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Droplet shape fluctuations in agitated emulsions—Beyond the dilute limit

Gad Frenkel

School of Engineering, Ruppin Academic Center, Emek-Hefer 40250, Israel

HIGHLIGHTS

- Formalism for dealing with droplet shapes in random velocity fields is developed.
- The interaction with the other droplets of the emulsion is taken into account.
- The formalism is used to investigate shape of droplets governed by surface tension.
- It is shown that the interaction between droplets suppresses deformation mode.
- The suppression occurs at low frequencies relative to the deformation–decay rate.

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ABSTRACT

The paper investigates the interaction between dispersed droplets in an emulsion under random stirring of the host fluid. The main interest is to examine the autocorrelation of the shape fluctuations in such randomly stirred host fluids, beyond the dilute limit regime. Keeping expressions to leading order in the density of droplets and in deviation of droplets shapes from spherical, the shape is expanded in spherical harmonic modes and the correlations of these modes are derived. The special case of droplets that are governed by surface tension is investigated in detail. The correlations of the deformations that are obtained for this special case are damped relative to the deformation correlations in the dilute regime. The dependence of the damping on frequency is also discussed.

1. Introduction

Single deformable objects such as droplets of one liquid dispersed 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 that each object deforms due to external stirring. The purpose here is not to restrict the general discussion to thermal velocity fields. While numerous authors have studied the fluctuations and diffusion of single deformable objects due to thermal agitation [3–10], 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 [11,12] 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 [13–16]. 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 shape fluctuations of a single object, to linear order in the density.

E-mail address: gadyf@ruppin.ac.il.

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The approach used in this paper is that of linear response. The two ingredients of the calculation are the equations relating the deformations of a single object to an external velocity field and the equations providing the velocity field due to a given deformation of a deformable object far away from its centre. The positions of the objects and the properties of the random stirring are given only in the statistical sense. The above ingredients are thus used to gain statistical information on the deformation of the object, e.g., correlation of the deformation.

The system considered has the following properties.

(a) The deformable objects are fluid (e.g., liquid droplets), 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. These are standard conditions, although some recent work suggests that slip conditions may be more appropriate for some liquid emulsions [17]. 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 [18,19]. However the description can be extended to Helfrich bending energy [20,21] 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, linearity implies that the Reynolds number is small and that the Stokes approximation to the Navier–Stokes equation is applicable. The actual velocity field is the sum of the imposed velocity field, $\vec{v}_{ext}$ (the velocity field that would have existed if the objects were absent and will also be referred to as the external or bare velocity field), 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,

$$\vec{v} = \vec{v}_{ext} + \vec{v}_\psi + \vec{v}_r. \tag{1}$$

(c) The external velocity field is assumed to be random with zero mean and correlations that depend only on distance and time difference. Furthermore, the dependence on the time difference is taken to be extremely short ranged (Dirac δ 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 such as thermal agitation [10]. The justification for using short range correlations in time will be further discussed, however, at a later stage. 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 approximated as very short ranged in time 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.

(d) Last, it is assumed that the objects are small in comparison to the typical distance between droplets and to the spatial correlation length of the velocity field.

Since small deviations are assumed 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

$$\frac{\rho}{R} + f(\Omega, t) - 1 = 0 \tag{2}$$

defines its surface, yielding for each spatial direction, Ω, 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 [22,23].

The random velocity field and the effect of the interaction between objects induce fluctuations in the values of the deformation coefficients describing each of the objects. Consider the autocorrelation of the deformation coefficient $f^i_{l,m}$ of the i'th object $\langle f^i_{l,m}(\omega) f^i_{l,-m}(-\tilde{\omega})\rangle$, where $f^i_{lm}(\omega)$ and $f^i_{lm}(\tilde{\omega})$ represent the Fourier transforms (FT) of the deformation coefficients of the i'th object with respect to time. The autocorrelation is expanded in orders of the number density of objects, n. To first order in the density n it is given by

$$\left\langle f^i_{l,m}(\omega) f^i_{l,-m}(-\tilde{\omega})\right\rangle = \mathcal{g}_{l,0}(\omega, \tilde{\omega}) + \tilde{\mathcal{g}}_{l,1}(\omega, \tilde{\omega}) \cdot n. \tag{3}$$

The first term on the right hand side of Eq. (3) 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