Shape fluctuations of a deformable body in a randomly stirred host fluid

Gad Frenkel and Moshe Schwartz
Raymond and Beverly Sackler Faculty of Exact Sciences, School of Physics and Astronomy, Tel Aviv University, Ramat Aviv 69978, Israel
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We consider a deformable body immersed in an incompressible fluid that is randomly stirred. Sticking to physical situations in which the body departs only slightly from its spherical shape, we investigate the deformations of the body. The shape is decomposed into spherical harmonic modes. We study the correlations of these modes for a general class of random flows that include, as a special case, the flow due to thermal agitation. Our results are general, in the sense that they are applicable to a large class of deformable bodies with energy that depends only on the shape of the body, and a general class of random flows.

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I. INTRODUCTION

In recent years we have witnessed a dramatic increase in the volume of research of deformable objects in a host liquid [1–13]. The immersed objects that are of scientific or industrial interest are very diverse, giving rise to a large number of subfields. The objects can be bulk objects, like droplets of a different liquid [11–16] or elastic colloidal particles [17,18], or they can be membranes separating various regions of the host liquid [19]. Membranes can be categorized as liquid membranes [1,5,11,12,20] or elastic membranes [21–23]. Liquid membranes are characterized by an energy that depends only on the shape of the membrane and liquid properties on the surface of the membrane like the local density of the membrane molecules [2,20,24]. Elastic membranes are characterized by additional fields like tangential strains, etc. While the description of the immersed objects is very detailed, the description of the host liquid is usually restricted to properties like viscosity and temperature [1,3,9,12,13,19]. The effect of temperature on diffusion and fluctuations of the deformations has been studied in numerous articles [1,3,10,12,13,19,25], giving the diffusion constant and correlations of the deformations of a single deformable body at equal times and at a general time separation. The effect of temperature has been recently studied even for the case of objects that are stretched by external shear [11]. While the effect of thermal agitation on deformable objects in solution is obviously very important, it is certainly not the only way in which the host liquid may be agitated. Clearly, in industrial and biological environments, the host liquid can be stirred, vibrated, pumped, etc. For instance, Wu and Libchaber have studied the effect of bacteria motion on the diffusion of small beads and have suggested that the source for the Brownian-like motion is the collective motion of the bacteria [26]. Further, nanoscale mechanical fluctuations of the red blood cell surface have been measured and shown to depend strongly on the biochemical environment and not only on temperature [27–30]. In fact, in many cases the resulting agitation is much more important to the behavior of the deformable objects than thermal agitation. The purpose of the present paper is to contribute to the understanding of such more general systems by viewing them in a unified way, such that the case of thermal agitation will be shown to be just a particular case and will be worked out as an example of the general approach.

Clearly, the actual treatment of a system of many interacting objects is extremely difficult. This is because such a system is not just a many-body system of objects interacting via the hydrodynamic interactions, but each object is characterized by an infinite number of degrees of freedom, corresponding to its possible deformations. All of these deformations interact. The problem is somewhat simplified if the deformation of the objects from spherical shape remains small and the fluid is in the linear regime, as in the case of the Stokes approximation to the Navier-Stokes equation.

Our final goal is to obtain the response of the composite system, of deformable interacting objects, to a given velocity field imposed on the liquid. The velocity field we have in mind may be fixed in time, like simple shear, or randomly fluctuating in time and space. Even in the first case the velocity field each object experiences must have a random part, due to the random passage of other objects nearby. Therefore effects of random flow are important to the understanding of such systems. First we wish to know the response of a single deformable object to a general random velocity field. Once the response of a single object is known, we can obtain the response of the full, many-body system, by using the response of each body as a source of an additional velocity field. In a former paper [10] we studied the motion of the center of a deformable object in the presence of a random flow and derived its mean-squared displacement (MSD). In this paper, we investigate the deformation degrees of freedom, completing thus the description of the response of a single object to a random velocity field. Decomposing the deformation into spherical harmonic modes, we consider the correlations between deformation modes. We find, among other things, that different modes are decoupled and that the correlation function does not depend on the parameter m of the $Y_{l,m}$ spherical harmonic (following from spherical symmetry). We also obtain a method to calculate these correlations as a function of time, build typical drop shapes from the correlation functions, and discuss several interesting cases.

The plan of this paper is as follows: In Sec. II we describe the system we have in mind and formulate the basic equations. In Sec. III we introduce the deformation coefficients and obtain their correlations for the simple case where the external velocity is uncorrelated in time. General correlation functions are considered in Sec. IV. We obtain the deformation correlations and simplify them for equal times. An algorithm for numerical computation of the correlations in the general case is described in the Appendix.

II. SYSTEM

Consider a single deformable body immersed in a host fluid.

(i) The deformable body is fluid, in the sense that the velocity field is well defined everywhere (both inside and outside the body). Each surface element moves with the velocity of the flow at its position:

$$\dot{\vec{r}}=\vec{v}(\vec{r}). \tag{1}$$

(ii) Both the body and the host fluid are incompressible, $\vec{\nabla}\cdot\vec{v}=0$.

(iii) The body is characterized by an energy that depends on its shape (i.e., changing the orientation or switching places of two surface particles while keeping the shape constant does not change the energy). The energy may be surface tension energy [31], Helfrich bending energy [20,24], etc. The shape of minimum energy is a sphere. Deformation of the shape changes the energy, exerts a force density on the liquid, and therefore generates an additional velocity field, denoted by $\vec{v}_\psi$.

(iv) We investigate the regime where the hydrodynamic equations are linear in the velocity (i.e., a velocity field induced by several sources is equal to the sum of the velocity fields that each source induces separately). For instance, if the flow is governed by the Navier-Stokes equation, then our assumption implies that the Reynolds number is small and that the Stokes approximation is applicable. The linearity implies that in our system the actual velocity field is the sum of the imposed velocity field $\vec{v}_{ext}$ (the velocity field that would have existed if the body was absent), and the velocity field induced by the deformations, $\vec{v}_\psi$,

$$\vec{v}=\vec{v}_{ext}+\vec{v}_\psi. \tag{2}$$

(v) We consider cases in which the external velocity field is random, with zero average. The velocity correlation function is known and depends only on distance and time difference. We also assume that the external velocity is small enough to allow the body to remain almost spherical.

III. DEFORMATIONS UNDER WHITE NOISE FLOW

Consider a spherical body which is slightly deformed. The equation

$$\frac{\rho}{R}+f(\Omega,t)-1=0 \tag{3}$$

defines its surface, yielding for each spatial direction Ω the distance, $\rho\equiv|\vec{r}-\vec{r}_0|$, of the surface from the center of the body $\vec{r}_0$. R is the radius of the undeformed sphere. The deformation function $f(\Omega,t)$ defines the shape. The deformation function is decomposed into spherical harmonics, $f(\Omega,t)=\Sigma_{l=1}^{\infty}\Sigma_{m=-l}^{l}f_{lm}(t)Y_{lm}(\Omega)$ (clearly the Y_{00} term can be absorbed in the definition of R). Our goal is to obtain the correlations between the deformation coefficients $f_{lm}(t)$.

The center of the body $\vec{r}_0$ is chosen to be the point around which the deformation coefficients with $l=1$ vanish: $f_{1m}=0$. A different definition of the center will introduce three additional equations for the deformation coefficients with $l=1$. We are not interested in those since in the first order the spherical harmonics with $l=1$ describe a translation of the body [10,14,2]. Let $\psi(\vec{r})$ be a three-dimensional scalar field, defined everywhere in such a way that the equation $\psi(\vec{r})=0$ describes the surface of the body [2,5]. The gradient of ψ is assumed to exist and not to vanish in the vicinity of $\psi(\vec{r})=0$. Straightforward manipulation of Eq. (1) gives rise to a continuity equation for ψ, presented here in a coordinate system that moves with the center of the body:

$$\dot{\psi}+\vec{v}_\psi\cdot\vec{\nabla}\psi=-(\vec{v}_{ext}-\dot{\vec{r}}_0)\cdot\vec{\nabla}\psi. \tag{4}$$

Assuming that $|\vec{v}_{ext}-\dot{\vec{r}}_0|$ is small on the surface and expressing the field ψ in the vicinity of the surface as

$$\psi=\frac{\rho}{R}+f(\Omega,t)-1, \tag{5}$$

the right-hand side of Eq. (4) is equal, in the first order, to $Q\equiv(1/R)\{\hat{\rho}\cdot[\vec{v}_{ext}-\dot{\vec{r}}_0]\}$ (see Ref. [2]), where $\hat{\rho}$ is a unit vector directed outwards from the center of the body. Since the minimum energy of the body is obtained for a spherical shape, the velocity induced by the body is zero when the sphere is undeformed. Therefore the leading order of the velocity $\vec{v}_\psi$ must be, in general, a linear functional of the deformation $f(\Omega,t)$. The term $\vec{v}_\psi\cdot\vec{\nabla}\psi$ is obtained in the leading order by taking $\vec{\nabla}\psi$ on the original sphere and $\vec{v}_\psi$ to first order in the deformation. Using Eq. (5) for ψ, the generic equation for the deformation coefficient f_{lm} must be of the form

$$\frac{\partial f_{lm}(t)}{\partial t}+\lambda_l f_{lm}(t)=-Q_{lm}(t), \tag{6}$$

where Q_{lm} is given by

$$Q_{lm}=\frac{1}{R}\int d\Omega\{\hat{\rho}\cdot[\vec{v}_{ext}-\dot{\vec{r}}_0]Y^*_{l,m}(\Omega)\} \tag{7}$$

and $\vec{v}_{ext}$ is evaluated on the undeformed body in the direction of the spatial angle Ω (for further details, see Ref. [10]). The above equation is a nonhomogeneous linear equation, where the nonhomogeneity is due to the random driving external flow. The homogeneous part of the equation does not depend on the external flow and represents the decay of deformations in the absence of external flow. Therefore there is no system of coordinates that may be preferred over others (even locally at a given point in space and time). This is the reason that the decay eigenvalues λ_l depend only on l and not on m. Otherwise the decay of a given shape depends on a choice of coordinates and this is impossible. An important point to notice is that different physical deformable objects, obeying the conditions outlined in Sec. II, differ only in the λ_l's.

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 we can simply use a projection operator on the transversal part of a general field:

$$\tilde{v}_{ext_i}(\vec{q}) \equiv \sum_j \left(\delta_{ij} - \frac{q_i q_j}{q^2} \right) u_j(\vec{q}), \tag{8}$$

where $\vec{u}$ is a general vector field and the bracketed term is the projection operator that removes the longitudinal part of $\vec{u}$, and therefore yields a general transversal velocity field $\tilde{\vec{v}}_{ext}$. Next, the correlations of the external velocity are easily expressed using the correlations of the general field $\vec{u}$. We are interested in cases were the system is statistically isotropic, homogeneous, and stationary. In these cases, the general field must obey:

$$\langle u_l(\vec{q},t_1) u_m(\vec{p},t_2) \rangle = \delta_{lm} \delta(\vec{q}+\vec{p}) \phi(q,t_2-t_1), \tag{9}$$

where δ_{lm} is the Kronecker delta, δ is the Dirac delta function, and ϕ is a general function of q and the time difference. In addition we assume that

$$\langle u_l(\vec{q},t) \rangle = 0. \tag{10}$$

In the rest of this section we consider a frequently used family of random flows in which the external velocity is uncorrelated in time,

$$\phi(q,t_2-t_1) = \tilde{\phi}(q)\, \delta(t_2-t_1). \tag{11}$$

Clearly, any random process of physical origin cannot have strict δ function correlations. Equation (11) above is a reasonable approximation for systems in which the decay times of the velocity correlations are much shorter than the decay times of the deformations, $1/\lambda_l$. In fact, even a weaker restriction suffices. Fourier components of the velocity field with wave vector $\vec{q}, \vec{v}_{\vec{q}}$, obeying $qR \ll 1$ are not relevant for the description of the deformation of the shape because they correspond to variations over length scales much larger than the size of the deformable object. Therefore it is enough that the q-dependent decay times of the correlations involving $\vec{v}_{\vec{q}}$ and $\vec{v}_{-\vec{q}}$ are short compared to the decay times of the deformations only for $qR > 1$. In these systems we can replace the exact correlation details with the effective delta function the strength of which is obtained by integrating the true correlation over time.

Transforming, we can calculate the correlation of the velocity at two points on the sphere, that are characterized by the directions $\hat{r}$ and $\hat{r}'$ relative to the center. This yields

$$\langle v^i_{ext}(\vec{r},t_1) v^j_{ext}(\vec{r}',t_2) \rangle = \int d\vec{q} e^{-i\vec{q}\cdot(\vec{r}-\vec{r}')} \left[\delta_{ij} - \frac{q_i q_j}{q^2} \right] \times \tilde{\phi}(q)\, \delta(t_2-t_1), \tag{12}$$

where $\vec{v}_{ext}(\vec{r},t)$ is the velocity at time t at place $\vec{r}$ on the surface and $\vec{r} \equiv \vec{r}_0(t) + R\hat{r}$. The average and correlations of Q_{lm} follow easily from the previous equations. The average of Q_{lm} is zero. The term $\hat{\rho}\cdot\dot{\vec{r}}_0$, in Eq. (7), does not contribute to any component of Q_{lm} except for those with $l=1$. In addition, the center $\vec{r}_0$ has been chosen to be the point around which the three deformation coefficients f_{lm} with $l=1$ are equal to zero. Therefore Q_{1m} is zero, and for any other l, $\dot{\vec{r}}_0$ can be dropped out of the expression for Q_{lm}.

Straightforward calculation of the Q_{lm} correlations, using its definition, Eq. (7), and the velocity correlations, Eqs. (8)–(11), yields

$$\langle Q_{lm}(t_1) Q_{l'm'}(t_2) \rangle = \mathbf{Q}_{ll'mm'}\, \delta(t_2-t_1), \tag{13}$$

where

$$\mathbf{Q}_{ll'mm'} \equiv \frac{1}{R^2} \int d\Omega \int d\Omega' \int d^3q Y^*_{lm}(\Omega) Y^*_{l'm'}(\Omega') \times \sum_{i,j=x,y,z} \left[\hat{r}_i \hat{r}'_j e^{-i\vec{q}\cdot(\hat{r}-\hat{r}')R} \left(\delta_{ij} - \frac{q_i q_j}{q^2} \right) \tilde{\phi}(q) \right]. \tag{14}$$

The spherical symmetry of the system implies that only for the terms for which $l'=l$ and $m'=-m$ differ from zero, and those terms do not depend on m. Hence

$$\mathbf{Q}_{ll'mm'} = \mathbf{Q}_{ll00} \delta_{l',l} \delta_{m',-m}. \tag{15}$$

The correlations of the deformation coefficients f_{lm} are obtained, using Eq. (6) and the correlations of Q_{lm}, by direct integration,

$$\lim_{t\to\infty} \langle f_{lm}(t) f_{l'm'}(t+\Delta t) \rangle = \int_{-\infty}^{0} dt_1 \int_{-\infty}^{\Delta t} dt_2 [\langle Q_{lm}(t_1) Q_{l'm'}(t_2) \rangle e^{\lambda_l t_1 + \lambda_{l'}(t_2-\Delta t)}], \tag{16}$$

where the limit $t\to\infty$ is taken to avoid the initial conditions of the deformation.

Finally, Eqs. (13), (15), and (16) yield

$$\langle f_{lm}(t) f_{l'm'}(t+\Delta t) \rangle_{t\to\infty} = \mathbf{Q}_{ll00} \frac{e^{-\lambda_l |\Delta t|}}{2\lambda_l} \delta_{l',l} \delta_{m',-m}. \tag{17}$$

For equal times, $\Delta t = 0$, these correlations, that are just the variances of the f_{lm}, are important because they contain the full statistical information about possible shapes of the objects. In Fig. 1 these correlations are used to generate typical

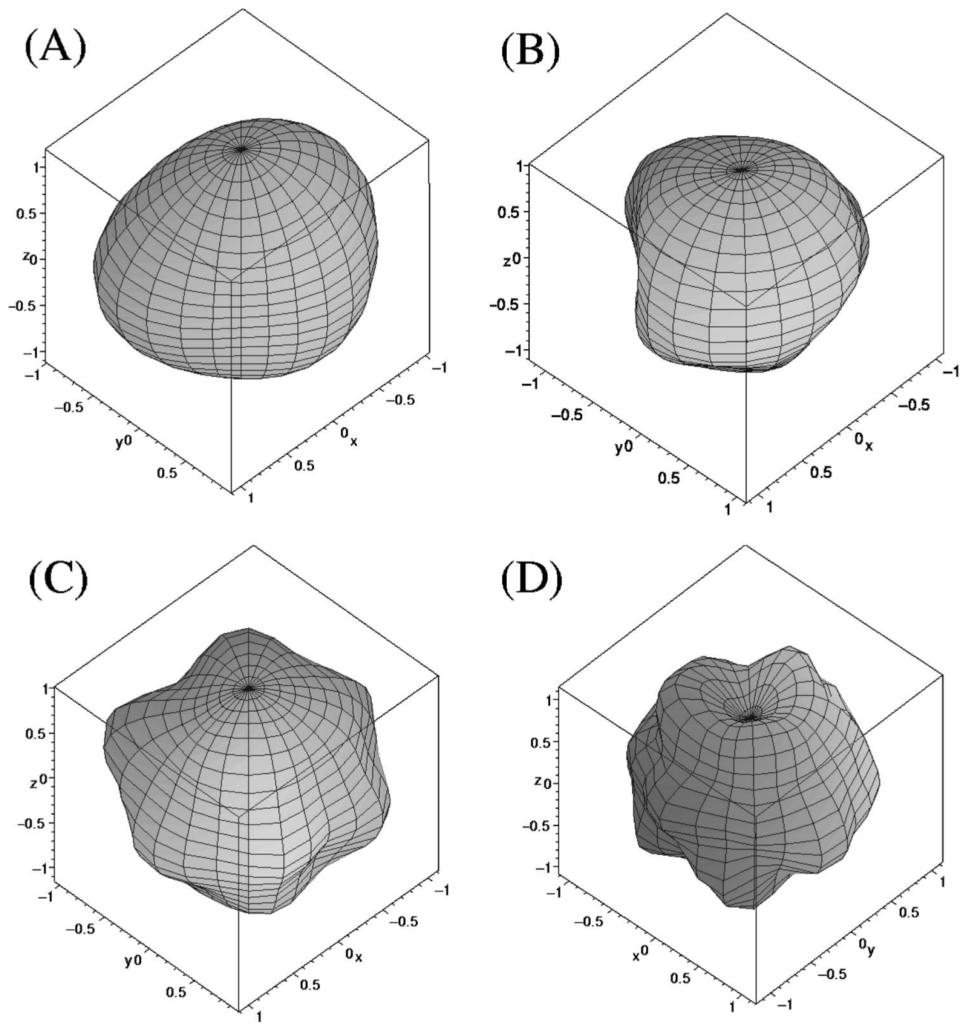

FIG. 1. A typical realization of a deformable body subjected to a random flow of the form $\phi(q) = C\delta(q\xi - 1)\delta(t)$ with (A) $R = 2\xi$, (B) $R = 4\xi$, (C) $R = 6\xi$, and (D) $R = 8\xi$.

shapes of droplets governed by surface tension and random flow given by $\phi(q,t) = C\delta(\xi q - 1)\delta(t)$. As can be seen, the surface of the body develops bumps. It is obvious that the typical size of these surface features depends on the ratio $\mu_1 = R/\xi$. As μ_1 increases, different surface elements become less and less correlated. Therefore, we expect to see features of smaller size (which correspond, clearly, to spherical harmonics of higher order). The smallest features correspond to deformation coefficients with $l \simeq \mu_1$ (or $l = 2$ if $\mu_1 \leqslant 2$). Figures 2 and 3 depict equal time correlations as function of l for two correlation functions of the form $\tilde{\phi}(q) = Cq^{-\alpha}G(\xi q)$, where $\alpha = \pm 2$, G is a function that has a cutoff at $\xi q = 1$ and ξ is the decay length scale. There are two independent dimensionless parameters, $\mu_1 = R/\xi$ and $\mu_2 = C/(R^{5-\alpha}\lambda_l^{min}$, where λ_l^{min} is the minimal value of the λ_l's ($l = 2 \cdots \infty$). In the case of a body characterized by surface tension energy, $\lambda_l \propto \lambda/\eta R$ where λ is the surface tension and η is the viscosity. We vary μ_1 and μ_2 by keeping $\xi = 1$ and $C\eta/\lambda = 1$ and changing R. As can be seen, there are two possibilities: either the $l = 2$ term dominates the curve or there is a maximum at $l_0 \approx R/\xi$. It is easy to see that the decay for $l \gg l_0$ is exponential. This suggests that there is a cutoff on the deformation coefficients at which the expansion can be terminated and therefore that the expansion we use here will be useful for systems in which μ_1 is small. The correlation functions depicted in Fig. 3 correspond, in the limit $R/\xi \ll 1$, to velocity correlations due to thermal agitation. Due to the importance of that problem we work out in the following a full analytic derivation of the time-dependent shape correlations. Such correlations were obtained in the past by various methods (e.g., equipartition of energy, Kirkwood equation, etc.). In a previous paper [10] we have shown that the correlation function for the external velocity due to thermal agitation has the form $\phi(q,t) = [K_B T/(2\pi)^3\eta][\delta(t)/q^2]$, where η is the viscosity of the fluid (notice that this holds for $q\xi \ll 1$ where ξ is of the order of the intermolecular distance. For $R/\xi \gg 1$ it is easy to show that the cutoff on q can be ignored). It is important to note that our velocity correlations are not obtained by matching our general result, Eq. (14), with previous results on correlations of deformations in the presence of thermal agitation. Indeed, we obtain the equilibrium velocity correlations in Ref. [10] from a general argument not related at all to the problem of deformable objects. We substitute the thermal velocity correlation given above into Eq. (14) and using the dimensionless parameter $\vec{y} = \vec{q}R$ we obtain

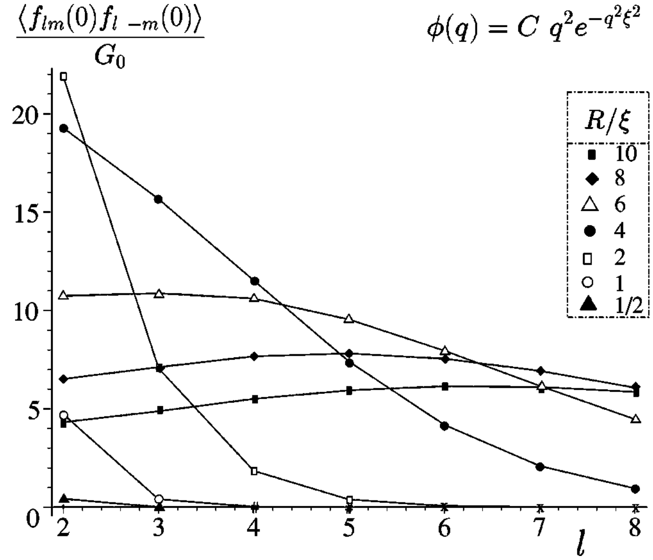

FIG. 2. The correlation of the deformation coefficients f_{lm} as a function of integer l, for $\phi(q) = Cq^2 e^{-q^2\xi^2}$ and several values of R/ξ ($G_0 \equiv \mu_2$). In the figure, # denotes points where $R/\xi = 1/2$ and $R/\xi = 1$ have the same value. x denote points where $R/\xi = 1/2,1,2$ values coincide.

$$\mathbf{Q}_{llm,-m} \equiv \sum_{i,j=x,y,z} \frac{1}{R^2} \int d\Omega \int d\Omega' Y^*_{lm}(\Omega) Y^*_{l,-m}(\Omega') \hat{r}_i \hat{r}'_j A_{ij}, \tag{18}$$

where

$$A_{ij} = \frac{1}{R} \int d\Omega_y \int y^2 dy e^{-\vec{y}\cdot(\hat{r}-\hat{r}')} \left[\delta_{ij} - \frac{y_i y_j}{y^2} \right] \left(\frac{K_B T}{(2\pi)^3 \eta} \frac{1}{y^2} \right). \tag{19}$$

We rotate the y coordinate system in such a way that its $\hat{z}$ axis is in the direction of $(\hat{r} - \hat{r}')$. Furthermore, we use the fact that $\int_{-\infty}^{\infty} dy \exp(-iyx) = 2\pi\delta(x)$ and calculate the rotated tensor,

$$\tilde{A}_{ij} = \frac{\pi^2}{R|\hat{r} - \hat{r}'|} \left(\frac{K_B T}{(2\pi)^3 \eta} \right) (1 + \delta_{i,\tilde{z}}) \delta_{i,j}. \tag{20}$$

Rotating the axes back we find that

$$\sum_{i,j} \hat{r}_i \hat{r}'_j A_{ij} = \frac{\pi^2}{R|\hat{r} - \hat{r}'|} \left(\frac{K_B T}{(2\pi)^3 \eta} \right) \times \left[\hat{r} \cdot \hat{r}' + \frac{\hat{r} \cdot (\hat{r} - \hat{r}')}{|\hat{r} - \hat{r}'|} \frac{\hat{r}' \cdot (\hat{r} - \hat{r}')}{|\hat{r} - \hat{r}'|} \right] = \frac{\pi^2}{2R} \left(\frac{K_B T}{(2\pi)^3 \eta} \right) \frac{3 \cos \gamma - 1}{\sqrt{2}\sqrt{1 - \cos \gamma}}, \tag{21}$$

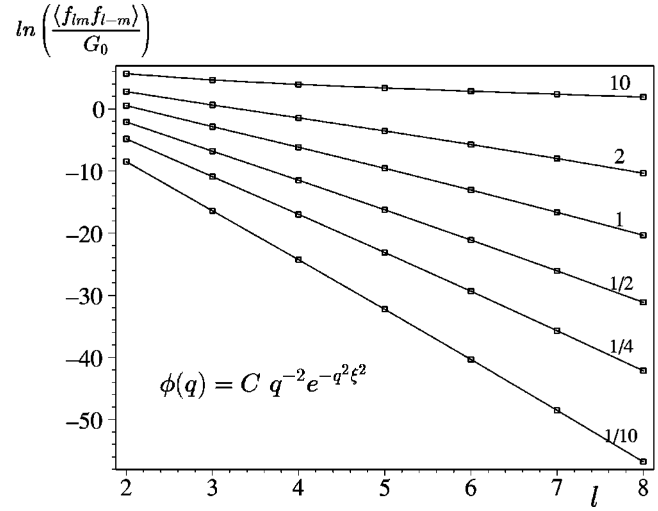

FIG. 3. The correlation of the deformation coefficients f_{lm} as a function of integer l, for $\phi(q) = Cq^{-2} e^{-q^2\xi^2}$ and several values of R/ξ. (The values are given above each corresponding line.) Note that for $R \gg \xi$ the curves coincide with the curves for thermal agitation ($G_0 \equiv \mu_2$).

where γ is the angle between $\hat{r}$ and $\hat{r}'$. Next, we develop $1/\sqrt{1 - \cos \gamma}$ in Legendre polynomials, use recurrence relations and the addition theorem

$$P_l[\cos(\theta)] = \frac{4\pi}{2l+1} \sum_{m=-l}^{l} (-1)^m Y_{lm}(\Omega) Y_{l-m}(\Omega'), \tag{22}$$

and find

$$\mathbf{Q}_{llm,-m} = \frac{2K_B T}{\eta R^3} \frac{(l+1)l}{(2l-1)(2l+1)(2l+3)}. \tag{23}$$

For a deformable body governed by surface tension [2],

$$\lambda_l = \frac{\lambda}{4\eta R} \frac{(l+2)(l+1)l(l-1)}{(l+\frac{3}{2})(l+\frac{1}{2})(l-\frac{1}{2})}. \tag{24}$$

Therefore, in the case of a body governed by surface tension under thermal agitation, Eqs. (17), (23), and (24) imply that the correlations of the deformations are given by

$$\langle f_{lm}(t) f_{l'm'}(t + \Delta t) \rangle = \frac{K_B T}{\lambda R^2} \frac{1}{(l-1)(l+2)} e^{-\lambda_l |\Delta t|} \delta_{l',l} \delta_{m',-m}. \tag{25}$$

Schwartz and Edwards [6] considered the special case of a deformable body in equilibrium at temperature T, using the Kirkwood equation. They found identical correlations in the deformations. That derivation, however, has been tailored for thermal agitation and cannot be generalized to take into account any other type of correlations in the host fluid.

So far we have shown how to implement Eq. (17) for flows that are uncorrelated in time to obtain the correlations in the deformations of the shape. In the following we generalize to correlations that are not instantaneous in time.

IV. GENERAL NOISE

In many cases white noise correlations are not sufficient to describe what really happens in the liquid, especially if the correlation time is of the order of other time parameters. Such is the case of a system of many droplets immersed in a host fluid. The random flow a droplet is subjected to results from the random motion and deformation of other droplets that pass nearby. It is obvious that in this case the approximation of the flow to be uncorrelated in time is not justified. Hence we need to generalize our description.

Suppose that $\phi(q,\Delta t)$ is a general function of q and the time differences Δt. The correlations of the external velocity are now extended in time. In order to calculate averages on the droplet at different times, we must now consider also the motion of the droplet. The definition of Q_{lm}, Eq. (7), implies that the correlations of the normal component of the external velocity field on the sphere (that to first order of the deformation is an adequate approximation of the external velocity field on the surface of the body) are

$$\langle Q_{lm}(t_1)Q_{l'm'}(t_2)\rangle = \frac{1}{R^2}\int d\Omega \int d\Omega' Y^*_{lm}(\Omega)Y^*_{l'm'}(\Omega') \times \sum_{i,j=x,y,z} [\hat{r}_i \hat{r}'_j \langle v^i_{ext}(\hat{r},t_1) v^j_{ext}(\hat{r}',t_2)\rangle]. \tag{26}$$

The correlation of the velocity at two points on the droplet, located in the directions $\hat{r}$ and $\hat{r}'$ and measured at different times, obviously depends on the displacement of the center $\Delta\vec{r}_0$. We first calculate the correlation for a general displacement of the center and then average the result according to the probability of finding the center at each point. To do this, we first express the velocity correlations by means of the Fourier transform of the velocity, use Eqs. (8) and (9), and obtain

$$\langle v^i_{ext}(\hat{r},t_1) v^j_{ext}(\hat{r}',t_2)\rangle = \int P(\Delta\vec{r}_0)d(\Delta\vec{r}_0)\int d^3q e^{-i\vec{q}\cdot(\Delta\vec{r}_0+R(\hat{r}-\hat{r}'))} \times\left(\delta_{ij}-\frac{q_iq_j}{q^2}\right)\phi(q,\Delta t), \tag{27}$$

where $\Delta\vec{r}_0=\vec{r}_0(t_1)-\vec{r}_0(t_2)$, $\Delta t=t_2-t_1$, $P(\Delta r_0)$ is the probability that the center will be displaced by $\Delta\vec{r}_0$ in the period of Δt.

It is obvious now that averaging over the center displacements will affect only the term $-i\vec{q}\cdot\Delta\vec{r}_0$ in the exponent since this is the only term that depends on $\Delta\vec{r}_0$. Consequently,

$$\langle v^i_{ext}(\hat{r},t_1) v^j_{ext}(\hat{r}',t_2)\rangle = \int d^3q\langle e^{-i\vec{q}\cdot\Delta\vec{r}_0}\rangle e^{-i\vec{q}\cdot(\hat{r}-\hat{r}')R} \times\left(\delta_{ij}-\frac{q_iq_j}{q^2}\right)\phi(q,\Delta t). \tag{28}$$

Assuming Gaussian distribution of the displacements of the center,

$$\langle e^{-i\vec{q}\cdot\Delta\vec{r}_0}\rangle = e^{-(q^2/6)\langle(\Delta\vec{r}_0)^2\rangle}. \tag{29}$$

In a previous paper [10], we considered the mean-squared displacement (MSD) of the center of a deformable body in a flow that is correlated in a general way. We found that the MSD in a period of time Δt obeys the equation

$$F(\Delta t) = 16\pi\int_0^{\Delta t} dt'\int_0^\infty e^{-(q^2/6)F(t')}\phi(q,t') \times[j_0(qR)+j_2(qR)]^2(\Delta t - t')q^2 dq, \tag{30}$$

where $F(\Delta t)\equiv\langle(\Delta\vec{r}_0)^2\rangle$ and $j_n(x)$ is the spherical Bessel function of order n. Therefore the correlation of the external velocity at two points on the surface, characterized by the directions $\hat{r}$ and $\hat{r}'$, measured at two different times with a time gap of Δt, is given by

$$\langle v^i_{ext}(\hat{r},t_1) v^j_{ext}(\hat{r}',t_2)\rangle = \int d^3q e^{-(q^2/6)F(\Delta t)} e^{-i\vec{q}\cdot(\hat{r}-\hat{r}')R} \times\left(\delta_{ij}-\frac{q_iq_j}{q^2}\right)\phi(q,\Delta t). \tag{31}$$

Equations (30) and (31) enable us to calculate the correlations of Q_{lm} [Eq. (26)]. Again, spherical symmetry implies that these correlations are nonzero only for $l'=l$ and $m'=-m$. Note that if the correlations were to be calculated at fixed $\Delta\vec{r}_0$ instead of fixed time difference, spherical symmetry would have been violated. Spherical symmetry holds in our case because the probability of having a given $\Delta\vec{r}_0$ for a given Δt, is a function of the absolute value of $\Delta\vec{r}_0$.

The correlations of the deformation coefficients can be obtained from their basic equation (6) using the correlations of Q_{lm},

$$\langle f_{lm}(t)f_{l'm'}(t+\Delta t)\rangle_{t\to\infty} = \int_{-\infty}^0 dt_1\int_{-\infty}^{\Delta t} dt_2\langle Q_{lm}(t_1)Q_{l-m}(t_2)\rangle \times e^{\lambda_l(t_1+t_2-\Delta t)}\delta_{l',l}\delta_{m',-m}. \tag{32}$$

Equations (26), (30), (31), and (32) form the calculation method for the correlations of the deformation coefficients. The method presented here may be hard to implement numerically, because of the many dimensional integration. In the Appendix we describe an algorithm that uses only one dimensional integration, thus making the computation task easier. This algorithm was used to obtain the results presented in the figures.

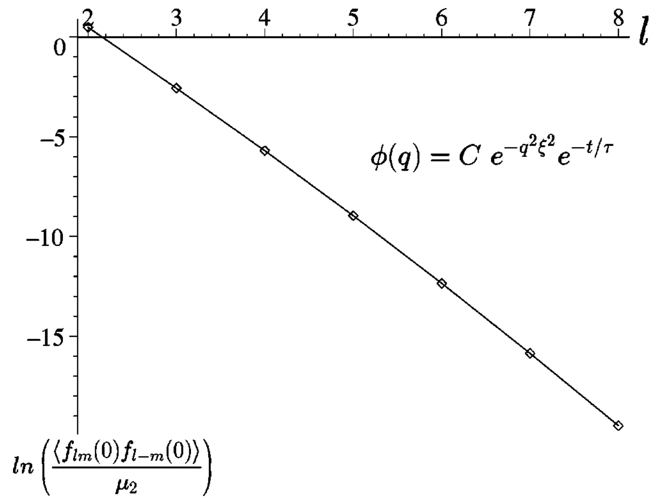

FIG. 4. The correlation of the deformation coefficients f_{lm} as a function of integer l, for a time-correlated velocity field $\phi(q,t) = Ce^{-q^2\xi^2}e^{-t/\tau}$ and $R/\xi = 1$.

The equal time correlations are given by

$$\langle f_{lm}(t) f_{l'm'}(t)\rangle_{t\to\infty} = \int_0^\infty dt' \langle Q_{lm}(0) Q_{l-m}(t')\rangle \frac{e^{-\lambda_l t'}}{\lambda_l} \delta_{l',l}\delta_{m',-m}. \tag{33}$$

Note that they still involve nonequal time correlation of the Q_{lm}'s. To demonstrate the applicability of the method presented above, we depict in Fig. 4 the equal time correlations as a function of l. This is done for a time correlated random flow that has a characteristic correlation length ξ and characteristic decay time τ, $\phi(q,t) = Ce^{-q^2\xi^2}e^{-t/\tau}$ and for droplets governed by surface tension. Note that the generalization to other deformable bodies that obey the conditions of Sec. II, is quite easy, because the dependence on the λ_l's is simple. The equal time correlations depend on three dimensionless parameters: $\mu_1 = R/\xi$, $\mu_2 = C\tau^2/R^5$ and $\mu_3 = \lambda\tau/\eta R$ (μ_3 has been written for the special case of an object governed by surface tension λ). We take $\mu_1 = \mu_2 = \mu_3 = 1$. The equal time correlations decay exponentially with increasing l, a behavior that is expected to hold for any correlation function for values of l that are larger than μ_1. This suggests that only the first few deformation coefficients are important to the dynamics of the system.

V. SUMMARY

We have constructed a method to calculate the correlations of the deformation coefficients $f_{l,m}$ that correspond to the decomposition of the shape into spherical harmonics, given the correlations in space and time of an external velocity field. We did it in two stages. The first was for external velocity fields that are uncorrelated in time: Eqs. (14) and (17). To demonstrate the applicability of our method we use it to calculate the correlations of the deformations in the case of thermal equilibrium. This is done by using the specific velocity correlations appropriate for that case. (The velocity correlations were obtained in the past from general considerations totally unrelated to the problem of deformable objects). In such a way, the problem of thermal deformation correlations is treated as a special case of our general approach. The second was for a general external velocity field that is correlated both in space and time: Eqs. (26), (30), (31), and (32). We discussed these correlations, used the results to construct numerically typical surface shapes, and considered the special case of thermal agitation. In addition, we pointed out from our numerical results that deformation coefficients with $l > \mu_1 \equiv R/\xi$ seem to decay exponentially and therefore are essentially unimportant. This suggests that working with spherical harmonics to investigate systems of deformable bodies is extremely useful for cases where μ_1 is small. For example, to describe a system of N spheres it might be enough to use only $12N$ deformation coefficients (that correspond to f_{lm} with $l = 2,3$) while for a description by points on the surface, the number of points needed to describe a single surface may be of the order of a hundred.

Our main motivation for developing this method was to build the basic tools for the treatment of a many object system, in which all the objects interact via the host fluid. The unknown response of the many object system to external flows requires the application of the method presented above to general random external flows. Therefore we have constructed the calculation method in such a way as if the correlation function $\phi(q,t)$ is externally given. However, in addition to the main motivation the results presented here can be applied directly to systems of a single deformable object in a random flow or to a system of objects in the dilute limit. In fact, we present here a theoretical prediction of the correlations of the deformations of a single deformable body that can be checked experimentally. The correlations of the velocity induced in one way or another can be measured in the absence of the object and then when the body is introduced its deformation can be recorded and the correlation analyzed. Moreover, the deformations of deformable objects of various radii can serve as some measure of velocity correlations in a liquid.

APPENDIX: ALGORITHM FOR THE CALCULATION OF THE DEFORMATION CORRELATIONS

The correlations of Q_{lm} [Eq. (14) or Eq. (26)] involve four dimensional integration while in the integrand, the correlations of the external velocity at two points [Eq. (12) or Eq. (31)] add a three-dimensional integration. As we can see, the method presented above is very hard to implement. Therefore we must find a way to lower the dimensionality of the integrals. The following algorithm illustrates a method to do so using the partial wave expansion

$$e^{-i\vec{q}\cdot(R\hat{\rho})} = \sum_{l=0}^{\infty}\sum_{m=-l}^{l} (-i)^l 4\pi j_l(qR) Y^*_{lm}(\Omega_q) Y_{lm}(\Omega), \tag{A1}$$

where Ω_q is the solid angle in the $\vec{q}$ direction and j_l is the spherical Bessel function. The result is a finite expression which is composed of a sum of terms where each term involves only one-dimensional integration. Unfortunately, although finite, this sum is too long to be presented here (see our website [32]).

Using the partial wave expansion, the correlations of the external velocity field on the surface are written explicitly using Eqs. (26) and (31) and the partial wave expansion as

$$\begin{aligned}\langle Q_{lm}(t)Q_{l'm'}(t+\Delta t)\rangle &= \sum_{I,J=x,y,z}\sum_{L,L'=0}^{\infty}\sum_{M=-L}^{L}\sum_{M'=-L'}^{L'}(-i)^L i^{L'}(4\pi)^2 \\ &\times\left(\int d\Omega Y^*_{lm}(\Omega)Y_{LM}(\Omega)\hat{r}_I\right) \\ &\times\left(\int d\Omega' Y^*_{l'm'}(\Omega')Y^*_{L'M'}(\Omega')\hat{r}'_J\right) \\ &\times\left[\int d\Omega_q Y^*_{LM}(\Omega_q)Y_{L'M'}(\Omega_q)\left(\delta_{I,J}-\frac{q_Iq_J}{q^2}\right)\right] \\ &\times\left(\int q^2 dq e^{-(q^2/6)F(\Delta t)}j_L(qR)j_{L'}(qR)\phi(q,\Delta t)\right).\end{aligned} \tag{A2}$$

We can perform the angular integrations, in Eq. (A2), over the solid angles Ω, Ω', and Ω_q by recalling the following facts:

(i) $\hat{r}_I$ and $\hat{r}_J$ can be expanded in spherical harmonics Y_{lm} with $l=1$.

(ii) $[\delta_{I,J}-(q_Iq_J)/q^2]$ can be expanded in spherical harmonics with $l=2$ and $l=0$.

(iii) The expanded expressions (that are composed of integrals of three spherical harmonics) are easily integrated using the Clebsch-Gordan coefficients. These integrals $\int Y^*_{l_1m_1}Y_{l_2m_2}Y_{l_3m_3}$ vanish unless $|l_1-l_3|\leq l_2\leq l_1+l_3$, the sum $l_1+l_2+l_3$ is even, and $m_2+m_3=m_1$. Hence in our case: the integral over Ω implies that $L=l\pm 1$ and $M-m=\{0,\pm 1\}$. The integral over Ω' implies that $L'=l'\pm 1$ and $M'+m'=\{0,\pm 1\}$, while from the integral over Ω_q it is easy to see that $L-L'=\{0\pm 2\}$ and $M-M'=\{0,\pm 1,\pm 2\}$.

Hence the expanded sum [32] will have a finite number of terms (about 100),

$$\begin{aligned}\langle Q_{lm}(t)Q_{l'm'}(t+\Delta t)\rangle &\equiv \sum_{\substack{L,L'\\M,M'\\I,J}}\left(\zeta^{l,l',m,m'}_{\substack{L,L'\\M,M'\\I,J}}\int q^2 dq e^{-(q^2/6)F(\Delta t)}\right. \\ &\left.\times j_L(qR)j_{L'}(qR)\phi(q,\Delta t)\right),\end{aligned} \tag{A3}$$

where ζ is a known algebraic expression and the integral over q is one dimensional.

In a previous article [10] we have shown how to calculate numerically the mean-squared displacement [Eq. (30)] using the differential equation:

$$\ddot{F}(t)=16\pi\int_0^{\infty}q^2 dq e^{-(q^2/6)F(t)}[j_0(qR)+j_2(qR)]^2\phi(q,t) \tag{A4}$$

with $F(0)=0$ and $\dot{F}(0)=0$ [where the latter holds for all cases except $\phi\propto\delta(t)$]. F can be obtained by direct step by step integration.

Finally, the correlation between the deformation coefficients $\langle f_{lm}(t)f_{l'm'}(t+\Delta t)\rangle$ are calculated using Eq. (32) for which the main contribution comes from $t'\approx t$ and $t''\approx t+\Delta t$. For equal time correlations, $\Delta t=0$, Eq. (33) for which the integration is one dimensional applies.

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