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Diffusiophoresis driven colloidal manipulation and shortcuts to adiabaticity

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Abstract

While compressing a colloidal state by optical means alone has been previously achieved through a specific time-dependence of the trap stiffness, realizing quickly the reverse transformation stumbles upon the necessity of a transiently expulsive trap. To circumvent this difficulty, we propose to drive the colloids by a combination of optical trapping and diffusiophoretic forces, both time-dependent. Forcing via diffusiophoresis is enforced by controlling the salt concentration at the boundary of the domain where the colloids are confined. The method takes advantage of the separation of time scales between salt and colloidal dynamics, and realizes a fast decompression in an optical trap that remains confining at all times. We thereby obtain a so-called shortcut to adiabaticity protocol where colloidal dynamics, enslaved to salt dynamics, can nevertheless be controlled as desired.

1. Introduction

Shortcut to adiabaticity protocols have been introduced as fast alternatives to otherwise time consuming transformations, where the parameters controlling a system are slowly (adiabatically) changed [1, 2]. Applied to quantum systems, they allow for manipulation on timescales shorter than the decoherence time, with applications in metrology and interferometry [2]. They are also useful when applied to classical devices, providing for instance a better control of micromechanical oscillators, with applications in faster atomic force imaging techniques [3]. Shortcut to adiabaticity protocols have consequently been studied and tested successfully in different systems [4–15]. In these approaches, one or two control parameters are engineered such that the equilibrium is reached in a chosen short time, using processes which are not necessarily heat-exchange-free nor isothermal. A beneficial feature of these methods lies in the multiplicity of admissible protocols, which allows to select those with desirable properties, in terms of robustness or optimality [2, 16, 17]. It is worth stressing that we use here the word ‘adiabaticity’ in the quantum mechanics inspired meaning of ‘slow enough’; it should thus not be confused with the thermodynamic meaning of being heat-exchange free [18].

The present work is devoted to manipulating trapped colloidal systems. It is related to the engineered swift equilibration (ESE) protocol presented in reference [19]. Following this framework, it will be here assumed that the colloidal dynamics is overdamped, which proves to be an experimentally very accurate approximation [18]. By appropriately tuning the trap stiffness as a function of time, a fast compression of a colloidal state was achieved, with a close to 100-fold acceleration compared to the bare relaxation time [19]. During the process, the probability distribution function of the colloid position is kept Gaussian while the trapping potential remains quadratic, facilitating reaching the equilibrium at the final time. Generalizations to the underdamped regime, and to two hydrodynamically interacting colloids have been worked out in references [20, 21]. Besides, variants have been proposed allowing for non-conservative forces [22], or transitions between non-equilibrium steady states [15].

A drawback of ESE protocols appears when comparing compression and expansion of systems involving confinement with optical tweezers. A state can be compressed by adequately choosing the time dependence

of the intensity of the trapping laser; increasing the speed of the transformation simply requires a larger laser intensity range [19]. On the other hand, expanding a trapped system in a quick fashion requires to create, transiently, an expulsive rather than confining potential [19, 20]. This is necessary to guarantee a fast displacement of the colloids toward their statistically relevant end position. In other words, the trap stiffness not only needs to follow a precise time dependence, but also to be negative during some time window. This is a difficult experimental challenge, for instance when trapping is realized with optical tweezers [23]. A partial solution to this shortcoming was proposed with the method worked out in reference [24], where protocols conditioned to using non-negative stiffness have been derived, optimizing furthermore the (mean) work performed by the operator. Here, we shall explore a different venue for effectively achieving transient negative stiffness. Our proposition is an alternative to the thermal bath engineering strategy put forward in reference [25]. There, an externally controlled noise is applied to the center of the trap, mimicking a time-dependent effective temperature that necessarily exceeds that of the bath in which the system is immersed (say water). The possibility to decompress the system under study without repulsive potentials ensues: advantage is taken of an enhanced diffusion. Yet, this technique does not suit for ESE compression [26]. On the contrary, the present combination of optical and phoretic forces is operational not only to allow for decompression, but can be as well transposed to compression. Our approach also differs from the optical feedback trap realizations proposed in [23], in so far as we do not operate any retro-action on the system, but proceed in a purely feed-forward manner.

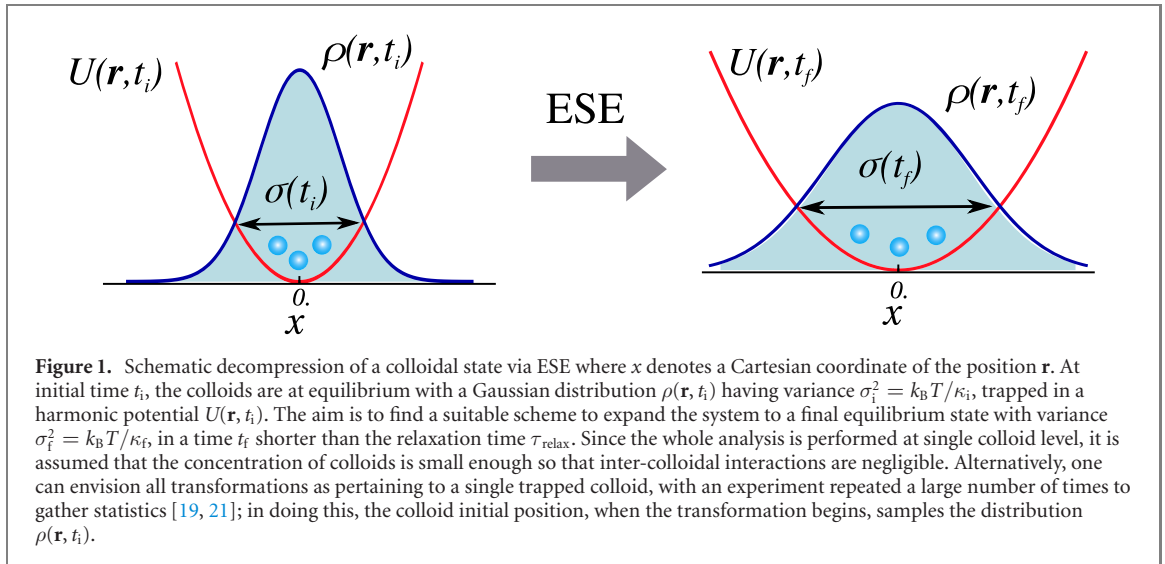
Since achieving fast decompression ESE paves the way toward possible application in designing or improving the performance of small-scale heat engines [27–30] and thermodynamic cycles in Brownian and active systems [30–37], we search for operational procedures of augmented ESE, to circumvent the decompression bottleneck. We propose here a protocol which takes advantage of the diffusiophoresis of colloids. Diffusiophoresis refers to the migration of colloids and macro-molecules immersed in a solution under a gradient of solute, that is driven by a direct surface–solute interaction [38, 39]. Recently, diffusiophoretic forces emerged in various microfluidic applications [40], for enhancing transport rate of flow [41] and designing novel types of active microswimmers [42–44], as well as for industrial applications such as underground oil and gas recovery [45]. Other applications include the effective and long-lasting cleaning or removal of particles and droplets from deep pores [46, 47], and enhanced oil recovery from deep wells [48–50]. A particular feature of diffusiophoresis emerges in electrolytes (i.e., ionic solutions where the solute is a salt) where the ions and the particle surface interact through electrostatic forces: the colloidal (mean) velocity becomes proportional to the gradient of the logarithm of the solute concentration, i.e., $\mathbf{V}_{DP} \propto \nabla \ln C$, at variance with neutral solutions where this dependency is linear in the concentration gradient [39]. This feature allows for efficient driving of colloids [51, 52], and guarantees significant forcing even in regime of small solute concentrations. It also opens the possibility of trapping colloids and macro-molecules via osmotic forces, as recently achieved [53, 54].

In this paper, we propose an ESE strategy that takes advantage of diffusiophoresis. As the phoretic force direction depends on both the solute gradient direction and the solute–surface interaction, creating a repulsive potential is achievable by appropriately tuning the solute concentration on the boundaries of the system as a function of time [54, 55]. Accordingly, our approach allows for both an accelerated expansion or an accelerated compression of a colloid state in harmonic or non-harmonic confinement. Our analysis restricts to low density systems, where colloidal dynamics does not result from colloid–colloid interactions, but from other types of forces. This logic can be pushed further, considering that a single colloid is being manipulated, in a repeated fashion in order to gather statistics and generate the distribution functions that will be object of interest here. So was the situation in the experiment reported in reference [19].

The rest of the paper organizes into six sections. We address the basics of ESE protocols in the overdamped regime and present its governing equations in section 2. Section 3 is devoted to the key equations of diffusiophoresis. Then, we combine the ESE method with diffusiophoresis to introduce the diffusiophoresis driven protocol in section 4 for Gaussian colloidal density and harmonic trapping potentials. In section 5, we discuss the consistency of these results. We consider non-Gaussian states and non-harmonic potentials in section 6, while concluding remarks are presented in section 7.

2. ESE for Gaussian distributions

The system under study is a (overdamped) Brownian colloidal particle in water, trapped in a harmonic potential $U = \kappa r^2/2$, where r is the distance to the trap center. This external potential is usually established by optical tweezers. We seek for a transition where the stiffness κ is changed from an initial value κ_i to a final value κ_f in a given time t_f . The idea is to find a suitable function for the stiffness $\kappa(t)$ such that, as demonstrated in figure 1, the system evolves from an equilibrium state with initial variance $\sigma_i^2 = k_B T / \kappa_i$ to the final value $\sigma_f^2 = k_B T / \kappa_f$. This should be achieved faster than the natural relaxation time $\tau_{\text{relax}} = \gamma / \kappa_f$,



that would be required to reach equilibrium after a sudden change of stiffness from κ_i to κ_f . Here, γ is the colloid drag coefficient, related to the diffusion coefficient of the colloid D_c through $\gamma = k_B T / D_c$, where k_B and T represent the Boltzmann constant and the water bath temperature, respectively. The system is in equilibrium at initial and final times, but not during the process. The bath temperature is constant. Note that with the above relations, the relaxation time can also be written $\tau_{\text{relax}} = \sigma_f^2 / D_c$: it corresponds to the time needed to explore diffusively a region of space of linear size σ_f .

2.1. The formalism

The overdamped Langevin equation describes the dynamics of the colloid at position \mathbf{r} relative to the trap center at position $\mathbf{0}$, as

$$\gamma \dot{\mathbf{r}} = -\nabla U(\mathbf{r}, t) + \xi(t) = -\kappa(t)\mathbf{r} + \xi(t), \quad (1)$$

where $\xi(t)$ is a random force, a white noise with 0 mean and correlation $\langle \xi_i(t) \xi_j(t') \rangle = 2\gamma k_B T \delta_{ij} \delta(t - t')$. This very treatment, neglecting colloidal–colloidal interactions, thus holds for dilute enough systems, a situation often met experimentally. Accordingly, the colloidal density $\rho(\mathbf{r}, t)$ is governed by the Fokker–Planck equation:

$$\partial_t \rho(\mathbf{r}, t) = \nabla \cdot \left(D_c \nabla \rho(\mathbf{r}, t) + \frac{1}{\gamma} \nabla U \rho(\mathbf{r}, t) \right). \quad (2)$$

Control is achieved here by changing of the stiffness $\kappa(t)$ such that the density remains Gaussian at all times

$$\rho(\mathbf{r}, t) = \left(\frac{\alpha(t)}{\pi} \right)^{n/2} \exp(-\alpha(t)r^2), \quad (3)$$

where n refers to the system's dimension. The inverse variance $\alpha(t)$ can be chosen arbitrarily, fulfilling the equilibrium boundary conditions $\alpha(0) = \kappa_i / (2k_B T)$, $\alpha(t_f) = \kappa_f / (2k_B T)$, to which we add the smoothness conditions $\dot{\alpha}(0) = \dot{\alpha}(t_f) = 0$, experimentally more friendly than discontinuities (which are, strictly speaking, possible and do indeed arise when optimal features are sought [16, 24, 56]). Non-Gaussian distributions will be addressed in section 6. We do not need to specify the colloid's radius, which enters implicitly in the friction coefficient γ .

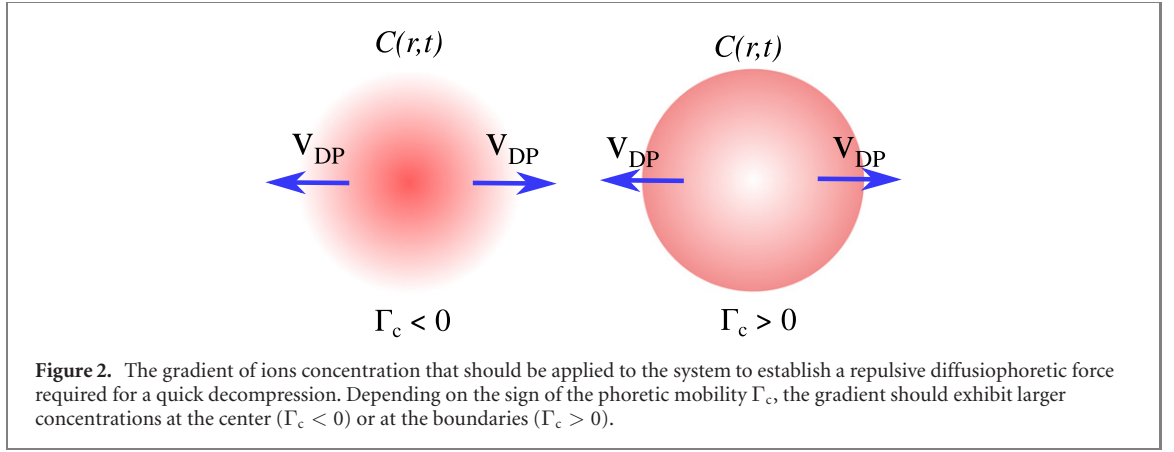
In order to obtain the time-dependent forcing $\kappa(t)$, we follow reference [19] and substitute equation (3) into the Fokker–Planck equation (2), and obtain:

$$\frac{n\dot{\alpha}(t)}{2\alpha(t)} - \dot{\alpha}(t)r^2 = \frac{\kappa(t)}{\gamma} (n - 2\alpha(t)r^2) - 2\alpha(t)D_c(n - 2\alpha(t)r^2), \quad (4)$$

which holds for any position r . This implies that

$$\kappa(t) = \frac{\dot{\alpha}(t)}{2\alpha(t)} \gamma + 2k_B T \alpha(t), \quad (5)$$

irrespective of dimension n . This indicates how to tune the trapping laser intensity with time. This led to an operational protocol in reference [19], whereby a colloidal state was compressed with a close to 100-fold gain in the time required to reach the desired equilibrium.



Before discussing the difficulty met when transposing this idea from compression to decompression, we point out the relevant dimensionless parameter quantifying acceleration. It is given by the ratio of time scales $\mathcal{A} = \tau_{\text{relax}}/t_f = \sigma_f^2/(D_c t_f)$ and measures the desired acceleration factor. Noting that $\dot{\alpha}$ has to be negative for a decompression processes, it follows from equation (5) that for sufficiently fast decompressions ($\mathcal{A} \gg 1$), the potential should become transiently ‘expulsive’ ($\kappa < 0$), irrespective of the choice of $\alpha(t)$. While feedback-based protocols have been proposed to this aim [23], achieving time-dependent and transiently repulsive confinements remains experimentally problematic, which motivates us to propose an alternative.

3. Diffusiophoresis for manipulating colloids

Diffusiophoresis is an out of equilibrium phenomenon by which colloids immersed in a solution migrate in response to a gradient of the solute concentration $\nabla C(\mathbf{r}, t)$. The solute gradient combined with direct interaction of particle–solute results in an osmotic pressure gradient in the thin interacting layer around the colloid. This pressure gradient creates a fluid flow and, consequently, colloid motion with velocities proportional to the solute gradient [38, 39]. Moreover, an interesting feature occurs in the case of electrolytes (a salt as a solute) and charged colloids, when the salt–colloid interaction potential has electrostatic nature. In this particular case, the thickness of the interacting layer (diffuse Debye layer) is inversely proportional to the ions concentration. This leads to a colloids’ velocity that is proportional to the gradient of logarithm of the ions concentration [38, 39]

$$\mathbf{V}_{\text{DP}} = \Gamma_c \nabla \ln C(\mathbf{r}, t), \quad (6)$$

where Γ_c is the phoretic mobility of colloidal particles. This quantity is related to the interaction potential between colloids and salt, and can be positive or negative¹. Accordingly, the diffusiophoretic force exerted on the colloid and the corresponding potential energy have the same log-dependency [57],

$$\mathbf{F}_{\text{DP}} = \gamma \Gamma_c \nabla \ln C(\mathbf{r}, t), \quad U_{\text{DP}} = -\gamma \Gamma_c \ln C(\mathbf{r}, t). \quad (7)$$

According to this relation, the direction and strength of \mathbf{F}_{DP} can be tuned by the concentration gradient and the phoretic mobility Γ_c . This feature yields the possibility of creating a repulsive force required for an ESE decompression protocol. Therefore, to make the colloids migrate away from the center of the system (i.e., to realize an expansion), a concentration gradient in the direction toward (respectively away from) the center should be applied for the case of negative (respectively positive) phoretic mobilities. It means that, as

¹ The phoretic mobility in symmetric $Z:Z$ electrolytes (with cations and anions of charges $\pm Ze$), is given by

$$\Gamma_c = \frac{\epsilon}{4\pi\eta} \left(\frac{k_B T}{Ze} \right)^2 \left[\Lambda \zeta - 2 \ln \left(1 - \tanh^2 \frac{\zeta}{4} \right) \right],$$

in the thin Debye layer limit. Here, ϵ and η are the dielectric constant and the viscosity of the solution, respectively, and $\zeta \equiv Ze\zeta/(k_B T)$ represents the scaled zeta-potential of the colloid. The first term originates from the difference of ionic diffusivities $\Lambda = (D_+ - D_-)/(D_+ + D_-)$. It can be negative or positive, depending on the sign of Λ as well as of the zeta-potential. However, the second term is always positive regardless of sign of zeta-potential. Therefore, when the diffusion coefficients of anions and cations are equal, i.e. $\Lambda = 0$, the phoretic mobility Γ_c is always positive. For different diffusion coefficients of anions and cations, one needs to replace D_s in the diffusion equation (8) by $2D_+ D_-/(D_+ + D_-)$, as long as the thin Debye layer approximation remains valid [39].

depicted in figure 2, the concentration on the boundaries $\phi(t)$ should be decreased (with respect to the concentration at the center) for the case of negative phoretic mobilities, and be increased for positive phoretic mobilities. Interestingly, thanks to the log-sensing, when time and position dependencies of the salt concentration $C(\mathbf{r}, t)$ factorize, $\nabla \ln C = \nabla C/C$ becomes independent of time. This happens in the long time limit when salt diffuses out of the system, so that $C \rightarrow 0$ [51, 53], with the surprising consequence that the force acting on the colloid is maintained while the solute at its origin fades away.

To complete the diffusiophoresis discussion, we note that for small colloidal charges (Debye–Hückel approximation) and in the thin Debye layer limit (i.e., with extension much smaller than colloid size), the concentration of positive and negative ions outside the Debye layer are approximately equal and governed by the diffusion equation [39]

$$\partial_t C(\mathbf{r}, t) = D_s \nabla^2 C(\mathbf{r}, t). \quad (8)$$

This is supplemented with initial and boundary conditions as follows:

$$C(\mathbf{r}, 0) = C_0, \quad C(\mathbf{r}, t)|_B = \phi(t), \quad (9)$$

where C_0 is the initial uniform ions concentration. Here, B refers to the boundaries of the system. For simplicity, we assume that the diffusion coefficient of positive and negative ions are equal and given by D_s ¹.

Finally, we note that the simplest way to establish a concentration gradient is to set a vanishing concentration $\phi(t)$ such that $\phi(t \geq 0) = 0$. Any choice of $\phi(t)$ that verifies $\phi = 0$ for $t \geq 0$ is possible. For example, one option is setting $\phi(t)$ *suddenly* to zero at $t = 0$ from a given value C_0 . As alluded to above, this leads after a salt-characteristic relaxation time (τ_s) to a time-position factorized salt concentration, with an associated time-independent forcing, see appendix A. However, although the resulting procedure is admissible for accelerating decompression, the final state is not controllable: the resulting stiffness is system size dependent, but cannot be tuned, as discussed in appendix A for a 1D system. Moreover, the configuration under study establishes a salt gradient toward the center $\mathbf{r} = \mathbf{0}$. Hence, as sketched in figure 2, this method can be used to decompress colloids with negative phoretic mobility (and compress colloids with positive phoretic mobility), but it is not a versatile approach for shortcutting adiabaticity, and we search for another route, where $\phi(t)$ will be deduced rather than imposed.

4. Diffusiophoresis driven ESE: Gaussian states

We next combine optical and diffusiophoretic drivings to establish our ESE protocol. The colloidal system is subject to optical confinement that can be time-dependent, and is furthermore in an electrolyte that can be controlled through the boundaries of the system, as performed in references [53, 55]. It might be thought that such a surface driving through the boundary concentration $\phi(t)$ does not provide enough command over the system, since salt invariably diffuses in the bulk of the solution, but we will see below that the present setting nevertheless offers interesting means to shape shortcutting protocols.

The total potential acting on the colloid is composed of a harmonic optical and a diffusiophoretic contribution, as

$$U = U_{\text{op}} + U_{\text{DP}} = \frac{1}{2} \kappa_{\text{op}}(t) r^2 - \gamma \Gamma_c \ln C(\mathbf{r}, t). \quad (10)$$

Both contributions are time dependent; in line with our underpinnings, we set the constraint $\kappa_{\text{op}} > 0$ for the optical contribution. In addition, we assume isotropy (so that C only depends on $r = |\mathbf{r}|$) and analyticity. The concentration, extremum at $\mathbf{r} = \mathbf{0}$, can then be expanded at small r to yield

$$\begin{aligned} U &= \frac{1}{2} \left(\kappa_{\text{op}}(t) - \gamma \Gamma_c \partial_r^2 \ln C(r, t) \Big|_{r=0} \right) r^2 + \mathcal{O}(r^4) \\ &\simeq \frac{1}{2} \kappa(t) r^2, \end{aligned} \quad (11)$$

(up to a position-independent shift in the potential origin, which is not relevant here) with the diffusiophoretic contribution to stiffness

$$\kappa_{\text{DP}}(t) \equiv \kappa(t) - \kappa_{\text{op}}(t) = -\gamma \Gamma_c \partial_r^2 \ln C(r, t) \Big|_{r=0}. \quad (12)$$

We demand that the total stiffness $\kappa(t)$ follows the rule given by equation (5), for a chosen $\alpha(t)$. While the sum $\kappa_{\text{DP}} + \kappa_{\text{op}}$ is hence known, the choice of κ_{op} enjoys some flexibility, and has to be specified in a convenient way. The simplest choice is arguably $\kappa_{\text{op}} = \kappa_i$, the only temporal dependant term in the total stiffness being borne by $\kappa_{\text{DP}}(t)$. Obtaining a steady non vanishing value for κ_{DP} requires that $\phi(t > t_f) = 0$:

otherwise, salt diffuses to reach a uniform concentration across the system, which cancels out the diffusiophoretic force. However, demanding *a priori* a vanishing $\phi(t > t_f)$ is a somewhat nontrivial constraint (see below). We will follow a different strategy where the signal $\phi(t)$ is the end product of the calculation, rather than an input; we will rather use the freedom of having a time-dependent optical potential, such that the diffusiophoresis provides a negative total stiffness (repulsive force), and the optical tweezers tune the initial and final states.

From equation (5) and the diffusion equation (8) which relates ∂_r^2 to ∂_t , we get the following constraint for the concentration at the center (see appendix B for details)

$$-\frac{\gamma\Gamma_c}{nD_s}\partial_t \ln C(0, t) = \frac{\dot{\alpha}(t)}{2\alpha(t)}\gamma + 2k_B T\alpha(t) - \kappa_{\text{op}}(t). \quad (13)$$

Solving for $C(0, t)$ yields

$$C(0, t) = C_0 \left(\frac{\alpha}{\alpha_i} \right)^{-\frac{nD_s}{2\Gamma_c}} \exp \left(-\frac{nD_s}{\gamma\Gamma_c} \int_0^t (2k_B T\alpha(\tau) - \kappa_{\text{op}}(\tau)) d\tau \right). \quad (14)$$

Finally, the concentration $\phi(t)$ at the boundary ($r = R$), is extracted from a Taylor expansion of the density at $r = 0$:

$$\begin{aligned} \phi(t) = C(r = R, t) &= \sum_{j=0}^{\infty} \frac{1}{j!} R^j \partial_r^j C(r, t)|_{r=0} \\ &= \sum_{j=0}^{\infty} \frac{1}{(2j)!} R^{2j} \partial_r^{2j} C(\mathbf{r}, t)|_{r=0} \\ &= \sum_{j=0}^{\infty} \frac{R^{2j}}{(2j)! D_s^j} \left(\prod_{m=1}^j \frac{2m-1}{2m-2+n} \right) \partial_t^j C(0, t). \end{aligned} \quad (15)$$

Here, we have made use of

$$\partial_r^{2j} C(\mathbf{r}, t)|_{r=0} = \left(\prod_{m=1}^j \frac{2m-1}{2m-2+n} \right) \frac{1}{D_s^j} \partial_t^j C(0, t), \quad (16)$$

which is obtained by taking successive derivatives of the diffusion equation (8) and invoking $\partial_r^j C|_{r=0} = 0$ (for j odd).

To summarize the procedure, the operator starts by the choice of $\alpha(t)$, which is proportional to the inverse variance of colloidal density (more precisely, $\sigma^2 = (2\alpha)^{-1}$). Then, a suitable function for the optical stiffness $\kappa_{\text{op}}(t)$ should be chosen, see the next section. Next, by calculating $C(0, t)$ from equation (14), we subsequently obtain $\phi(t)$ from equation (15), truncating the infinite summation appropriately. There is at this point no guarantee that $\phi(t)$ and, consequently, $C(\mathbf{r}, t)$ be positive. Working for the parameter range where both functions are non-negative sets a limitation to the approach, that will be assessed. To this end, we will provide in the next section a phase portrait for $\phi(t)$ as a function of the acceleration factor \mathcal{A} ($= \tau_{\text{relax}}/t_f$) and the ratio of initial and final stiffnesses κ_i/κ_f .

There are three time-scales in our problem: the relaxation time of the colloidal particle $\tau_{\text{relax}} = \sigma_f^2/D_c$, the operation time t_f , and the salt diffusion time $\tau_s = R^2/D_s$ (where R denotes the system size, see also appendix A). Due to the size asymmetry, the diffusion coefficient of ions is much larger than that of colloidal particles ($D_s \gg D_c$). Moreover, we cannot suppress the Brownian diffusion of ions (salt). Therefore, the final time t_f should be larger than τ_s in order for the diffusion of ions not to perturb the final state reached at t_f : any remaining non equilibrium ionic dynamics for $t > \tau_s$ would impinge on the colloidal state and make it non stationary. Therefore, we impose that $\tau_s < t_f \ll \tau_{\text{relax}}$, which means that $R^2/D_s \ll \sigma_f^2/D_c$. This sets an upper bound for the system size R , which controls the time scale for salt dynamics, and should be kept low for our approach to be operational [58].

As a final remark, we stress that we have cut the infinite Taylor expansion equation (15). To assess the possible resulting loss of precision for the whole scheme, we shall test the consistency of the results. To this end, having the boundary condition for salt concentration, $\phi(t)$, we solve the diffusion equation (8) numerically and obtain $C(\mathbf{r}, t)$. We then check that $\phi(t)$ is reasonably close to $C(\mathbf{r} = \mathbf{R}, t)$. Next, having $C(\mathbf{r}, t)$, we can calculate stiffnesses ($\kappa(t)$ and $\kappa_{\text{DP}}(t)$) and compare them with the target functions, as given by equations (5) and (12). Finally, we need to evaluate the final variance $\sigma_f^2 = \int r^2 \rho(\mathbf{r}, t_f) d^n \mathbf{r}$ which should be equal to $k_B T/\kappa_f$.

5. Results in one dimension: Gaussian states

5.1. Characterizing the acceptable protocols

Before putting the scheme to the test for a one dimensional confined system $-L/2 \leq x \leq L/2$, we discuss the choice of the inverse variance of the colloid's position through $\alpha(t)$, with which the whole procedure starts. Any function that verifies the initial and final conditions of a prescribed α with vanishing first derivative, can be taken. The simplest choice is polynomial in time and reads

$$\alpha^{(1)}(t) = \frac{1}{2k_B T} (\kappa_i + (\kappa_f - \kappa_i)(3s^2 - 2s^3)), \quad (17)$$

where $s = t/t_f$. Here again, other choices that fulfill the boundary conditions are possible and a more suitable variant turns out to be

$$\alpha^{(2)}(t) = \frac{1}{2k_B T} (\kappa_i + (\kappa_f - \kappa_i)(1716s^7 - 9009s^8 + 20020s^9 - 24024s^{10} + 16380s^{11} - 6006s^{12} + 924s^{13})). \quad (18)$$

Indeed, the protocol stemming from $\alpha^{(1)}$ is less accurate than its $\alpha^{(2)}$ -counterpart: the reason for this lies in the smoothness of $\alpha^{(2)}(t)$, rather than $\alpha^{(1)}(t)$, at initial and final times, as illustrated in figure 3: the first 6 derivatives of $\alpha^{(2)}$ do vanish at initial and final times, as compared to the first derivative only with $\alpha^{(1)}$. This smoothness leads to a soft running of concentration before t_f . Then, the ions are offered more time to reach the uniform state before finishing the process, reducing the ion diffusion effect on the colloid's final state. This results in an enhanced stability of the whole scheme, as explicit comparison for both protocols reveals, see appendix C for the $\alpha^{(1)}$ calculation.

Besides, it proves convenient to take

$$\kappa_{\text{op}}(t) = 2k_B T \alpha(t), \quad (19)$$

so that from equation (5) we get

$$\kappa_{\text{DP}} = \frac{\dot{\alpha}(t)}{2\alpha(t)} \gamma. \quad (20)$$

Note that this choice, in the quasi-static limit where $t_f \rightarrow \infty$, would lead to a consistent adiabatic protocol without the need of any diffusiophoretic assistance [59]. In doing so, it can be considered that the non adiabatic contribution to the protocol is entirely borne by the diffusiophoretic drive. Then, from equation (14), $C(0, t)$ takes the simple form

$$C(0, t) = C_0 \left(\frac{\alpha}{\alpha_i} \right)^{-\frac{nD_s}{2\Gamma_c}}. \quad (21)$$

Equation (5) subsequently indicates how the total stiffness κ acting on the colloid should depend on time. This stiffness has an optical and a diffusiophoretic contribution. The optical contribution has been chosen to be $\kappa_{\text{op}} = 2k_B T \alpha(t)$, and it remains to be seen what is the corresponding time dependence for κ_{DP} . This is a rather subtle task, since what is supposed to be experimentally controllable is the salt concentration $\phi(t)$ at the boundary of the domain (assumed for simplicity to be position independent while in principle, it could also exhibit a non trivial spatial dependence). Thus, we look for the $\phi(t)$ that would guarantee that U_{DP} in equation (7) is quadratic close to the origin, and with curvature $\kappa_{\text{DP}}(t)$.

Before calculating $\phi(t)$, we discuss how fast the procedure can be run. As pointed out in section 4, enforcing the rule given in equation (5) may give rise to negative concentrations, particularly so when high speed is sought: this sets a lower bound for the operating time t_f when a given compression ratio κ_i/κ_f is targeted or conversely, an upper bound for κ_i/κ_f when t_f is fixed. These bounds are shown in figures 4(a) and (b). The green regions show the accessible final states where $\phi(t)$ is positive during the whole process $0 \leq t \leq t_f$. However, the states in the white regions are not achievable since they lead to a negative $\phi(t)$ at least once in the time interval $0 \leq t \leq t_f$.

Figure 4 illustrates that the extent of the allowed region is smaller for negative phoretic mobility than for positive ones. This is related to the sketch of figure 2, and the fact that when $\Gamma_c < 0$, the smaller salt concentrations should be in the vicinity of the domain's boundary (inwards salt gradient), see also panels (c) and (d) in figure 4. The state point for panel (c) lies on the boundary of the allowed region in panel (a) (see the black dot in panel (a)) and it is indeed observed that the time profile $\phi(t)$ in panel (c) does vanish for $t \simeq t_f/2$. The gist of our proposal is to compute the driving $\phi(t)$ to be imposed as the salt concentration at the boundary. Figures 4(c) and (d) provide us with an explicit answer for the illustrative parameters

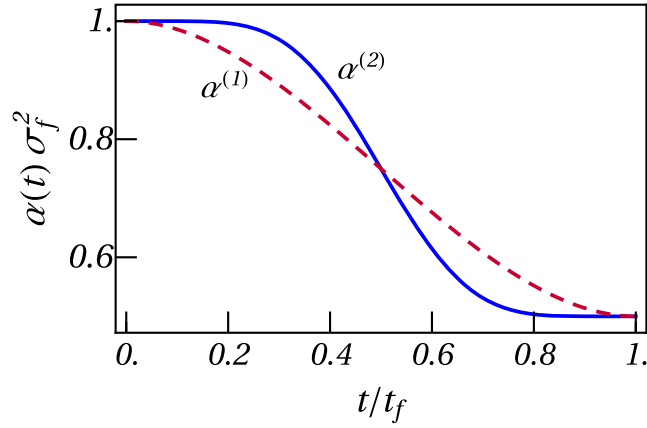


Figure 3. Two different choices for $\alpha(t)$: $\alpha^{(1)}$ (in red) and $\alpha^{(2)}$ (in blue) as a function of rescaled time t/t_f . The difference in their behavior for small $t \simeq 0$ and for $t \simeq t_f$ is at the root of their distinct efficiency.

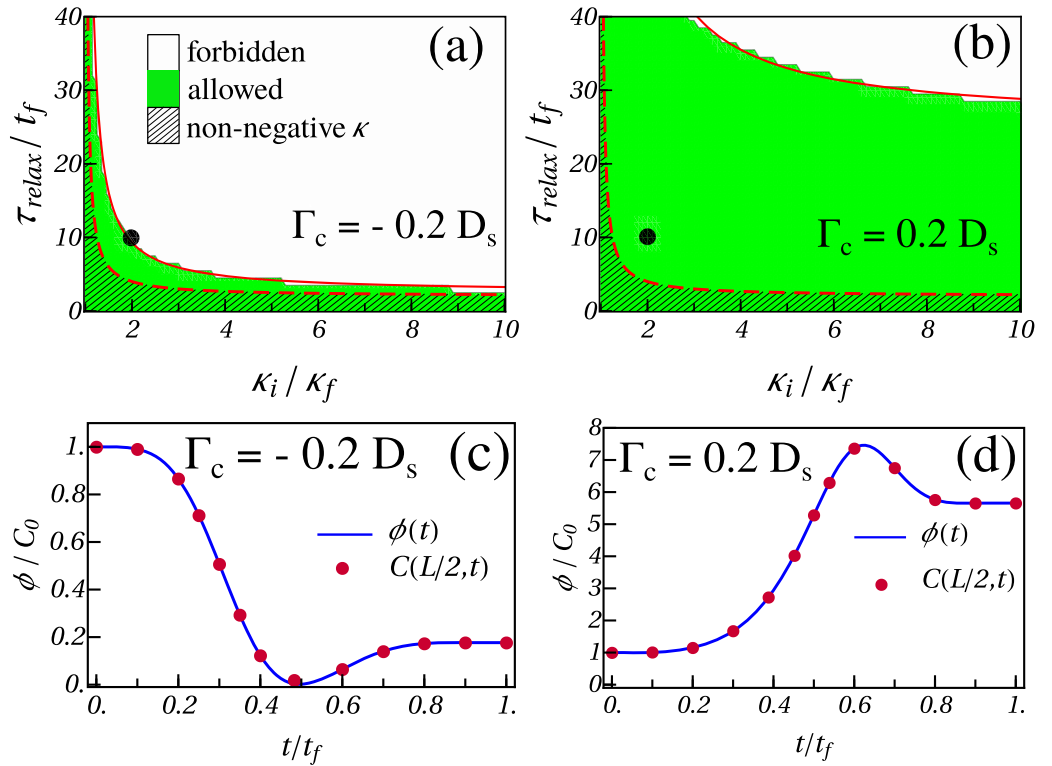


Figure 4. (a)–(b) Phase plots of accessible state points as a function of $\mathcal{A} = \tau_{\text{relax}}/t_f$ and the compression factor κ_i/κ_f , for different phoretic mobilities: (a) $\Gamma_c = -0.2 D_s$ and (b) $\Gamma_c = 0.2 D_s$. The green regions show the values for which $\phi(t)$ is positive at all times, i.e. in the whole interval $[0, t_f]$. Conversely, the white regions are for those states for which $\phi(t)$ takes a negative value at least once during the protocol. The plots correspond to $\alpha(t) = \alpha^{(2)}(t)$ given by equation (18) and optical stiffness $\kappa_{\text{op}} = 2k_B T \alpha(t)$, with $D_c = 0.002 D_s$, and a system size $L = 10 \sigma_f$. The hatched region shows the accessible states obtained by any ESE protocol with an ever non-negative stiffness (given by equation (34) in reference [24]). The non-hatched green region therefore helps visualizing the benefits of the present protocols over that of reference [24]. Panels (c)–(d) show the boundary salt concentration $\phi(t)$ for the same parameters, for $\mathcal{A} = 10$ and $\kappa_i/\kappa_f = 2$, corresponding to the black dots in panels (a)–(b). This quantity is supposed to be externally controlled in the experiment; it is our driving field. The blue curves show the results obtained by the Taylor expansion (15), truncated at order $j = 16$: it is the computed driving concentration to be applied to the system's boundary. On the other hand, the red dots show the concentration obtained by numerically solving the diffusion equation (8), i.e., $C(\pm L/2, t)$, imposing $\phi(t)$ at the boundary. The two functions should coincide, as they do here; a lack of accuracy or self-consistency in the treatment would lead to a mismatch.

chosen. When $C(0, t)$ is known, we use the truncated expansion (15) to get the boundary concentration $\phi(t)$; truncation at order $j = 16$ leads to an acceptable balance between accuracy and simplicity.

In figure 4, the separatrix between the green (allowed) and white (forbidden) regions determines a ‘speed limit’ of the expansion [18]. This separatrix is the locus of state points for which the required salt concentration $\phi(t)$ becomes negative. The fact that $\phi(t_0) = 0$ for some time t_0 (with $0 \leq t_0 \leq t_f$) gives a

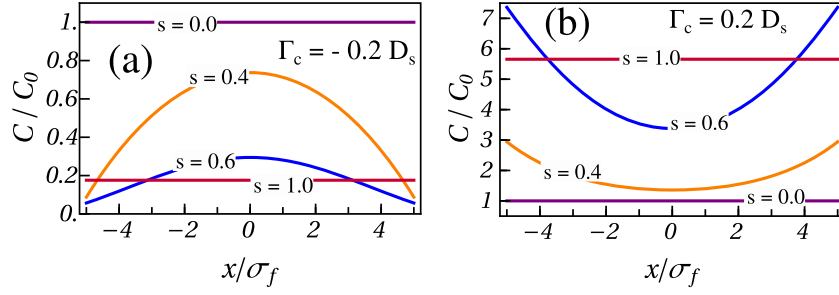


Figure 5. Salt concentration $C(x, t)$ at different scaled times $s = t/t_f$ for two phoretic mobilities (a) $\Gamma_c = -0.2 D_s$ (b) $\Gamma_c = 0.2 D_s$ obtained by solving the diffusion equation (8) numerically, imposing the boundary condition given by $\phi(t)$ in figures 4(c)–(d).

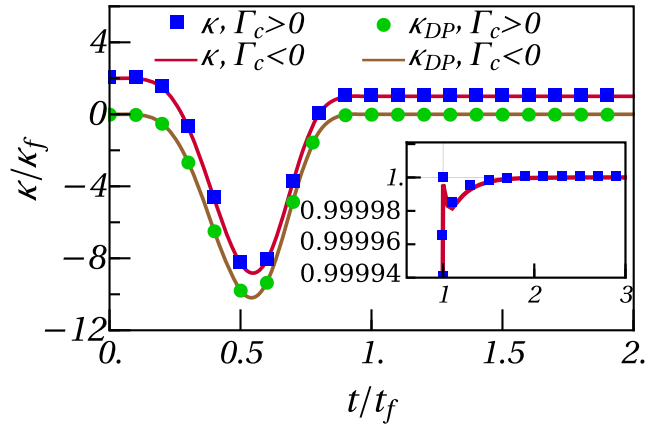


Figure 6. Total (κ) and diffusiophoretic (κ_{DP}) stiffnesses calculated by solving the diffusion equation (8) numerically, as a function of time for the same parameters as figure 4, i.e. in particular $\Gamma_c = \pm 0.2 D_s$. The inset shows the total stiffness at late times. The deviation of κ from the target value after finishing the process is negligible.

relation between \mathcal{A} and κ_f , of the form

$$\mathcal{A} \simeq \frac{C_1}{\kappa_f - \kappa_i} + C_2, \quad (22)$$

where the constants C_1 and C_2 depend on the choice of α and the phoretic mobility. This yields the boundary line between the allowed and forbidden regions in figure 4.

5.2. Checking consistency

To assess the protocol quality, we need to check for the results consistency. Indeed, our calculation of the driving field $\phi(t)$ relies on an expansion that connects information pertaining to the system center to information at the boundary. We thus solve numerically the diffusion equation for salt density, and obtain the concentration profile $C(x, t)$, from which all quantities of interest follow, from colloidal forces to observables like the colloidal position variance σ^2 . A first stringent test is to verify that the target $\phi(t)$, worked out analytically, coincides with the salt concentration at the boundary, measured from the numerical solution $C(x, t)$. This self-consistency requirement is well obeyed in figures 4(c) and (d), see the blue curves and red dots. This confirms that the proposed $\alpha(t)$ is a suitable choice, and that we have retained enough terms in the Taylor truncation. Appendix C indicates that self-consistency is somewhat better when working with $\alpha^{(2)}$ rather than $\alpha^{(1)}$.

The numerically computed ions concentration profiles $C(x, t)$ are shown in figure 5. The concentration is uniform at the initial and final times. In between, a gradient sets in, which is toward the center for $\Gamma_c < 0$ and away from the center for $\Gamma_c > 0$, as sketched in figure 2.

The next results that should be bench-marked are the stiffnesses. The total stiffness $\kappa(t)$ and diffusiophoretic stiffness κ_{DP} are shown in figure 6. Starting from $\kappa_i = 2\kappa_f$, we see that $\kappa(t)$ reaches its target value κ_f at $t = t_f$. It is observed that $\kappa_{DP}(t)$, which originates from the salt concentration gradient, vanishes at the initial and final times because of the uniform salt distribution. In between, it is negative as embodied in equation (12). Besides, by construction, we have chosen that not only κ but also κ_{op} be directly related to $\alpha(t)$. Hence, the dynamics of κ and κ_{op} are independent of the sign of the mobility Γ_c , as

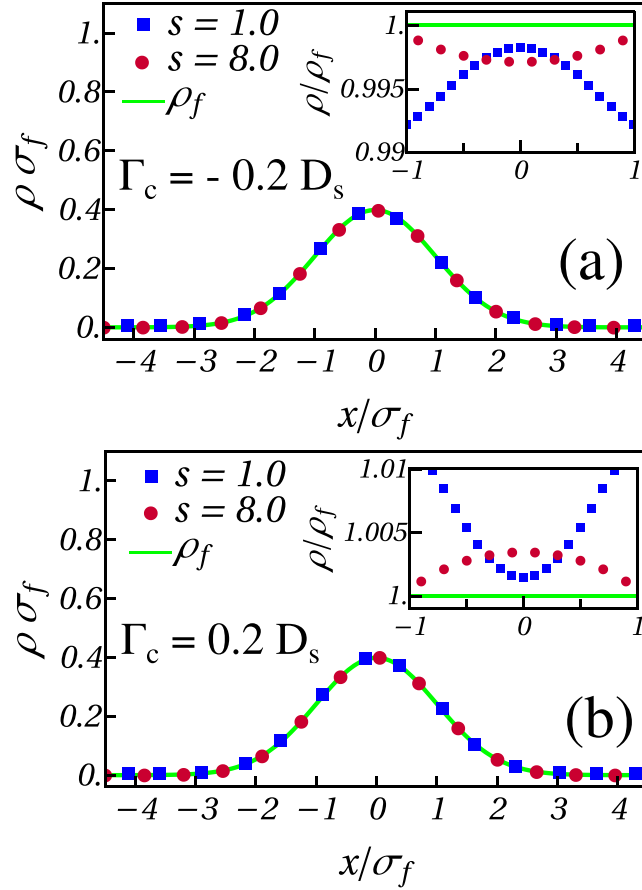


Figure 7. Colloidal density $\rho_{\text{num}}(x, t)$ calculated by solving the Fokker–Planck equation (2) numerically, at the final time $s = t/t_f = 1.0$ and some time after finishing the process ($s = 8.0$). The driving potential acting on the colloids, U in equation (10), is obtained from solving numerically the salt diffusion equation (8). The achieved density overlaps with the target Gaussian distribution $\rho_f = \rho(x, t_f)$ given by equation (3). At other times, the agreement is equally good. The panels correspond to (a) $\Gamma_c = -0.2D_s$ and (b) $\Gamma_c = 0.2D_s$ for $\alpha(t)$ and $\kappa_{\text{op}}(t)$ given by equations (18) and (19), respectively. The insets represent ρ_{num}/ρ_f near the system center. The densities differ slightly from the target Gaussian distribution.

illustrated in figure 6. Note from figure 5 that the required salt profiles, however, do strongly depend on the sign of Γ_c . Furthermore, a more careful inspection with the inset plots reveals that at $t = t_f$, the salt profile is not completely steady, which results in an undesired force on the colloids: as discussed in section 4, the diffusion of ions for $t > t_f$ leads to a small error for the measured stiffness, compared to the target value. A perfect protocol would lead to $\kappa = \kappa_f$ exactly at t_f , without any variation afterward. Note however the smallness of the mismatch (distance to unity) in the inset of figure 6.

Having the concentration profile $C(x, t)$, one can solve numerically the Fokker–Planck equation, equation (2), to obtain the colloid density $\rho_{\text{num}}(x, t)$ in order to compare with the target Gaussian distribution $\rho(x, t)$ given by equation (3). The results for the density in different times are demonstrated in figure 7. The density keeps its Gaussian distribution during the whole process. One observes that at the time t_f , and also after that $t > t_f$, the density differs slightly from the final target distribution. However, as the inset figures show, the differences are small.

Finally, we compute the variance of the colloid $\sigma^2(t)$ to ensure that the target expansion is realized. Recall that $\sigma^2 = (2\alpha)^{-1}$ and that the time dependence of α is chosen from the outset. The variance is plotted as a function of time for two phoretic mobilities $\Gamma_c = \pm 0.2D_s$ in figure 8. The variance calculated numerically, i.e. $\int_{-\infty}^{+\infty} x^2 \rho_{\text{num}}(x, t) dx$, is close to the target one $1/(2\alpha(t))$; however, it deviates slightly at long times. This is due to the density mismatch displayed in figure 7. It turns out that σ^2 is a rather sensitive probe for the small imperfections of the protocol, and reveals possible defects more clearly than the quantities reported hitherto. At any rate, the difference between the desired and observed variances is less than a few percents at all time, and stabilizes to a value very close to the target variance. Besides, although we have assumed that the colloid density is Gaussian, the non-quadratic terms in the potential, equation (7), may cause non-Gaussianity. In appendix D, we quantify the deviations from Gaussian distribution and show that they also remain relatively small.

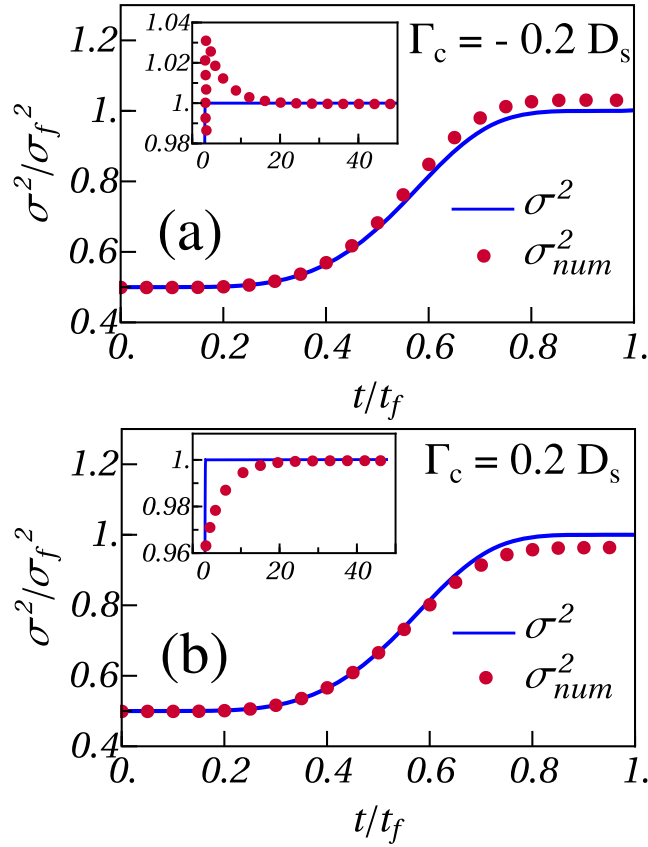


Figure 8. Variance σ^2 of the colloidal position distribution as a function of time. (a) $\Gamma_c = -0.2D_s$ and (b) $\Gamma_c = 0.2D_s$ for $\alpha(t) = \alpha^{(2)}(t)$ and $\kappa_{op}(t)$ given by equations (18) and (19). Same parameters as in figure 4. The variance calculated numerically, i.e., $\sigma_{num}^2 = \int x^2 \rho_{num}(x, t) dx$, is compared to the target $1/[2\alpha(t)]$; it deviates slightly from this quantity, with a maximum mismatch at t_f . The insets show the evolution for longer times, where a stable value is reached for $t \simeq 20t_f$, which shows a good match with the target value.

6. Manipulating non-Gaussian states

The previous sections were devoted to shortcutting the dynamics of a system confined in a harmonic potential, enforcing the colloidal state to be Gaussian at all times. We address here the situation of a non-harmonic confinement. A non-Gaussian distribution ensues. We will consider a quartic confining potential $U_{eq} \sim x^4$ as an illustrative example. At equilibrium, we then have $\rho_{eq} \sim \exp(-\alpha x^4)$. The aim is to drive the system from an initial state with variance σ_i to a final state with increased variance σ_f , in a processing time t_f , arbitrarily shorter than the relaxation time $\tau_{relax} = \sigma_f^2/D_c$. To this end, we again follow the ESE procedure, as done in section 4 and in reference [19], working out in one dimension, and driving the system through the control of salt concentration $\phi(t)$ at its boundaries ($x = \pm L/2$). We will demand that the colloid density be at all times of the form

$$\rho(x, t) = \frac{\alpha^{1/4}(t)}{2\Gamma(5/4)} \exp(-\alpha(t)x^4), \quad (23)$$

where $\Gamma(x)$ represents the Gamma function and the prefactor of the exponential stems from normalization. Variance and kurtosis of the distribution are

$$\sigma^2 = \frac{N}{2\sqrt{\alpha}}, \quad \mathcal{K} = \frac{\langle (x - \langle x \rangle)^4 \rangle}{\sigma^4} - 3 \simeq 0.812, \quad (24)$$

where $N = \Gamma(3/4)/(2\Gamma(5/4)) \simeq 0.678$ is a numerical factor. It can be shown that a suitable driving potential reads

$$U(x, t) = \frac{1}{4}\delta(t)x^4 + \frac{1}{2}\kappa(t)x^2. \quad (25)$$

This potential should match the equilibrium potential in x^4 at the initial and final times, so that $\kappa(0) = \kappa(t_f) = 0$. The function $\alpha(t)$ can be chosen arbitrarily, fulfilling the equilibrium boundary conditions $\alpha(0) = \delta_i/(4k_B T)$ and $\alpha(t_f) = \delta_f/(4k_B T)$. Thus, the variance of final state is

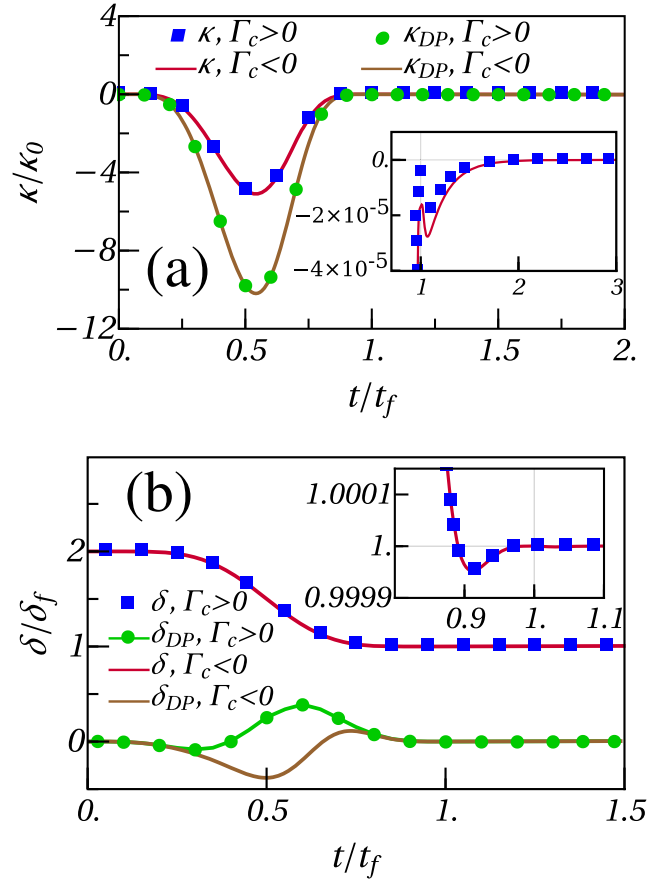


Figure 9. Non-Gaussian situation. Total and diffusiophoretic stiffnesses, calculated by solving the diffusion equation (8) numerically, as a function of rescaled time t/t_f , for the same parameters as in figure 4 and two phoretic mobilities $\Gamma_c = \pm 0.2D_s$ where $\kappa_0 = Nk_B T/\sigma_f^2$ and $\delta_f = N^2 k_B T/\sigma_f^4$. Here, $\alpha = \alpha^{(2)}$ as given in equation (18), from which $\kappa_{op}(t)$, and δ_{op} follow, see equations (27) and (31). The insets show a close-up of the total stiffnesses near the final time. The deviation of κ and δ from the target value after finishing the process is negligible.

$\sigma_f^2 = N\sqrt{k_B T/\delta_f} \equiv N\sigma_0^2$. Moreover, We choose $\alpha(t)$ such that $\dot{\alpha}(0) = \dot{\alpha}(t_f) = 0$ to have a smooth function at initial and final times. As with the harmonic situation, we restrict to continuous functions.

Substituting $\rho(x, t)$ and $U(x, t)$ into the Fokker–Planck equation (2) gives:

$$\frac{\dot{\alpha}}{4\alpha} - \dot{\alpha}x^4 = D_c(-12\alpha x^2 + 16\alpha^2 x^6) + \frac{1}{\gamma}(3\delta x^2 + \kappa) - \frac{4\alpha}{\gamma}x^3(\delta x^3 + \kappa x), \quad (26)$$

which is valid for any position x . This implies that

$$\kappa(t) = \frac{\gamma}{4} \frac{\dot{\alpha}(t)}{\alpha(t)}, \quad \delta(t) = 4k_B T \alpha(t). \quad (27)$$

Therefore, choosing an appropriate function for $\alpha(t)$ also sets the stiffness coefficients $\kappa(t)$ and $\delta(t)$, through equation (27). A decompression results in having $\dot{\alpha} < 0$, so that $\kappa(t) < 0$. As a result, the potential U in (25) features a double-well form for $0 < t < t_f$ [19] (α and thus δ are positive).

The density of bathing salt solution $C(x, t)$ satisfies the diffusion equation (8) with boundary condition given by equation (9). Here again, the goal is to work out the salt concentration at the boundary of the system, that will lead to the diffusiophoretic force in the bulk that is precisely of the form leading to the desired dynamics in equation (23). As before, the colloidal dynamics is enslaved to that of salt through the Fokker–Planck equation, while salt itself obeys pure diffusion.

With both optical and diffusiophoretic potentials, the total potential acting on the colloid is:

$$U = U_{op} + U_{DP} = \frac{1}{2}\kappa_{op}x^2 + \frac{1}{4}\delta_{op}x^4 - \gamma\Gamma_c \ln C(x, t). \quad (28)$$

We thus consider here that the non linearity of the optical trapping leads to a quartic term in U , in addition to the standard harmonic term. While this is routine experimental practice for the quadratic term in κ_{op} , we also assume that the quartic term can be tuned over time by the operator, which may prove difficult. The present analysis, compared to that of the previous sections, is for this reason more speculative. In line with

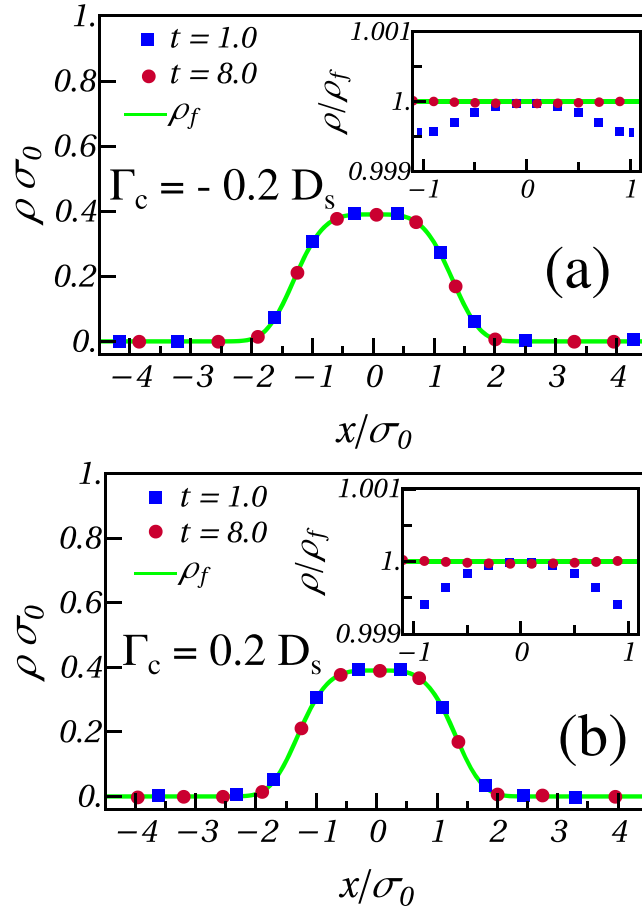


Figure 10. Non-Gaussian situation. Colloidal density $\rho_{\text{num}}(x, t)$ calculated by solving the Fokker–Planck equation (2) numerically, for different times. ρ_f shows the target non-Gaussian distribution given by equation (23). $\alpha^{(2)}(t)$, $\kappa_{\text{op}}(t)$, and $\delta_{\text{op}}(t)$ are given by equations (18), (33) and (35), respectively. The other parameters are the same as in figure 4. The insets represent the ratio ρ/ρ_f with a zoom onto the center $x = 0$. The densities very slightly differ from the target distribution.

the previous analysis, we take $\kappa_{\text{op}}(t)$ and $\delta_{\text{op}}(t)$ as non-negative for all times. In addition, we assume isotropy and analyticity, which implies here $\partial_x^j \ln C|_{x=0} = 0$ for j odd. Thus, the expansion near the system center gives

$$U \simeq \frac{1}{2} \left(\kappa_{\text{op}} - \gamma \Gamma_c \partial_x^2 \ln C(x, t)|_{x=0} \right) x^2 + \frac{1}{4} \left(\delta_{\text{op}} - \frac{1}{6} \gamma \Gamma_c \partial_x^4 \ln C(x, t)|_{x=0} \right) x^4. \quad (29)$$

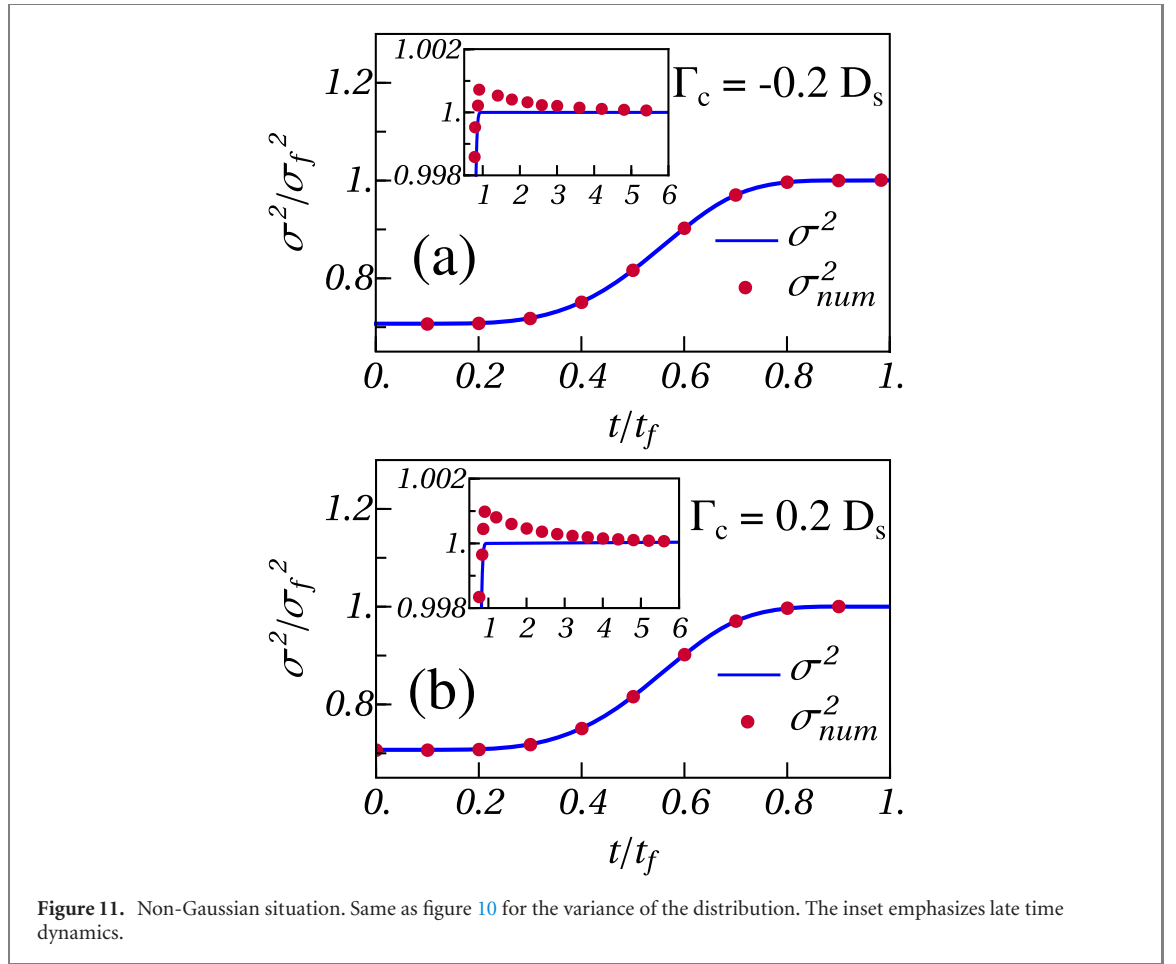
Then, the diffusiophoretic stiffnesses are

$$\begin{aligned} \kappa_{\text{DP}}(t) &\equiv \kappa(t) - \kappa_{\text{op}}(t) = -\gamma \Gamma_c \partial_x^2 \ln C(x, t)|_{x=0}, \\ \delta_{\text{DP}}(t) &\equiv \delta(t) - \delta_{\text{op}}(t) = -\frac{1}{6} \gamma \Gamma_c \partial_x^4 \ln C(x, t)|_{x=0}. \end{aligned} \quad (30)$$

Combining with equation (27) and using the diffusion equation (8), we obtain the following differential equation for the density at the center $C(0, t)$ and optical stiffnesses:

$$\begin{aligned} \kappa_{\text{DP}}(t) &= -\frac{\gamma \Gamma_c}{D_s} \partial_t \ln C(0, t) \\ &= -\kappa_{\text{op}}(t) + \frac{\gamma}{4} \frac{\dot{\alpha}(t)}{\alpha(t)} \\ \delta_{\text{DP}}(t) &= -\frac{1}{6} \frac{\gamma \Gamma_c}{D_s^2} \left(-2(\partial_t \ln C(0, t))^2 + \partial_t^2 \ln C(0, t) \right) \\ &= -\delta_{\text{op}}(t) + 4k_B T \alpha(t). \end{aligned} \quad (31)$$

For a chosen $\alpha(t)$, the total quadratic and quartic stiffnesses κ and δ are known, while κ_{op} or δ_{op} are arbitrary function with no other constraint than being positive.



Obtaining $\phi(t)$: as in the Gaussian case, there is some flexibility in the choice of $\alpha(t)$ and also $\kappa_{\text{op}}(t)$. We take a similar function for $\alpha(t)$ as equation (18), that led previously to accurate protocols:

$$\alpha^{(2)}(t) = \frac{1}{4k_B T} \left(\delta_i + (\delta_f - \delta_i)(1716s^7 - 9009s^8 + 20020s^9 - 24024s^{10} + 16380s^{11} - 6006s^{12} + 924s^{13}) \right), \quad (32)$$

with again $s = t/t_f$. As far as $\kappa_{\text{op}}(t)$ is concerned, it proves convenient to choose

$$\kappa_{\text{op}}(t) = -\kappa(t) = -\frac{\gamma}{4} \frac{\dot{\alpha}(t)}{\alpha(t)}. \quad (33)$$

Solving equation (31) for $C(0, t)$ and $\delta_{\text{op}}(t)$ yields

$$C(0, t) = C_0 \left(\frac{\alpha(t)}{\alpha_i} \right)^{-\frac{D_s}{2\Gamma_c}} \quad (34)$$

and

$$\delta_{\text{op}}(t) = 4k_B T \alpha(t) + \frac{1}{12} \frac{k_B T}{D_c D_s} \left(\frac{\dot{\alpha}^2}{\alpha^2} \left(1 - \frac{D_s}{\Gamma_c} \right) - \frac{\ddot{\alpha}}{\alpha} \right). \quad (35)$$

An interesting feature of the choice made for κ_{op} is that we recover the exact same dynamics for $C(0, t)$ as for Gaussian states, and thus we can recycle the corresponding results, in particular the computation of $\phi(t)$. Indeed, our choice results in

$$\kappa_{\text{DP}} = \frac{\gamma}{2} \frac{\dot{\alpha}}{\alpha}, \quad (36)$$

which coincides with equation (20). As a consequence, the phase diagram in figure 4, together with the underlying salt density profile shown in figure 5 also apply here, at the expense of replacing the compression factor κ_i/κ_f by δ_i/δ_f . Differences between the harmonic and non-harmonic confinement will nevertheless appear when considering the force applied onto the colloids, as discussed next.

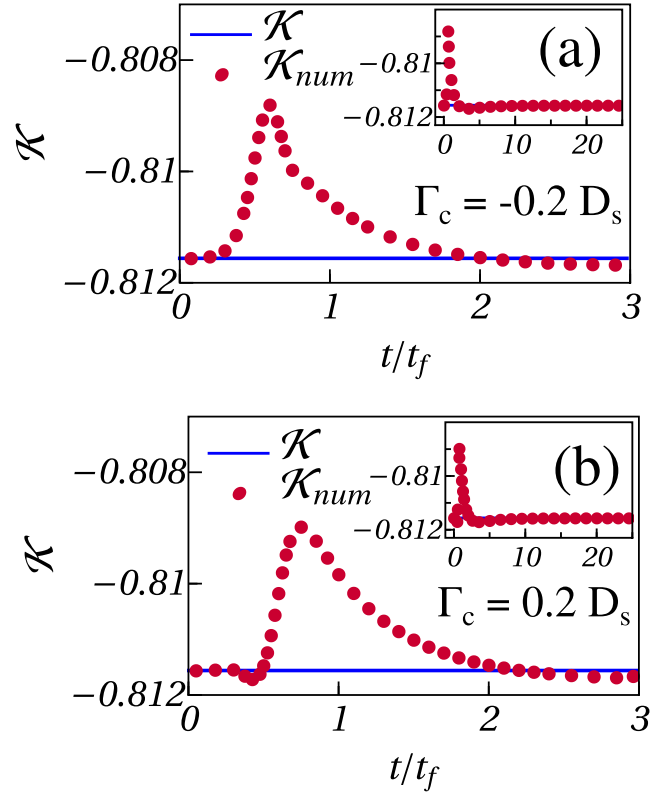


Figure 12. Same as figure 11, for the reduced fourth moment of the distribution (kurtosis).

Note that we have assumed that δ_{op} should remain positive. This is not guaranteed, since the second term on the rhs of equation (35) may take negative values. Yet, remembering that α is of order σ_f^{-4} , the ratio of the second and the first terms in equation (35) is of order $\mathcal{A}^2 D_c / (10 D_s)$. As in typical electrolytes $D_c / D_s \sim 10^{-3}$, we conclude that $\delta_{\text{op}} > 0$ as long as $\mathcal{A} < 100$, or more generally $\mathcal{A} < \sqrt{10 D_s / D_c}$. It should also be emphasized that as compared to the Gaussian situation, the present method is experimentally more challenging from the optical trapping point of view, since not only the quadratic stiffness κ_{op} needs to be controlled in time, but also the quartic stiffness δ_{op} .

The stiffnesses $\kappa(t)$ and $\delta(t)$ are displayed in figures 9(a) and (b). Once the diffusion equation has been solved for salt dynamics, the stiffnesses follow from equations (27) and (31). We can see that as required, not only κ but also κ_{DP} (and thus $\kappa_{\text{op}} = \kappa - \kappa_{\text{DP}}$), do vanish both at $t = 0$ and $t = t_f$. Besides, κ_{DP} and κ are negative in the meantime, in order to generate the double-well form for the potential, required to achieve the decompression. This is purely driven by diffusio-phoresis. The fact that $\kappa > \kappa_{\text{DP}}$ signals that the optical contribution κ_{op} is always positive. As with harmonic confinement, we have chosen the time dependence of κ and κ_{op} to be blind to the sign of mobility Γ_c . Thus, κ_{DP} inherits from this property. A similar comment holds for $\delta(t)$, while one needs to study δ_{DP} to discriminate positive from negative Γ_c . The inset of figure 9 indicates that a residual late time dynamics slightly impinges on the forcing, and causes a small error in the measured stiffnesses.

We next turn to the colloid density $\rho_{\text{num}}(x, t)$, obtained from solving numerically the Fokker–Planck equation, equation (2). The results are shown in figure 10. The density keeps the desired non-Gaussian distribution during the whole process, as shown at t_f in the figure. The agreement with the prescribed density is striking and outperforms what had been obtained at Gaussian level. For times exceeding t_f , a slight drift exists, as displayed in the inset. To ensure that the target expansion is realized, we calculate numerically the variance of the colloid $\sigma_{\text{num}}^2(t) = \int_{-\infty}^{+\infty} x^2 \rho_{\text{num}}(x, t) dx$. The results are plotted as a function of time for two phoretic mobilities $\Gamma_c = \pm 0.2 D_s$ in figure 11. The variance measured from the solution to the Fokker–Planck equation appears very close to the target variance, $\sigma^2 = N / (2\sqrt{\alpha(t)})$. The maximum error is observed for $t \simeq t_f$ and is below 0.1%; the mismatch then quickly decays to zero (see the inset). The plots confirm that we achieve a better control of non-Gaussian states than Gaussian ones (comparing figures 8 and 11). The reason is that the present treatment accounts for non harmonic contributions to the potential U , while they are discarded in sections 4 and 5. Finally, for completeness, we evaluate the kurtosis of the colloidal density. This quantity, as shown in figure 12, is nearly constant and changes in time only on

a small scale, such that $\mathcal{K} \simeq 0.8116 \pm 0.003$. This confirms that the colloidal density follows the target self-similar non-Gaussian distribution given by equation (23) during the whole procedure. The kurtosis remains negative for both positive and negative mobilities, as a consequence of light tails in the target distribution equation (23).

7. Discussion and conclusion

By a proper combination of optical and phoretic forcing, we have proposed a family of decompression protocols for manipulating colloidal states. These protocols are free of the difficulty that plagues an all optical method, namely the fact that the trap stiffness in the vicinity of its center should become negative, expelling colloids away, in some time window. Here, diffusiophoretic forces provide the required deconfining forces; they are monitored by an externally controlled salt buffer, that bathes the exterior of the system under study. In other words, we drive our system by the (time-dependent) salt concentration $\phi(t)$ at its boundary, as it was realized in a series of experimental works [51, 53]. It is a rather non trivial result that in doing so, we achieve the precise bulk forcing that is required to manipulate the colloidal states under consideration. We take advantage here of the fast salt dynamics, as compared to the slower colloidal response time. This is the key factor that allows to decompress the colloidal state with a significant speedup compared to the intrinsic response time. We have shown that the method is operational not only in a situation of harmonic confinement, but also in a quartic external potential.

After having derived analytically the appropriate drive $\phi(t)$, we have put our predictions to the test by first solving exactly the salt diffusion equation from which the diffusiophoretic forces acting on the colloids follow; these forces are then injected into the Fokker–Planck equation ruling colloidal dynamics. An excellent agreement has been reported between the resulting colloidal densities, and their (time-dependent) targets. For times larger than the protocol duration t_f , all evolution has to stop, and it is indeed what has been observed. The colloidal dynamics is enslaved to that of salt through the Fokker–Planck equation, while salt itself obeys pure diffusion, irrespective of colloidal arrangement. This is an approximation, valid for a low colloidal density. This is implicitly the limit in which we worked, since all colloid–colloid interactions are neglected. An experimental way to achieve such a limit is to work with a single colloid, and gather statistics by repeating the experiment [19, 21]. A simpler alternative deals with trapping a colloidal suspension, under the proviso that the maximal volume fraction remain small.

For simplicity, we have presented results in a one dimensional system, but we checked that the idea is also operational in two dimensions. Besides, it is possible to use a similar method to compress the system rather than decompress it. While the added value of our protocol for compression is less clear, since an all optical device may achieve a similar result in a possibly more direct way, the present idea may nevertheless be used for speeding compression if the laser intensity cannot exceed some threshold, to avoid damaging the colloids.

The present contribution enriches the field of shortcut to adiabaticity protocols [2], extending its underpinnings to non optical forcings in soft matter systems. While our method is approximate, we have tested its accuracy. We also charted out the configurations (state points) that are amenable to our treatment, and accessible for a given acceleration factor \mathcal{A} . Indeed, some state points (outside the green region in figure 4) are not accessible. Yet, we did not attempt at optimality in any sense. There is thus room for improved protocols, that would for instance enlarge the accessible region in parameter space (green regions in figure 4), or aim at the best stability with respect to a slight miscalibration of initial parameters, through a careful choice of the—to a large extent arbitrary—function $\alpha(t)$.

An open question deals with the possible challenges raised by the experimental realization of the present ideas, starting with the precise time control of the driving concentration $\phi(t)$, that may require some experimental ingenuity. Besides, the question of optical stiffness anisotropy should be addressed. For instance, we have assumed the coincidence of the point of zero optical force, with that of zero diffusiophoretic force. We briefly discuss in appendix E the consequences of a misalignment, with the conclusion that the protocols is relatively immune to a modest mismatch between the centers of optical and diffusiophoretic drivings. Besides, one needs to change the salt concentration in a buffer that bathes from the exterior the system of interest, without perturbing hydrodynamically the colloidal bead's motion. When addressing these questions, one should keep in mind that we made no attempt at optimizing the proposed protocols, which leaves a useful flexibility that can be taken advantage of to minimize a given artifact. Optimization here also amounts to selecting the relevant time-dependence for the positional variance, through the choice of the function $\alpha(t)$. We also note that *a priori* detrimental salt concentration fluctuations, that could affect the diffusiophoretic drive, can be reduced by increasing the initial uniform salt concentration C_0 . It is also quite clear that implementing the non-Gaussian protocol leads to enhanced difficulties, since the joint control of quadratic and quartic stiffness coefficients is required.

Data availability statement

No new data were created or analysed in this study.

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Appendix A. Fast expansion beyond ESE

As discussed in section 3, fast decompression is possible making use of diffusiophoretic forces in an otherwise time-independent optical potential, i.e., $\kappa_{\text{op}} = \kappa_i$. To obtain a non vanishing diffusiophoretic contribution to the confining stiffness, we need $\phi(t \geq 0) = 0$. Without loss of generality, we consider again a one-dimensional system. The analytical solution of the diffusion equation (8) in 1D for a system confined in $-L/2 \leq x \leq L/2$ reads

$$\begin{aligned} C(x, t) &= \frac{4}{L} \sum_{n=0}^{\infty} (-1)^n \cos(\lambda_n x) e^{-\lambda_n^2 D_s t} \left(\frac{1}{\lambda_n} + \lambda_n D_s \int_0^t e^{\lambda_n^2 D_s \omega} \phi(\omega) d\omega \right) \\ &= \phi(t) - \frac{4}{L} \sum_{m=1}^{\infty} (-1)^m \frac{1}{D_s^m} \sum_{n=0}^{\infty} \frac{(-1)^n}{\lambda_n^{2m+1}} \cos(\lambda_n x) \left(\frac{d^m \phi(t)}{dt^m} + e^{-\lambda_n^2 D_s t} \frac{d^m \phi(t)}{dt^m} \Big|_{t=0} \right), \end{aligned} \quad (\text{A.1})$$

where $\lambda_n = (2n+1)\pi/L$ and $\phi(t) = C(\pm L/2, t)$ is the boundary concentration, playing the role of the driving field in our analysis and monitored by the experimentalists. Considering $\phi(t \geq 0) = 0$, we have for $t \gg \tau_s \equiv L^2/D_s$, $C(x, t) \propto \cos(\pi x/L) \exp(-\pi^2 D_s t/L^2)$ which features factorized x and t dependencies. The diffusiophoretic potential follows as

$$U_{\text{DP}}(x) = -\gamma \Gamma_c \log \left(\cos \left(\frac{\pi x}{L} \right) \right). \quad (\text{A.2})$$

Close enough to $x = 0$, this can be Taylor expanded, and U_{DP} therefore emulates harmonic trapping with stiffness

$$\kappa_{\text{DP}} = \gamma \Gamma_c \frac{\pi^2}{L^2}, \quad (\text{A.3})$$

which only depends on the system size L . The time-scale required to reach such a state is τ_s .

The potential energy close to the trap center, with optical and diffusiophoretic terms, is then approximated by $U \simeq \frac{1}{2}(\kappa_{\text{op}} + \gamma \Gamma_c \pi^2/L^2)x^2$. The density $\rho(x, t)$ evolves according to the Fokker-Planck equation (2) which for the one dimensional system, at long times $t \gg \tau_s$, and near the center $|x| \ll L$, reduces to

$$\partial_t \rho(\mathbf{r}, t) \simeq \partial_x \left(D_c \partial_x \rho(x, t) + \left(\frac{\kappa_{\text{op}}}{\gamma} + \Gamma_c \frac{\pi^2}{L^2} \right) x \rho(x, t) \right). \quad (\text{A.4})$$

With an initial Gaussian density $\rho(x, 0)$ (variance σ_i^2), $\rho(x, t)$ remains Gaussian in a harmonic trap, and as long as $|x| \ll L$ the solution reads

$$\rho(x, t) = \sqrt{\frac{g(t)}{2\pi\sigma_f^2}} e^{-g(t) \frac{x^2}{2\sigma_f^2}}, \quad (\text{A.5})$$

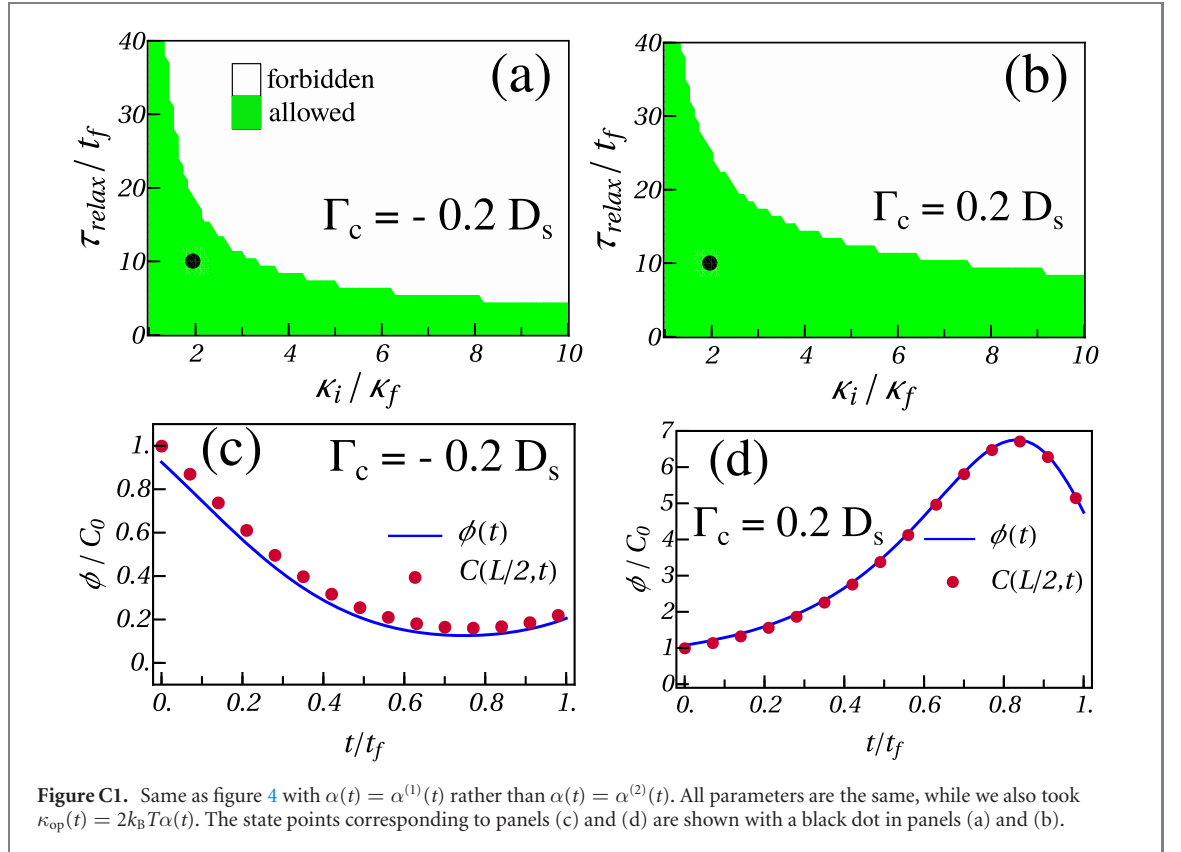
where $g(t) = \kappa_i / \left((\kappa_i + (\kappa_f - \kappa_i)e^{-2D_s t/\sigma_i^2}) \right)$. Particularly, we have

$$\rho(x, t \rightarrow \infty) = \sqrt{\frac{1}{2\pi\sigma_f^2}} e^{-\frac{x^2}{2\sigma_f^2}}, \quad (\text{A.6})$$

with $\sigma_f^2 = k_B T / \kappa_f$. Linearization of relation (A.2) is thus justified as long as $\sigma_f \ll L$.

We conclude here that we have realized a fast decompression, on a time scale τ_s , where nevertheless the final position variance is determined by L , the system size, through the effective stiffness

$$\kappa_f = \kappa_{\text{op}} + \kappa_{\text{DP}} = \kappa_{\text{op}} + \gamma \Gamma_c \frac{\pi^2}{L^2}. \quad (\text{A.7})$$



Moreover, we can see that κ_{DP} takes negative values only for $\Gamma_c < 0$, which would not allow to decompress a system with $\Gamma_c > 0$. Therefore, the present method lacks control and generality, and another route has been explored in the main text.

Appendix B. Constraint on $C(0, t)$

In this appendix, we present the calculations behind equation (13). The assumptions of isotropicity and analyticity of $C(\mathbf{r}, t)$ imply $\partial_r^j C|_{r=0} = 0$ for j odd, so that expansion of $C(\mathbf{r}, t)$ at $r = 0$ gives

$$C(\mathbf{r}, t) = C(0, t) + \partial_r^2 C(\mathbf{r}, t)|_{r=0} \frac{r^2}{2} + \partial_r^4 C(\mathbf{r}, t)|_{r=0} \frac{r^4}{4!} + \mathcal{O}(r^6), \quad (\text{B.1})$$

which reveals that $\frac{1}{r} \partial_r C|_{r=0} = \partial_r^2 C|_{r=0}$ (although $\partial_r C|_{r=0} = 0$). Then, for $\nabla^2 C$ in n -dimensional system at $r = 0$ we have

$$\begin{aligned} \nabla^2 C|_{r=0} &= \partial_r^2 C(\mathbf{r}, t)|_{r=0} + \frac{n-1}{r} \partial_r C(\mathbf{r}, t)|_{r=0} \\ &= n \partial_r^2 C(\mathbf{r}, t)|_{r=0}. \end{aligned} \quad (\text{B.2})$$

Substituting this relation into the diffusion equation (8) gives

$$\partial_t C(\mathbf{r}, t)|_{r=0} = D_s n \partial_r^2 C|_{r=0}. \quad (\text{B.3})$$

Thus, we have

$$\begin{aligned} \partial_r^2 \ln C(\mathbf{r}, t)|_{r=0} &= \frac{1}{C} \partial_r^2 C(\mathbf{r}, t) \Big|_{r=0} - \frac{1}{C^2} (\partial_r C(\mathbf{r}, t))^2 \Big|_{r=0} \\ &= \frac{1}{n D_s} \frac{1}{C} \partial_t C(\mathbf{r}, t)|_{r=0} = \frac{1}{n D_s} \partial_t \ln C(0, t). \end{aligned} \quad (\text{B.4})$$

Combining this equation with equations (12) and (5), we obtain a differential equation for the concentration at the center $C(0, t)$ as

$$-\frac{\gamma \Gamma_c}{n D_s} \partial_t \ln C(0, t) = \frac{\dot{\alpha}(t)}{2 \alpha(t)} \gamma + 2 k_B T \alpha(t) - \kappa_{op}(t). \quad (\text{B.5})$$

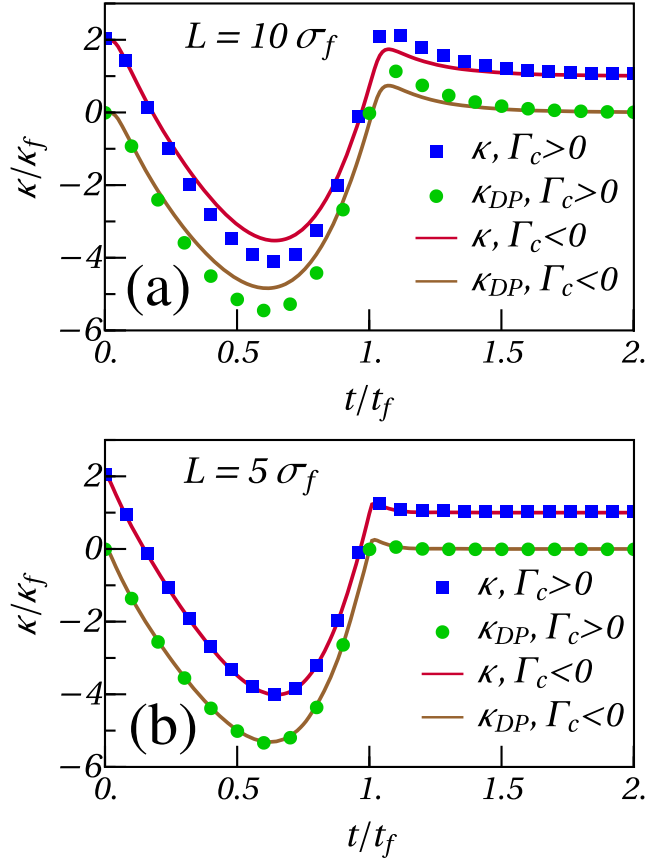


Figure C2. The stiffnesses $\kappa(t)$ and $\kappa_{DP}(t)$, as a function of time for parameters $\mathcal{A} = 10$, $\kappa_i = 2\kappa_f$, $D_c = 0.002D_s$, $\kappa_{op}(t) = 2k_B T \alpha^{(1)}(t)$ and for two phoretic mobilities: $\Gamma_c = \pm 0.2D_s$; here, $\alpha(t) = \alpha^{(1)}(t)$. The two graphs are for different sizes (a) $L = 10\sigma_f$ and (b) $L = 5\sigma_f$. Note the undesired time-dependence of the quantities displayed, for $t > t_f$. The data collapse in panel (b) is indicative of a better accuracy of the protocol, as confirmed by the near constancy of the stiffnesses for $t > t_f$.

Appendix C. Results for $\alpha^{(1)}(t)$

In the main text, we presented results for the situation where the inverse variance of colloidal positions, encoded in the function $\alpha(t)$, is given by $\alpha^{(2)}$ in equation (18). We work out here the method with another choice ($s = t/t_f$),

$$\alpha(t) = \alpha^{(1)}(t) = \frac{1}{2k_B T} (\kappa_i + (\kappa_f - \kappa_i)(3s^2 - 2s^3)), \quad (C.1)$$

that highlights the importance of the steadiness of α , essentially near the end point $t = t_f$. Here also, we suppose $\kappa_{op}(t) = 2k_B T \alpha(t)$. The resulting phase portrait, the counterpart of figure 4, is shown in figure C1.

Two comments are in order. First and for $\Gamma_c < 0$, the $\alpha^{(1)}$ protocol extends the reach of the method as compared to the $\alpha^{(2)}$ protocol. The reverse conclusion holds for $\Gamma_c > 0$. Second, and more relevant, the accuracy does not fare favorably to that shown in panel (c) of figure 4. With positive mobilities (panel (d)) we observe a better collapse of the analytically computed $\phi(t)$ shown with the line, and the symbols resulting from the numerical resolution of the salt diffusion equation. In panel (c), the lack of accuracy is in particular already visible at $t = 0$, where ϕ slightly departs from C_0 , the salt buffer concentration. This is a consequence of our Taylor expansion truncation, not only to get $\phi(t)$, but also the potential U , that may feature non-harmonic contributions. These shortcomings are made more apparent when looking at the dynamics of the resulting stiffness κ , or κ_{DP} as in figure C2. It is seen that for $t > t_f$, these quantities are not steady. Besides, the effect of the sign of the mobility Γ_c , at variance with the programmed solution (see figure 6), is another illustration of the imperfections at stake here. As expected and observed in comparing the two graphs in figure C2, the protocol works better when applied to a smaller system.

Appendix D. Non-Gaussian features of would-be Gaussian states

In sections 4 and 5, we have supposed that the colloid density remains Gaussian during the protocol. In this appendix, we question the validity of this assumption. To this end, we calculate numerically the kurtosis,

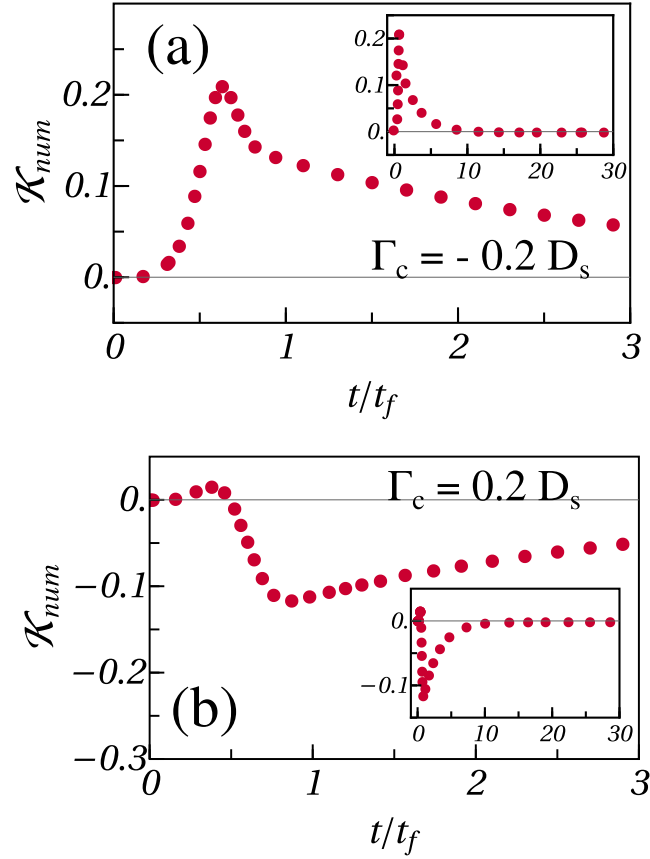


Figure D1. Kurtosis of the colloidal position distribution as a function of time for the target Gaussian states. The panels correspond to (a) $\Gamma_c = -0.2 D_s$ and (b) $\Gamma_c = 0.2 D_s$ for $\alpha(t)$ given by equation (18) and the same parameters as in figure 4. The insets show the dynamics for long times.

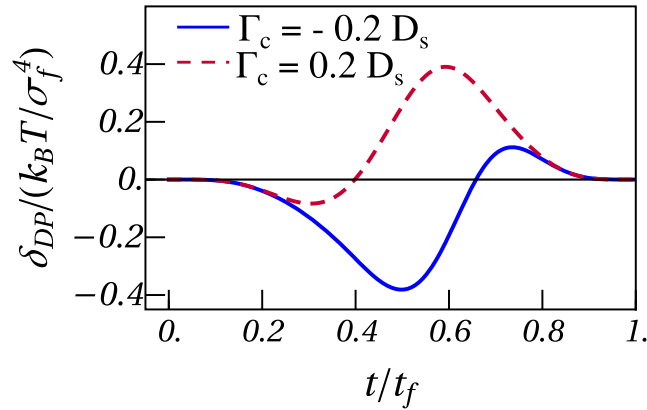
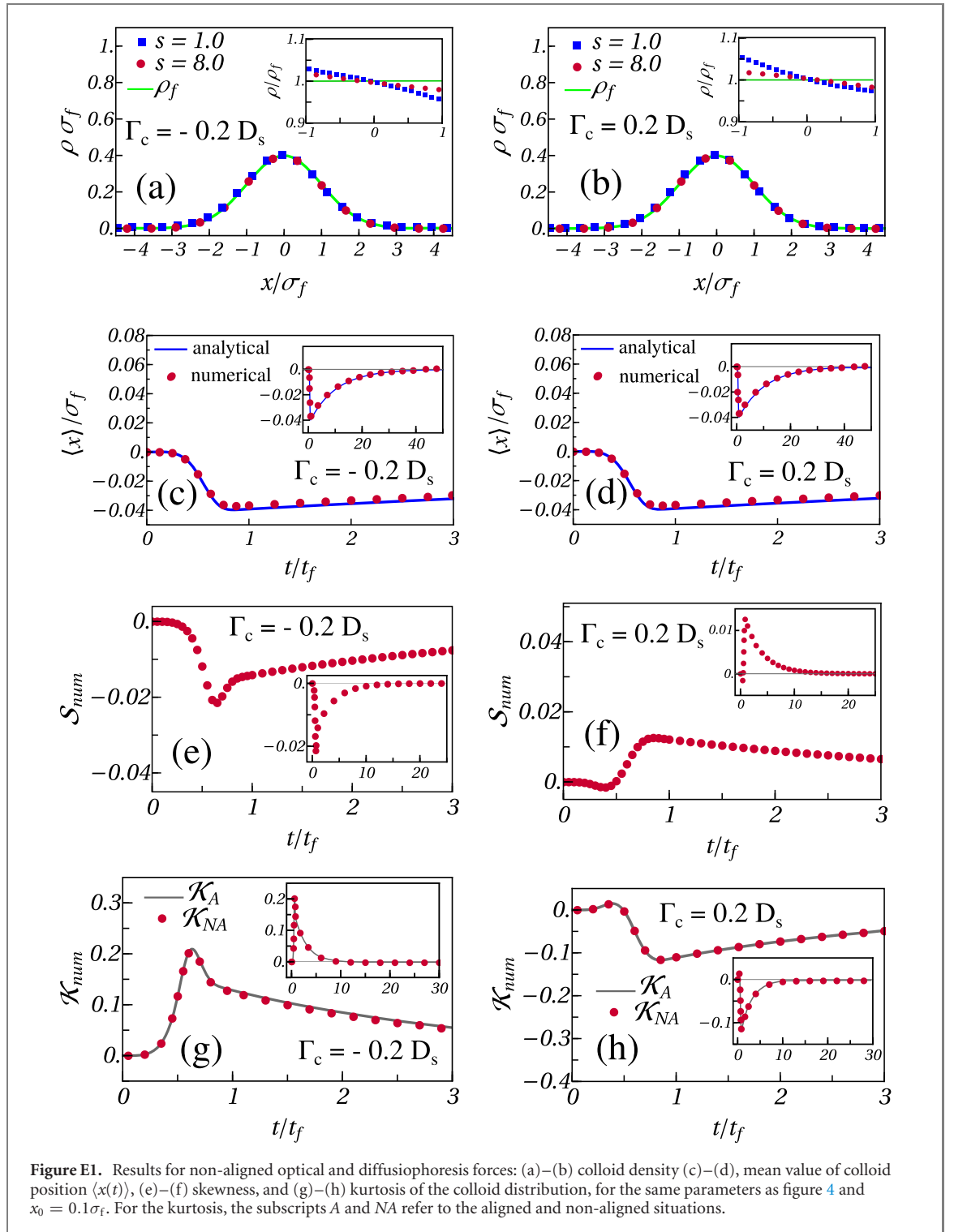


Figure D2. Quartic stiffness coefficient δ_{DP} as a function of time for the Gaussian states for two phoretic mobilities. The panels correspond to the same parameters as in figure 4 and $\alpha(t)$ given by equation (18).

which measures the non-Gaussianity of the colloidal density. While the skewness remains zero by symmetry, the kurtosis does not strictly vanish, at variance with an exact Gaussian density. The numerically calculated result $\mathcal{K}_{\text{num}}(t) = \int_{-\infty}^{+\infty} (x - \langle x \rangle)^4 \rho_{\text{num}}(x, t) dx / \sigma^4 - 3$ is shown in figure D1. This non-zero kurtosis originates from the quartic term in the total potential energy and leads the colloid density to deviate from Gaussian distribution. However, the maximum kurtosis is around 0.2 which is rather small.

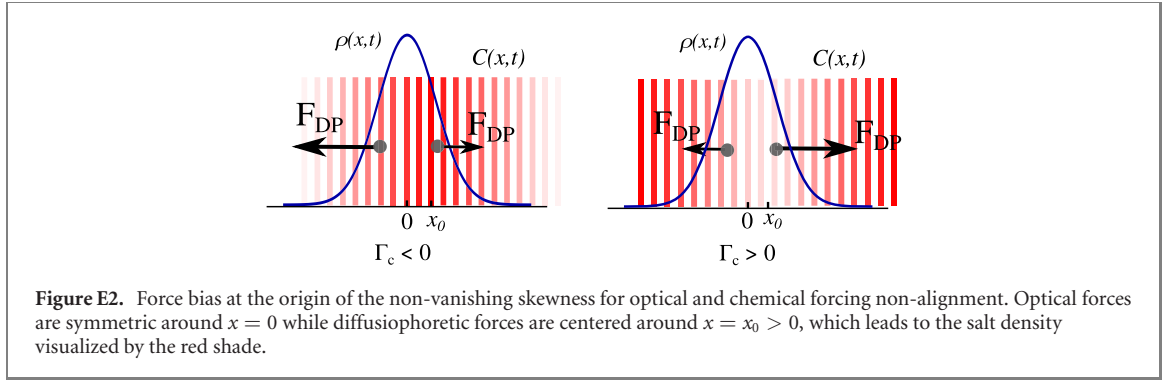
Furthermore, the kurtosis depends on the phoretic mobility. This is explained by considering the quartic term in the potential energy ($\delta_{DP} x^4 / 4$), where $\delta_{DP}(t) = -\frac{1}{6} \gamma \Gamma_c \partial_x^4 \ln C(x, t)|_{x=0}$. As shown in figure D2, unlike the quadratic term, $\delta_{DP}(t)$ depends on the phoretic mobility. For $\Gamma_c < 0$, δ_{DP} is mostly negative. This pushes the colloids further toward the exterior (enhanced expulsive contribution). Thus, there will be an



overpopulation at larger distances and a positive kurtosis. For $\Gamma_c > 0$, the argument is reversed (mostly positive δ_{DP} , mostly negative kurtosis).

Appendix E. Consequences of misaligning of the optical and diffusiophoretic forces

We address here a possible experimental limitation, coming from the fact that it may prove difficult to have a perfect coincidence between the minimum of the optical potential created, and the point of symmetry of the salt driving, where the diffusiophoretic forces vanish. We therefore assume that the optical and diffusiophoretic forces are centered at $x = 0$ and $x = x_0 \neq 0$, respectively. Then, the total potential energy is



given by

$$U = \frac{1}{2} \kappa_{\text{op}}(t) x^2 - \gamma \Gamma_c \ln C(x - x_0, t) \simeq \frac{1}{2} \kappa(t) x^2 - \kappa_{\text{DP}}(t) x_0 x, \quad (\text{E.1})$$

where $\kappa(t) = \kappa_{\text{op}} + \kappa_{\text{DP}}$, as in the main text, with

$$\kappa_{\text{DP}}(t) = -\gamma \Gamma_c \partial_{x,x}^2 \ln C(x, t)|_{x=x_0}. \quad (\text{E.2})$$

This results in a time-dependent trap center, located at $x = \kappa_{\text{DP}}(t) x_0 / \kappa(t)$. The assumptions of isotropicity and analyticity of $C(x, t)$ still hold with $\partial_r^j C|_{x=x_0} = 0$ for j odd. This potential leads to a Gaussian colloidal position distribution with a time-dependent mean value $\langle x(t) \rangle$:

$$\rho(x, t) = \sqrt{\frac{\alpha(t)}{\pi}} e^{-\alpha(t)[x - \langle x(t) \rangle]^2}. \quad (\text{E.3})$$

Substituting $\rho(x, t)$ into the Fokker–Planck equation, equation (2), in addition to the rule given by equation (5) for $\kappa(t)$, we obtain a differential equation governing $\langle x \rangle$ as

$$\gamma \partial_t \langle x(t) \rangle = \kappa_{\text{DP}}(t) x_0 - \kappa(t) \langle x(t) \rangle. \quad (\text{E.4})$$

Solving for $\langle x(t) \rangle$ with initial condition $\langle x(t=0) \rangle = 0$ yields

$$\langle x(t) \rangle = \int_0^t d\tau \frac{\kappa_{\text{DP}}(\tau) x_0}{\gamma} e^{-\int_0^\tau d\tau' \kappa(\tau')/\gamma}. \quad (\text{E.5})$$

Since non-aligning does not change the evolution of $\kappa(t)$ in equation (5), the protocol remains the same as presented in the main text for the aligned case with $\kappa_{\text{op}}(t) = 2k_B T \alpha(t)$ and $\alpha(t)$ given in equation (19). Hence, the variance does not change and corresponds to figure 8. We consider the changes of other moments. The colloidal density ρ_{num} is obtained by solving numerically the Fokker–Planck equation (2). The result is shown in figures E1(a)–(b) for the same parameters as in figure 4 and $x_0 = 0.1\sigma_f$. It is seen, especially in the inset plots, that the other moments, i.e., the mean value, skewness and kurtosis, are subject to changes. Then, we evaluate $\langle x \rangle$ either from equation (4), or by calculating numerically $\langle x \rangle_{\text{num}} = \int_{-\infty}^{\infty} x(t) \rho_{\text{num}} dx$. As figures E1(c)–(d) and also the inset plots in panels (a)–(b) show, $\langle x \rangle$ starts from zero at initial time and reaches a maximum value. According to equation (E.5), the maximum value of deviation from initial trap center is given by $\langle x \rangle_{\text{max}} = \kappa_{\text{DP}}(t^*) x_0 / \kappa(t^*)$ at t^* , when both the time dependent trap center x_{trap} ($\nabla U(x_{\text{trap}}, t) = 0$) and the mean value overlap. At the end of the protocol when $t = t_f$, $\langle x \rangle$ differs slightly from the target trap center at $x = 0$. This deviation is as a result of the inertia of the colloid: although the ions concentration is uniform at $t = t_f$ and the diffusiophoretic force vanishes, the colloid is on average slightly away from the optical potential minimum at $x = 0$, which results in a further drift.

To investigate if the colloid density remains Gaussian, we calculate the skewness $\mathcal{S}_{\text{num}} = (x - \langle x \rangle)^3 / \sigma^3$ and the kurtosis. The skewness is shown in figures E1(e) and (f). Non-alignment leads to a non-vanishing value, which is expected because of the mismatch between the center of diffusiophoretic forcing at x_0 and the optical center at $x = 0$. Moreover, the skewness depends on the phoretic mobility such that it is positive for $\Gamma_c > 0$ and negative for $\Gamma_c < 0$. The sketch in figure E2 explains that a negative skewness ensues for a positive x_0 when $\Gamma_c < 0$, while the converse holds for $\Gamma_c > 0$. Finally, as figures E1(g) and (h) shows and as expected, non-alignment does not affect the kurtosis. The kurtosis here (red symbols) is almost the same as the one for the aligned case (shown by a solid gray curve).

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- [57] Diffusiophoretic motion is globally force-free [61]; by ‘diffusiophoretic force’, we refer here to the ‘osmotic-pushing’ [61], which takes its origin in the thin diffuse-layer in the vicinity of the colloids. Within the present overdamped description, it is the sum of all forces acting on every colloid that does vanish: solvent friction, osmotic push, and optical forcing.
- [58] In practice, this limit does not seem too much demanding. Indeed, the ratio D_s/D_c is on the order of 103 for a micron-sized colloid. Thus, R/σ_f should not exceed 30. Since σ_f is a measure of the spread of colloidal positions, R should be larger than a few σ_f in order not to interfere with the colloidal displacements, but less than $30\sigma_f$.
- [59] As in the rest of the text, we use here adiabatic in the quantum mechanics inspired meaning of ‘slow enough’. Confusion should be avoided with the thermodynamic meaning of being heat-exchange free [18].
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