

Chapter 1

Introduction

Crystallisation is one of the oldest forms of phase separation used by humanity [1], put simply, it is the formation and growth of a new structured phase within a disordered bulk phase. This has applications in a number of industries such as pharmaceuticals, food production, and electronics [2]; where the extraction of dilute materials can help improve product quality while keeping production costs low. The industrialisation of crystallisation has allowed engineers to reliably and efficiently induce the crystal formation within a bulk phase. However, while on a large scale crystallisation is seemingly an understood physical process, just a small amount of investigation into the literature reveals that at a micro scale there is yet to be unifying theory that can accurately explain the process of crystallisation [3]. Of which there two primary areas of focus, nucleation and crystal growth. The latter focusing on how an already stable crystal grows and how it takes on its final shape; whereas the former is more concerned with which factors contribute to its formation in order to control crystal growth.

The focus of this research is partly based around investigating the impact of a focused electromagnetic field can influence the nucleation process by inducing shear flow within the bulk phase. This in hope that the localised EM field can be used to study different phenomena in nucleation in order to develop a better understanding of the underlying mechanisms.

1.1 Nucleation

Nucleation is an example of a binary phase separation, where a dilute phase is miscible in a bulk phase, more often called the solute and solvent respectively. Because of thermodynamics, the two can only remain in equilibrium while below a specific concentration (C_{eq}) - below which the chemical potential μ for a miscible solution is greater than the potential required to separate the two phases. Once C_{eq} is exceeded there is a chemical potential difference driving the solution to separate the two phases. Since different combinations of solute and solvent will have different equilibrium concentrations, researchers often instead measure the ratio between the solute and solvent by using 'supersaturation', one form of measuring supersaturation is shown below [4]:

$$S(T) = \frac{C_{sol}}{C_{eq}(T)} \quad (1.1)$$

Where C_{sol} is just the solute concentration, $C_{eq}(T)$ is the equilibrium concentration at temperature T . While the solution remains supersaturated there is a chemical potential driving force for the solute to coalesce and separate from the solution as an ordered solid, the first formation of the crystal is referred to as the nucleus and understanding its formation has been the focus of researchers for decades now. Typically, for an industrial crystallisation process the working principle is based on controlling and manipulating the supersaturation of the system.

1.1.1 Primary & Secondary nucleation

From an industrial perspective, the nucleation process can be broadly categorised into either primary or secondary nucleation. The former being described as the formation of an initial nucleus within the bulk phase; absent of any external stimuli primary nucleation is considered stochastic and is typically undesirable for industrial applications where short and consistent residence times are desirable. At a small scale one can estimate the nucleation rate by making repeated measurements of sample solutions and seeing how many have nucleated after a given time, giving us a Poisson distribution.

$$P(t) = 1 - \exp[-JV(t - t_g)] = \frac{M^*(t)}{M} \quad (1.2)$$

Where J is the nucleation rate, t_g is the 'growth time', V is the volume of the individual samples, and $M^*(t)$ & M are the number of nucleated samples and the total number of samples used respectively. While this is useful for studying the effects of different parameters at a small scale, for industrial applications there are so many additional factors at play that can throw off this estimation.

In contrast, secondary nucleation is the result of a initial seed crystal inducing further nucleation within the bulk solution [5], in absence of a seed crystal nucleation is near impossible. The research of Secondary Nucleation is fascinating for its own merits; is it speculated that the supersaturation barrier is non-existent [6], and furthermore there are a multitude of mechanisms that can induce secondary nucleation making it imperative that researchers can control the nucleation rate at an industrial scale.

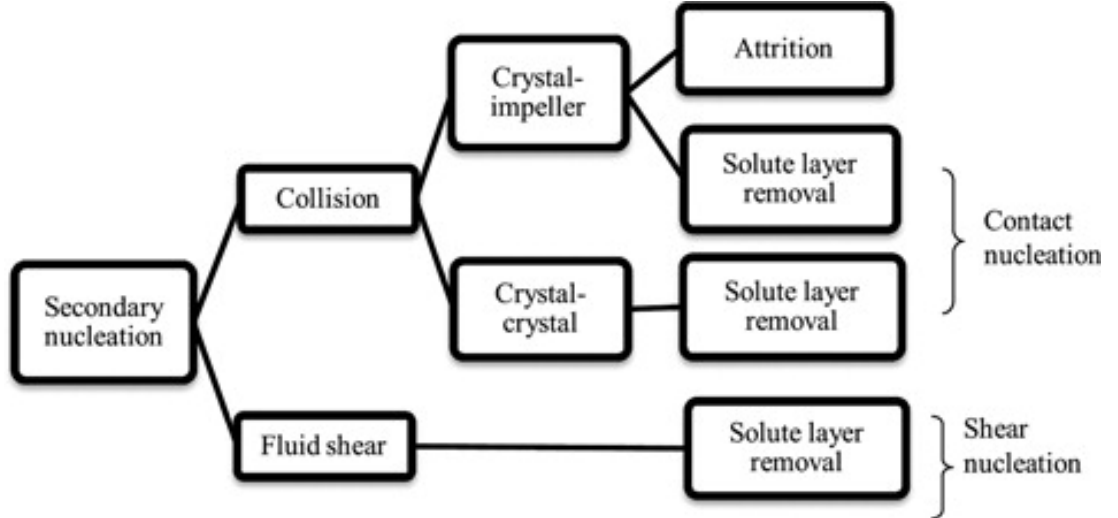


Figure 1.1: Secondary Nucleation mechanisms, classified by Agrawal and Paterson [7]

1.2 Nucleation Theories

1.2.1 Classical Nucleation Theory (CNT)

Sometimes referred to as 'Gibbs Nucleation Theory' the original theory was first formed from the works of Volmer and Weber, and Frenkel [8,9]. While initially it was more focused into describing droplet formation in condensing vapours it was extrapolated to describe crystallisation. The central premise of classical theory is that nucleation occurs stochastically throughout the bulk phase due to collisions between individual solute molecules, ions, or atoms; at the same time the bulk phase is resistant to the formation of a new phase. The competition between these random collisions and the bulk solution can be used to predict the probability of a newly formed nucleus.

Consider a supersaturated solution, after some time enough individual sub units collide, forming a nucleus of volume $4\pi r^3/3$. The newly formed phase has a lower chemical potential than the surrounding solution, reducing the free energy of the system; at the same time, the formation of a new interface is resisted by the bulk phase due to surface tension. The net free energy of the system for a nucleus of radius r is given as [10]:

$$\Delta G = \frac{-4\pi r^3}{3v} k_B T \ln(S) + 4\pi r^2 \sigma_{inf} \quad (1.3)$$

Where v is the approximate volume of an individual molecule, k_B is the Boltzmann constant, and σ_{inf} is the interfacial tension of the bulk solution. Plotting the free energy of the system against nucleus size reveals a critical size above which the gain in free energy exceeds the interfacial tension.

The maximum value of ΔG_{tot} is the free energy barrier that any newly formed nucleus needs to overcome in order to stabilise. The nucleation rate (the volume of new crystalline material formed per unit time), is therefore commonly defined as being dependent on the energy barrier ΔG^* :

$$J = A \exp \left[-\frac{\Delta G^*}{k_B T} \right] \quad (1.4)$$

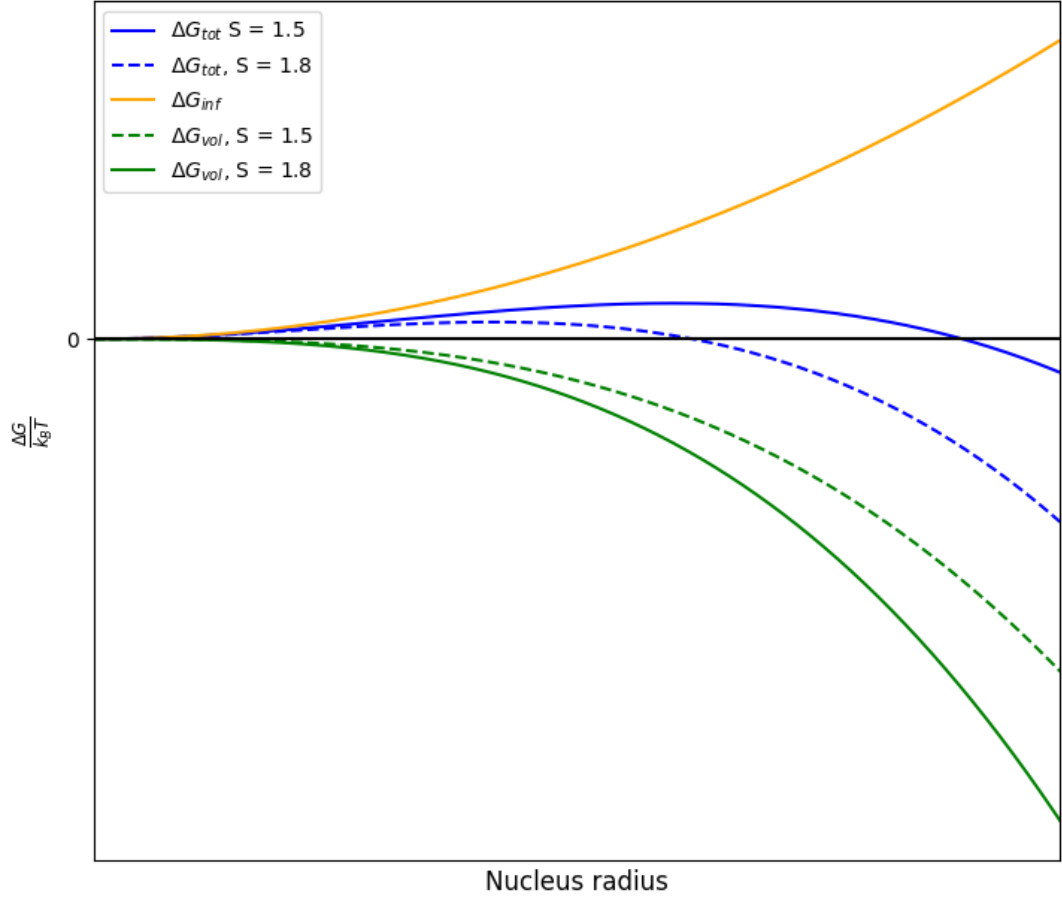


Figure 1.2: Free energy diagram of a newly formed nucleus according to the Classical Nucleation Theory. The total free energy (blue) is due to the competition between the volume free energy gain (green) and the interfacial free energy cost (orange). Dotted lines are for a higher supersaturation than the solid lines, the interfacial energy cost is independent of supersaturation.

Where A is a pre-factor that can be fine tuned to the exact demands of the system (typically involving Zeldovich Factor, Z) the free energy barrier can be found by finding the turning point of ΔG_{tot} .

CNT is often regarded as a good description of the macro system, its obvious that for all crystallization systems there is an inherent energy barrier that dictates the nucleation rate. Where it falters is in its predictive ability, both in estimating nucleation rates [11,12], and in the structure of newly formed nuclei [13,14]. Recent

studies suggest classical nucleation is merely one of many possible pathways that can be taken to produce a structured crystalline phase. Prompting the development of alternative theories to better describe the nucleation process.

1.2.2 Two Step Nucleation

The two step nucleation theory is an extension to the CNT, colloid simulations showed that short range (such as in proteins [15, 16]) interactions allow for the formation of a liquid-liquid metastable phase from which a new solid phase could form [10, 17]. *in situ* techniques for studying nucleation several papers reported the presence of stable liquid-like clusters that formed prior to nucleation [15, 18, 19]. It can be understood by Oswald’s rule [20], which says that any crystallising system does not immediately take the path to the lowest possible energy state but instead first transitions to the state with the smallest free energy barrier, further transitions can still occur but this pathway minimises the overall free energy cost. If we consider the free energy diagram from before (fig. ??) we can consider a case where the supersaturation is low ($S \approx 1.01$) then a dense liquid droplet will have a lower chemical potential than the surrounding fluid but the crystalline phase will have a subsequently lower chemical potential than the dense liquid phase. Initially it was suspected that the formation of the dense liquid phase was a result of stochastic density fluctuations in the system; further simulations suggest that the the local bond order is a stronger driving force than the local density [21].

The current research into two step nucleation (or now more commonly referred to as multi-step nucleation theory) is developing a robust framework to describe what nucleation pathway will occur given the initial conditions. Reviews of all currently documented nucleation pathways identified highlighted the fact the need for a the development of *in situ* techniques that can induce nucleation locally but can also reliably identify the nucleation pathway across a broad range of experimental conditions [3, 10].

1.3 Crystallisation methods

1.3.1 Cooling Crystallisation

For some binary mixtures the supersaturation is heavily dependent on the solution temperature, therefore a simple method of producing crystals is by cooling the mixture to induce crystal formation. At ambient temperatures the solute concentration is too high to be fully incorporated into the solution ($S \gg 1$), after heating however the solute is fully dissolved into the solution ($S < 1$). Now as the mixture is allowed to cool to room temperature the supersaturation will increase until crystal formation begins, the rate of cooling drastically influencing the size and number of crystals produced.

If dT/dt is high then the final product will consist of large crystal and be low in number, as the nucleation rate is directly related to the supersaturation only a handful of nuclei can form before the remaining solute grows onto the surface. If dT/dt is low then the final product will consist of smaller crystals and be far more numerous, as the supersaturation is so large that the new nuclei are forming continuously. Between these two extremes, one can define the meta-stable zone width, a region in the temperature-concentration phase space where both nucleation and crystal growth can be reliably controlled. The lower limit being given by the solubility curve of the binary mixture, and the upper limit being defined by the metastable zone.

The viability of cooling crystallisation is dependent on the meta-stable zone width, too narrow and the process is difficult to control, too wide and the crystal growth rate may be insufficient for the desired outcome.

1.3.2 Evaporative Crystallisation

In situations where control of the final product size or shape is not the main focus, often the cheapest method of producing a crystalline product is simply to allow the solvent evaporate and separate from the solute. Depending on the total volume of solvent to evaporate this process can take on time scale of several days to complete.

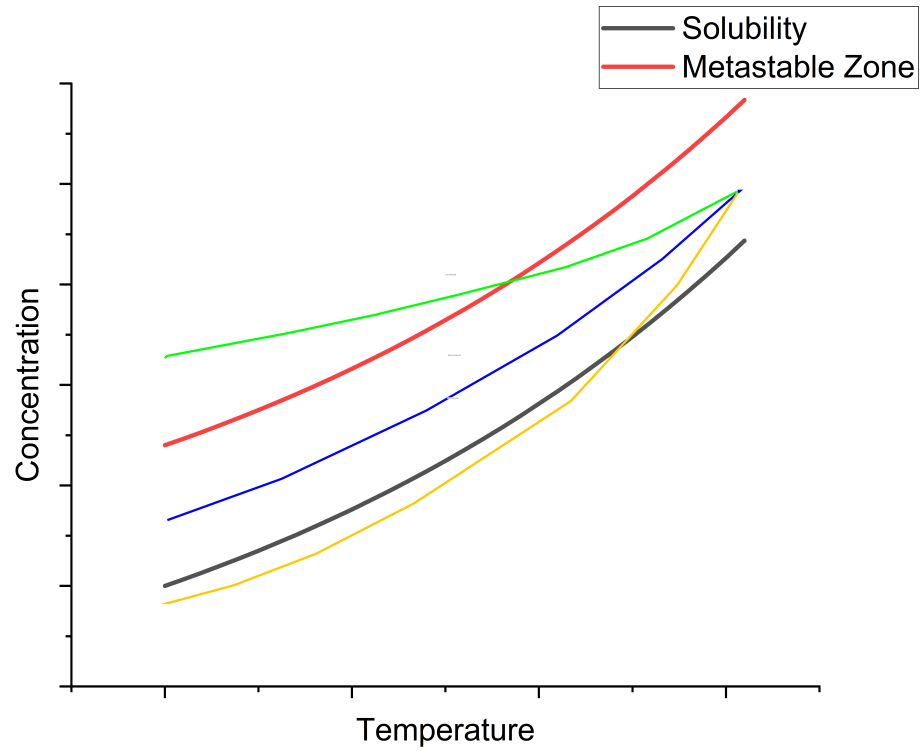


Figure 1.3: Typical Concentration vs Temperature plot with the solubility curve (black) and metastable zone (red). 3 different cooling curves are shown as well: a high rate of cooling (orange) shows the mixture quickly dropping below the solubility curve resulting in no new nuclei forming; a low rate of cooling (green) shows the mixture exceeding the metastable zone, where now nucleation occurs freely; and a typical cooling rate (blue) shows how a typical cooling crystalliser will operate, sitting in the middle of the other two curves.

1.4 Optical Tweezers

1.4.1 Background

Optical tweezing has been a field of applied optics ever since the 1970s when Ashkin [22] first showed that focused light was capable of trapping micron sized particles due to light exerting 'radiation pressure'. The working principle was that a light source such as a laser could trap small objects within a 2D plane, as long as the light source had an approximately Gaussian profile. Soon after, Ashkin showed that the introduction

of a microscope objective would allow one to focus the light source to a diffraction limited point that would stably trap small objects within a confined volume [23]. This allowed Ashkin and others to study biological material and would later be used to probe microscopic properties such as the formation of colloidal aggregates [24] to the drag forces exerted by a pure vacuum [25, 26]. Due to the predictable behaviour of light, optical tweezers have become essential for measuring and exerting precise forces on the magnitude of pico-newtons allowing one to probe the material properties of the smallest materials.

1.4.2 Literature related to laser induced nucleation

From as early as 1996 it has been known that laser irradiation is a viable method of inducing nucleation within a supersaturated solution [27], the first reported case was notable as it used a $1.064\ \mu\text{m}$ laser meaning it was unlikely to be a photo-chemical reaction but rather a physical one. Future research has found nucleation can be induced by 1 of 3 routes involving direct laser induction: firstly, Non-photochemical laser induced nucleation (NPLIN) where the solution is irradiated with a pulsed laser [27–29], several papers have debated the exact mechanism that induces NPLIN [28, 30]. Two suggested hypothesis are: an optical Kerr effect, where the solute molecules are aligned to lower the nucleation barrier [30]; a dielectric polarisation effect, in which solute clusters are stabilised within the electric field which drastically increases the likelihood of nucleation [31]. Both hypothesis have their limitations and there has yet to be a single theory that explains NLPIN thoroughly. In either case the mean pulse intensity needs to be kept relatively low (on the order of $0.1 - 0.01\text{GW}/\text{cm}^2$), as high intensity pulses lead to a completely different nucleation mechanism.

High intensity laser induced nucleation (HILIN), where the pulse intensity is on the order of several PW/cm^2 is far simpler a mechanism to explain in comparison to NPLIN. The production of nuclei can be wholly associated to a cavitation process within the target solution, where the laser focus results in thermo-cavitation and the subsequent pressure change leads to a nucleation event around the focus of the laser [, ,]. There is still not a general consensus on how cavitation influences the local supersaturation [,

], nor is there a clear understanding of what properties of the crystal can be controlled [,]. It has been suggested that in theory any solution can undergo HILIN [], proving such a theory requires a strong understanding of the phenomena both before and after cavitation occurs.

Lastly, there is trapping induced nucleation, this is where optical tweezers come into play, the optical trap has been shown to have different effects on supersaturated solutions depending on where it has been focused. When focusing on the cover slip, supersaturated solutions of glycine and D_2O were shown to create a dense liquid droplet of glycine and water [,], applying DLS analysis showed that the dense liquid region was populated by clusters that would consolidate together upon being focused by the optical trap []. Molecular simulations of glycine solutions showed that these clusters are unstable when using pure glycine below the saturation point [] suggesting that the clusters are formed due to glycine reaction products. When the optical trap was moved from the cover slip to the air-solution interface nucleation would occur before a dense liquid region could form []. Repeated experiments where the laser is focused on the air-solution interface have lead to a variety of different nucleation events. In some instances the nucleation occurs spontaneously after a short period of time []; whereas allowing a solution to age results in the formation of amorphous precursors that when irradiated will nucleate immediately []. The precursors are only seen when the solution is irradiated by an optical tweezer and the growth rate can be controlled somewhat by varying the laser power []. Notably the only work has been done with simply irradiating the solution with a trapping potential, there has not been an attempt to introduce a trappable object into the solution, the trapping potential has been used to influence the growth of a crystal front [].

1.4.3 Optical Rotation

For any electromagnetic field it is possible to transfer both linear and angular momentum []; more accurately the field is said to have both orbital and spin momentum. Though there is some debate on how to decompose the total momentum into these two components [], for this thesis we do not need to calculate the exact quantities and

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will instead look at the broader effects of both components. The orbital momentum can be understood as the shape of the wavefront of the particular field in question, for simple Gaussian beams the wavefronts are uniform and equally spaced resulting in the typical radiation pressure that Ashkin and co demonstrated [1]. However, higher order modes of a Gaussian beam (such as the Laguerre-Gaussian modes) have non-uniform wave fronts meaning the orbital momentum has both angular and linear components; depending on the relative size of the target particle one can induce rotation, or orbiting [2]. Spin angular momentum (SAM) is attributed to the spin density of the field, early research has shown that the spin density is non-zero for any beam but while the SAM can easily be zero for homogeneous scatterers, this has sparked debate if SAM is even a physical quantity as it does not aid in the transport of energy directly [3]. This paradox is resolved by representing the wave as an array of finite loops that all together cancel one-another out when the medium is homogeneous, when inhomogeneity is introduced (such as by being refracted by a birefringent medium) these circular loops no longer cancel out resulting in a non-zero SAM [4], depending on the material a number of rotational effects can occur.

For a perfectly homogeneous non-absorbing sphere the total SAM transferred from a circularly polarised beam is zero; if, however, the wave front of the beam is helical - for example using a Laguerre-Gaussian beam [5] - one can control the rotational motion via transfer of OAM. Due to the structure of the LG beam the individual rays are not perpendicular with the propagation direction; as a trapped particle will experience a constant torque while being pulled towards the centre of the trap focus. This was demonstrated in [6] where Copper Oxide micro particles rotated with a frequency of around 2 Hz due to the orbital AM from a LG beam; this was enhanced further by circularly polarising the trapping beam. Typically for a non-absorbing particle the polarization state of the beam has a negligible impact on the angular momentum transferred, this is not the case for particles with a high absorption efficiency as the combined handedness of the LG beam and polarisation vector can lead to an enhanced transfer of SAM. However, by far the best method for inducing rotation is using birefringent materials.

Birefringence is a material property often seen in crystalline materials, if the crystal lattice has a different structures when viewed at different orientations then light will be refracted differently depending on its polarisation. For circularly polarised light the inhomogeneity results in a high degree of SAM being transferred to the target object [], this has been exploited to rotate microspheres as fast as 1000 Hz while suspended in a bulk medium [] as well as a means of measuring the local temperature and shear response of said medium [,]. Calculating the optical torque applied to a birefringent material is given via:

$$\begin{aligned}\tau_{opt} = & -\frac{\epsilon}{2\omega_{laser}}E_0^2\sin(kd(\Delta n))\cos 2\theta\sin 2\phi \\ & +\frac{\epsilon}{2\omega_{laser}}E_0^2(1-\cos(kd(\Delta n))\sin 2\phi)\end{aligned}\quad (1.5)$$

Where θ is the angle between the particle's orientation vector and the propagation direction of the EM field, and ϕ is the phase shift in the EM field. The first term represents the 'orientational' torque which is due to the target particle being aligned along the EM field, when aligned $\theta = 0$ meaning the entire term is negligible for particle's with a stable orientation. The second term is due purely to the polarisation of the optical trap, for circularly polarised light $\phi = \pi/4$ thus maximising the torque transferred to the target particle. Birefringence can also be induced if the target particle has an anisotropic shape, in particular if the particle shape is elongated along one major axis; the go to particle shape is a silica dimer (two spheres tangentially attached) due to silica's stability and strong adhesion. Using a silica dimer research groups have achieved a rotation frequency in the realm of several GHz [] in a vacuum.

1.4.4 Shear induced Nucleation

It has long been known that solution nucleation is influenced by shearing of the fluid, from stirring agents in the container (i.e. propellers) to the container boundary layers, however there has yet to be a clear mechanism by which said shearing effects the nucleation event. Theoretical research into shear induced nucleation suggests that there should be a slight increase in the nucleation rate at low shear rates, reaching a

maximum increase in nucleation rate, and then at higher shear rates the nucleation rate begins to drop off. This has been shown theoretically for both simple colloidal [32–34] and ice crystal formation [35]; however, no experimental work into these systems has been conducted to prove this is the case. There is some experimental evidence for this phenomena in simple salt and protein solutions - though the authors emphasise that mechanical agitation cannot be ruled out - there has not been an exhaustive study into the shearing effects apart from in glycine solutions. In [33] it was found that a shear rate of around 3000 s^{-1} was the maximum shear rate that would yield the highest nucleation rate. Using the theoretical model established in [32, 36] which modifies the CNT to account for the effects of a nucleus undergoing shearing, accounting for the fact that a nucleus' growth is undergoing competition between flow-mediated molecular transport and the strain applied by the flow field which inhibits the growth of the nucleus. The central conclusion (from both the theoretical and experimental results) is that there is an optimal shear rate in which the nucleation rate is maximised. However, a question that arises from this result, if there is an optimal shear rate in which molecular transport is maximised and strain is minimised, then surely there should also be a shear rate in which the molecular transport and strain are equal - allowing one to suspend a nucleus at a constant radius. In this scenario, the molecular transport would prevent the nucleus from dissolving, but the strain would prevent the nucleus from growing. This however would require one to be able to apply a continuous shear rate to a targeted nucleus with high precision, there is also no model for an individual nucleus undergoing growth.

1.5 Aims and Objectives

Overall the aim of the PhD is to study viability of using micro-rotors to generate localised fluid flow around the beam focus. The results are reported in chapter 3, this is then succeeded by experimental work where we instead using a galvano-mirror to generate shear flow. While overall unsuccessful the addition of a moving beam focus showed that the growth of a nucleus can be localised around the trap focus.

In addition, computer simulations studying the dynamics of asymmetric dimers and

their applications as complex micro-rotors are discussed in chapter 4. Lastly two novel methods of measuring rotational motion are discussed in chapter 5; firstly via a novel detection fibre method that allows for instantaneous measurements of the orientational behaviour of optically trapped ellipsoids/dimers; and secondly we create a simulative quadrant photo diode that replicates laboratory results, utilising linear regression techniques we measure the change in orientation in order to measure the optical torque applied to a non-birefringent particle.

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