Shape Fluctuations in Randomly Stirred Dilute Emulsions

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I. INTRODUCTION

Single deformable objects such as droplets of one liquid immersed in another liquid fluctuate in shape in response to random external stirring \[1\]. The purpose of this paper is to investigate the effect of interaction between such deformable objects \[2\] on the way they deform due to fluctuations of the velocity field in the fluid in which they are immersed. Numerous authors have studied the fluctuations and diffusion of single deformable objects due to thermal agitation \[3, 4, 5, 6, 7, 8, 9, 10\]. Nevertheless, there are clearly other ways in which systems are agitated. In industrial and biological environments, the host liquid is often stirred, shaken or pumped in ways which are very different from thermal agitation. The list of examples is not restricted to artificial processes. It also includes natural processes such as Brownian motion of small beads induced by the collective motion of bacteria \[11\] and nano-scale mechanical fluctuations of the red blood cell surface that have been measured and shown to depend strongly on the biochemical environment and not only on temperature \[12, 13, 14, 15\]. For this reason the external velocity field agitating the system is taken to be more general than that corresponding just to thermal motion. The article provides thus the general equations describing the effect of a finite density of deformable objects on the diffusion and shape fluctuations of a single object, to linear order in the density.

The system considered has the following properties.

(a) The deformable objects are fluid, in the sense that the velocity field is well defined everywhere (both inside and outside the object). No slip and no penetration conditions are assumed at the interface of the deformable object. Hence, each surface element moves with the velocity of the flow at its position. In addition, both the objects and the host fluid are
incompressible. The objects are characterized by an energy that depends on their shape (i.e. changing the orientation or switching places of two surface particles while keeping the shape constant does not change the energy). The main example considered in this paper is surface tension \[16, 17\]. However the description can be extended to Helfrich bending energy \[18, 19\] and other cases where the shape of minimum energy is nearly spherical. Deformation of the shape changes the energy, exerts a force density on the liquid and therefore generates an additional velocity field, denoted by $\vec{v}_{\psi}$.

(b) The hydrodynamic equations of the host liquid are linear in the velocity (i.e. a velocity field induced by several sources is equal to the sum of the velocity fields induced by each source separately). For instance, if the flow is governed by the Navier Stokes equation, then the linearity implies that the Reynolds number is small and that the Stokes approximation is applicable. The actual velocity field is the sum of the imposed velocity field, $\vec{v}_{\text{ext}}$ (the velocity field that would have existed if the objects were absent), the velocity field induced by the deformations of the object under consideration, $\vec{v}_{\psi}$, and $\vec{v}_{r}$ which is the velocity field created by the rest of the deformable objects,

$$\vec{v} = \vec{v}_{\text{ext}} + \vec{v}_{\psi} + \vec{v}_{r}. \quad (1)$$

(c) The external velocity field is assumed to be random. The correlations are assumed depend only on distance and time difference. Furthermore, the dependence on the time difference is taken to be extremely short ranged (Dirac $\delta$ function in the time difference). In principle, equations for the dependence of the shape correlations on the density of deformable objects can be worked out for any dependence of the velocity correlations on time. These are very complicated, however, and the above choice of the dependence of the external velocity correlations on time simplifies matters considerably and is certainly realistic in many cases. It is important to note that the results obtained here are not used to determine the external velocity correlations. Those correlations are just taken as a given input. For example, in the special case of thermal agitation the velocity correlation was calculated from first principles \[10\] and only then used to calculate the diffusion constant and deformation characteristics of a deformable object immersed in the liquid. In addition the external velocity is assumed to be small enough to allow the body to remain almost spherical.

Since we assume small deviations from the spherical shape it is only natural to describe the surface shape of the objects using spherical harmonics. Consider a spherical body which
is moving and is slightly deformed. The equation

\[ \frac{\rho}{R} + f(\Omega, t) - 1 = 0 \]  

defines its surface, yielding for each spatial direction, \( \Omega \), the distance, \( \rho \equiv |\vec{r} - \vec{r}_0| \), of the surface from the centre of the body, \( \vec{r}_0 \). \( R \) is the radius of the undeformed sphere. The deformation function, \( f(\Omega, t) \), defines the shape and may be expanded in spherical harmonics,

\[ f(\Omega, t) = \sum_{l=1}^{\infty} \sum_{m=-l}^{l} f_{lm}(t) Y_{lm}(\Omega) \]  

(clearly the \( Y_{00} \) term can be absorbed in the definition of \( R \)). The goal is to obtain the correlations between the deformation coefficients, \( f_{lm}(t) \). The centre of the object, \( \vec{r}_0 \), is chosen to be the point around which the deformation coefficients with \( l = 1 \) vanish: \( f_{1m} = 0 \). A different definition of the centre will introduce three different equations for the deformation coefficients with \( l = 1 \). These are not interesting, as far as the shape is concerned, since in the first order of the deformation the spherical harmonics with \( l = 1 \) describe a rigid translation of the object \[20, 21\].

The random velocity field and the effect of the interaction between objects induce fluctuations in the values of the deformation coefficients describing any of the objects. Consider the autocorrelation of the deformation coefficient \( f_{i,m} \) of the \( i \)’th object

\[ \langle f_{i,m}(\omega)f_{i,-m}(\tilde{\omega}) \rangle \]  

where \( f_{i,m}(\omega) \) and \( f_{i,m}(\tilde{\omega}) \) represents the Fourier transforms (FT) of the deformation coefficients of the \( i \)’th object in respect to time. The autocorrelation is expanded in orders of \( n \). To first order in \( n \) it is given by

\[ \langle f_{i,m}(\omega)f_{i,-m}(\tilde{\omega}) \rangle = G_{i,0}(\omega, \tilde{\omega}) + G_{i,1}(\omega, \tilde{\omega}) \cdot n \]  

The first term on the right hand side of the equation (3) above gives the shape correlations of a single object, that has been described previously \[1\]. The second term represents the correction to the shape correlations due to a small but finite density of deformable objects. The purpose of this article is to obtain that correction.

The paper is organized as follows. Section II deals with a single deformable object in random flow. The aim of this section is to introduce the basic definitions of flow and present the zero-order terms in the expansion of the shape correlations in the density of objects \( n \). In section III the first order terms of the shape correlation functions are derived. Section IV deals with the special case of identical droplets that are governed by surface tension in a random flow. In order to improve readability, Parts of the derivation of the first order terms
and the velocity field induced by a deformable body governed by surface tension are left to the appendix.

II. A SINGLE OBJECT IN RANDOM FLOW

The response of a single object immersed in a host liquid to an external random flow has been described in previous work [1]. The results are sketchily repeated here for the benefit of the reader as the general equations obtained here are to be exploited in the next section by replacing the external velocity field by the velocity field seen by the object when a finite density of deformable objects is immersed in the liquid. The latter velocity field is the sum of the imposed velocity field and the velocity fields induced by all the other deformable objects. The correlations of the deformation coefficients as well as diffusion constant of the centre will be obtained here in terms of the correlations of the external velocity. The diffusion constant will be needed in the next section to obtain the first order correction in the density of the shape correlations.

The no-slip and no-penetration conditions yield an equation of evolution for the deformation coefficients [20],

\[ \frac{\partial f_{lm}(t)}{\partial t} + \lambda_l f_{lm}(t) = -Q_{lm}(t). \]  

(4)

The effect of the velocity field induced by the deformable object itself is represented by the second term on the left hand side above. The \( \lambda_l \)'s characterize the way in which a deformation with definite \( l \) decays to zero in the absence of an external velocity and other objects. Different physical systems are characterized by different sets of \( \lambda_l \) [4, 8, 20, 22].

The term on the right hand side, \( Q_{lm} \), is given by

\[ Q_{lm} = \frac{1}{R} \int d\Omega \left\{ \hat{\Omega} \cdot \left[ \vec{v}_{ext} - \hat{r}_0 \right] Y_{l,m}^*(\Omega) \right\}, \]

(5)

where the external velocity field, \( \vec{v}_{ext} \), is evaluated on the undeformed body and \( \hat{\Omega} \) is a unit vector in the direction of the spatial angle \( \Omega \) (for further detail see [10]). The velocity of the centre \( \hat{r}_0 \) contributes only to \( Q_{1,m} \). In addition, the definition of the centre implies that

\[ Q_{1,m} = 0, \]

(6)

for all \( m \).

It is convenient to write the correlation function of the external velocity field in momentum
space. This is so because the random velocity field is transversal when the fluid is incompressible. Consequently, in real space, the flow must always be correlated in a very complex way. On the other hand, in momentum space, the transversal part of a general field is easily obtained:

\[ v_{\text{ext}}^i(\vec{q}) \equiv \sum_j \left( \delta_{ij} - \frac{q_i q_j}{q^2} \right) u^j(\vec{q}), \]  

where \( \vec{u} \) is a general vector field and \( i \) and \( j \) denote Cartesian components. The bracketed term is the projection operator that removes the longitudinal part of \( \vec{u} \), and therefore yields a general transverse velocity field \( \vec{v}_{\text{ext}} \). Next, the correlations of the external velocity are easily expressed using the correlations of the general field \( \vec{u} \).

\[ \langle u^l(\vec{q}, t_1) u^m(\vec{p}, t_2) \rangle = \delta_{lm} \delta(\vec{q} + \vec{p}) \phi(q, |t_2 - t_1|), \]  

where \( \phi \) is a general function of \( q \) and the time difference (with the only limitation that its Fourier transform in the time difference is non-negative). As was mentioned before, this investigation is restricted to cases where the external velocity is uncorrelated in time,

\[ \phi(q, t) = \tilde{\phi}(q) \delta(t), \]  

and where the mean of the velocity field vanishes, \( \langle u^l(\vec{q}, t) \rangle = 0. \)

Using the above, the diffusion coefficient of the centre of the deformable object is obtained

\[ D = \frac{8\pi^3}{3} \int_0^\infty q^2 dq \tilde{\phi}(q) J^2(qR), \]  

where \( J(x) = j_0(x) + j_2(x) \) and \( j_n(x) \) is the spherical Bessel function of order \( n \). The correlations of the deformation coefficients, \( f_{l,m} \), are given by

\[ \langle f_{lm}(t) f_{l'm'}(t + \Delta t) \rangle_{t \to \infty} = Q_{ll'} \frac{\delta_{ll'} e^{-\lambda_l |\Delta t|}}{2\lambda_l} \delta_{m',-m}, \]  

where

\[ Q_{ll'} \equiv \int \frac{d\Omega}{R^2} \int d\Omega' \int d^3q Y^*_l(\Omega) Y^*_{l'}(\Omega') \left[ \tilde{\Omega}_l \tilde{\Omega}_l' e^{-i\vec{q} \cdot (\vec{r} - \vec{r}')} R \left[ \delta_{ij} - \frac{q_i q_j}{q^2} \right] \tilde{\phi}(q) \right]. \]  

The contribution to the autocorrelation is presented here for completeness after being Fourier transformed in time,

\[ G_{l,0}(\omega, \tilde{\omega}) = \frac{\delta(\omega - \tilde{\omega})}{4\pi^2(\lambda_l^2 + \omega^2)} Q_{ll}. \]
III. FIRST ORDER CORRECTION

The aim of this section is to obtain the correction to the autocorrelation of the object’s shape, which is linear in the density of objects $n$. The full autocorrelation of deformation coefficients is obtained by replacing $\vec{v}_{ext}$ in equation (4) by the sum of the external velocity and the velocity induced by the other objects,

$$\langle f_{i,m}^*(\omega)f_{i,-m}(-\omega) \rangle = \frac{1}{\lambda_l + i\omega} \frac{1}{\lambda_l - i\omega} \frac{1}{(2\pi)^4 R^2} \int d\Omega_1 \int d\Omega_2 \int d^3q_1 \int d^3q_2 \int d\omega_1 \int d\omega_2 Y_{l,m}^*(\Omega_1)Y_{l,-m}^*(\Omega_2)e^{i\vec{q}_1 \cdot \hat{\Omega}_1}e^{i\vec{q}_2 \cdot \hat{\Omega}_2}G^i(\vec{q}_1, \omega - \omega_1)G^i(\vec{q}_2, -\omega - \omega_2)\left(\nu_{ext}^i(\vec{q}_1, \omega_1) + \nu^\alpha_i(\vec{q}_1, \omega_1)\right)\left(\nu_{ext}^\beta(\vec{q}_2, \omega_2) + \nu^\beta_i(\vec{q}_2, \omega_2)\right).$$

In this and in all the following equations, the Einstein summation convention is applied to the Cartesian components $\alpha, \beta$. In addition, $G^i(\vec{q}, \omega)$ appearing in the equation above, is defined as the temporal FT of

$$G^i(\vec{q}, t) = \exp(i\vec{q} \cdot \vec{r}_0(t)).$$

The first order correction in the density has two contributions. The first contribution is obtained by neglecting $\vec{v}_r$ in both brackets on the right hand side of eq. (14) but taking the $G$’s to first order in $n$. This results in a contribution $\mathcal{O}_{l,1}^{(1)}$ given by

$$n\mathcal{O}_{l,1}^{(1)}(\omega, \vec{\omega}) = -n\delta(\omega - \vec{\omega})\frac{1}{\lambda_l^2 + \omega^2} \frac{1}{(2\pi)^2 R^2} \int d\Omega_1 \int d\Omega_2 \int d^3qY_{l,m}^*(\Omega_1)Y_{l,-m}^*(\Omega_2)\left(\delta_{l,1}\hat{\Omega}_1 \cdot \vec{q} - \frac{q^\alpha q^\beta}{q^2}\right)\int dt e^{-i\omega t} e^{-\frac{q^2}{4}\mathcal{F}_0(t)} \phi(q, t) \frac{q^2}{6}\mathcal{F}_1(t).$$

In this general form $\mathcal{F} = \mathcal{F}_0 + \mathcal{F}_1 \cdot n$ is the total mean square displacement (MSD) of the center of mass. The above contribution vanishes, however, for cases where the bare velocity field is uncorrelated in time, eq. (9). Consider the integral over $t$ on the right hand side of eq. (9). The Dirac delta function in $\phi(q, t)$, eq. (9), sets $t = 0$. Since for $t = 0$ the MSD must vanish in any order of the expansion in $n$, $\mathcal{F}_1(0) = 0$ and the right hand side of eq. (16) vanishes.

The second contribution arises from those terms in (14) linear in $\vec{v}_r$, taking the MSD to zero order in $n$. In the linear approximation and for small deformations, the velocity field created by the deformable objects can be written as a sum of prefactors, $\mathcal{O}_{l,m}^i$ that multiply
the deformation modes of the j’th object. Considering a set of identical objects, it is obvious that \( O_{l,m}^j \) is identical for all \( j \). Thus, the superscript is dropped and \( \vec{v}_r \) is written as

\[
\vec{v}_r(\vec{r}, t) = \sum_{j \neq i} \sum_{l,m} O_{l,m}(\vec{r} - \vec{r}_0^j(t)) f_{l,m}^j(t).
\]  

The prefactors \( O_{l,m} \) are model dependent and are calculated in appendix B for deformable objects that are governed by surface tension. Using the above expression for \( \vec{v}_r \), the second contribution is given by

\[
G_{(2)}^{l,m}(\omega, \tilde{\omega}) \equiv -2 \mathcal{R} \left\{ \frac{1}{\lambda_l^2 + \omega^2} \frac{1}{2\pi^2 R^8} \int d\Omega_1 \int d\Omega_2 \int d^3 q_1 \int d^3 q_2 \right. 
\]

\[
Y_{l,m}^*(\Omega_1) Y_{l,-m}^*(\Omega_2) e^{i\vec{q}_1 \cdot \hat{\Omega}_1} e^{i\vec{q}_2 \cdot \hat{\Omega}_2} \sum_{l',m'} O_{l',m'}(\vec{q}_1) \int d\Omega_3 Y_{l',m'}^*(\Omega_3) 
\]

\[
e^{-i \vec{q}_2 \cdot \hat{\Omega}_3} R(\delta_{\gamma,\beta} - \frac{q_2^\gamma q_2^\beta}{q_2^2}) \tilde{\phi}(q_2) \left( S_q - 1 \right) \}
\]

where \( \mathcal{R}\{x\} \) denote the real part of \( x \) (for detailed derivation of equation (18) see appendix A).

Once \( O_{l,m} \) and \( \lambda_l \) of a specific system and the correlations of the bare velocity field are known, the correction to the shape correlations can be calculated using the above equations. In what follows, the actual use of the general equations is demonstrated for a specific system of deformable objects that are governed by surface tension.

### IV. DROPLETS GOVERNED BY SURFACE TENSION

To determine the shape correlations we need \( \tilde{\phi}(q) \) that describes the effect of external agents on the system. We need the set of the \( \lambda_l \)’s describing the decay of a deformation of angular momentum \( l \) of a given membrane in the absence of the bare velocity field. We also need the \( O_{l,m} \)’s that describe how the velocity field in the liquid responds to the deformation of a single object. The above quantities depend on the properties of the deformable objects and of the liquid in which they are immersed.

The system to be considered in the following is a system of membranes governed by surface tension. Namely, the energy of the membrane \( U_S \) is given by \( U_S = \lambda S \) where \( S \) is the surface area. The viscosity, \( \eta \), is assumed to be uniform inside and outside the objects. (The qualitative behaviour for different viscosities is not changed for a finite reasonable range of
viscosity ratios [24] and using identical viscosities eliminates the boundary conditions, thus simplifying the calculations considerably). For that case the $\lambda_l$’s were derived in the past [20],

$$\lambda_l = \frac{\lambda}{4\eta R} \frac{(l + 2)(l + 1)l(l - 1)}{(l + \frac{3}{2})(l + \frac{1}{2})(l - \frac{1}{2})}. \quad (19)$$

The $O_{l,m}$’s, for the same system, are needed only for large distances from the centre of the object inducing the velocity by its deformations, because the density of deformable objects is low. The leading non trivial contribution to the $O_{l,m}$ is calculated in appendix B and is given by

$$O_{l,m}(\vec{r}) = \delta_{l,2} \frac{\lambda}{\eta} \left( \frac{R}{r} \right)^2 \frac{4}{5} Y_{2,m}(\Omega) \hat{\Omega}, \quad (20)$$

where $\hat{\Omega}$ is the unit vector in the direction of $\vec{r}$.

Now, the integration over $\Omega_1$, $\Omega_2$ and $\Omega_3$ in eq. (18) can be easily done using the partial wave expansion,

$$e^{-i\vec{q} \cdot (R \hat{\Omega})} = \sum_{l=0}^{\infty} \sum_{m=-l}^{l} (-i)^l 4\pi j_l(qR) Y_{l,m}(\Omega) Y_{l,m}(\Omega). \quad (21)$$

These integrations produce a long but finite set of terms that is not presented here because of its length and complexity. To facilitate the integration over the $q$’s, the following tactics is used. The above expression is split into three parts by defining the following Fourier transforms:

$$A_{m,m'}(\vec{r}_1) = \int d\Omega_1 \int d^3q_1 \frac{e^{i\vec{q}_1 \cdot \hat{\Omega}_1 R}}{i\omega + \lambda_2 + q_1^2 D} O_{l,m'}(\vec{q}_1) Y_{l,m}(\Omega_1) \hat{\Omega}_1^m e^{i\vec{q}_1 \cdot \vec{r}_1} (2\pi)^{\frac{3}{2}}, \quad (22)$$

$$B_{m,m'}(\vec{r}_2) = \int d\Omega_2 \int d\Omega_3 \int d^3q_2 e^{i\vec{q}_2 \cdot (\Omega_2 + \Omega_3) R} (\delta_{\gamma,2} - \frac{q_2^\gamma q_2^\beta}{q_2^2}) \hat{\phi}(q_2) Y_{l,-m}(\Omega_2) Y_{l,m'}(\Omega_3) \hat{\Omega}_2^\gamma \hat{\Omega}_3^\beta e^{i\vec{q}_2 \cdot \vec{r}_2} (2\pi)^{\frac{3}{2}}, \quad (23)$$

and

$$C(\vec{r}_3) = \int d^3q_3 \left(S_{\vec{q}_3} - 1\right) \frac{e^{i\vec{q}_3 \cdot \vec{r}_3}}{(2\pi)^{\frac{3}{2}}} \quad (24)$$
Combining the above together, the first correction to the shape correlation for a dilute system is obtained by changing the order of integration and performing first the integration over the $q$'s,

$$nG_{l,1}(\omega, \tilde{\omega}) = 2R \left\{ \frac{\delta(\omega - \tilde{\omega})}{(\lambda^2 + \omega^2)R^3(2\pi)^2} \int d^3r \sum_{m'} A_{m,m'}(\vec{r})B_{m,m'}(\vec{r})C(-\vec{r}) \right\}. \quad (25)$$

Note that since the system is supposed to be invariant under rotations, the structure factor can depend only on the absolute value of $q$ and $C$ must depend only on the absolute value of $r$.

An additional property of the shape correlations is that the expression:

$$\frac{1}{2l+1} \sum_{m} \langle f_{l,m}(\omega)f_{l,m}^*(-\tilde{\omega}) \rangle$$

transforms as a scalar. Moreover, due to the rotational symmetry, the shape correlation does not depend on $m$ and therefore,

$$\langle f_{l,m}(\omega)f_{l,m}^*(-\tilde{\omega}) \rangle = \frac{1}{2l+1} \sum_{m'} \langle f_{l,m'}(\omega)f_{l,m'}^*(-\tilde{\omega}) \rangle. \quad (26)$$

In this way the coordinate system is easily rotated without loss of generality.

Last, note that the expression for $C(r)$ is, up to a prefactor of $n(2\pi)^{-\frac{3}{2}}$, no other than the pair distribution function. The pair distribution function $C$ is chosen, as a good approximation in the dilute regime, to be the pair distribution function of hard spheres system in the dilute limit,

$$C(r) = \begin{cases} 0 & \text{if } r < 2R \\ n^2 & \text{if } r > 2R. \end{cases} \quad (27)$$

A straightforward but tedious derivation, shows that the shape correlations for all $l$ except $l = 2$ vanish and for $l = 2$ it is given by the following expression:

$$G_{l,1}(\omega, \tilde{\omega}) = -\Re \left\{ \delta_{l,2} \frac{512\sqrt{2}\delta(\omega - \tilde{\omega})}{3\sqrt{\pi}(\lambda^2 + \omega^2)} \frac{\lambda}{\eta R^4} \int_{2R}^{\infty} dr \int_{0}^{\infty} dq \right. \left. \frac{3\cos(qR)qR - 3\sin(qR) + \sin(qR)q^2 R^2}{(i\omega + \lambda_2 + q^2D)q^4} \int_{0}^{\infty} dq_2 \tilde{\phi}(q_2) \sin(qr) \sin(q_2r) \right. \left. \frac{(-9\cos(q_2R)q_2R + 9\sin(q_2R) - 4\sin(q_2R)q_2^2 R^2 + \cos(q_2R)q_2^3 R^3)^2}{q_2^7} \right\}. \quad (28)$$

The integration over $q$ can be done analytically but will not be presented here due to the length of the expression. Note also that the integration over $r$ must be done last. Once $\tilde{\phi}$ is
FIG. 1: The correlations of the deformation coefficient with \( l = 2 \), \( \langle f_{2,m}^{i}(\omega)f_{2,m}^{i}(-\tilde{\omega})^* \rangle \), to zeroth order (dotted line) and first order (continuous line) in the density of objects. The units along the y axis are relative and the units along the x axis \( \omega \) are normalized by the decay rate \( \lambda_2 \).

known, the result can be calculated analytically for special cases or numerically for others. A specific example for the use of eq. (28) is depicted in fig. 1, for a correlation function that has the form of the Yukawa potential. The Fourier transform of the correlation function is given by

\[
\tilde{\phi}(q_2) = \frac{\phi_0}{(a^2 + q_2^2)},
\]

(29)

where \( a = \frac{1}{R^2} \) is used in this example. In addition \( \frac{\lambda_2 R^2}{D} = 1 \) is used. The correlation of the deformation coefficients, \( f_{l,m}^{i} \), with \( l = 2 \) is depicted to zeroth order (dotted line) and first order (continuous line) in the density of objects. The correlation of the deformation coefficients must have the form,

\[
\langle f_{l,m}^{i}(\omega)f_{l,m}^{i}(-\tilde{\omega})^* \rangle = \delta(\omega - \tilde{\omega})\Gamma_l(\omega),
\]

(30)

where \( \Gamma \) is a general function that depends on \( l \) and \( \omega \). In general, the correction that is linear in density, \( nG_1 \), must be small relatively to the zeroth order term \( G_0 \). In this example however, the density \( n \) is chosen to be large enough to observe changes. The correlations of deformation coefficients with \( l \neq 2 \) do not change to first order in the density of the deformable objects. As can be seen, the deformation modes with \( l = 2 \) are suppressed by the interaction between the droplets at low frequencies, \( \omega < \lambda_2 \). This is expected due to the
retarded response of each droplet to the external velocity field. This decay rate $\lambda_2$ introduces a new time scale that controls the decay of fluctuations produced by the external field. At low frequencies, lower than the time it takes the deformation modes to decay; the velocity field, induced by neighbouring droplets, responds effectively to the external velocity field and thus decreases the $f_{l,m}$ correlation at low frequencies.

**APPENDIX A: ON THE CALCULATION OF THE DEFORMATION CORRELATIONS**

This appendix derives the contribution of the terms involving $v_{ext}^\alpha(\vec{q},\omega_1)v_{r}^\beta(\vec{q}_2,\omega_2)$ and $v_{ext}^\alpha(\vec{q},\omega_1)v_{ext}^\beta(\vec{q}_2,\omega_2)$ in eq. (14) (i.e. $G_{l,1}^{(2)}$). These two terms are complex conjugates of each other. Thus only the first term will be considered and the correction $G_{l,1}^{(2)}$ is given by twice the real part of the answer. In order to keep the expressions to first order in the deformation and density of objects, expressions must be kept linear in $\vec{v}_r$ and $f_{l,m}$. First, the average is broken into two parts, the velocity correlation and the average over expressions containing $\vec{r}_0$ (This approximation was justified and used a number of times in the past [10, 25]).

$$G_{l,1}^{(2)}(\omega,\tilde{\omega}) = -\frac{1}{(\lambda_l + i\omega)(\lambda_l - i\tilde{\omega})R^2} R^2 \langle 2\pi \rangle$$

$$\int d\omega_1 \int d\omega_2 Y_{l,m}(\Omega) Y_{l,-m}(\Omega_2) e^{i\vec{q}\cdot\vec{r}_0} e^{i\Omega_2 R} \langle G^\alpha_l(\vec{q},\omega - \omega_1) G^\beta_l(\vec{q}_2, -\tilde{\omega} - \omega_2) \rangle$$

$$\sum_{j\neq i} \sum_{l',m'} O^\alpha_{l',m'}(\vec{q}) \int dt e^{-i\omega_1 t} e^{-i\tilde{\omega}_1 t} \int d\omega_3 e^{i\omega_3 t} \frac{1}{(\lambda_l + i\omega_3)R^2} R^2 \int d\Omega_3 \int d^3q_3 Y_{l',m'}^*(\Omega_3)$$

$$\langle v_{ext}^\alpha(\vec{q}_3,\omega_3 - \omega_1) v_{ext}^\beta(\vec{q}_2,\omega_2) \rangle$$

The Use of the expression for the correlation of the bare velocity, eq. (8), and the definition of $G^\alpha_l$, eq (15), yield
\[ G_{t,1}^{(2)}(\omega, \tilde{\omega}) = -\frac{1}{(\lambda_t + i\omega)(\lambda_t - i\tilde{\omega})R^2} (2\pi)^{-6} \int d\Omega \int d\Omega_1 \int d^3q \int d^3q_2 \]

\[ \int d\omega_1 \int d\omega_2 Y_{l,m}^{*}(\Omega) Y_{l,-m}^{*}(\Omega_1) e^{i\vec{q}\cdot \vec{\epsilon}_\Omega R} e^{i\vec{q}_2\cdot \vec{\epsilon}_{\Omega_1} R} e^{\alpha_i \vec{r}_{\Omega}} \sum_{j \neq i} \sum_{l',m'} O_{l',m'}^\alpha(q) \]

\[ \int d\omega_1 \int d\omega_2 \frac{1}{\lambda_{l'} + i\omega_3} \int d\Omega_3 Y_{l',m'}^{*}(\Omega_3) e^{-i\vec{q}_2\cdot \vec{\epsilon}_{\Omega_3} R} e^{\gamma_i \vec{r}_{\Omega_3}} (\delta_{\gamma, \beta} - \frac{q_2^2 \gamma_2^3}{q_2^2}) \phi(q_2, \omega_2) \]

\[ \left\langle G^i(\vec{q}, \omega - \omega_1) G^i(\vec{q}_2, -\omega - \omega_2) G^j(-\vec{q}, \omega_1 - \omega_3) G^j(-\vec{q}_2, \omega_3 + \omega_2) \right\rangle \]

where

\[ \left\langle \exp(i\vec{q} \cdot (\vec{r}_0(t_1) - \vec{r}_0(t_3))) \exp(i\vec{q}_2 \cdot (\vec{r}_0(t_2) - \vec{r}_0(t_4))) \right\rangle \]

The integration over \( \omega_1 \) can be done using \( \int d\omega_1 e^{i\omega_1(t_1-t_3)} = 2\pi \delta(t_1-t_3) \). Decoupling the average into a product of averages one finds that

\[ \sum_{j \neq i} \left\langle \exp\left(i\vec{q} \cdot (\vec{r}_0(t_1) - \vec{r}_0(t_3)) \right) \exp\left(i\vec{q}_2 \cdot (\vec{r}_0(t_2) - \vec{r}_0(t_4)) \right) \right\rangle \]

\[ = \sum_{j \neq i} \left\langle \exp\left(i\vec{q} \cdot (\vec{r}_0(t_1) - \vec{r}_0(t_2)) \right) \right\rangle \left\langle \exp\left(-i\vec{q} \cdot (\vec{r}_0(t_1) - \vec{r}_0(t_4)) \right) \right\rangle \]

\[ \left\langle \exp\left(i(\vec{q} + \vec{q}_2) \cdot (\vec{r}_0(t_2) - \vec{r}_0(t_4)) \right) \right\rangle \left\langle \exp\left(-i(\vec{q} + \vec{q}_2) \cdot (\vec{r}_0(t_4) - \vec{r}_0(t_4)) \right) \right\rangle \]

\[ = \exp\left(-\frac{q^2}{6} \mathcal{F}(t_1-t_2) - \frac{q_2^2}{6} \mathcal{F}(t_1-t_4) - \frac{(\vec{q} + \vec{q}_2)^2}{6} \mathcal{F}(t_2-t_4) \right) \]

\[ \frac{1}{N} \sum_i \sum_{j \neq i} \left\langle \exp\left(i(\vec{q} + \vec{q}_2) \cdot \vec{r}_0(t_4) \right) \exp\left(-i(\vec{q} + \vec{q}_2) \cdot \vec{r}_0(t_4) \right) \right\rangle, \]

where \( N \) is the number of deformable objects in the system. The reason for the decoupling of exponents is that the driving external velocity correlation decays both in time and with distance. Because of the low density and the short range repulsion (as of hard spheres in our approximate description to the first order in the density of objects) the exponents can be only weakly correlated.

The addition and subtraction of the \( i \)'th object to the sum over \( j \) results in

\[ \sum_{j \neq i} \left\langle \exp(i\vec{q} \cdot (\vec{r}_0(t_1) - \vec{r}_0(t_3)) \right) \exp(i\vec{q}_2 \cdot (\vec{r}_0(t_2) - \vec{r}_0(t_4))) \right\rangle = \]

\[ e^{-\frac{q^2}{6} \mathcal{F}(t_1-t_2)} e^{-\frac{q_2^2}{6} \mathcal{F}(t_1-t_4)} e^{-\frac{(\vec{q} + \vec{q}_2)^2}{6} \mathcal{F}(t_2-t_4)} \left(S_{\vec{q}+\vec{q}_2} - 1 \right). \]
By combining all the above together, using variable transformation and performing the integrations over \( \omega_3, \omega_2 \) and \( t_4 \), the shape correlation are derived,

\[
G_{l_1}^{(2)}(\omega, \tilde{\omega}) = -2R \left\{ \delta(\omega - \tilde{\omega}) \frac{1}{\lambda_l^2 + \omega^2} \frac{1}{(2\pi)^2} \int d\Omega \int d\Omega_2 \int d^3\mathbf{q} \int d^3\mathbf{q}_2 \right\}
\]

\[
Y_{l,m}^*(\Omega)Y_{l,-m}^*(\Omega) \sum_{\lambda} R_\lambda^\alpha(\hat{\mathbf{q}}) \int d\Omega_3 Y_{\lambda',m'}(\Omega_3) e^{-i\mathbf{q}_2 \cdot \hat{\mathbf{e}}_{\Omega_3} R_\gamma^\lambda}
\]

\[
(\delta_{\gamma,\beta} - \frac{q^2_2 q^2_2}{q^2_2}) \int_0^\infty dt_1 \int_{-\infty}^\infty dt_2 e^{-i\omega(t_1-t_2)} \phi(q_2, t_2) e^{-\lambda_l t_1} e^{-\frac{q^2}{8} \mathcal{F}(t_1)} e^{-\frac{q^2}{8} \mathcal{F}(t_2)} \left( S_{\hat{q}+\hat{q}_2} - 1 \right),
\]

from which (18) is obtained.

APPENDIX B: THE VELOCITY FIELD GENERATED BY DEFORMATION FOR THE SURFACE TENSION CASE

Consider a liquid droplet governed by surface tension that is immersed in a host liquid. Assume that the viscosities inside the droplet and in the host liquid are equal. The deformation of the shape of the object changes its energy and in response induces a force density that acts on the fluid. The force density creates in its turn an additional velocity field, denoted here as \( \mathbf{v}_\psi \),

\[
\mathbf{v}_\psi(\mathbf{r}) = \frac{1}{\eta} \int S(\mathbf{r} - \mathbf{r}') \cdot \mathbf{F}(\mathbf{r}') d^3\mathbf{r}',
\]

where \( \mathbf{F} \) is the force density created by the object and \( S \) is the Oseen tensor that is given by

\[
S_{i,j}(\mathbf{r}) = \frac{1}{8\pi} \left( \frac{\delta_{i,j}}{r} + \frac{r_i r_j}{r^3} \right)
\]

It is easy to calculate the force density using simple field theory. Let \( \psi(\mathbf{r}) \) be a three dimensional scalar field, defined everywhere in such a way that the equation \( \psi(\mathbf{r}) = 0 \) describes the surface of the object \( \mathcal{S} \). The gradient of \( \psi \) is assumed to exist and not to vanish in the vicinity of \( \psi(\mathbf{r}) = 0 \). Under the additional assumption that the deformation of
the object do not produce over-hangs, $\psi(\mathbf{r})$ is written using the deformation function, $f(\Omega)$, given in eq. (2).

$$\psi = \frac{r}{R} + f(\Omega, t) - 1. \quad (B3)$$

In [20] the force density created by a deformed objects governed by surface tension is given by,

$$\mathbf{F}(\mathbf{r}) = -\lambda (\nabla \cdot \hat{n}) \delta(\psi(\mathbf{r})) \nabla \psi(\mathbf{r}), \quad (B4)$$

where $\hat{n} = \frac{\mathbf{V}_\psi}{|\nabla \psi|}$ is a unit vector in the direction normal to the surface of the deformable object.

In addition, the following symbols are used for the angular parts of the gradient and Laplacian,

$$D = \hat{\theta} \frac{\partial}{\partial \theta} + \hat{\phi} \frac{1}{\sin(\theta)} \frac{\partial}{\partial \phi} \quad (B5)$$

and

$$D^2 = \frac{1}{\sin(\theta)} \frac{\partial}{\partial \theta} \left( \sin(\theta) \frac{\partial}{\partial \theta} \right) + \frac{1}{\sin^2(\theta)} \frac{\partial^2}{\partial \phi^2}, \quad (B6)$$

where $\hat{\theta}$ and $\hat{\phi}$ are unit vectors of $\theta$ and $\phi$. The velocity field $\mathbf{v}_\psi$ is obtained as a function of $f(\Omega', t)$ by the use of equations (B1) and (B3),

$$\mathbf{v}_\psi(\mathbf{r}) = -\frac{\lambda R}{8\pi \eta} \int d\Omega' \left[ \left( \frac{2}{X} + \frac{\mathbf{X} \cdot \hat{\Omega}'}{X^3} \mathbf{X} - \frac{2}{X^3} R\mathbf{X} + 6 \frac{(\mathbf{X} \cdot \hat{\Omega})^2}{X^5} R\mathbf{X} \right) f(\Omega') \right.$$

$$- \left( \frac{2Df(\Omega')}{X} + \frac{2\mathbf{X} \cdot (Df(\Omega'))}{X^3} \mathbf{X} \right) \left( \hat{\Omega}' + \frac{Df(\Omega')}{1-f} \right) - \left( \hat{\Omega}' + \frac{\mathbf{X} \cdot \hat{\Omega}'}{X^3} \right) D^2 f(\Omega') \left] \right| (1-f)^2 d\Omega'.$$

Keeping the above expression to the first order of the deformation $f$ results with

$$\mathbf{v}_\psi(\mathbf{r}) = \frac{\lambda R}{8\pi \eta} \int d\Omega' \left[ \left( \frac{2\hat{\Omega}'}{X} + \frac{2\mathbf{X} \cdot \hat{\Omega}'}{X^3} \mathbf{X} - \frac{2}{X^3} R\mathbf{X} + 6 \frac{(\mathbf{X} \cdot \hat{\Omega})^2}{X^5} R\mathbf{X} \right) f(\Omega') \right.$$

$$- \left( \frac{2Df(\Omega')}{X} + \frac{2\mathbf{X} \cdot (Df(\Omega'))}{X^3} \mathbf{X} \right) \left( \hat{\Omega}' + \frac{Df(\Omega')}{1-f} \right) - \left( \hat{\Omega}' + \frac{\mathbf{X} \cdot \hat{\Omega}'}{X^3} \right) D^2 f(\Omega') \left] \right| (1-f)^2 d\Omega'.$$
\[ \vec{X} \equiv \vec{r} - R \hat{\Omega}' \]

where \( f \) is replaced by its expansion in spherical harmonics \( f(\Omega, t) = \sum_{l=2}^{\infty} \sum_{m=-l}^{l} f_{l,m}(t)Y_{l,m}(\Omega) \). The spherical harmonics are eigenvalues of \( D^2 \). In contrast, \( D \) mixes different harmonics. The density of objects is assumed to be low and therefore the distance \( r \) to the closest droplet is typically much larger than the radius of the droplet \( R \). Hence, the velocity is expanded to the first nontrivial order in \( \frac{R}{r} \). In addition, a rotated coordinate system is used in which \( \vec{r}' = r\hat{z}' \), where \( \hat{z}' \) is the \( z \) direction in the rotated coordinate system. In that system,

\[ \vec{v}_\psi(r\hat{z}', t) = \frac{\lambda}{\eta} \left( \frac{R}{r} \right)^2 \sqrt{\frac{2}{5\pi}} f'_{2,0} \hat{z}', \quad (B9) \]

where \( f'_{2,0} \) is the deformation coefficient of \( Y'_{2,0} \) in the rotated coordinate system. The velocity field in a general direction is given by a rotation of the coordinate system. The transformation under rotation of the spherical harmonics (and thus of \( f_{2,0} \)) is given by the addition theorem,

\[ P_n(\cos(\gamma)) = \frac{4\pi}{2n+1} \sum_{m=-n}^{n} Y_{n,m}(\theta_1, \varphi_1)Y_{n,m}^*(\theta_2, \varphi_2), \quad (B10) \]

where \( \gamma \) is the angle between the directions \( \hat{\Omega}_1 \) and \( \hat{\Omega}_2 \) that corresponds to \( (\theta_1, \varphi_1) \) and \( (\theta_2, \varphi_2) \). Note that the calculation of \( f'_{2,0} \) involves integration over all angles that produces a dependence of the induced velocity on the angles at the original coordinate system. Thus the induced velocity in a general direction is given by,

\[ \vec{v}_\psi(\vec{r}) = \frac{\lambda}{\eta} \left( \frac{R}{r} \right)^2 \frac{4}{5} \sum_{m=-2}^{2} Y_{2,m}(\Omega)f_{2,m}(t)\hat{r}, \quad (B11) \]

where \( \hat{r} \) is a unit vector in the direction of \( \vec{r} \). \( O_{l,m} \) is obtained by comparing eq. (17) with eq. (B11),

\[ O_{l,m} = \delta_{l,2} \frac{\lambda}{\eta} \left( \frac{R}{r} \right)^2 \frac{4}{5} Y_{2,m}(\Omega) \hat{r}. \quad (B12) \]

Hence, the only spherical harmonics modes that contribute to the velocity field, far from the object, are the terms with \( l = 2 \).

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