

3. Results

We study the effective slip over microcavities partially filled with a second fluid using the parameters summarized in Tab. 1. Here, six pairs of fluids are considered as in the experiment (Solomon *et al.* 2014), leading to a wide range of $\tilde{\mu}_2/\tilde{\mu}_1$ from 31.7 to 3.83×10^{-3} . The filling fraction is initialized to $\delta = 0.94$ (corresponding to a depth $d_0 = 0.47H$) to allow for some sloshing of the lubricant. The velocity ratio $\chi = \tilde{U}_c/(\dot{\gamma}\tilde{H})$ is set constant for all the fluid pairs and shear rates to reduce the number of parameters and focus on the single physical effects mentioned above. This also implies that the capillary number is varied by changing the outer fluid viscosity. As an example, for $\dot{\gamma} = 800 \text{ s}^{-1}$, $\tilde{H} = 20 \mu\text{m}$, and $\tilde{U}_c = 2.63 \text{ m/s}$, the velocity ratio $\chi \approx 164$ (which is indeed much greater than 1), and the corresponding Ca increases from 1.92×10^{-3} to 1.59 for the different viscosities considered.⁸ We further modify Ca at a fixed χ to study the effect of interface deformation. Finally, the effect of the

⁸We estimate the dimensional shear rate at the upper boundary of our computational domain from the experimental Hele-Shaw setup in Wexler *et al.* (2015) (2 mL/min through a cross-section of $180 \mu\text{m} \times 7 \text{ mm}$). Assuming a parabolic velocity profile, the reference velocity at $\tilde{H} = 20 \mu\text{m}$ is $\tilde{U} = 0.016 \text{ m/s}$, leading to a shear rate of $\dot{\gamma} = 800 \text{ s}^{-1}$. The characteristic contact line velocity is calculated as $\tilde{U}_c = \tilde{\sigma}/\tilde{\mu}_2$, taking $\tilde{\sigma} = 0.02 \text{ kg/s}^2$ and $\tilde{\mu}_2 = 0.0076 \text{ kg/ms}$, as in Kronbichler & Kreiss (2017)

Table 1: Parameters for the outer (subscript 1) and lubricant (subscript 2) fluids in the present study.

$\tilde{\mu}_1 [\text{kg/ms}]$	$\tilde{\mu}_2 [\text{kg/ms}]$	$\tilde{\mu}_2/\tilde{\mu}_1$	δ	χ	Ca	θ_s (deg)
0.0024	0.0760	31.7	0.94	164	0.02 ~ 5	80 or 105 or 76 ~ 84
0.0024	0.0076	3.17	0.94	164	0.02 ~ 5	80 or 105 or 76 ~ 84
0.0152	0.0076	0.5	0.94	164	0.02 ~ 5	80 or 105 or 76 ~ 84
0.1504	0.0076	5.05×10^{-2}	0.94	164	0.02 ~ 5	80 or 105 or 76 ~ 84
0.8942	0.0076	8.50×10^{-3}	0.94	164	0.02 ~ 5	80 or 105 or 76 ~ 84
1.9850	0.0076	3.83×10^{-3}	0.94	164	0.02 ~ 5	80 or 105 or 76 ~ 84

static contact angle is investigated by considering $\theta_s = 80^\circ$ (leading to a convex meniscus), $\theta_s = 105^\circ$ (concave meniscus), and $\chi u_c(\theta) = 0$ for $\theta_s \in [76^\circ, 84^\circ]$ to mimic some contact angle hysteresis.

3.1. Motions at the contact line

We precompute the contact line velocity χu_c as function of contact angle θ for the range of parameters listed in Tab. 1, using the nanoscale phase-field model described in Sec. 2.2. Numerically, the non-dimensional height of the local box is $H' = 36$, with grid size $h = 36/128$ and time step $\Delta t = 0.5$. Steady state results obtained after 4000 time steps are plotted in Fig. 2. Here, the solid lines correspond to static contact angle $\theta_s = 80^\circ$ measured from the outer fluid side. The non-zero contact line velocity at $\theta \neq \theta_s$ shows the tendency of the contact line to reach its equilibrium position. For the results presented next, we have also used static contact angles $\theta_s = 105^\circ$, corresponding to menisci protruding into the cavity, and $\theta_s \in [76^\circ, 84^\circ]$, modelling a contact angle hysteresis of 8° . Keeping the rest of the parameters unchanged, the contact line velocities for $\theta_s = 105^\circ$ and the case with hysteresis are obtained by shifting the curves pertaining each viscosity ratio horizontally to the modified static angles, see Fig. 2 (right) for an example.

As we vary $\tilde{\mu}_2/\tilde{\mu}_1$ over four orders-of-magnitude, Fig. 2 reveals a non-trivial dependence of the contact line dynamics. On the one end, χu_c changes rapidly with θ for very viscous lubricants, almost diverging for $\theta < 60^\circ$ in the case of $\tilde{\mu}_2/\tilde{\mu}_1 = 31.7$; on the other end, as the lubricant becomes less and less viscous, the $\chi u_c(\theta)$ relations eventually collapse onto one curve. Qualitatively, reduction of χu_c for decreasing $\tilde{\mu}_2/\tilde{\mu}_1$ is expected as we normalize the flow using the shear in the outer fluid; in other words, it is easier (hence requires less velocity) to displace a less viscous fluid (*i.e.* deviating θ from θ_s). In addition, the change of the curvature of the function $\chi u_c(\theta)$ can be inferred from the reciprocity of the two fluids (*i.e.* $-\chi u_c$ instead of χu_c and $180 - \theta$ instead of θ for the same $\tilde{\mu}_1/\tilde{\mu}_2$)⁹. Quantitatively, the present model has been compared favorably with Cox' law (Cox 1986), especially for small angle deviations (Kronbichler & Kreiss

⁹This is merely a qualitative argument, as it does not preserve the static angle unless $\theta_s = 90^\circ$.

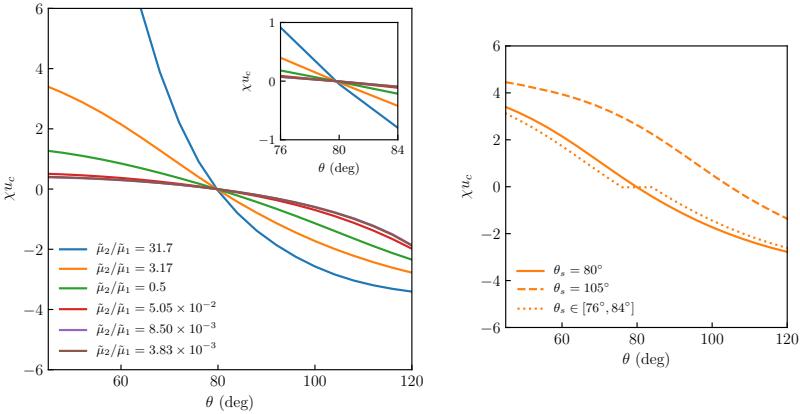


Figure 2: (color online) Relations between the apparent contact angles and the contact line velocities for $\theta_s = 80^\circ$ under various viscosity ratios, precomputed using the contact line model described in Sec. 2.2. Inset shows a close-up at small angle deviations, whereas the panel on the right illustrates how we model different static angles and contact angle hysteresis.

2017). Since this is the regime where the fluids normally operate at, we expect our model to accurately capture the small-scale contact line motions.

Finally, we note that the slope of the contact line velocity profiles near the static contact angle, θ_s , (cf. Fig. 2 inset) plays an important role in the wetting of the cavity under external shear. As we will discuss later, the difference of the contact line velocity with the viscosity ratio completely alters the robustness of lubricant infused cavities.

3.2. Effective slip above the cavities

Now, we present steady-state results of the effective slip length, defined in Eq. (4), obtained by solving the governing Eqs. (1–3) for the setup depicted in Fig. 1, using the two-phase Stokes solver described in Sec. 2.3. The overall results are compiled and plotted in Fig. 3, divided into the following two categories. First, we discuss the results obtained fixing the interface shape and pinning the contact point at the cavity corner, and compare with existing theories (denoted as “fix./pin.”). In a later section, we present results with depinned interface, *i.e.* contact line not at the cavity corner, obtained both fixing the interface (“fix./depin.”) and letting it move according to the multiscale model presented above (“depin.”).

3.2.1. Fixed interfaces pinned at the corners

Partly as a validation of our numerical methods, we first consider interface of fixed shapes pinned at the cavity tips. These are obtained by imposing in

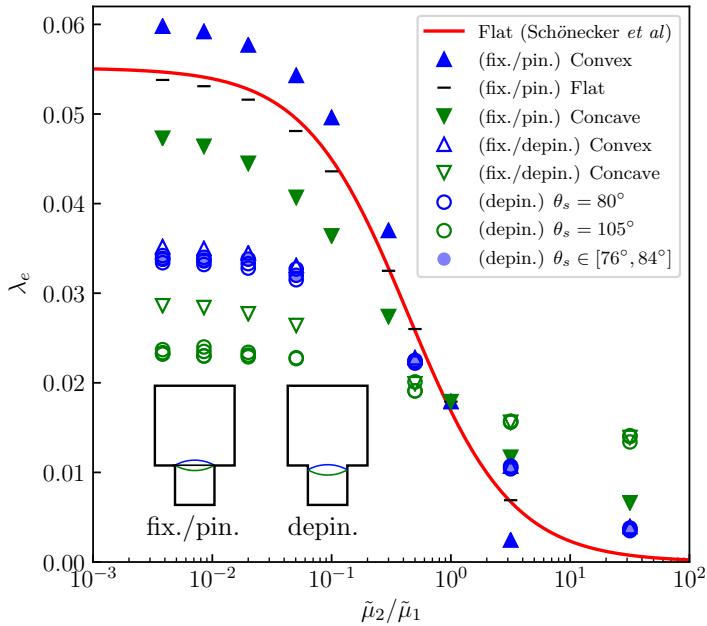


Figure 3: (color online) Effective slip as function of viscosity ratio under various static contact angles, filling fractions, and capillary numbers. The bars represent flat and fixed interfaces fully covering the cavity ($\delta = 1$), where the analytical result from Schönecker *et al.* (2014) is also plotted (red line). The filled symbols, upper blue and lower green triangles, stand for convex ($\theta_s = 80^\circ$) or concave ($\theta_s = 105^\circ$) interfaces pinned at the cavity tip in the zero capillary limit (*i.e.* fixed interface). The open symbols are the steady state solutions at $\delta = 0.94$ for different θ_s and Ca. The capillary number Ca is not indicated as it does not affect the results noticeably.

the simulation flat/circular menisci fully covering the cavities, indicated by bars/filled triangles in Fig. 3. Comparing with the analytical model taking into account finite dissipation in the cavity (Schönecker *et al.* 2014), a close agreement is observed over the broad $\tilde{\mu}_2/\tilde{\mu}_1$ spectrum examined. Specifically, the results show a continuous decrease of the effective slip as the viscosity ratio increases; the rate of variation is logarithmic for $0.1 < \tilde{\mu}_2/\tilde{\mu}_1 < 10$, it begins to saturate for $\tilde{\mu}_2/\tilde{\mu}_1 \lesssim 0.1$, and it is practically zero for $\tilde{\mu}_2/\tilde{\mu}_1 > 10$.

We further examine the curvature dependence of the effective slip length, using $\theta_s = 80^\circ$ and 105° as two representative curvatures for weakly convex and concave interfaces respectively. As shown in Fig. 3, for $\tilde{\mu}_2/\tilde{\mu}_1 < 1$, weakly convex interfaces have larger slip than weakly concave ones, consistently with

previous analytical and experimental studies (Davis & Lauga 2009; Karatay *et al.* 2013). The difference of the effective slip between convex and concave menisci increases in the limit of zero viscosity ratio; this is approximately 25% bigger in the convex case. On the other hand, when $\tilde{\mu}_2/\tilde{\mu}_1 > 1$, the relative magnitude flips: the concave interfaces have a larger slip length than the flat ones, while the convex interfaces can even have negative slip, adding more drag to the flow. The reason for this asymmetry is rather straightforward. Similar to the reasoning in Sbragaglia & Prosperetti (2007), the increased shear stress modified by a more viscous fluid will reduce the local slip, even more so when the interface bows into the channel, hence a smaller λ_e for the convex meniscus than for the concave one.

Lastly, we remark that the dependence of the effective slip length on the curvature is non-trivial. Previous studies have shown the existence of a critical contact angle beyond which the effective slip becomes negative ($\theta_s \lesssim 30^\circ$ by our definition) (Davis & Lauga 2009; Karatay *et al.* 2013; Hyvälöuma & Harting 2008). The angles we consider here are far from that range.

3.2.2. Interfaces depinned from the corners.

Next, we allow the interface to deform and slide on the cavity walls under external shear, removing the constraint of edge pinning considered earlier. The data pertain the steady state configuration, reached for shorter times at smaller Ca and verified to be unaffected by any numerical perturbations. Specifically, the effective slip length obtained initializing the filling ratio of the cavity to $\delta = 0.94$ are displayed with open or round symbols in Fig. 3.

First, we note that the effective slip length of partially filled cavities differs appreciably from the fully covered ones, regardless of the contact angle and the capillary number. For very low ($\tilde{\mu}_2/\tilde{\mu}_1 < 0.1$) and very high ($\tilde{\mu}_2/\tilde{\mu}_1 > 10$) viscosity ratios, the difference is at least a factor of 2. Meanwhile, within the cases considered for depinned interfaces, the effect of the viscosity ratio on the slip length is weaker than it is for pinned interfaces. The overall variation of λ_e is reduced. These observations suggest that the filling fraction of the cavity may be the main factor determining the effective slip.

To examine possible relationships between λ_e and δ , we display in Fig. 4 the effective slip under various filling fractions, for both convex ($\theta_s = 80^\circ$) and concave ($\theta_s = 105^\circ$) interfaces, at $\tilde{\mu}_2/\tilde{\mu}_1 = 0.05$ and 3.17 . For this extensive parameter study, fixed interface shapes are imposed, corresponding to the zero capillary limit (minimum-energy interface) to speed up the simulations. Indeed, having a small capillary number does not affect the result, as shown in Fig. 3 where the slip length for the “fix./depin.” cases are not significantly different from the depinned cases at low Ca.

As shown in Fig. 4, the effective slip length clearly depends on the filling fraction: as the meniscus recedes from the cavity tip, λ_e quickly decreases or increases depending on $\tilde{\mu}_2/\tilde{\mu}_1$; the variation is the sharpest in the early stage ($0.8 \lesssim \delta < 1$), while it is nearly negligible as δ further reduces. Plotting λ_e

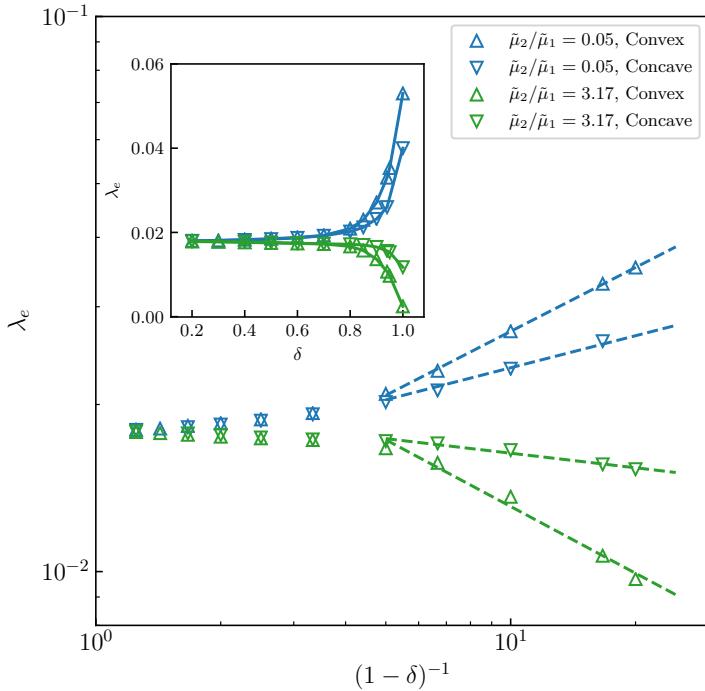


Figure 4: Effective slip of partially filled cavities for convex (80°) and concave (105°) interfaces for $\tilde{\mu}_2/\tilde{\mu}_1 = 0.05$ (blue triangles) and 3.17 (green triangles) in the zero capillary limit. The main figure shows power law relations of the slip length when plotted against the inverse “void fraction” $(1 - \delta)^{-1}$, indicated by the dashed lines (linear least squares fits for $(1 - \delta)^{-1} > 5$, or equivalently $\delta > 0.8$). The inset shows the sharp reduction/increase of the slip length as the meniscus recedes.

against $(1 - \delta)^{-1}$, which may be interpreted as an inverse “void fraction” of the cavity, we find a power law relation between the effective slip and the filling fraction. Indicated by the dashed lines in Fig. 4, the effective slip behaves as $\lambda_e \sim (1 - \delta)^{-c}$ for $\delta > 0.8$, where c is a constant related to the viscosity ratio and the overall geometry. Specifically, using linear least squares, we find $c \approx 0.38$ (convex) and ≈ 0.19 (concave) for $\tilde{\mu}_2/\tilde{\mu}_1 = 0.05$, and $c \approx -0.40$ (convex) and ≈ -0.09 (concave) for $\tilde{\mu}_2/\tilde{\mu}_1 = 3.17$. At equal viscosities, the $\lambda_e - (1 - \delta)^{-1}$ relations display cone-like patterns with the spreading angle function of both the viscosity ratio and the meniscus curvature. At lower filling ratios, all the points converge to the value of the slip length of the single-phase cavity. We remark that a theoretical determination of c is likely difficult, as the governing equation here is biharmonic (Crowdy 2017a; Ng & Wang 2009). Nevertheless,

our results clearly illustrate the pronounced dependence of the effective slip on the interface displacement, already when small, for transverse grooves.

We note from above that the slip λ_e varies in opposite directions depending on $\tilde{\mu}_2/\tilde{\mu}_1$. This is also shown in Fig. 3, where the effective slip of the depinned interfaces intercepts the red line (*i.e.* the results for a flat interface) at $\tilde{\mu}_2/\tilde{\mu}_1 = 1$ for both contact angles under consideration. Specifically, the effective slip λ_e is the same for $\delta = 0.94$ and $\delta = 1$, if $\tilde{\mu}_2 = \tilde{\mu}_1$; when $\tilde{\mu}_2 < \tilde{\mu}_1$, the slip is larger for $\delta = 1$; when $\tilde{\mu}_2 > \tilde{\mu}_1$, on the contrary, it is larger for $\delta = 0.94$. This crossover thus suggests an additional viscosity dependence of the effective slip coupled with the filling fraction of the cavities.

To quantitatively compare the effect of the viscosity ratio on λ_e at $\delta = 0.94$, we display the tangential shear stress τ_{xy} for flat, convex, and concave menisci in Fig. 5. Here, τ_{xy} is evaluated either on the cavity tip (at $y = 0$) or along the fluid-fluid interface (at $y = -0.03H$), and it is normalized by the unit tangential shear stress τ_∞ imposed above the floor (at $y = H$). As shown in Fig. 5, the normalized shear stress decreases as we reduce $\tilde{\mu}_2/\tilde{\mu}_1$ for all the cases, consistent with enhanced slip at lower lubricant viscosities; however, τ_{xy}/τ_∞ does not converge to zero as it would have been if the cavities were fully covered. Close comparison of Fig. 5(c) and (d) also explains the flipping of the relative magnitude of λ_e between convex and concave interfaces noted above: the shear stress is less for convex interfaces when $\tilde{\mu}_2/\tilde{\mu}_1 < 1$, while it is less (on average) for the concave ones at $\tilde{\mu}_2/\tilde{\mu}_1 > 1$. Moreover, Fig. 5 reveals that the distribution of the local shear for partially filled cavities is non-uniform. When $\tilde{\mu}_2/\tilde{\mu}_1 < 1$, τ_{xy}/τ_∞ always retains its minimum value at $x = 0$, and increases gradually towards the walls (at $x = \pm 0.25$); when $\tilde{\mu}_2/\tilde{\mu}_1 > 1$, the shear stress profiles can have several local minima/maxima depending on the protrusion angle. Such non-uniformity is most prominent when the interface is convex. In general, both the viscosity of the two fluids and the geometry of the liquid-infused cavities appear to influence τ_{xy}/τ_∞ .

We remark that constant shear stress along substrate surfaces is sometimes assumed in theoretical models to obtain analytical solutions (Schönecker *et al.* 2014). Although it is verified for fully-covered flat cavities (see Fig. 4 in Schönecker *et al.* (2014)), our results suggest that it is inaccurate for partially filled ones, even along the fluid-fluid interface, see Fig. 5(b). Since liquid-infused substrates are not always fully-covered in practice (Wexler *et al.* 2015), our simulations suggest this assumption be relaxed when developing more comprehensive models.

Finally, we discuss the role of capillary and hysteresis, referring back to the circular symbols in Fig. 3. These data are obtained via the multiscale contact line model for capillary numbers $\text{Ca} = 0.02 \sim 5$ and contact angles $\theta_s = 80^\circ, 105^\circ$, or $76^\circ \sim 84^\circ$ at initial filling fraction $\delta = 0.94$. Surprisingly, we find virtually no influence of the contact angle hysteresis on the effective slip length for the entire range of viscosity ratios considered. The filled circles, corresponding to $\theta_s \in [76^\circ, 84^\circ]$, lie closely on top of the blue open circles

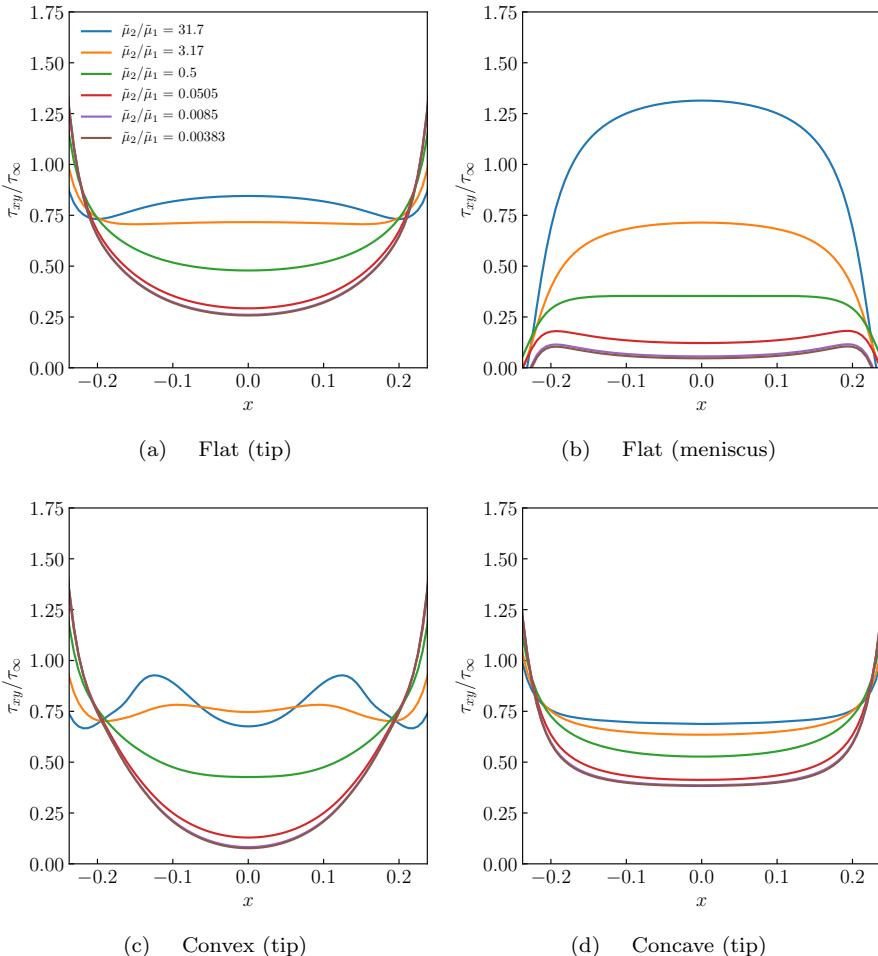


Figure 5: Normalized tangential stress evaluated on the plane of the cavity tip (a, c, d) or along the menisci (b) for various viscosity ratios. In all the cases, the menisci are fixed and depinned from the corners, corresponding to filling fraction $\delta = 0.94$.

denoting $\theta_s = 80^\circ$. Our results thus provide evidence that small scale roughness on the substrate surface, due to the material itself or the fabrication precision, does not necessarily increase the overall drag over the cavities. Indeed, as discussed in Schönecker *et al.* (2014), the effective slip length is a far-field effect determined by the mean velocity above the substrate. Since a small contact angle hysteresis does not alter significantly the interface profile nor its wetting

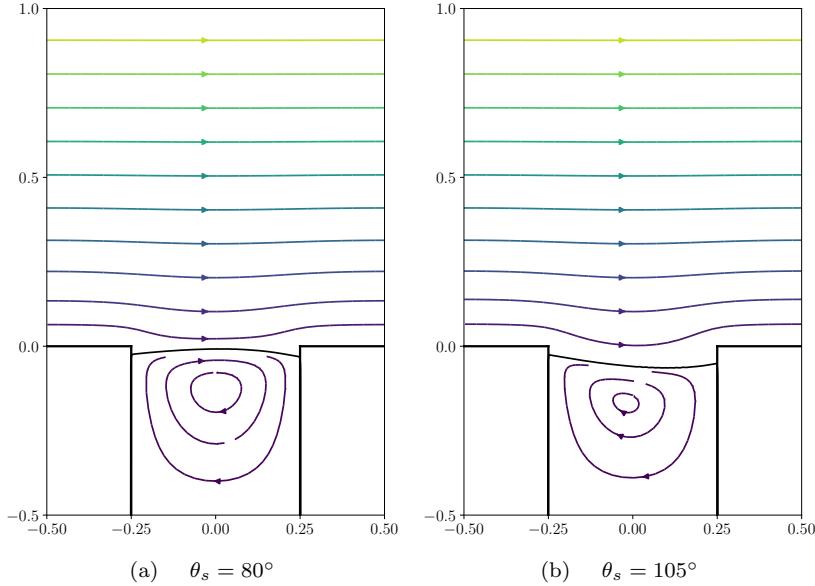


Figure 6: (color online) Typical shapes of stable interfaces and streamlines for the flow over a partially filled cavity. These examples correspond to the steady state configurations, for (a) $\theta_s = 80^\circ$ and (b) $\theta_s = 105^\circ$, at $\tilde{\mu}_2/\tilde{\mu}_1 = 0.5$ and $\text{Ca} = 0.02$.

behavior, these changes are expected to be quickly smeared out away from the substrate.

The above reasoning applies only to the cases when the capillary number is small. Further increasing the capillary number, hence the shear, can eventually deform the fluid interface to an extent that a stable configuration may not be attainable. In the remaining, we will consider the lubricant-infused surface under extreme shear rates. As we examine the possible consequence under various conditions, a seemingly counter-intuitive technical solution will be suggested.

3.3. Possible drainage of the lubricant

First, we examine typical interface profiles, both convex and concave, under moderate shear levels, see Fig. 6. Specifically, we consider the viscosity ratio $\tilde{\mu}_2/\tilde{\mu}_1 = 0.5$, the capillary number $\text{Ca} = 0.02$, and the initial contact angle $\theta_0 = \theta_s$. The steady-state solutions are taken at $t = 5$ in units of $1/\tilde{\gamma}$. As illustrated in the figure, the flow, while circulating inside the cavity, is already parallel at $y \approx 0.5H$. The deformation of the interfaces is almost negligible comparing to the initial conditions, only the contact points displacing slightly in opposite directions due to the shear. These two configurations are examples

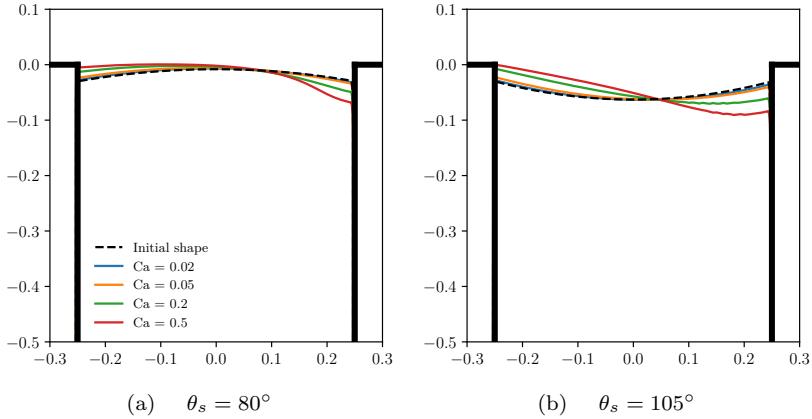


Figure 7: Interface profiles under increasing capillary numbers for viscosity ratio $\tilde{\mu}_2/\tilde{\mu}_1 = 5.05 \times 10^{-2}$ at $t = 5$.

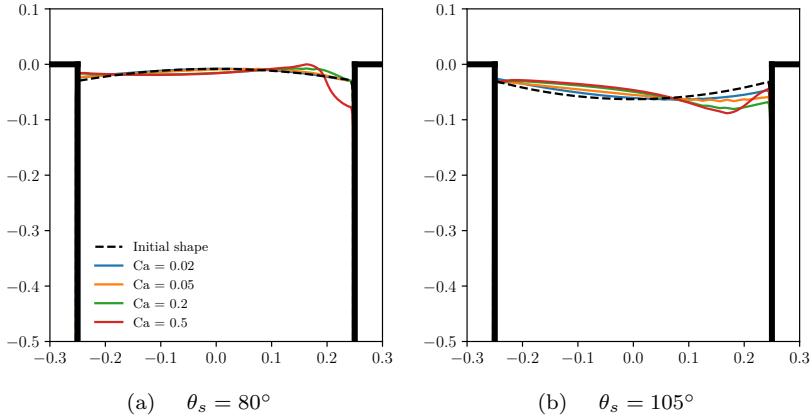


Figure 8: Interface profiles under increasing capillary numbers for viscosity ratio $\tilde{\mu}_2/\tilde{\mu}_1 = 0.5$ at $t = 5$.

of lubricant-infused cavities in working condition. The overall small change of the interface shapes is the reason for the weak shear dependence of the effective slip length discussed in Sec. 3.2.

To test the robustness of the LIS under stronger shear, we successively increase the capillary number from 0.02 to 5, keeping the other parameters unchanged (see Tab. 1). The resulting interface profiles are visualized in Figs.

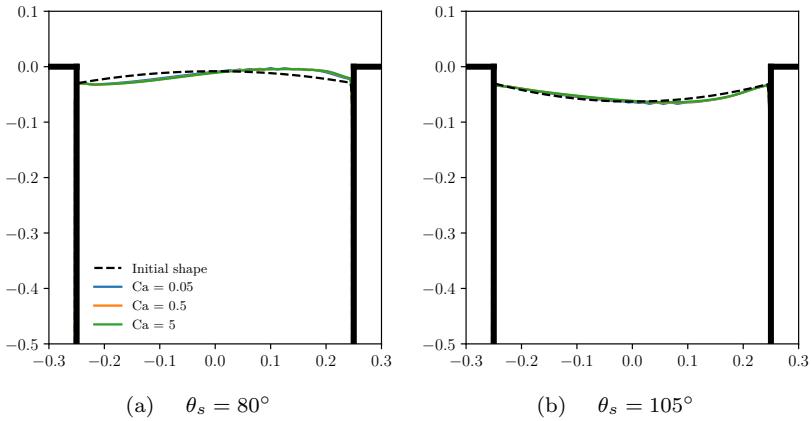


Figure 9: Interface profiles under increasing capillary numbers for viscosity ratio $\tilde{\mu}_2/\tilde{\mu}_1 = 3.17$ at $t = 5$.

7–9. As expected, increasing Ca generally leads to larger deformations of the interface. For $\tilde{\mu}_2/\tilde{\mu}_1 = 5.05 \times 10^{-2}$ (Fig. 7), the upstream contact point continuously moves towards the tip of the cavity, indicating a draining motion of the lubricant driven by the shear. For $\tilde{\mu}_2/\tilde{\mu}_1 = 0.5$ (Fig. 8), the downstream contact point responds more instead, almost leading to the interface rupture when Ca = 0.5. However, as we further increase the viscosity ratio, increasing the shear has no visible effect on the interface. For $\tilde{\mu}_2/\tilde{\mu}_1 = 3.17$ (Fig. 9), we barely observe any additional deformation even increasing Ca by two orders of magnitude. The lubricant stays firmly in the cavity regardless of the external shear.

Following these observations, we map the results pertaining all the viscosity ratios and capillary numbers considered in the phase diagram in Fig. 10. Here, three regimes are defined, which we label as stable, marginal, and unstable. For partially filled cavities with initial filling fraction $\delta = 0.94$ (*i.e.* initial depth $d_0/H = 94\%$ measured from the contact points), we consider cases where the final depth varies within $94 \pm 2\%$ as *stable*; if the final depth varies between $94 \pm 4\%$, which is very close to the cavity tip but still below, we consider the configuration as *marginal*; lastly, if one contact point has already/nearly hit the cavity tip, or if the interface is clearly disrupted (see e.g. Fig. 8), we consider the case as *unstable*. A similar, but simplified, criterion has also been chosen in Seo *et al.* (2018) for the onset of gas pocket instability in turbulent flows. We evaluate the final depths either in the steady states, or at $t = 5$ if a steady state has not been reached.

As shown in Fig. 10, the robustness of the LIS exhibits a rather complex dependence on the capillary number and the viscosity ratio. On the lower

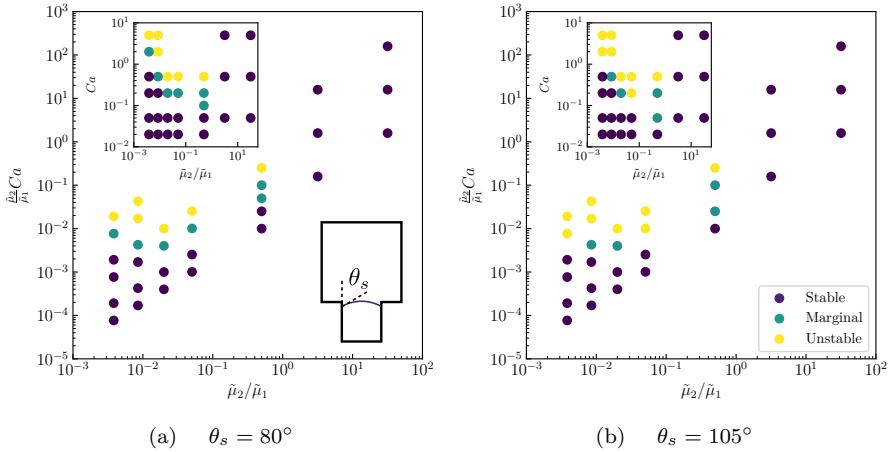


Figure 10: Phase diagram in the cavity capillary–viscosity ratio plane ($\tilde{\mu}_2/\tilde{\mu}_1$, $\tilde{\mu}_2/\tilde{\mu}_1 \text{Ca}$) showing the robustness of the lubricant-infused cavities under various capillary numbers and viscosity ratios. The inset reports the same data as function of the outer capillary number Ca .

viscosity side, *i.e.* $\tilde{\mu}_2/\tilde{\mu}_1 \lesssim 0.1$, lubricants of both convex and concave interfaces becomes unstable above a critical capillary number. Mapping the data on the $(\tilde{\mu}_2/\tilde{\mu}_1, \tilde{\mu}_2/\tilde{\mu}_1 \text{Ca})$ plane, our results suggest $\text{Ca}_{crit} \approx 0.01\tilde{\mu}_1/\tilde{\mu}_2$. That is, the critical capillary number, defined with the outer fluid, is inversely proportional to the viscosity ratio $\tilde{\mu}_2/\tilde{\mu}_1$; it is harder to drain a less viscous lubricant outside the cavity fixing the outer fluid. Note that similar results were also observed experimentally for longitudinal grooves, where less viscous lubricants are found to remain over a longer distance within the grooves (Liu *et al.* 2016). Our simulations thus point towards the same direction in the design of transverse LIS against shear-driven failures.

As the viscosity ratio further increases, however, the existence of a critical capillary number, Ca_{crit} , is no longer clear: at $\tilde{\mu}_2/\tilde{\mu}_1 = 0.5$, the equivalent $\text{Ca}_{crit} \approx 0.1\tilde{\mu}_1/\tilde{\mu}_2$, which would be an order-of-magnitude higher than before; while for $\tilde{\mu}_2/\tilde{\mu}_1 > 1$, no Ca_{crit} has been found within a reasonable range of capillary numbers. This shear-induced failure can be associated with draining in the cavities, and we propose that its mechanism be linked to the dewetting of the lubricant and to its viscosity.

Recalling the $\chi u_c(\theta)$ relations for various viscosity ratios in Fig. 2, the profiles collapse onto one curve for $\tilde{\mu}_2/\tilde{\mu}_1 < 0.05$, suggesting that it is the cavity capillary number, $\tilde{\mu}_2/\tilde{\mu}_1 \text{Ca}$, that determines the onset of failure. This is confirmed by testing one additional case with $\tilde{\mu}_2/\tilde{\mu}_1 = 0.02$ and the same dependency $\chi u_c(\theta)$ as in the other cases (thus not extracted from the phase-field

simulations); for this case, we indeed obtain the same $\text{Ca}_{\text{crit}} \approx 0.01\tilde{\mu}_1/\tilde{\mu}_2$, consistent with the other viscosity ratios in the same range (see Fig. 10). When $\tilde{\mu}_2/\tilde{\mu}_1$ increases to above 0.5, Fig. 2 shows a continuous deviation of the wetting relations. The slope of the $\chi u_c(\theta)$ curve near the static angle θ_s increases (in magnitude) rapidly with $\tilde{\mu}_2/\tilde{\mu}_1$, making it more difficult for the contact line to deform, hence reducing the drainage of the lubricant towards the cavity corner. Since the capillary number is limited by the shear rates and proportional to the scale of the micron-scale texture, substrates impregnated by very viscous lubricants are in practice very difficult to fail.

The above phenomenological mechanism suggests that, taking the lubricant viscosity as a design parameter, the intermediate viscosity ratios (e.g. $\tilde{\mu}_2/\tilde{\mu}_1 = 0.01 \sim 1$ depending on the specific condition) are to be avoided in the application of LIS. This is consistent with previous experiments of longitudinal grooves towards the lower viscosity branch (Liu *et al.* 2016); more viscous lubricants, on the other hand, seem to ensure higher robustness.

Finally, we note that the critical capillary numbers reported here should be considered as an estimate, since, in practice, draining of the lubricant will also depend on the physical/chemical conditions near the cavity corner. However, we do not expect these practical limitations to influence the qualitative insight obtained from our simulations.

4. Conclusions

In this paper, motivated by applications of micro-engineered liquid-infused surfaces, we study the drag reduction and robustness of the flow over an array of two-dimensional transverse grooves partially filled with an immiscible lubricant.

We use a multiscale numerical framework to model the wetting of the two fluids at the cavity walls as well as the deformation of the interface under the shear. In particular, we combine two separate simulation methods at different scales: (*i*) nanoscale phase field simulations for the contact line dynamics, and (*ii*) micron-scale Stokes flows simulations using information from (*i*) as a modified boundary condition, assuming self-similarity of the velocity field in the vicinity of the moving contact line. We believe the approach is however more general and it could be extended to include molecular dynamics simulations modelling the surface chemistry and roughness at the nanoscale.

We examine the effective slip λ_e in order to quantify the steady-state drag reduction of the LIS. Specifically, we fix the geometry of the cavity and vary the lubricant-to-outer-fluid viscosity ratio $\tilde{\mu}_2/\tilde{\mu}_1$, the capillary number Ca , the static contact angle θ_s , and the filling fraction of the cavity δ . The main results are summarized as follows.

1. λ_e depends primarily on δ ; the filling rate is therefore the main factor determining the effective slip.
2. Lower $\tilde{\mu}_2/\tilde{\mu}_1$ leads to reduced drag; the reduction is however less pronounced comparing to fully covered cavities. We relate this effect to

the shear stress profiles τ_{xy} along the cavity tip, and show that τ_{xy} is non-uniform (contrary to the fully covered cases).

3. The effect of the contact angle on the effective slip length is different for different viscosity ratios and the filling fractions.
4. The effect of the contact angle hysteresis and of the capillary number on λ_e is negligible, except
5. when Ca increases above a critical value Ca_{crit} , and the LIS can possibly fail. For an initial filling fraction $\delta = 0.94$, the critical capillary number $\text{Ca}_{crit} \approx 0.01\tilde{\mu}_1/\tilde{\mu}_2$ for $\tilde{\mu}_2/\tilde{\mu}_1 \lesssim 0.1$. For very viscous lubricants (e.g. $\tilde{\mu}_2/\tilde{\mu}_1 > 1$), on the other hand, the cavity remains impregnated due to their generally larger contact line velocity.

As a final remark, we note that this problem is characterised by a large number of control parameters, including e.g. geometry, static contact angle, surface chemistry, so that this study can be extended in a number of non-trivial ways. In addition, from a purely hydrodynamic point of view, the flow above the cavity may affect the contact line motion: it may therefore be relevant to study the response of the flow in the cavity to temporally varying shear and vortices relatively far from it.

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Paper 7

7

Implementation note on a minimal discrete-element lubrication/contact dynamics model for dense suspensions

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We describe and summarize a class of minimal numerical models emerged from recent development of simulation methods for dense particle suspensions in over-damped linear flows. The main ingredients include (i) a frame-invariant, short-range lubrication model for spherical particles, and (ii) a soft-core, stick/slide frictional contact model activated when particles overlap. We implement a version of the model using a modified velocity-Verlet algorithm that explicitly solves the N -body dynamical system in $\mathcal{O}(cN)$ operations, where c is a kernel constant depending on the cutoff of particle interactions. The implementation is validated against literature results on jamming transition and shear thickening suspensions from 40% to 64% volume fractions. Potential strategies to extend the present methodology to non-spherical particles are also suggested for very concentrated suspensions.

1. Introduction

The behaviour of systems involving the motion of small particles in a suspending fluid covers a wide range of phenomena of interest to both scientists and engineers. Dense suspensions, where the volume fraction of solid particles becomes comparable to or even higher than that of the fluid (see Figure 1), have particularly rich and sometimes unexpected rheologies, such as yielding, shear thinning, continuous shear thickening (CST), or discontinuous shear thickening (DST) (Mewis & Wagner 2012; Denn & Morris 2014; Guazzelli & Pouliquen 2018; Morris 2020). Apart from being theoretically intriguing, these complex behaviours often have major practical implications. For instance, while it makes sense for the cement industry to manufacture suspensions that do not shear thicken, the same feature becomes an advantage for designing flexible body armor.

Despite the practical importance, theoretical development of suspension rheology remains challenging and only a few analytical solutions have been found in the dilute regime, see e.g. Einstein (1906); Batchelor & Green (1972b). This is partially due to the lack of a precise knowledge or control of the

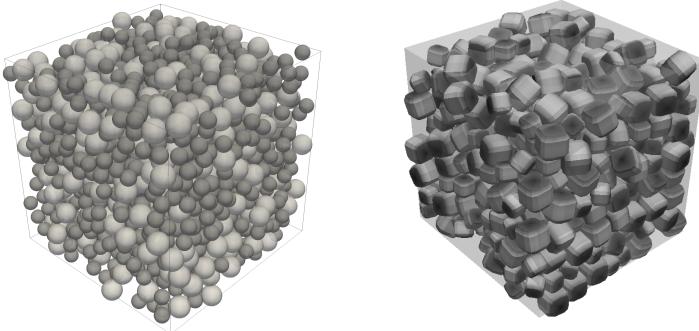


Figure 1: Visualizations of dense particle suspensions. (left) A bidisperse suspension of 2000 spheres at 55% volume fraction. (right) A random organization of 500 superballs at 50% volume fraction.

various interactions at the particle level and partially due to the mathematical difficulties involved in many-body problems. On the other hand, solving a system of interacting particles appears relatively straightforward in an algorithmic perspective. In fact, the last decades have seen tremendous advancement in both numerical simulations and computer hardware. In the context of rheology and soft condensed matter, some of the numerical models that have been developed include molecular dynamics (MD) (Alder & Wainwright 1959; Verlet 1967), dissipative particle dynamics (DPD) (Hoogerbrugge & Koelman 1992; Groot & Warren 1997), Stokesian dynamics (Brady & Bossis 1988), and minimal discrete element methods (M-DEM) (Mari *et al.* 2014; Cheal & Ness 2018), to name a few. The objective of the present note is thus to provide a brief summary of the latest development, specifically on the discrete-element lubrication/contact (DLCD) models that recently emerged from a blending of hydrodynamics and granular physics, and comment on some implementation details.

The paper is organized as follows. In Sec. 2, we summarize the mathematical formulation, relevant physical parameters and their rheological characterization. A brief note on the numerical integration of the equations of motion is also provided. In Sec. 3, we validate the implementation against two benchmark cases in the literature, *i.e.* jamming transition and shear thickening. Finally, we end with a short discussion on potential extensions of the present methodology to non-spherical particles.

2. The DLCD model

In this section, we briefly summarize the mathematical formulation and essential physical parameters of the DLCD model. Detailed descriptions can be found

in Seto *et al.* (2013); Mari *et al.* (2014); Cheal & Ness (2018) and references therein.

2.1. Mathematical formulation

The translational and rotational dynamics of a rigid particle is governed by the Newton-Euler equations,

$$\sum_M \mathbf{F}_i^M = m_i \frac{d\mathbf{u}_i}{dt}, \quad (1a)$$

$$\sum_M \mathbf{T}_i^M = \mathbf{I}_i \frac{d\boldsymbol{\omega}_i}{dt} + \boldsymbol{\omega}_i \times (\mathbf{I}_i \boldsymbol{\omega}_i), \quad (1b)$$

where \mathbf{F}_i and \mathbf{T}_i denote the force and torque exerted on the center-of-mass of particle i ; m_i and \mathbf{I}_i are its mass and moment-of-inertia tensor in the body frame (scalar for spheres); \mathbf{u}_i and $\boldsymbol{\omega}_i$ denote its translational and angular velocities, respectively. The force and torque are summed over various modes of particle-fluid and particle-particle interactions that include the following single-body or pairwise *hydrodynamic*, *contact*, and *physico-chemical* contributions

$$\sum_M \mathbf{F}_i^M = \mathbf{F}_i^S + \sum_j^{N_L} \mathbf{F}_{i,j}^L + \sum_j^{N_C} \mathbf{F}_{i,j}^C + \sum_j^{N_R} \mathbf{F}_{i,j}^R + \sum_j^{N_A} \mathbf{F}_{i,j}^A, \quad (2a)$$

$$\sum_M \mathbf{T}_i^M = \mathbf{T}_i^S + \sum_j^{N_L} \mathbf{T}_{i,j}^L + \sum_j^{N_C} \mathbf{T}_{i,j}^C + \sum_j^{N_R} \mathbf{T}_{i,j}^R + \sum_j^{N_A} \mathbf{T}_{i,j}^A. \quad (2b)$$

Their specific functional forms, for the case of spheres, are listed below.

1. Stokes' drag (acting on each particle i)

$$\mathbf{F}_i^S = -6\pi\mu a_i(\mathbf{u}_i - \mathbf{U}_i^\infty), \quad (3a)$$

$$\mathbf{T}_i^S = -8\pi\mu a_i^3(\boldsymbol{\omega}_i - \boldsymbol{\Omega}_i^\infty), \quad (3b)$$

where μ is the dynamic viscosity of the underlying fluid, a_i the particle radius, \mathbf{U}_i^∞ and $\boldsymbol{\Omega}_i^\infty$ the undisturbed translational and angular velocities of the fluid at the particle position. A linear flow satisfies the relation, $\mathbf{U}^\infty = \boldsymbol{\Omega}^\infty \times \mathbf{x} + \mathbb{E}^\infty \cdot \mathbf{x}$, where \mathbf{x} denotes the position vector, and \mathbb{E}^∞ is the rate-of-strain tensor (Batchelor 1967).

2. Lubrication (acting on each lubricating pair i, j)

$$\begin{aligned} \mathbf{F}_{i,j}^L &= -(X_{ii}^A \mathbb{P}_n + Y_{ii}^A \mathbb{P}_t)(\mathbf{u}_i - \mathbf{u}_j) \\ &\quad + Y_{ii}^B(\boldsymbol{\omega}_i \times \mathbf{n}_{ij}) + Y_{ji}^B(\boldsymbol{\omega}_j \times \mathbf{n}_{ij}), \end{aligned} \quad (4a)$$

$$\mathbf{F}_{j,i}^L = -\mathbf{F}_{i,j}^L, \quad (4b)$$

$$\mathbf{T}_{i,j}^L = -Y_{ii}^B(\mathbf{u}_i - \mathbf{u}_j) \times \mathbf{n}_{ij} - \mathbb{P}_t(Y_{ii}^C \boldsymbol{\omega}_i + Y_{ij}^C \boldsymbol{\omega}_j), \quad (4c)$$

$$\mathbf{T}_{j,i}^L = -Y_{ji}^B(\mathbf{u}_i - \mathbf{u}_j) \times \mathbf{n}_{ij} - \mathbb{P}_t(Y_{ji}^C \boldsymbol{\omega}_i + Y_{jj}^C \boldsymbol{\omega}_j), \quad (4d)$$

where \mathbf{n}_{ij} denotes the unit normal vector pointing from particle i to particle j , $\mathbb{P}_n = \mathbf{n}_{ij} \mathbf{n}_{ij}$ and $\mathbb{P}_t = \mathbb{1} - \mathbf{n}_{ij} \mathbf{n}_{ij}$ represent the normal and

tangential projection matrices and the X 's and Y 's are scalar resistances depending on μ , a_i , a_j , and the gap between the two particles; see Appendix A for the detailed expressions and Appendix B for their numerical treatment.

3. Contact force (acting on each overlapping pair i, j)

$$\mathbf{F}_{i,j}^C = -k_n \mathbf{h}_{ij} - \gamma_n \mathbb{P}_n(\mathbf{u}_i - \mathbf{u}_j) - k_t \boldsymbol{\xi}_{ij}, \quad (5a)$$

$$\mathbf{F}_{j,i}^C = -\mathbf{F}_{i,j}^C, \quad \text{with} \quad |k_t \boldsymbol{\xi}_{ij}| \leq \mu_c |k_n \mathbf{h}_{ij} + \gamma_n \mathbb{P}_n(\mathbf{u}_i - \mathbf{u}_j)|, \quad (5b)$$

$$\mathbf{T}_{i,j}^C = a_i k_t (\mathbf{n}_{ij} \times \boldsymbol{\xi}_{ij}), \quad (5c)$$

$$\mathbf{T}_{j,i}^C = a_j k_t (\mathbf{n}_{ij} \times \boldsymbol{\xi}_{ij}). \quad (5d)$$

where $\mathbf{h}_{ij} = h_{ij} \mathbf{n}_{ij}$ denotes the signed normal surface gap between particles i and j (positive when overlapping), $\boldsymbol{\xi}_{ij}$ the signed tangential stretch (see Appendix C for definition), k_n the normal spring constant, γ_n the damping constant, k_t the tangential spring constant, and μ_c the friction coefficient. The condition in Eq. (5b) states the Coulomb's law of friction.

4. Electrostatic repulsion (acting on each repulsing pair i, j)

$$\mathbf{F}_{i,j}^R = -F_{er} \bar{a} / a_1 \exp(-\kappa h_{ij}) \mathbf{n}_{ij}, \quad (6a)$$

$$\mathbf{F}_{j,i}^R = -\mathbf{F}_{i,j}^R. \quad (6b)$$

where F_{er} is the force scale for the electrostatic repulsion, $\bar{a} = 2a_i a_j / (a_i + a_j)$ the harmonic mean radius of two interacting particles i and j , and κ the inverse Debye length.

5. Van der Waals attraction (acting on each attracting pair i, j)

$$\mathbf{F}_{i,j}^A = \frac{A \bar{a} \mathbf{n}_{ij}}{12(h_{ij}^2 + \epsilon^2)}, \quad (7a)$$

$$\mathbf{F}_{j,i}^A = -\mathbf{F}_{i,j}^A. \quad (7b)$$

where A is the Hamaker constant and $\epsilon = 0.1 \bar{a}$ a regularization term (Singh *et al.* 2019).

Note that the hydrodynamic force and torque given above are strictly valid for spheres. The validity of these expressions for non-spherical particles depends on (i) the dominance of the non-hydrodynamic interactions over the hydrodynamic ones and (ii) the departure of the particle shape from a sphere. In addition, the torque due to non-hydrodynamic forces has the general form of $\mathbf{T} = \mathbf{r} \times \mathbf{F}$, where \mathbf{r} is the lever arm vector. Therefore, extra care must be taken when applying the above formulation directly to non-spherical particles.

2.2. Model parameters

The preceding equations are formulated in dimensional form with a unit system flexibly chosen for the convenience of simulations (e.g. $a_1 = 1$ [Length], $\dot{\gamma} = 10^{-2}$ [1/Time], etc.). For the model output to correspond *physically* to a dense suspension of inertialess, rigid particles, the following asymptotic conditions must be satisfied as close as possible.

1. A vanishing Stokes number, $\text{St} = \rho\dot{\gamma}a_1^2/\mu \ll 1$.

The Stokes number controls the effect of particle inertia on the particle dynamics in viscous flows. This can be readily seen by inserting $F \sim \mu a U$ in the force balance $F \approx \rho a^3(\delta U/\delta\tau)$, and comparing $\delta\tau$ with $1/\dot{\gamma}$ at $\delta U \sim U$. Empirically, we find $\text{St} \sim \mathcal{O}(10^{-2})$ is often sufficient for the particle inertia to be negligible.

2. A vanishing stiffness-scaled shear rate, $\hat{\dot{\gamma}} = \dot{\gamma}a_1/\sqrt{k_n/(\rho a_1)} \ll 1$.

$\hat{\dot{\gamma}}$ describes the particle “hardness”. It follows from the scaling analysis that two particles of same radius a , at overlap distance δ , have a contact area $A \approx \pi a \delta$. Given the collision force $F \approx k_n \delta$, the characteristic velocity is then $U \sim \sqrt{P/\rho} \sim \sqrt{k_n/(\rho \pi a)}$. This provides a collision time scale $\tau \sim a/\sqrt{k_n/(\rho \pi a)}$ that can be compared with $1/\dot{\gamma}$. Empirically, we find $\hat{\dot{\gamma}} \sim \mathcal{O}(10^{-4})$ is sufficient for the particles to be considered as hardspheres.

3. A vanishing non-dimensional relaxation time, $\hat{\tau} = \gamma_n \dot{\gamma}/k_t$ or $\mu a_1 \dot{\gamma}/k_t \ll 1$.

$\hat{\tau}$ describes the non-dimensional relaxation time associated with a contact. Both normal and tangential contacts have relaxation times; here, we require the latter to be much less than 1, since $k_t < k_n$ typically. The second definition for $\hat{\tau}$ above uses the normal lubrication force as the dashpot in case $\gamma_n = 0$, cf. Eqs. (4a, A.1a, A.2a).

Apart from the above conditions, the behaviour of the simulated suspension is determined by the volume fraction ϕ and any force-rescaled shear rate $\hat{\dot{\gamma}}_r = 6\pi\mu a_1^2/F$ if an additional non-hydrodynamic force scale exists in the system, e.g. friction or electrostatic repulsion. Physically, $\hat{\dot{\gamma}}_r$ is usually introduced to invoke a rate-dependent rheology.

2.3. Stress tensor and bulk rheology

The bulk stress tensor is calculated as

$$\Sigma = 2\mu E^\infty + \frac{1}{V} \left(\sum_i^N \mathbb{S}_i^S + \sum_{i,j}^{N_l} \mathbb{S}_{ij}^L + \sum_{i,j}^{N_c} \mathbb{S}_{ij}^C + \sum_{i,j}^{N_r} \mathbb{S}_{ij}^R + \sum_{i,j}^{N_a} \mathbb{S}_{ij}^A \right), \quad (8)$$

where $V = L_x L_y L_z$ is the volume of the simulation box, and \mathbb{S} 's denote the stresslets due to various interactions. In the conventional coordinate system where x, y, z denote the streamwise, velocity-gradient, and vorticity directions, respectively, the individual terms in Eq. (8) corresponding to a simple shear

flow are given by

$$\mathbb{E}^\infty = \begin{bmatrix} 0 & \dot{\gamma}/2 & 0 \\ \dot{\gamma}/2 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}, \quad (9a)$$

$$\mathbb{S}_i^A = (20\pi\mu a_i^3/3)\mathbb{E}^\infty, \quad (9b)$$

$$\mathbb{S}_{mn}^L = (F_m^L r_n + F_n^L r_m)/2, \quad (9c)$$

$$\mathbb{S}_{mn}^C = F_m^C r_n, \quad (9d)$$

where \mathbf{r} is the separation vector pointing from particle i to particle j , and Eq. (9d) applies to other interparticle forces as well. Note that, the calculation of the lubrication stresslet, Eq. (9c), involves a simplification: the $\mathcal{O}(\delta)$ and isotropic terms are neglected; see Radhakrishnan (2018) for the derivation. Note also that, the subscript in Eq. (9b) refers to particle i , while the subscripts in Eqs. (9c–9d) denotes the tensor notation.

Once the stress tensor is obtained, the shear stress σ , normal stress differences N_1 and N_2 , and particle pressure Π can be readily calculated from the following definitions,

$$\sigma = \Sigma_{12}, \quad (10a)$$

$$N_1 = \Sigma_{11} - \Sigma_{22}, \quad N_2 = \Sigma_{22} - \Sigma_{33}, \quad (10b)$$

$$\Pi = -\text{Tr}(\Sigma)/3. \quad (10c)$$

The relative viscosity and non-dimensional particle pressure are defined as $\eta_r = \sigma/(\mu\dot{\gamma})$ and $\eta_n = \Pi/(\mu\dot{\gamma})$, respectively.

2.4. Numerical integration

The governing equations presented in Sec. 2.1 can be integrated in time explicitly using the modified velocity-Verlet algorithm (Groot & Warren 1997),

$$\mathbf{x}_i^{(n+1)} = \mathbf{x}_i^{(n)} + \Delta t \mathbf{v}_i^{(n)} + \frac{\Delta t^2}{2} \boldsymbol{\alpha}_i^{(n)}, \quad (11a)$$

$$\mathbf{v}_i^{(n+1/2)} = \mathbf{v}_i^{(n)} + \frac{\Delta t}{2} \boldsymbol{\alpha}_i^{(n)}, \quad (11b)$$

$$\boldsymbol{\alpha}_i^{(n+1)} = \mathcal{F} \left\{ \mathbf{x}_i^{(n+1)}, \mathbf{v}_i^{(n+1/2)} \right\}, \quad (11c)$$

$$\mathbf{v}_i^{(n+1)} = \mathbf{v}_i^{(n)} + \frac{\Delta t}{2} \left(\boldsymbol{\alpha}_i^{(n)} + \boldsymbol{\alpha}_i^{(n+1)} \right), \quad (11d)$$

where $\mathbf{x}_i^{(n)} = (x, y, z)_i^{(n)}$, $\mathbf{v}_i^{(n)} = (u, v, w)_i^{(n)}$, and $\boldsymbol{\alpha}_i^{(n)}$ denote the position, velocity, and acceleration vectors of particle i , respectively, at time $t = n\Delta t$, and \mathcal{F} denotes the force functional as in Eq. (2a). For spherical particles, the same update procedure applies to particle orientations, angular velocities, and angular accelerations (see Sec. 4 for non-spherical particles).

To comply with simple shear flows at fixed volume, the Lees-Edwards boundary condition is imposed on particle positions and their u velocity components to remove the wall effects and reduce the size of the computational box (Lees &

Edwards 1972). Physically, it results in a homogeneous suspension with a net momentum flux in the y direction if the system is far from equilibrium, and it reads

$$x = \begin{cases} (x + L_x - x_{sh}) \bmod L_x & \text{if } y > L_y, \\ (x + L_x + x_{sh}) \bmod L_x & \text{if } y < 0, \\ (x + L_x) \bmod L_x & \text{otherwise,} \end{cases} \quad (12a)$$

$$y = (y + L_y) \bmod L_y, \quad (12b)$$

$$z = (z + L_z) \bmod L_z, \quad (12c)$$

$$u = \begin{cases} u - u_{sh} & \text{if } y > L_y, \\ u + u_{sh} & \text{if } y < 0, \end{cases} \quad (12d)$$

where

$$x_{sh} = \dot{\gamma}L_y t \bmod L_x, \quad u_{sh} = \dot{\gamma}L_y, \quad (13)$$

define the position and velocity shifts at time t .

The above explicit integration scheme is second-order accurate in time and requires the time step Δt to be smaller than the smallest physical time scale of the process, cf. Sec. 2.2. Empirical experience suggests $\Delta t\dot{\gamma} \approx 10^{-6}$. Alternatively, the equations of motion can be solved by matrix inversion upon setting the left-hand-side of Eq. (1) to zero, thus obtaining particle velocities in the quasi-static limit (Mari *et al.* 2014). The latter approach is equivalent to the one adopted here provided $St \ll 1$, see e.g. Ness *et al.* (2018).

Finally, we note that the force calculation, Eq. (11d), is usually the most time-consuming step in a Verlet integration. One straightforward technique to speed up the computation is to construct a near-neighbour list (NNL) for each particle and only calculate the force between particle pairs therein. This way, the operation count for a complete update reduces from being quadratic to being linear with the number of particles.¹⁰ Practically, the algorithm requires $\mathcal{O}(cN)$ operations, where c is a kernel constant mainly depending on the cutoff distance of particle interactions.¹¹

3. Validations

In this section, we validate our numerical implementation of the DLCD model with two benchmark rheologies, *i.e.* jamming transition and shear thickening.

¹⁰The overhead due to the construction of a NNL normally does not exceed the cost reduction for dense suspensions.

¹¹The lubrication cutoff is usually chosen as $0.05\min(a_1, a_2)$ for dense suspensions, see Cheal & Ness (2018).

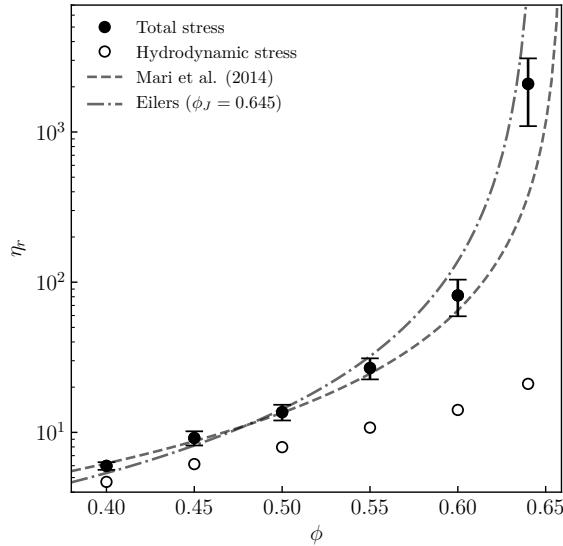


Figure 2: Relative viscosity versus volume fraction for suspensions of frictionless spheres.

3.1. Jamming transition

We simulate 200 initially randomly distributed spherical particles in a cubic box. A monodisperse suspension is considered for the lowest volume fraction $\phi = 40\%$, while bidisperse suspensions with radius ratio $a_2/a_1 = 1.4$ in equal volumes are used for higher volume fractions to prevent ordering. The random seeds are generated by the protocol of Mari and Seto.¹² The particles interact via Stokes drag, lubrication, and collision forces without any friction or physico-chemical interactions. The parameters used for this case are summarized in Table 1.¹³

Table 1: Summary of parameters.

N	St	$\dot{\gamma}$	$\hat{\tau}$	μ_c	$t\dot{\gamma}$
200	$10^{-2} \sim 10^{-3}$	$7 \times 10^{-6} \sim 10^{-4}$	$5 \times 10^{-8} \sim 10^{-6}$	0	10

Figure 2 shows the relative viscosity for six volume fractions ranging from 40% to 64%. The data pertain temporal averages and standard deviations calculated in the standard way over a strain of 10. Clearly, a jamming transition indicated by a diverging η_r as ϕ increases is observed in favorable comparison

¹²See https://github.com/rmari/LF_DEM for details.

¹³Numerical convergence is checked by halving $\dot{\gamma}$ or doubling k_n , where roughly the same results are obtained.

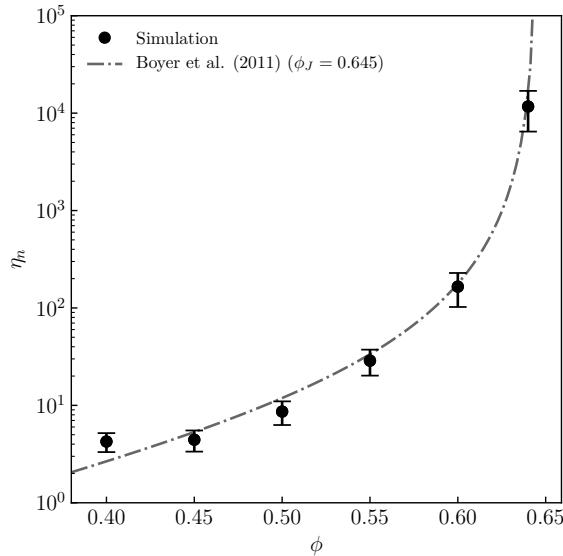


Figure 3: Particle pressure (normalized) versus volume fraction for suspensions of frictionless spheres.

with both previous numerical simulations and an empirical correlation. Specifically, the power-law fitting of Mari *et al.* (2014) satisfies $\eta_r = 1.4(1 - \phi/\phi_J)^{-1.6}$, with the jamming volume fraction $\phi_J = 0.66$; while the Eilers' correlation has the form $\eta_r = [1 + (5\phi/4)/(1 - \phi/\phi_J)]^2$, where we take $\phi_J = 0.645$. Our data fall well within the two limits, suggesting a jamming volume fraction at approximately 65% for frictionless spheres. Note that, the exact value of ϕ_J is sensitive to the particle overlap (numerically) or surface roughness (experimentally), as the contact stress dominates over the hydrodynamic one at high volume fractions. For spheres, the latter becomes less than 10% of the total stress for $\phi \gtrsim 0.6$, see Figure 2.

We also examine the non-dimensional particle pressure of the same suspension and compare our results with the theoretical model of Boyer *et al.* (2011). As illustrated in Figure 3, an even steeper divergence of η_n with ϕ is observed, suggestive of a strong tendency for the system to dilate. The constitutive law derived by Boyer *et al.* (2011), supported by their own experiment, is given as $\eta_n = [\phi/(\phi_J - \phi)]^2$. Taking $\phi_J = 0.645$, an overall excellent agreement is observed except for the lowest volume fraction $\phi = 40\%$, where our data slightly over-predicts the particle pressure. In general, the comparisons above verify our implementation of the DLCD model in the case of jamming transition of frictionless spheres.

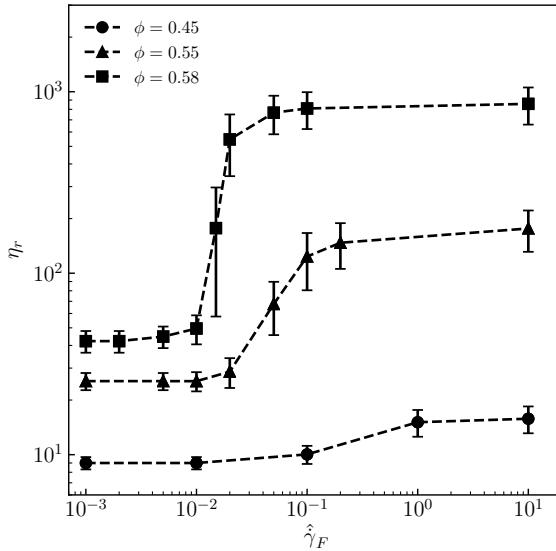


Figure 4: Continuous and discontinuous thickening of suspensions of frictional spheres. $N = 500$, $\mu_c = 0.5$.

3.2. Shear thickening

As contact becomes more important at higher particle concentrations, we simulate three frictional suspensions displaying continuous and discontinuous shear thickening rheologies. Specifically, we implement the critical-load friction model introduced in Mari *et al.* (2014) that activates the friction force only when the normal collision force exceeds a critical value. Figure 4 reproduces the results of Mari *et al.* (2014). Here, the relative viscosity is plotted against a non-dimensional shear rate defined by the ratio of the Stokes drag and the threshold friction, cf. Sec. 2.2. At 45% volume fraction, the suspension viscosity increases gradually with the shear due to increased particle contact. The slope steepens for $\phi = 0.55$; while at $\phi = 0.58$, we observe an abrupt increase of the mean relative viscosity, accompanied by large fluctuations, indicating a switch from CST to DST. Accurate prediction of the onset of DST remains a theoretical challenge (Morris 2020). Our results demonstrate the present implementation of the DLCD model can be used to probe such rheologies in details.

4. Extension to non-spherical particles

Finally, we turn our attention to suspensions of non-spherical particles. While spheres represent the simplest geometry convenient for theoretical and numerical studies, suspensions in reality are almost certainly composed of non-spherical

particles. For very dilute suspensions, this may not be an important issue as the macroscopic behaviours are usually not very sensitive to the exact particle shape. However, for dense suspension, a completely different rheology or phase transition may be expected as the lubrication intensifies and particle contact increases, see e.g. Damasceno *et al.* (2012); Royer *et al.* (2015); Trulsson (2018). To simulate suspensions in the latter case, various levels of simplifications may be made depending on the specific particle shape and volume fraction. Here, we present a minimal model that captures the essential effects, mostly suitable for spherical aggregates or polyhedra at high concentrations, extended from the DLCD model.

Assuming collisions and frictions are the most significant factor to the suspension rheology, we can approximate the hydrodynamic interactions using the same functional forms as presented in Sec. 2.1. That is, each particle experiences a Stokes drag based on some hydraulic radius, \hat{a} , and each neighbouring pair interacts via the lubrication force as if they were spheres. The exact value of \hat{a} depends on the shape of the particle, e.g. $\hat{a} = L/2$ for a cube of length L ; in general, this is an approximation that shall not greatly affect the bulk behaviour. In a similar spirit, the radii used in the lubrication calculation can be taken as \hat{a} or the inverse of the local curvature. We note that more rigorous treatments of the hydrodynamic interactions have been proposed, though it is rather complicated to implement and is limited to spheroids, see e.g. Claeys & Brady (1993). Our simplified approach is similar to the mean-field description developed for wet foams, which has also been employed in studies of frictionless particles (Durian 1997; Marschall *et al.* 2019).

For non-spherical particles, an additional complication arises due to the generally nontrivial rigid-body dynamics, where particle orientations matter and the moment-of-inertia, \mathbf{I}_i , is a tensor. This motivates us to use a quaternion-based, predictor-corrector direct multiplication (PCDM) scheme, as follows

$$\mathbf{x}_i^{(n+1)} = \mathbf{x}_i^{(n)} + \Delta t \mathbf{v}_i^{(n+1/2)}, \quad (14a)$$

$$\mathbf{v}'_i^{(n+1)} = \mathbf{v}_i^{(n+1/2)} + \frac{\Delta t}{2} \boldsymbol{\alpha}_i^{(n)}, \quad (14b)$$

$$\boldsymbol{\omega}_i^{(n+3/4),b} = \boldsymbol{\omega}_i^{(n+1/2),b} + \frac{\Delta t}{4} \boldsymbol{\beta}_i^{(n),b}, \quad (14c)$$

$$\boldsymbol{\omega}_i^{(n+3/4)} = q^{(n+1/2)} \boldsymbol{\omega}_i^{(n+3/4),b} \left(q^{(n+1/2)} \right)^{-1}, \quad (14d)$$

$$q'^{(n+1)} = \left[\cos \left(\frac{\|\boldsymbol{\omega}_i^{(n+3/4)}\| \Delta t}{4} \right), \sin \left(\frac{\|\boldsymbol{\omega}_i^{(n+3/4)}\| \Delta t}{4} \right) \frac{\boldsymbol{\omega}_i^{(n+3/4)}}{\|\boldsymbol{\omega}_i^{(n+3/4)}\|} \right] q^{(n+1/2)}, \quad (14e)$$

$$\boldsymbol{\omega}'_i^{(n+1),b} = \boldsymbol{\omega}_i^{(n+1/2),b} + \frac{\Delta t}{2} \boldsymbol{\beta}_i^{(n),b}, \quad (14f)$$

$$\boldsymbol{\omega}'_i^{(n+1)} = q'^{(n+1)} \boldsymbol{\omega}'_i^{(n+1),b} \left(q'^{(n+1)} \right)^{-1}, \quad (14g)$$

$$\boldsymbol{\alpha}_i^{(n+1)} = \mathcal{F} \left\{ \boldsymbol{x}_i^{(n+1)}, \boldsymbol{v}'_i^{(n+1)}, q'^{(n+1)}, \boldsymbol{\omega}'_i^{(n+1)} \right\}, \quad (14\text{h})$$

$$\boldsymbol{T}_i^{(n+1)} = \mathcal{T} \left\{ \boldsymbol{x}_i^{(n+1)}, \boldsymbol{v}'_i^{(n+1)}, q'^{(n+1)}, \boldsymbol{\omega}'_i^{(n+1)} \right\}, \quad (14\text{i})$$

$$\boldsymbol{T}_i^{(n+1),b} = \left(q'^{(n+1)} \right)^{-1} \boldsymbol{T}_i^{(n+1)} q'^{(n+1)}, \quad (14\text{j})$$

$$\boldsymbol{\beta}_i^{(n+1),b} = \left(\boldsymbol{I}_i^b \right)^{-1} \left(\boldsymbol{T}_i^{(n+1),b} - \boldsymbol{\omega}'_i^{(n+1),b} \times \boldsymbol{I}_i^b \boldsymbol{\omega}'_i^{(n+1),b} \right), \quad (14\text{k})$$

$$\boldsymbol{\omega}_i^{(n+3/2),b} = \boldsymbol{\omega}_i^{(n+1/2),b} + \Delta t \boldsymbol{\beta}_i^{(n+1),b}, \quad (14\text{l})$$

$$q^{(n+3/2)} = \left[\cos \left(\frac{\|\boldsymbol{\omega}'_i^{(n+1)}\| \Delta t}{2} \right), \sin \left(\frac{\|\boldsymbol{\omega}'_i^{(n+1)}\| \Delta t}{2} \right) \frac{\boldsymbol{\omega}'_i^{(n+1)}}{\|\boldsymbol{\omega}'_i^{(n+1)}\|} \right] q^{(n+1/2)}, \quad (14\text{m})$$

$$\boldsymbol{\omega}_i^{(n+3/2)} = q^{(n+3/2)} \boldsymbol{\omega}_i^{(n+3/2),b} \left(q^{(n+3/2)} \right)^{-1}, \quad (14\text{n})$$

$$\boldsymbol{v}_i^{(n+3/2)} = \boldsymbol{v}_i^{(n+1/2)} + \Delta t \boldsymbol{\alpha}_i^{(n+1)}, \quad (14\text{o})$$

where $q = q_0 + \boldsymbol{q}$ is the quaternion, $\boldsymbol{\beta}$ is the angular acceleration, and superscripts prime and b denote prediction and body-frame values, respectively. Clearly, the integration scheme becomes much more cumbersome when the particles are non-spherical.

For collision detection, we employ the classical GJK algorithm that efficiently computes the Euclidean distance between a pair of convex sets (Gilbert *et al.* 1988). The resulting bulk rheology can be calculated in the same way as in Sec. 2.3, *i.e.* summing the first moment of various force fields. Validation of the present algorithm will be presented in upcoming publications.

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Appendix

A. Lubrication resistances

The scalar resistances introduced in Eqs. (4a–4d) follow those in Kim & Karrila (2013); Cheal & Ness (2018), and are given as

$$X_{ii}^A = a_i (R_{xii1}/\delta + R_{xii2} \log(1/\delta)), \quad (\text{A.1a})$$

$$Y_{ii}^A = a_i R_{yia} \log(1/\delta), \quad (\text{A.1b})$$

$$Y_{ii}^B = a_i^2 R_{yib} \log(1/\delta), \quad (\text{A.1c})$$

$$Y_{ji}^B = a_j^2 R_{yjib} \log(1/\delta), \quad (\text{A.1d})$$

$$Y_{ii}^C = a_i^3 R_{yiic} \log(1/\delta), \quad (\text{A.1e})$$

$$Y_{ij}^C = Y_{ji}^C = a_i^3 R_{yijc} \log(1/\delta), \quad (\text{A.1f})$$

$$Y_{jj}^C = a_j^3 R_{yjjc} \log(1/\delta), \quad (\text{A.1g})$$

where $\delta = 2h_{ij}/(a_i + a_j)$ is the non-dimensional surface gap between particles i and j , and the coefficients R 's are functions of the size ratio $\lambda = a_j/a_i$ and μ . Specifically, they are calculated as

$$R_{xii a1} = (6\pi\mu) \frac{2\lambda^2}{(1+\lambda)^3}, \quad R_{xii a2} = (6\pi\mu) \frac{\lambda(1+7\lambda+\lambda^2)}{5(1+\lambda)^3}, \quad (\text{A.2a})$$

$$R_{yia} = (6\pi\mu) \frac{4\lambda(2+\lambda+2\lambda^2)}{15(1+\lambda)^3}, \quad (\text{A.2b})$$

$$R_{yiib} = (-4\pi\mu) \frac{\lambda(4+\lambda)}{5(1+\lambda)^2}, \quad (\text{A.2c})$$

$$R_{yjib} = (-4\pi\mu) \frac{\lambda^{-1}(4+\lambda^{-1})}{5(1+\lambda^{-1})^2}, \quad (\text{A.2d})$$

$$R_{yiic} = (8\pi\mu) \frac{2\lambda}{5(1+\lambda)}, \quad (\text{A.2e})$$

$$R_{yijc} = (8\pi\mu) \frac{\lambda^2}{10(1+\lambda)}. \quad (\text{A.2f})$$

$$R_{yjjc} = (8\pi\mu) \frac{2\lambda^{-1}}{5(1+\lambda^{-1})}. \quad (\text{A.2g})$$

B. Numerical treatment of the lubrication at small and large distances

We note that the lubrication force and torque are singular at contact, with $X \sim 1/\delta$ and $Y \sim \log(1/\delta)$, see Appendix A. The singularity derives from assuming perfectly smooth particles in the mathematical sense. In practice, particle contact is inevitable due to surface roughness; thus, we allow small overlap to occur invoking both lubrication and contact forces. Numerically, the lubrication force is saturated below $h_{inner} = 0.001a_1$ and truncated above $h_{outer} = (0.05 \sim 0.2)a_1$, where a_1 denotes the smallest particle radius. The outer range is introduced to reduce the computational cost and is adjusted by examining the radial distribution function at each volume fraction.¹⁴

¹⁴The lubrication formulation has been simplified comparing to Jeffrey & Onishi (1984); Jeffrey (1992) by neglecting terms of $\mathcal{O}(1)$ or higher order. Therefore, the lubrication outer cutoff cannot be arbitrarily large. See Radhakrishnan (2018) for the detailed algebra.

C. Tangential stretch for contacting particles

Following Appendix B, another consequence of the particle roughness is the initiation of frictional contact. Here, we adopt the standard stick/slide model for the calculation of the friction force, as given in Eqs. (5a–5b) (Cundall & Strack 1979; Luding 2008). Specifically, the tangential stretch vector is calculated as

$$\xi_{ij}(t) = \begin{cases} \int_{t_0}^t -P_t[(\mathbf{u}_i - \mathbf{u}_j) + (a_i \boldsymbol{\omega}_i + a_j \boldsymbol{\omega}_j) \times \mathbf{n}_{ij}] dt', & \text{if } |\xi_{ij}| < |\xi_{max}|, \\ \xi_{max}, & \text{otherwise,} \end{cases} \quad (\text{C.1})$$

where ξ_{max} is maximal tangential stretch allowed by the Coulomb's law of friction (Eq. 5b), and t_0 is the moment a frictional contact is established. We choose $k_t = (2/7)k_n$ as given in Cheal & Ness (2018).

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