Statistical mechanics of bubbly liquids

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(Received 22 August 1995; accepted 8 December 1995)

The dynamics of bubbles at high Reynolds numbers is studied from the viewpoint of statistical mechanics. Individual bubbles are treated as dipoles in potential flow. A virtual mass matrix of the system of bubbles is introduced, which depends on the instantaneous positions of the bubbles, and is used to calculate the energy of the bubbly flow as a quadratic form of the bubbles' velocities. The energy is shown to be the system's Hamiltonian and is used to construct a canonical ensemble partition function, which explicitly includes the total impulse of the suspension along with its energy. The Hamiltonian is decomposed into an effective potential due to the bubbles' collective motion and a kinetic term due to the random motion about the mean. An effective bubble temperature—a measure of the relative importance of the bubbles' relative to collective motion—is derived with the help of the impulse-dependent partition function. Two effective potentials are shown to operate: one due to the mean motion of the bubbles, dominates at low bubble temperatures, where it leads to their grouping in flat clusters normal to the direction of the collective motion, while the other, temperature-invariant, is due to the bubbles' position-dependent virtual mass and results in their mutual repulsion. Numerical evidence is presented for the existence of the effective potentials, the condensed and dispersed phases, and a phase transition. © *1996 American Institute of Physics*. [S1070-6631(96)00404-0]

I. INTRODUCTION

The dynamics of bubbly liquids—a Newtonian liquid filled with a dispersed gas phase in the form of bubbles—is of interest in a variety of engineering problems. Bubbly flows are common in the energy-conversion, oil and chemical industries, in natural gas distribution networks, and in any flow in which rapid pressure variations can lead to phase change, e.g., cavitation, or where sound waves can be strongly modified by bubble clouds. A major theoretical and technical problem associated with bubbly flows is predicting, and thus controlling, the flow regime, and then within the flow regime understanding the transport of heat, mass, momentum, and sound.

Continuum modeling of gas/liquid flows in the region of dispersed bubbly flow is an active area of research and has been approached through a variety of different methods. One of these has been developed by Batchelor,[1] Delhaye and Achard,[2] Hinch,[3] Voinov and Petrov,[4] Banerjee and Chan,[5] Drew,[6] Biesheuvel and van Wijngaarden,[7] and Pauchon and Banerjee,[8] among others. These authors have applied averaging—in time, space, in time and space, over an ensemble, etc.—to the continuum-mechanical equations describing the exact motion of each phase at each point. The system of resulting averaged equations is closed with the help of constitutive relations determined from a list of variables that are supposed to influence the phase interactions. The resulting equations have several unknown coefficients that have to be determined in some way, e.g., from experiments. The models that different investigators have proposed usually differ in the choice of closure relationships.

Another, variational, approach has been developed by Geurst[9,10] and Pauchon and Smereka.[11] These authors have chosen to volume-average the energy of a two-phase flow and then treat the volume-averaged energy as the Lagrangian by applying to it a generalized form of Hamilton's variational principle. Pauchon and Smereka[11] have shown the variational and averaging approach to be complementary: the variational approach is capable of providing explicit analytical forms for the constitutive equations of the averaging approach. However, as Smereka[12] has pointed out, although Geurst's model seems to have a sound theoretical basis, it turns out to be ill-posed in the dilute limit; Geurst and other authors argued that the ill-posedness is associated with the assumption of an isotropic arrangement of bubbles. As a result, much effort has been devoted to studying the related issues of clustering of bubbles and voidage wave propagation in bubbly liquids. Sangani and Didwania[13,14] and Smereka[12] carried out computer simulations of bubbles in an ideal fluid. In both studies, it has been found that, if initially given similar velocities, the bubbles would tend to form clusters positioned broadside to the direction of motion; Smereka found that the clustering would be inhibited, however, if the variance of the initial velocities of bubbles was sufficiently large.

A kinetic-theory-like approach to the problem of bubbly liquids has been investigated by Biesheuvel and Gorissen[15] and van Wijngaarden and Kapteyn.[16] These workers have obtained effective equations by taking moments of an N-particle probability density function for bubble positions and velocities and focused their attention on the propagation of void fraction disturbances in bubble flows.

In this paper, we present a formulation of the problem of bubbly flows from a statistical mechanics point of view. This idea was hinted at in the work of Smereka,[12] but not devel-

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oped nor explored. Indeed, it is an interesting and open question as to whether this continuum problem of bubble motion has a statistical description of the same form as atoms or molecules. On the one hand, classical statistical mechanics is a powerful means of studying macroscopic bodies. It provides the molecular basis of macroscopic properties by explaining the connection between the observed values of a thermodynamic function and the properties of the molecules making up the system. On the other hand, bubbles, as considered in this paper (of constant spherical shape and size, monodisperse, inviscid and described as dipoles in potential flow) are fundamentally different from molecules as "elementary particles" of statistical mechanics in one important aspect, namely, they do not have mass; rather, their motion results in flow of the underlying liquid of nonzero density. Thus, "virtual" or "added" mass of the flow is critical to the behavior of the bubbles as a statistical system. Like a molecular system, the total energy and momentum of the flow are shown to be integrals of the bubbles' motion. The interactions among the bubbles occur through the velocity field in the fluid generated by the bubbles' motion, and will be seen to be of the form similar to repulsive r^{-6} interactions in molecular systems.

By formulating and examining a bubbly flow as a statistical ensemble we can predict, discuss, and quantify patterns of collective behavior of a macroscopically large number of bubbles as consequences of the distinctive features of bubbles treated as individual particles. We are able to discern factors influencing the flow's structure and properties, and describe them in thermodynamic terms, such as interaction potentials, temperature, phase transition, etc. Finally, in a fashion similar to classical atomic systems, the validity of our predictions and calculations will be tested by simulating the bubbly flow numerically and by analyzing the results of the simulations.

In Sec. II, we approximate a bubbly flow at high Reynolds numbers as potential flow and derive its total energy. We apply Lagrange's formalism to the energy in order to derive the equations of motion for the bubbles. In Sec. III, we show that the bubbly flow is a Hamiltonian system and define the canonical ensemble partition function, along with the "temperature" for such a flow in much the same way as is done for atomic systems. Unlike atomic systems, however, the virtual mass for the bubbly flow is dependent on the positions of all the bubbles, and as a result the partition function for an ensemble of bubbles explicitly depends on the total impulse of the flow determined in a frame of reference in which the liquid would be motionless, were it not for the presence of the moving bubbles. As the relative position of the bubbles changes, so does the "mass" of the system, and therefore the center-of-mass or collective motion is coupled to the internal degrees of freedom. These two distinctive features—the coordinate-dependent mass and the momentum-dependent partition function—together give rise to a number of unusual effects not found in atomic systems. In particular, the coordinate-dependent mass results in the clustering of bubbles in the direction normal to that of their mean motion, and also in an effective repulsive potential that can prohibit the clustering at sufficiently high values of the bubble temperature (defined as a measure of the bubbles' chaotic motion). Also in Sec. III, we predict the value of the temperature at which a phase transition occurs between the clustered and dispersed states. In Sec. IV, we develop a molecular-dynamics-like method for simulating bubbly flows and present and discuss numerical results: snapshots of representative bubble configurations for 27 and 64 bubbles at different volume fractions and at different bubble temperatures; the influence of viscous dissipation and gravity; evidence of the effective repulsive potential and the phase transition between the clustered and random phases. These results are shown to support the statistical mechanical description.

II. EQUATION OF MOTION FOR A DISPERSION OF BUBBLES

A. Kinetic energy of a bubbly flow

We shall make use of Lagrange's formalism in order to derive an equation of motion for bubbles in a dispersed flow. In this framework, the first step is to obtain an expression for the Lagrangian $\mathscr{L}$. In this problem, the kinetic energy of the fluid $\mathscr{T}$ will be found to be quadratic in the vector of the bubbles' velocities $\mathbf{U}$, and thus the Lagrangian is the kinetic energy $\mathscr{T}$ minus the potential energy $\mathscr{E}$. We shall set the potential energy to zero; this restriction, of course, can be relaxed if so desired. Thus, $\mathscr{L}=\mathscr{T}$, and we determine the kinetic energy as a function of the bubbles' positions and velocities. In order to do so, we model the bubbly flow as a dispersion of a finite number, N, of monodisperse spherical bubbles (maintained spherical by a presumed large interfacial tension), characterized by a constant internal pressure moving in an unbounded fluid at high Reynolds number ($Re=Ua/\nu \gg 1$) subject to gravitational, external, or fluctuating pressure forces. Because the Reynolds number is large and the bubble surface is a free surface, the flow outside the bubbles is approximately inviscid and irrotational. Hence, the fluid velocity can be written as the gradient of a velocity potential, ϕ, satisfying Laplace's equation,

$$\nabla^2\phi=0, \tag{1}$$

everywhere in the fluid, with no flux boundary conditions on the bubble surfaces,

$$\mathbf{n}\cdot\nabla\phi=\mathbf{n}\cdot\mathbf{U}^{\nu},$$

where $\mathbf{n}$ is the unit outward normal from the surface $\partial\Omega^{\nu}$ of bubble ν and $\mathbf{U}^{\nu}$ is that bubble's velocity. This is, of course, an approximation, but it is known from the work of Moore[17,18] that the flow outside a moving bubble is to a very good approximation irrotational, with the vorticity confined to a thin $O(Re^{-1/2})$ boundary layer at the bubble surface and to a narrow $O(Re^{-1/4})$ wake. Furthermore, Kok[19,20] has shown both theoretically and experimentally that the motion of two interacting bubbles can also be predicted by the appropriate two bubble solution of Laplace's equation.

The total kinetic energy of the fluid is (cf. Secs. 2.7–2.10, 6.2, and 6.4 of Batchelor[21])

$$\mathscr{T}=\frac{1}{2}\rho\int_{\partial\Omega^{\infty}}\phi\mathbf{U}^{\infty}\cdot\mathbf{n}^{\infty}dS-\frac{1}{2}\rho\sum_{\nu}\mathbf{U}^{\nu}\cdot\int_{\partial\Omega^{\nu}}\phi\mathbf{n}^{\nu}dS, \tag{2}$$

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where $\mathbf{U}^\infty$ denotes the fluid velocity at infinity and ρ the density of the fluid. The first integral in (2) is taken over a closed boundary at infinity and is, therefore, equal to zero as long as we assume that the fluid's motion is entirely due to the bubbles; indeed, we treat the bubbles as dipoles in this study (cf. Appendix), i.e., the potential ϕ falls off as r^{-2} and the liquid velocity $\mathbf{U}^\infty$ falls off as r^{-3} at large r and thus the first integral in (2) vanishes. The solution of (1) is linear in the bubbles' velocities $\mathbf{U}$ and therefore the kinetic energy can be recast, with the help of the virtual mass matrix $\mathbf{M}$, into a quadratic form:

$$\mathscr{T} = \tfrac{1}{2}\rho\tau\mathbf{U}\cdot\mathbf{M}\cdot\mathbf{U}, \tag{3}$$

where $\tau = (4/3)\pi a^3$ is the volume of a spherical bubble of radius a and $\mathbf{U}$ is a vector of the individual bubble velocities $\mathbf{U}^\nu$. The derivation of the mass matrix is given in detail in the Appendix.

We also find it convenient to write the total energy of the flow as

$$\mathscr{T} = \tfrac{1}{2}\rho\tau\mathbf{U}^\mu\cdot\mathbf{M}^{\mu\nu}\cdot\mathbf{U}^\nu, \tag{4}$$

where the Greek indices denote the individual bubbles and, if repeated, imply a summation; $\mathbf{M}^{\mu\nu}$ are the coupling submatrices of the mass matrix $\mathbf{M}$ (cf. Appendix). Finally, the individual bubbles' momenta are defined as

$$\mathbf{P}^\mu = \rho\tau\mathbf{M}^{\mu\nu}\cdot\mathbf{U}^\nu, \tag{5}$$

and the total impulse of the suspension (i.e., the linear momentum that needs to be imparted to the fluid in order to generate from rest the flow due to motion of the bubbles with translational velocities $\mathbf{U}^\mu$) is

$$\mathbf{P}_t = \sum_{\mu=1}^{N} \mathbf{P}^\mu. \tag{6}$$

The total impulse $\mathbf{P}_t$ is an integral of the motion (cf. Art. 119 of Lamb[22]), as is the total energy $\mathscr{T}$.

B. Lagrangian formulation of the problem

From expression (3) for the total kinetic energy, we can derive the equation of motion for the bubbles. Let $\mathbf{R}$ denote the vector of the bubbles' centers, $\mathbf{F}^g$ the external forces such as gravity, and $\mathbf{F}^v$ the viscous forces. Then, recalling Lagrange's equation, the equation of motion of the bubbly suspension is

$$\frac{d}{dt}\left(\frac{\partial\mathscr{L}}{\partial\dot{\mathbf{R}}}\right) = \frac{\partial\mathscr{L}}{\partial\mathbf{R}} + \mathbf{F}^g + \mathbf{F}^v. \tag{7}$$

Lamb[22] and, more recently, Hinch and Nitsche[23] have shown that the Lagrangian generalized force,

$$\mathbf{F}^L = \frac{d}{dt}\left(\frac{\partial\mathscr{L}}{\partial\dot{\mathbf{R}}}\right) - \frac{\partial\mathscr{L}}{\partial\mathbf{R}},$$

is the same as the pressure force exerted by the fluid on the bubbles $\mathbf{F}^p$. Thus, the equation of motion (7) is equivalent to the force balance on the massless bubbles,

$$\mathbf{F}^p + \mathbf{F}^g + \mathbf{F}^v = 0. \tag{8}$$

[Bubble mass could be added to (8) or (7) without difficulty. The conclusions of the work would not be affected for small bubble mass. Similarly, angular momentum could be added for nonspherical particles.]

From (3), together with the fact that $\mathscr{L} = \mathscr{T}$, we compute the necessary quantities:

$$\frac{\partial\mathscr{L}}{\partial\mathbf{R}} = \frac{\partial\mathscr{T}}{\partial R_k} = \frac{\partial}{\partial R_k}\left(\frac{1}{2}\rho\tau U_i M_{ij}(R_k)U_j\right) = \frac{1}{2}\rho\tau U_i\frac{\partial M_{ij}}{\partial R_k}U_j$$

and

$$\frac{\partial\mathscr{L}}{\partial\dot{\mathbf{R}}} = \frac{\partial\mathscr{T}}{\partial U_k} = \frac{1}{2}\rho\tau(U_i M_{ik} + M_{kj}U_j) = \rho\tau M_{kj}U_j,$$

due to the fact that the mass matrix is symmetric. Furthermore,

$$\frac{d}{dt}\left(\frac{\partial\mathscr{T}}{\partial U_k}\right) = \rho\tau(\dot{M}_{kj}U_j + M_{kj}\dot{U}_j),$$

with

$$\dot{M}_{kj} = \frac{\partial M_{kj}}{\partial R_l}\dot{R}_l = \frac{\partial M_{kj}}{\partial R_l}U_l.$$

Hence,

$$\frac{d}{dt}\left(\frac{\partial\mathscr{T}}{\partial U_k}\right) = \rho\tau\left(\frac{\partial M_{kj}}{\partial R_l}U_l U_j + M_{kj}\dot{U}_j\right),$$

and the equation of motion (7) becomes

$$\rho\tau M_{kj}\dot{U}_j = \rho\tau\left(\frac{1}{2}U_i\frac{\partial M_{ij}}{\partial R_k}U_j - \frac{\partial M_{kj}}{\partial R_l}U_l U_j\right) + F_k^g + F_k^v. \tag{9}$$

Equation (9) is the governing equation for determining the bubbles' motion. We see clearly that the virtual mass matrix does indeed act as the mass of the system.

Equation (9) defines the dynamics of the bubbles and is sufficient to follow their motion for given initial conditions under the action of prescribed forces. Before doing so, we turn to a consideration of the bubbles as a statistical ensemble and investigate whether this dynamical system has thermodynamics akin to a molecular system.

III. BUBBLY FLOW AS A STATISTICAL ENSEMBLE

Consider the bubbly flow of Sec. II in the absence of gravity and fluid viscosity. It proves possible then to treat the total energy of the bubbly flow,

$$\frac{1}{2\rho\tau}\mathbf{P}\cdot\mathbf{M}^{-1}\cdot\mathbf{P},$$

as the system's Hamiltonian, $\mathscr{H}(\mathbf{P},\mathbf{R})$, with $\mathbf{P}$ and $\mathbf{R}$ as the corresponding generalized momenta and coordinates. Indeed, $\mathscr{H}$ satisfies Hamilton's equations,

$$\dot{\mathbf{R}} = \frac{\partial\mathscr{H}}{\partial\mathbf{P}}, \tag{10}$$

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$$\dot{\mathbf{P}} = -\frac{\partial \mathscr{H}}{\partial \mathbf{R}}, \tag{11}$$

which are equivalent to the equation of motion (9) with $\mathbf{F}^g = \mathbf{F}^v = 0$, and thus the inviscid, neutrally buoyant bubbly flow is a Hamiltonian system (with $6N$ degrees of freedom) to which the standard methods of statistical mechanics can be applied.

A. Effective energies

We begin by decomposing the full energy of the bubbly flow into effective potential and kinetic energies. First, we write each bubble's individual impulse, defined by (5), as follows:

$$\mathbf{P}^\mu = \mathbf{p}^\mu + \frac{1}{N}\mathbf{P}_t,$$

where the deviation from the mean, $\mathbf{p}^\mu$, can be regarded as the "chaotic" impulse of bubble μ. The total energy of the flow is then

$$\mathscr{T} = \frac{1}{2\rho\tau}\left(\frac{1}{N^2}\mathbf{P}_t \cdot \sum_{\mu,\nu} (\mathbf{M}^{-1})^{\mu\nu} \cdot \mathbf{P}_t + \mathbf{p}\cdot\mathbf{M}^{-1}\cdot\mathbf{p} + \frac{2}{N}\mathbf{P}_t \cdot \left(\sum_{\mu} (\mathbf{M}^{-1})^{\mu\nu}\cdot\mathbf{p}^\nu\right)\right), \tag{12}$$

where $\mathbf{p}$ denotes the full vector of the bubbles' chaotic impulses. The first term on the right-hand side of (12) is due to the collective motion of the bubbles with the same velocity $\mathbf{u}_0 = (\rho\tau N)^{-1}\mathbf{P}_t \cdot \Sigma_{\mu,\nu}(\mathbf{M}^{-1})^{\mu\nu}$; it depends only on the positions of the bubbles' centers, $\mathbf{R}$, and thus can be thought of as an "effective potential energy." The sum of the second and third terms in (12) we shall call the effective kinetic energy; it accounts for the individual, or chaotic, portion of the bubbles' motion. It is easy to see that the chaotic impulses $\mathbf{p}$ along with the positions of the bubbles' centers $\mathbf{R}$ can play the role of the generalized coordinates that satisfy Hamilton's equations of motion.

Smereka[12] has defined effective energies by decomposing the bubble's velocity, rather than its impulse, as follows:

$$\mathbf{U}^\mu = \mathbf{u}_0 + \mathbf{u}^\mu,$$

such that

$$\mathbf{M}^{\mu\nu}\cdot\mathbf{u}^\nu = 0,$$

$$\rho\tau\mathscr{M}\cdot\mathbf{u}_0 = \mathbf{P}_t,$$

where

$$\mathscr{M} = \sum_{\mu,\nu} \mathbf{M}^{\mu\nu}.$$

Then, recalling (4), the total energy of the flow is

$$\mathscr{T} = \frac{1}{2\rho\tau}\mathbf{P}_t\cdot\mathscr{M}^{-1}\cdot\mathbf{P}_t + \frac{\rho\tau}{2}\mathbf{u}^\mu\cdot\mathbf{M}^{\mu\nu}\cdot\mathbf{u}^\nu. \tag{13}$$

Again, the first term on the right-hand side of (13) is due to the collective motion of the bubbles with the same velocity $\mathbf{u}_0$; it depends only on the position of the bubbles' centers $\mathbf{R}$, and thus Smereka has termed it the effective potential energy; the second term is the corresponding effective kinetic energy.

Smereka used the decomposition (13) to show that the effective potential energy $(2\rho\tau)^{-1}\mathbf{P}_t\cdot\mathscr{M}^{-1}\cdot\mathbf{P}_t$ approaches a minimum when the bubbles arrange themselves in flat clusters positioned broadside to the direction of the collective motion. For the purposes of the present study, we shall use the "Hamiltonian" decomposition (12), rather than (13). As given by (12), the total energy of the flow satisfies Hamilton's equations (10) and (11), if $\mathbf{p}$ is used as the generalized momentum instead of $\mathbf{P}$, and thus equation (12) can and will be used to construct the canonical ensemble partition function. The decomposition of Smereka in (13) is not in the standard form for statistical mechanics and cannot be used to construct the partition function.

Another result of Smereka[12] is that the variational principle of minimal potential energy is equivalent to the principle of maximal virtual mass. To show that bubbles in collective motion with no random velocities will indeed tend to increase their virtual mass we consider the following situation.

Suppose that initially the bubbles are randomly positioned in space but all have the same velocity, e.g., (0,0,1). They are then released and we are interested in the change in the added-mass coefficient C_M defined as the following norm on the mass matrix:

$$C_M \equiv \frac{\mathbf{U}\cdot\mathbf{M}\cdot\mathbf{U}}{\mathbf{U}\cdot\mathbf{U}}.$$

If we write the Lagrangian of the system as $\mathscr{L} = 1/2\rho\tau C_M U^2$, then Lagrange's equation (7) can be transformed to

$$\dot{C}_M = \frac{1}{2}\left(\frac{\partial C_M}{\partial R_i}\right)U_i - C_M\frac{\dot{U}_i U_i}{U^2}. \tag{14}$$

Initially, when $\mathbf{U} = [0,0,1;0,0,1;\ldots;0,0,1]$, the first term on the right-hand side of (14) is proportional to the change in C_M corresponding to a rigid translation of the whole suspension along the z-axis, and therefore is zero. The randomly distributed bubbles will begin to develop velocities in the two other orthogonal directions, x and y. In order to do so, energy will have to be extracted from the bubbles' collective motion, the bubbles will slow down in the z-direction, to conserve the total energy and impulse, and thus $\dot{U}_i$ and U_i will be anti-parallel, i.e., $\dot{U}_i U_i$ will be negative and $\dot{C}_M$ positive, which means that C_M will initially tend to increase. In other words, since the bubbles have to decelerate in the z-direction while their total impulse in that direction must be conserved, their virtual mass must increase. Smereka[12] recognized the analogy between potential flow outside the bubbles and the effective conductivity of a material where the liquid has a unit conductivity and the spherical bubbles are insulators (cf. Appendix). He was able to show that the only way for the bubbles to increase their virtual mass would be to organize themselves in clusters positioned broadside to the direction of the collective motion; this situation corresponds to the minimal effective conductivity of the matrix in

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the direction of the mean flow. Thus, following Smereka, we also conclude that the bubbles' collective motion will lead to their clustering. In the next section we investigate how this clustering manifests itself when the bubbles are viewed as a thermodynamic system.

B. Canonical ensemble partition function for bubbly flow

We now apply the apparatus of equilibrium statistical mechanics by treating the suspension of N bubbles as a canonical ensemble. Denoting the Hamiltonian of the ensemble by $\mathscr{H}$, the partition function, within a constant factor, is given by

$$Q=\int \exp[-\beta\mathscr{H}(\mathbf{R},\mathbf{P})-\boldsymbol{\gamma}\cdot\mathbf{P}_t(\mathbf{R},\mathbf{P})]\; d\mathbf{R}\; d\mathbf{P}. \tag{15}$$

The integration in (15) is performed over the entire phase space. The form (15) follows from the fact that there are only seven additive integrals of motion of a classical system: the energy, linear momentum, and angular momentum. Since we have no angular momentum for spherical bubbles, only two constants β and $\boldsymbol{\gamma}$ are needed (cf. Sec. 4 of Landau and Lifshitz[24]). Here β will be seen to play the role of the inverse temperature and $\boldsymbol{\gamma}/\beta$ will be the average velocity of the bubbles.

The necessity of including the total impulse of the bubbly flow, $\mathbf{P}_t$, in the expression for the canonical ensemble partition function is dictated by the fact that the "center-of-mass" motion is coupled to the internal degrees of freedom. As the relative coordinates change so too does the mass, leading to the coupling with the total impulse.

Let us denote for convenience the effective potential energy as $\mathscr{U}$ and the effective kinetic energy as $\mathscr{K}$, such that

$$\mathscr{U}=\frac{1}{2\rho\tau N^2}\mathbf{P}_t\cdot\sum_{\mu,\nu}(\mathbf{M}^{-1})^{\mu\nu}\cdot\mathbf{P}_t \tag{16}$$

and

$$\mathscr{K}=\frac{1}{2\rho\tau}\left(\mathbf{p}\cdot\mathbf{M}^{-1}\cdot\mathbf{p}+\frac{2}{N}\mathbf{P}_t\cdot\left(\sum_{\mu}(\mathbf{M}^{-1})^{\mu\nu}\cdot\mathbf{p}^{\nu}\right)\right).$$

The potential energy $\mathscr{U}$ is purely $\mathbf{R}$-dependent, just as in the classical case of a gas of particles interacting via a potential. However, due to the fact that the virtual mass of the bubbly suspension is position-dependent, the kinetic energy $\mathscr{K}$ is both $\mathbf{R}$- and $\mathbf{p}$-dependent and differs from that of a classical statistical mechanical system.

Depending on the choice of the momentum generalized coordinates—one can choose either the bubbles' full impulses $\mathbf{P}$ or their chaotic impulses $\mathbf{p}$—the following two alternative expressions for the partition function are possible:

$$Q=\int \exp\left[-\frac{\beta}{2\rho\tau}\mathbf{P}\cdot\mathbf{M}^{-1}\cdot\mathbf{P}-\boldsymbol{\gamma}\cdot\sum_{\mu}\mathbf{P}^{\mu}\right]d\mathbf{R}d\mathbf{P} \tag{17}$$

or

$$Q=\int \exp[-\beta\mathscr{U}-\boldsymbol{\gamma}\cdot\mathbf{P}_t]\times\left(\int\exp\left[-\beta\mathscr{K}-\boldsymbol{\gamma}\cdot\sum_{\mu}\mathbf{p}^{\mu}\right]d\mathbf{p}\right)d\mathbf{R}. \tag{18}$$

Upon integrating (17) and (18) by parts with respect to an arbitrary component of the vector $\mathbf{P}$ or $\mathbf{p}$ and denoting by angle brackets averaging over the ensemble, we obtain

$$\frac{\beta}{\rho\tau}\langle\mathbf{P}\cdot\mathbf{M}^{-1}\cdot\mathbf{P}\rangle-\boldsymbol{\gamma}\cdot\left\langle\sum_{\mu}\mathbf{P}^{\mu}\right\rangle=3N \tag{19}$$

and

$$\frac{\beta}{\rho\tau}\left\langle\mathbf{p}\cdot\mathbf{M}^{-1}\cdot\mathbf{p}+\frac{1}{N}\mathbf{P}_t\cdot\left(\sum_{\mu}(\mathbf{M}^{-1})^{\mu\nu}\cdot\mathbf{p}^{\nu}\right)\right\rangle-\boldsymbol{\gamma}\cdot\left\langle\sum_{\mu}\mathbf{p}^{\mu}\right\rangle=3N. \tag{20}$$

At thermal and mechanical equilibrium of the ensemble with its surroundings,

$$\left\langle\sum_{\mu}\mathbf{p}^{\mu}\right\rangle=0,\quad\left\langle\sum_{\mu}\mathbf{P}^{\mu}\right\rangle=\mathbf{P}_t,$$

and it follows from (19) and (20) that

$$\beta^{-1}=\frac{1}{3\rho\tau N}\left\langle\mathbf{p}\cdot\mathbf{M}^{-1}\cdot\mathbf{p}+\frac{1}{N}\mathbf{P}_t\cdot\left(\sum_{\mu}(\mathbf{M}^{-1})^{\mu\nu}\cdot\mathbf{p}^{\nu}\right)\right\rangle \tag{21}$$

and

$$\frac{\boldsymbol{\gamma}}{\beta}=\frac{1}{\rho\tau N}\left\langle\frac{1}{N}\sum_{\mu,\nu}(\mathbf{M}^{-1})^{\mu\nu}\cdot\mathbf{P}_t+\left(\sum_{\mu}(\mathbf{M}^{-1})^{\mu\nu}\cdot\mathbf{p}^{\nu}\right)\right\rangle=\frac{1}{\rho\tau N}\left\langle\sum_{\mu}(\mathbf{M}^{-1})^{\mu\nu}\cdot\mathbf{P}^{\nu}\right\rangle=\frac{1}{N}\left\langle\sum_{\mu}\mathbf{U}^{\mu}\right\rangle. \tag{22}$$

Thus $\boldsymbol{\gamma}/\beta$ is equal to the average velocity of all the bubbles in the flow, showing that $\boldsymbol{\gamma}\cdot\mathbf{P}_t/\beta$ is the energy of the collective motion.

That $\boldsymbol{\gamma}/\beta$ must be proportional to the average velocity of the bubbles in the ensemble could also be deduced from the following argument, adapted from Sec. 14 of Hill.[25] We consider here a generalized ensemble which can, with probabilities p_k, be found in a number of states with energies E_k; this formulation can be generalized to account for continuous states. We associate the thermodynamic internal energy E with the ensemble average $\bar{E}$ (the averaging denoted by the overbar):

$$E\leftrightarrow\bar{E},$$

where

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$$\bar{E}=\sum_{k,\mathbf{P}_t} p_k E_k\,,$$

$$p_k=\frac{\exp(-\beta E_k-\boldsymbol{\gamma}\cdot\mathbf{P}_t)}{Q}, \tag{23}$$

$$Q=\sum_{k,\mathbf{P}_t}\exp(-\beta E_k-\boldsymbol{\gamma}\cdot\mathbf{P}_t).$$

In differential form,

$$d\bar{E}=\sum_{k,\mathbf{P}_t} dp_k E_k\,, \tag{24}$$

where the E_k are constants. We now use (23) to eliminate E_k in (24),

$$d\bar{E}=-\frac{1}{\beta}\sum_{k,\mathbf{P}_t}(\boldsymbol{\gamma}\cdot\mathbf{P}_t+\ln p_k+\ln Q)dp_k\,. \tag{25}$$

Since

$$\bar{\mathbf{P}}_t=\sum_{k,\mathbf{P}_t} p_k\mathbf{P}_t\,,$$

(25) becomes

$$\begin{aligned} d\bar{E}&=-\frac{1}{\beta}\sum_{k,\mathbf{P}_t}(\ln p_k dp_k)-\frac{1}{\beta}\boldsymbol{\gamma}\cdot d\bar{\mathbf{P}}_t \\ &=-\frac{1}{\beta}d\left(\sum_{k,\mathbf{P}_t}(p_k\ln p_k)\right)-\frac{1}{\beta}\boldsymbol{\gamma}\cdot d\bar{\mathbf{P}}_t\,, \end{aligned} \tag{26}$$

where we have employed $\Sigma dp_k=0$. With the further association $\mathbf{P}_t\leftrightarrow\bar{\mathbf{P}}_t$, (26) is seen to be just the statistical version of the thermodynamic equation,

$$dE=TdS-\boldsymbol{\Pi}\cdot d\mathbf{P}_t\,, \tag{27}$$

where E, T, and S are, as usual, the internal energy, temperature, and entropy of the system, whereas $\boldsymbol{\Pi}$ is the generalized force appropriate to $\mathbf{P}_t$ (treated here as a generalized coordinate). By comparing (26) and (27) we have

$$\boldsymbol{\Pi}\leftrightarrow\frac{\boldsymbol{\gamma}}{\beta}.$$

It can further be seen that $\boldsymbol{\Pi}$, along with $\boldsymbol{\gamma}/\beta$, must have units of velocity. In our problem, there is only one characteristic velocity, namely the average velocity of the bubbles. Thus, it is reasonable, in agreement with (22), to assume $\boldsymbol{\gamma}/\beta$ proportional to $\bar{\mathbf{U}}\equiv N^{-1}\langle\Sigma_\mu\mathbf{U}^\mu\rangle$.

Now, given an ensemble at specified values of β and $\boldsymbol{\gamma}$, we can always rotate the coordinate system, simultaneously rescaling the time, such that in the transformed frame of reference, $\bar{\mathbf{U}}=\mathbf{e}_c$, where $\mathbf{e}_c$ is a unit velocity vector pointing in the direction of the collective motion, this direction being the same in all appropriately rotated ensembles. In other words,

$$\boldsymbol{\gamma}=\beta\mathbf{e}_c\,, \tag{28}$$

and the partition function (15) can be rewritten as

$$Q=\int\exp(-\beta[\mathscr{H}(\mathbf{R},\mathbf{P})+\mathbf{e}_c\cdot\mathbf{P}_t(\mathbf{R},\mathbf{P})])d\mathbf{R}d\mathbf{P}.$$

Finally, we define the temperature T of the system as the inverse of β, as given by (21):

$$T\equiv\frac{1}{\beta}.$$

The bubble temperature T can take on values from zero to infinity; it provides a measure of the relative importance of the chaotic to collective motion of the bubbles in the flow.

C. Effective potentials

We shall now show that the coordinate-dependent virtual mass matrix of the bubbly flow, along with the bubbles' collective motion, gives rise to two effective inter-bubble potentials. Recalling (28), we begin by evaluating the integral in (17) over the **P**-subspace of the phase space:

$$Q=\left(\frac{2\pi\rho\tau}{\beta}\right)^{3N/2}\int(\det\mathbf{M})^{1/2}\exp\left(\frac{\beta\rho\tau\mathbf{e}_c\cdot\mathscr{M}\cdot\mathbf{e}_c}{2}\right)d\mathbf{R}, \tag{29}$$

where we recognize the factor

$$\left(\frac{2\pi\rho\tau}{\beta}\right)^{3N/2}$$

as the partition function Q^{id} of an ideal gas of N particles of mass $\rho\tau$ each, normalized by the volume occupied by the suspension V_c raised to the Nth power. Recall that $\mathscr{M}$ is the sum of the coupling submatrices $\mathbf{M}^{\mu\nu}$ of the mass matrix.

The integral in (29), also called the excess part Q^{ex} of the partition function, can be rewritten as

$$Q^{ex}=V_c^{-N}\int\exp[-\beta(\mathscr{U}_1+\mathscr{U}_2)]\,d\mathbf{R}, \tag{30}$$

where the two effective potentials $\mathscr{U}_1$ and $\mathscr{U}_2$ are defined as

$$\beta\mathscr{U}_1=-\tfrac{1}{2}\ln(\det\mathbf{M}) \tag{31}$$

and

$$\mathscr{U}_2=-\frac{\rho\tau}{2}\mathbf{e}_c\cdot\mathscr{M}\cdot\mathbf{e}_c\,. \tag{32}$$

To understand the behavior of the effective potential $\mathscr{U}_1$, it will be useful to consider a dilute suspension in which only two of the N bubbles, μ and ν, are appreciably close to each other, while the remainder are sufficiently distant—from μ, ν, and each other. Then, in the matrices $\mathbf{M}_1$ and $\mathbf{M}_2$ [from (A12) and (A13) in the Appendix], $\nabla\nabla r_{\mu\nu}^{-1}$ and $\nabla\nabla r_{\nu\mu}^{-1}$ will contain all the significant interactions. Furthermore, if we normalize the distance between μ and ν by the bubble radius a and choose a coordinate system such that in it $\mathbf{r}_{\mu\nu}=(r,0,0)$, then it is straightforward to compute the determinant of the full mass matrix of the system:

$$\begin{aligned} &\det\mathbf{M}=\frac{\det\mathbf{M}_2}{\det(\mathbf{M}_1-\mathbf{I})}=\frac{1}{2^6}\left(1-\frac{9}{2}\left(\frac{1}{r}\right)^6+O\left(\left(\frac{1}{r}\right)^{12}\right)\right), \\ &\ln(\det\mathbf{M})\sim\text{ const}-\frac{9}{2}\left(\frac{1}{r}\right)^6,\quad\text{for large values of } r, \end{aligned} \tag{33}$$

and thus

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$$\beta\mathscr{U}_1 \sim \frac{9}{4}\left(\frac{1}{r_{\mu\nu}}\right)^6, \quad \text{for large } r_{\mu\nu}. \tag{34}$$

The coordinate dependence of the virtual mass matrix acts as a repulsive, r^{-6}, potential.

The other effective potential, $\mathscr{U}_2$, bears a very close resemblance to the effective potential energy of the bubbly flow as defined in (16) or by Smereka;[12] the absolute value of $\mathscr{U}_2$ increases as the bubbles form clusters positioned normal to the direction of their collective motion. However, the role played by $\mathscr{U}_1$ in the partition function Q differs fundamentally from that played by $\mathscr{U}_2$, in that it is insensitive to changes in the inverse temperature β. Thus, the influence of $\mathscr{U}_2$, unlike that of $\mathscr{U}_1$, is expected to diminish with increasing temperature.

D. Phase transition

The results of the previous section suggest that the spatial arrangement of bubbles in a suspension will strongly depend on the relative importance of the two effective potentials: the one due to the collective motion acts to make the bubbles cluster, while the effective potential due to the coordinate-dependent virtual mass is repulsive. Because $\beta\mathscr{U}_1$ is independent of temperature, while $\beta\mathscr{U}_2$ is proportional to $1/T$, the repulsive potential dominates at high temperature, while the collective dominates at low temperatures. Thus, one expects a "phase transition" to occur. A clustered bubbly flow ("condensed," or "frozen" phase) started at a low temperature will become more and more random as the temperature increases until at a certain value of T it becomes completely random ("gaseous," or "melted" phase).

We shall now attempt to predict the temperature of the phase transition (or, at least, an upper bound on it) for the bubbly suspension. We treat the suspension as a rarefied gas, in which not more than one pair of bubbles may be assumed to be interacting significantly at any one time, and obtain an expression for the second virial coefficient, B, as a function of the inverse temperature β for this gas. Recall the corresponding equation of state (Landau and Lifshitz[24]):

$$P = \frac{N}{\beta V}\left(1 + \frac{NB(\beta)}{V}\right), \tag{35}$$

where the gas has been assumed to consist of N particles, P is the pressure, and V is the volume occupied by the gas. The second virial coefficient can be evaluated as (Landau and Lifshitz[24])

$$B(\beta) = \frac{1}{2}\int (1 - e^{-\beta U^{12}})dV, \tag{36}$$

where U^{12} is the energy of interaction of the two particles and the integration is performed over the entire volume V. One can expect the phase transition to occur in the vicinity of the point where the isotherm exhibits singular behavior, i.e., where the derivative of the pressure with respect to the volume, obtained from the equation of state (35), vanishes. Differentiating (35) with respect to V yields for the value of the second virial coefficient corresponding to the phase transition temperature,

$$B(\beta_{pt}) = -\frac{V}{2N}. \tag{37}$$

The interaction energy U^{12} can be written as the sum of the potentials $\mathscr{U}_1$ and $\mathscr{U}_2$ from (31) and (32):

$$U^{12} = -\frac{1}{2\beta}\ln(\det \mathbf{M}) - \frac{\rho\tau}{2}\mathbf{e}_c \cdot \mathscr{M} \cdot \mathbf{e}_c .$$

Assume now that the collective motion of the bubbles takes place in the z-direction, i.e., that $\mathbf{e}_c = (0,0,e_c)$. The evaluation of $\mathbf{e}_c \cdot \mathscr{M} \cdot \mathbf{e}_c$ together with the result (33) for $\ln(\det\mathbf{M})$ yield (to the leading order in the small quantity a/r):

$$U^{12} = \frac{1}{\beta}\left(\ln 8 + \frac{9}{4}\left(\frac{a}{r}\right)^6\right) - \frac{\pi\rho e_c^2 a^3}{24}\left(N - \frac{3}{2}\pi\left(\frac{a}{r}\right)^3\right).$$

The evaluation of the integral in (36) produces (again, we retain the leading order terms only)

$$B(\beta) = V\left(8\ \ln\ 8 - N\pi\beta\frac{\rho e_c^2 a^3}{3}\right),$$

and from (37) we have the following estimate of the phase transition temperature:

$$\beta_{pt} = \frac{3(8\ln 8 + (2N)^{-1})}{\pi\rho e_c^2 a^3 N}.$$

In the above expression, we recognize a^3N as a quantity proportional to the void fraction of the flow $f = 4/3\pi a^3 N$. Assuming N large, it is easy to see that

$$\beta_{pt} = \frac{32\ln 8}{\rho e_c^2 f},$$

or, alternatively,

$$\frac{T_{pt}}{\rho e_c^2} = \frac{f}{32\ln 8} \approx 0.015 f. \tag{38}$$

Thus the phase transition temperature, in the approximation considered, is simply proportional to f. Suspensions with larger bubble concentrations are expected to undergo the transition between the clustered and disordered phases at higher temperatures.

In the next section, we shall test the validity of the above arguments by analyzing the results of numerical simulations of bubbles in a potential flow.

IV. NUMERICAL METHOD AND RESULTS

To simulate the motion of bubbles in a suspension, we place at the origin a cell containing N bubbles, and periodically replicate the cell to fill all space (cf. Appendix). At the beginning of a simulation run, the bubbles are assigned initial positions and velocities. The equation of motion (9) is numerically integrated to produce the bubbles' trajectories, along with data on their velocities, accelerations, etc., so that we will be able to follow the total energy and impulse of the system—these quantities must be conserved at all times. As the simulation evolves in time, we expect the influence of the initial coordinates and velocities to diminish so that the resulting distribution will be determined solely by the nature of

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external and internal forces acting on the bubbles. In the course of a run, the bubbles' trajectories along with the effective kinetic and potential energies and other quantities of interest are computed. The stabilization of the effective energies signals equilibration, and all necessary statistics can then be computed.

A. Dynamics of a collision of a pair of bubbles

We have already covered the derivation of the equation of motion. As far as its numerical integration is concerned, there are numerous codes and packages available, and we shall not discuss this aspect in detail. However, one issue still needs to be resolved—that of a possible collision of a pair of bubbles. Here we follow Sangani and Didwania.[14] In an inviscid flow bubbles can come arbitrarily close to each other. The viscous forces are small at large Reynolds numbers and therefore cannot prevent the bubbles from touching one another. Thus the bubbles can come in contact and the following two possibilities arise: they will either coalesce if the surface tension forces are large compared to the inertia forces, *in the absence of surface-active impurities*, or, if the latter condition is not satisfied, as numerous experiments indicate, the bubbles will bounce away almost instantaneously. As reported by Kok,[20] if the concentration of the impurities is not too high, the observed trajectories of the bubbles are in a very good agreement with those obtained theoretically using the potential flow approximation for the fluid velocity. On the other hand, if the concentration of impurities is moderate or high, wake formation affects the dynamics of the pair of bubbles, and the potential flow approximation will have to be modified to include wake effects.

When a pair of bubbles—labeled for convenience 1 and 2—undergoes a collision, the total impulse and kinetic energy of the liquid remain unchanged. Suppose that a very short range force comes into play when these two bubbles approach each other and that this force is directed along the separation vector $\mathbf{d}$ given by

$$\mathbf{d}=\frac{1}{2a}(\mathbf{r}_1-\mathbf{r}_2).$$

We then may think that an equal and opposite impulse is applied to the two bubbles,

$$\frac{d\mathscr{I}}{dt}=F_c\delta(t-t_c)\mathbf{d},$$

for time t close to the collision time t_c. Here, F_c is the magnitude of the impulse. Integrating with respect to time in the immediate vicinity of t_c leads to a change in the impulse of bubble 1 during the collision process $\triangle\mathscr{I}=F_c\mathbf{d}$. The change in the impulse of bubble 2 is equal in magnitude and opposite in the direction to that of bubble 1, and the impulse change of all the other bubbles in the cell is zero. Thus the total impulse of the system is conserved and we need only calculate the change in the kinetic energy of the system.

The impulse due to the motion of bubble μ is

$$\mathbf{P}^\mu=\mathbf{M}^{\mu\nu}\cdot\mathbf{U}^\nu,$$

and the total energy of the system can be written as

$$\mathscr{T}=\sum_\mu \mathscr{T}^\mu=\frac{1}{2}\rho\tau\sum_\mu \mathbf{U}^\mu\cdot\mathbf{P}^\mu.$$

We therefore require that the sum of $\mathscr{T}^\mu$ over all the bubbles does not change during the collision, i.e.,

$$\sum_\mu \triangle\mathscr{T}^\mu=0. \tag{39}$$

During the collision, the velocities of the bubbles change linearly with the magnitude of the impulse F_c; thus,

$$\frac{2}{\rho\tau}\triangle\mathscr{T}^\mu=\triangle(\mathbf{U}^\mu\cdot\mathbf{P}^\mu)$$
$$=F_c(\mathbf{U}^\mu\cdot\hat{\mathbf{P}}^\mu+\hat{\mathbf{U}}^\mu\cdot\mathbf{P}^\mu)+F_c^2(\hat{\mathbf{U}}^\mu\cdot\hat{\mathbf{P}}^\mu), \tag{40}$$

where $\hat{\mathbf{U}}^\mu$ is the change in the velocity of the bubble μ and $\mathbf{U}^\mu$ is the velocity before the collision. The same convention applies to $\mathbf{P}^\mu$ and in particular we have

$$\hat{\mathbf{P}}^1=-\hat{\mathbf{P}}^2=\mathbf{d},$$

whereas for all $\mu>2$, $\hat{\mathbf{P}}^\mu=0$. Now, substituting (40) into (39) yields two values of F_c, one of which equals zero and corresponds to no collision, and the other corresponds to the magnitude of the total impulse during the collision,

$$F_c=-\frac{\Sigma_\mu(\mathbf{U}^\mu\cdot\hat{\mathbf{P}}^\mu+\hat{\mathbf{U}}^\mu\cdot\mathbf{P}^\mu)}{\sum_\mu \hat{\mathbf{U}}^\mu\cdot\hat{\mathbf{P}}^\mu}. \tag{41}$$

Thus, in dynamic simulations we first move the bubbles to the point where they collide and determine $\hat{\mathbf{U}}^\mu$ by solving for the velocity potential $\hat{\phi}$ given the impulses $\hat{\mathbf{P}}^\mu$ associated with each bubble. Substituting for $\hat{\mathbf{U}}^\mu$ in (41) allows us to determine F_c and hence the values of $\mathbf{P}^\mu$ and $\mathbf{U}^\mu$ for all the bubbles immediately after the collision.

The fact that generally the velocities of all the bubbles will change in the aftermath of the collision of just two bubbles can be explained as follows. The fluid and the bubbles are assumed to be incompressible, and thus the information about the collision is instantaneously transmitted to all the bubbles in the flow. (Physically, it is transmitted at the sound speed, which is infinite for the incompressible media.) Thus all bubbles will change their velocities in such a manner so that both the total impulse and the kinetic energy of the suspension will be conserved.

B. Bubbly flow in the absence of external forces

To study configurations of inviscid, neutrally buoyant bubbles ($\mathbf{F}^g=\mathbf{F}^v=0$) at different values of the bubble temperature, we conducted numerical experiments with 27 and 64 bubbles placed in a cubic unit cell at different values of the void fraction. The initial positions of the bubbles varied from closely packed clusters oriented normally to the z-direction (close to a minimum of the effective potential energy, as the collective motion in all experiments was also directed along the z-axis) to bubbles randomly distributed throughout the cell; the initial velocities—from purely collective motion, all with the same velocity, to completely random motion, with negligible resultant collective motion. As a

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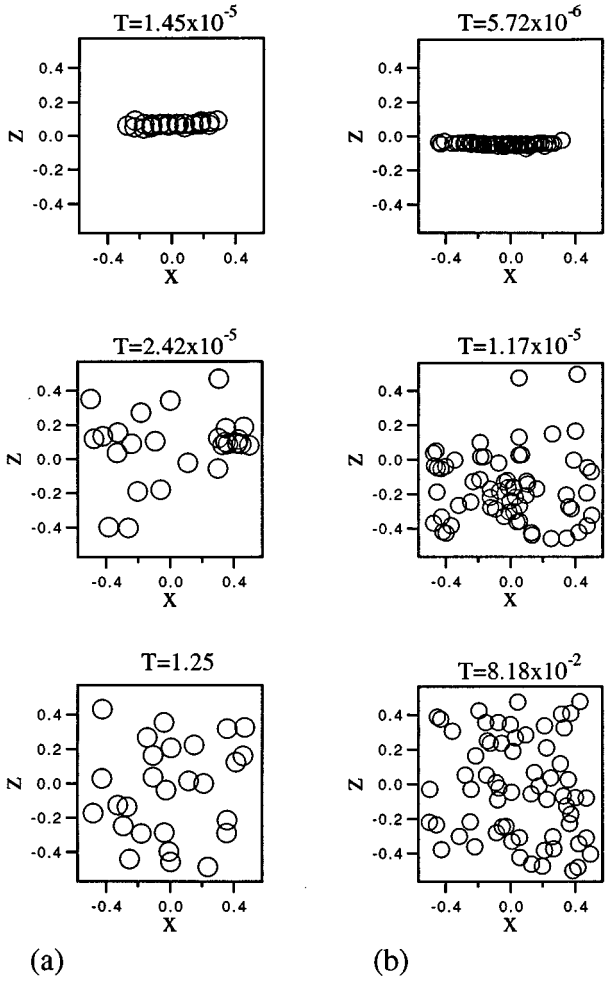

FIG. 1. Representative configurations of 27 (a) and 64 (b) bubbles, at a void fraction of 0.014, projected onto the (x,z) plane. As the temperature increases, the bubbles become less clustered.

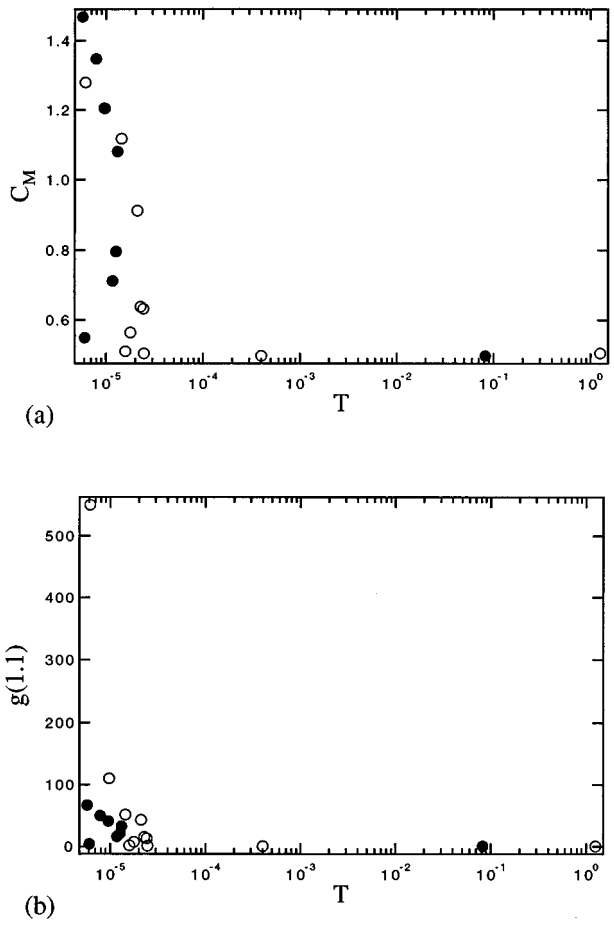

FIG. 2. Added-mass coefficient (a) and $g(1.1)$ (b) versus the bubble temperature, as obtained in numerical runs with 27 (empty circles) and 64 bubbles (filled circles) at a void fraction of 0.014.

result, the equilibrium temperatures in these experiments varied greatly. Figure 1 presents typical equilibrium configurations corresponding to different temperatures of 27 and 64 bubbles. At low temperatures the bubbles have very little kinetic energy and remain clustered as their configuration corresponds to a minimum of the effective potential energy (cf. Smereka[12]). The individual bubbles' motions are highly correlated and the added-mass coefficient is relatively large. As the temperature increases, the bubbles develop more chaotic motion until, at a certain point, the clusters disappear completely, the bubbles become randomly dispersed, and, appropriately, the added-mass coefficient takes on values close to 1/2; after this point, there are no changes in the spatial arrangement of the bubbles. This point can be described as one where the transition between the clustered and random phases becomes complete.

In Figs. 2 and 3, we have plotted the added-mass coefficient C_M and the value of the pair-distribution function at 1.1 bubble diameters, $g(1.1)$, versus the bubble temperature T for two different void fractions, $f=0.014$ and $f=0.11$. Both C_M and $g(1.1)$ can serve as measures of the relative degree of clustering of the bubbles: they increase as the bubbles collect themselves in clusters. The results in Figs. 2 and 3 suggest that the transition between the clustered and random phases takes place in a temperature interval near $T\approx 5\times 10^{-5}$ for $f=0.014$ and near $T\approx 5\times 10^{-4}$ for $f=0.11$. Because we use a microcanonical ensemble in our simulations, coexistence of the two phases is observed in this temperature interval; it would thus be appropriately called a "two-phase" region. Also, because of the relatively small number of bubbles in the ensemble, we observe that the values of C_M and $g(1.1)$ are not unique when plotted against the temperature (cf. Sec. 11.3 of Allen and Tildesley[26]). Nevertheless, at very low values of the temperature, e.g., less than 10^{-6}, the bubbles are always close-packed in a flat cluster, while at high enough temperatures, $T>10^{-2}$ say, they are always randomly distributed throughout space. These results are evidence for the existence of the two potentials (34) and (32). At low temperatures, the influence of the r^{-6} repulsive potential (34) is negligible and the bubbles form flat clusters due to the action of their collective potential given by (32). As the temperature grows, however, the repulsive potential gains more prominence and finally prohibits clustering of the bubbles (even though there is still significant collective motion).

It is interesting to see how well our earlier calculation of

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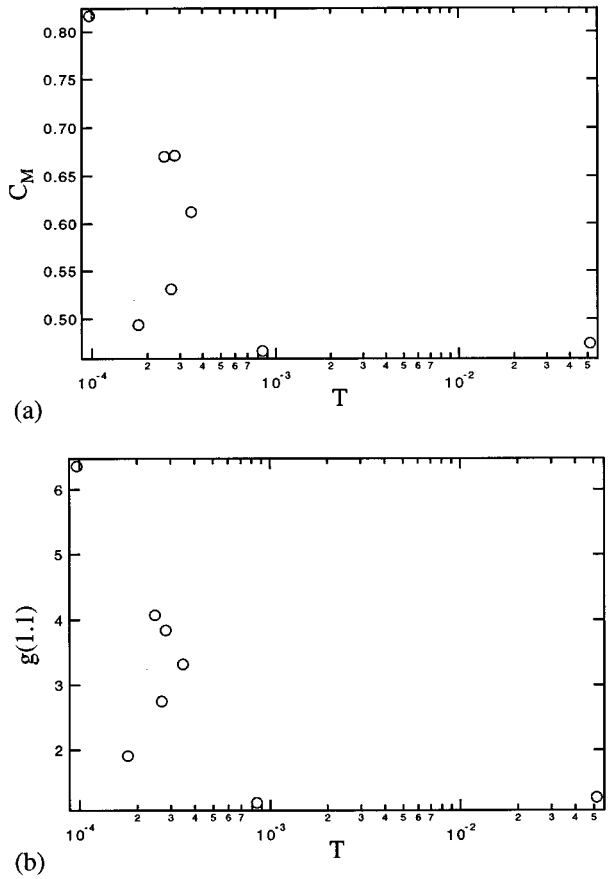

FIG. 3. Added-mass coefficient (a) and $g(1.1)$ (b) versus the bubble temperature for 27 bubbles at a void fraction of 0.11.

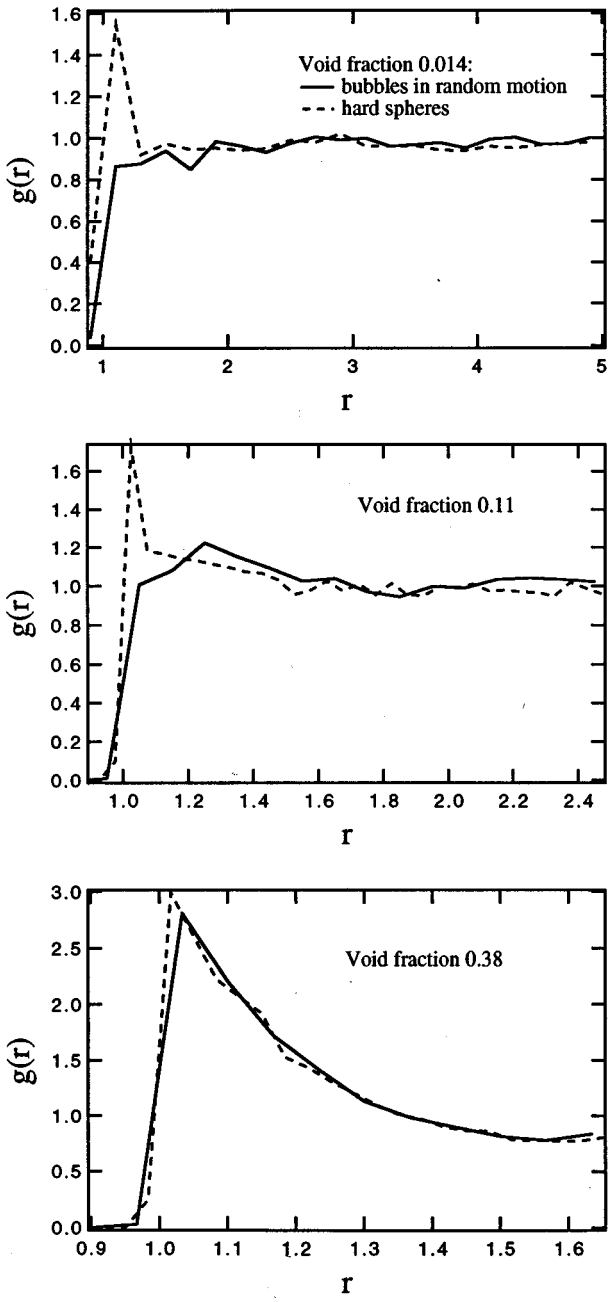

FIG. 4. Pair-distribution functions: random bubbles versus hard spheres. The bubbles exhibit repulsion resulting in the absence of a peak at contact.

the transition temperature (38) compares with the results in Figs. 2 and 3. For the void fraction $f=0.014$, (38) predicts the phase transition temperature $T_{pt}=2.1\times10^{-4}$, and for $f=0.11$, $T_{pt}=1.6\times10^{-3}$. Thus, in both cases, (38) seems to overpredict the phase transition temperature by a factor of 5. On the other hand, it should be noted that the qualitative prediction that T_{pt} is proportional to f holds. Also, one has to keep in mind that (38) should be viewed as an estimate of the upper bound on the phase transition temperature since we only considered the second virial coefficient.

In another series of runs, conducted at three different values of the void fraction, the bubbles were initially randomly distributed in space and assigned random velocities so that the total momentum of the flow in the unit cell was small compared to the characteristic value of momentum associated with the motion of one bubble. This ensured that the bubbles did not have any appreciable collective motion during the simulation; thus, their motion could be termed purely chaotic and it was hoped that there would be evidence of the r^{-6} repulsive potential that we discussed earlier.

We compare pair-distribution functions of bubbles without collective motion against those of hard spheres at three different void fractions: 0.014, 0.11, and 0.38 in Fig. 4. At all values of the void fraction the hard-sphere pair-distribution function has a noticeable peak at contact. On the other hand, the bubble distribution either completely lacks such a peak, or, at the largest void fraction where the bubbles have little freedom of movement, the peak is lower in magnitude. These results confirm our earlier predictions that an effective repulsive potential governs the bubbles' behavior when there is little collective motion.

We conclude this section by commenting on the results of our attempt to apply a Monte Carlo procedure to compute the partition function (30) for our bubbly flow as a canonical ensemble. Unfortunately, these experiments proved prohibitively computer-expensive since we needed on the order of a million of bubble configurations, and the inversion of a $3N\times3N$ matrix was required in order to compute the mass

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matrix for each configuration. When we gave the Monte Carlo routine the several thousand configurations that had been obtained from the molecular dynamics simulation, all but very few of these configurations were found suitable for a Monte Carlo calculation of Q, thus providing an indirect confirmation that the bubbly flow behaves as a thermodynamic system.

C. Bubbly flow with gravity and viscous drag

To study the influence of gravity (or, equivalently, buoyancy) as well as viscous dissipation on the structure and dynamics of a bubbly flow at high Re numbers, we used the following expressions for the forces on a single bubble μ:

$$\mathbf{F}^g = \rho\tau\mathbf{g}, \tag{42}$$

$$\mathbf{F}^v = 12\pi\eta a\mathbf{U}^\mu, \tag{43}$$

where η is the viscosity of the liquid. The viscous drag, as given by (43), is valid only as a leading order approximation (Smereka[12]). This approximation, however, will be sufficient for the purposes of the present discussion. [In general, viscous forces are obtainable from the Rayleigh dissipation function, $\mathbf{F}^v = -\partial F/\partial\dot{\mathbf{R}}$, where $F = \frac{1}{2}\dot{E}_v$, and $\dot{E}_v = 2\eta\int|\nabla\nabla\phi|^2 dV$ is the rate of energy dissipation for the inviscid flow. Equation (43) is the result for an isolated bubble.]

First, we discuss the influence of buoyancy, in the absence of viscous dissipation. In our numerical simulations, we have always found the bubbles, regardless of their initial positions and velocities, to aggregate in flat clusters normal to the direction of gravity. Why this should be the case can be explained by examining the following situation. Consider two bubbles separated by a certain distance rising in a liquid under the influence of gravity. Such bubbles will be subjected to a force of mutual attraction; this force will grow in magnitude as the bubbles approach each other. Simultaneously, the velocity of the bubbles' rise will grow and this too will result in a larger value of the force of attraction. If the bubbles collide, they will not come as far apart as their initial separation, while their velocities collinear to the direction of gravity will grow larger, which in turn will result in an increasing attractive force, and so forth. Eventually the two bubbles will form a cluster with the line joining their centers directed normally to gravity. We can add here that Kok[19] has shown that a pair of bubbles rising under buoyancy will always rotate to be oriented in the cross-stream direction, regardless of the pair's initial orientation. This point further strengthens the applicability of the above argument to our problem.

Bubbles under the action of gravity can also be characterized in thermodynamic terms given in Sec. III of this paper. As the bubbles accelerate in the direction of gravity, their collective velocity in this direction grows faster than the magnitude of their random motion, and thus the temperature of the suspension decreases resulting in a phase transition from randomness to flat clusters of bubbles normal to gravity.

Finally, the combined effect of buoyancy and viscous dissipation will also be that of eventual clustering of the bubbles, regardless of their initial spatial arrangement and velocity distribution. Due to viscous dissipation, the initial velocities whose direction will not coincide with that of gravity will decrease in magnitude until they become negligible. From that point on, all the bubbles will be moving in the direction of gravity with the same velocity that can be determined from the balance of viscous drag (43) and buoyancy (42). This motion will lead to the creation of flat clusters of bubbles normal to the direction of gravity. The corresponding numerical results have been found in agreement with the above discussion.

Thus, it can be concluded that buoyancy, acting on its own or in combination with viscous dissipation, leads to the formation of clusters of bubbles positioned broadside to the direction of gravity.

V. CONCLUSIONS

In this paper, we have shown that bubbly liquids at high Reynolds numbers can be modeled as a gas of particles obeying the laws of statistical mechanics. studying the dynamics of bubbly liquids at high Reynolds numbers. The inviscid, massless, neutrally buoyant bubbles are approximated as dipoles in potential flow of an ideal fluid. The integrals of the bubbles' motion—the kinetic energy and the linear momentum of the flow—are derived. The second-rank tensor that arises in the derivation plays the role of an added, or virtual, mass of the flow; it accounts for the dynamical role of the fluid brought into motion by the bubbles. The expressions for the integrals of motion are similar to those of ''classical'' systems of material particles in that the energy is a quadratic (and Hamiltonian) and the impulse a linear form of the vector of the bubbles' velocities.

These similarities are useful in that they let us conduct further analysis along the lines of such traditional methods as statistical mechanics and molecular dynamics. Moreover, they provide a background for highlighting and analyzing the important physical features that distinguish a collection of bubbles in potential flow from material particles in vacuum. These features are the following.

(i) The virtual mass of the bubbly suspension depends on the bubbles' relative spatial arrangement.

(ii) The collective motion of the bubbles relatively to the otherwise undisturbed underlying liquid is coupled to the internal degrees of freedom.

Treating a collection of dipoles in potential flow as a canonical ensemble, we account for the above features by including in the partition function not only the system's Hamiltonian, but also its total impulse in the frame of reference in which the fluid would stay motionless were it not for the presence of the bubbles. Similarly, for non-spherical bubbles or particles in potential flow, one would need to explicitly include the total angular momentum in the integral for the partition function, thus accounting for the influence of the total angular momentum on the internal degrees of freedom of the particles.

Having constructed the partition function, we proceed to treat the bubbly dispersion as an ensemble of particles interacting by means of additional effective thermodynamic po-

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tentials capturing the dynamics that distinguishes the bubbles from material particles in vacuum. In principle, the partition function is all that is needed to characterize fully the thermodynamic state of the ensemble. Though unable to compute the partition function for the bubbles with position-dependent mass, we can manipulate and analyze it such that conclusions can be made about the collective effects in the suspension.

In a fashion similar to that used in studying atomic systems, we define the temperature of the bubbly suspension as a measure of the bubbles' random motion. However, unlike classical systems, due to the position-dependent added mass of the bubbles, their random motion also results in a repulsive r^{-6} potential.

On the other hand, the collective motion of the bubbles, resulting in a non-zero total impulse of the suspension, also influences the bubbles' spatial arrangement because of its coupling to the internal degrees of freedom of the bubbles. By including the total impulse in the integral for the partition function we are able to describe the action of the collective motion as that of another effective potential. The action of this potential results in the formation of flat clusters of bubbles oriented normally to the vector of the total impulse of the suspension.

Finally, in our thermodynamic treatment, by computing the second virial coefficient for the ensemble of bubbles we estimate the temperature of the phase transition between the clustered and random phases.

The above theoretical results were verified by conducting molecular-dynamics-like simulations of bubbles as dipoles in potential flow.

This is the first instance we know of that has shown that a system of hydrodynamically interacting particles behaves as a thermodynamic system, obeying the same laws of classical statistical mechanics. This statistical mechanical approach may find use in modeling other hydrodynamic systems for which a Hamiltonian can be found.

ACKNOWLEDGMENT

The authors are grateful to Professor Zhen-Gang Wang for a number of useful discussions.

APPENDIX: MASS MATRIX FOR A BUBBLY FLOW AT HIGH REYNOLDS NUMBERS

1. Derivation for a finite number of bubbles

Following the method for solving Laplace's equation by Bonnecaze and Brady,[27,28] we write Laplace's equation (1) in its integral form,

$$\phi(\mathbf{x})-\phi^E(\mathbf{x})=\frac{1}{4\pi}\sum_\nu \int_{\partial\Omega^\nu}\left(\mathbf{F}\frac{1}{\lambda r}+\phi\nabla_y\frac{1}{r}\right)\cdot\mathbf{n}dS, \tag{A1}$$

where $\phi(\mathbf{x})$ is the potential field at $\mathbf{x}$, $\phi^E(\mathbf{x})$ is the imposed or external potential field at $\mathbf{x}$ in the absence of any bubbles, $\mathbf{F}$ is the flux, defined as $\mathbf{F}=-\lambda\nabla_y\phi$, λ being the conductivity of the medium (equal to 1 in the fluid and 0 inside a bubble), and the integral is over the surface of the ν bubble. The distance $r=|\mathbf{x}-\mathbf{y}|$ and $\nabla_y=\partial/\partial(\mathbf{y}-\mathbf{x})$. The bubble moments are defined by

$$q^\nu\equiv\int_{\partial\Omega^\nu}\mathbf{F}\cdot\mathbf{n}dS, \tag{A2}$$

$$\mathbf{S}^\nu\equiv\int_{\partial\Omega^\nu}(\mathbf{x}\mathbf{F}+\lambda\phi\mathbf{I})\cdot\mathbf{n}dS, \tag{A3}$$

which are the charge (monopole) and dipole, respectively, and where the position $\mathbf{x}$ in the moment definitions is defined relative to the center of the spherical bubble. Here and throughout, $\mathbf{I}$ is the usual notation for the identity matrix. Expanding the integral in (A1) in terms of bubble moments yields

$$\phi(\mathbf{x})-\phi^E(\mathbf{x})=\frac{1}{4\pi\lambda}\sum_\nu\left(q^\nu\frac{1}{r}+\mathbf{S}^\nu\cdot\nabla_y\frac{1}{r}+\ \dots\ \right),$$

where the expansion has been truncated at the dipole level and the moment propagators $(1/r,\nabla_y 1/r,\dots)$ are evaluated at $\mathbf{x}-\mathbf{R}^\nu$, where $\mathbf{R}^\nu$ is the center of bubble ν.

We can also act on the integral form of Laplace's equation (A1) with $\nabla=\nabla_x$ to obtain

$$\nabla\phi(\mathbf{x})-\nabla\phi^E(\mathbf{x})$$

$$=-\frac{1}{4\pi\lambda}\sum_\nu\left(q^\nu\nabla_y\frac{1}{r}+\mathbf{S}^\nu\cdot\nabla_y\nabla_y\frac{1}{r}+\ \dots\ \right). \tag{A4}$$

In our problem, $\mathbf{F}=-\mathbf{v}$, where $\mathbf{v}$ is the fluid velocity. The evaluation of the integrals in (A2) and (A3) yields

$$q^\nu=0, \tag{A5}$$

$$\mathbf{S}^\nu=\int_{\partial\Omega^\nu}\phi\mathbf{n}dS-\tau\mathbf{U}^\nu. \tag{A6}$$

Before going further, we need to address the issue of including multipoles of order higher than two. Accounting for the quadrupoles, octupoles, etc. will surely enhance the precision of the bubble dynamics simulations. However, as such a modification will inevitably result in a manifold slowing down of the speed of computer calculations, one must carefully consider whether the inclusion of higher order moments will affect the results significantly. The bubble problem is analogous to the effective conductivity problem with the bubbles having zero conductivity, and for this reason, as Bonnecaze and Brady[27] have shown, the inclusion of multipoles of order higher than the dipole will lead to insignificant changes in the final results. In earlier work, we have consid-

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ered the two-bubble problem, and arrived at the same conclusion. Having said that, we shall now truncate the expansion (A4) at the dipole level, thus obtaining, with the help of (A5) and (A6),

$$\nabla\phi(\mathbf{x})-\nabla\phi^E(\mathbf{x})$$
$$=-\frac{1}{4\pi}\sum_{\nu}\left(\int_{\partial\Omega^\nu}\phi\mathbf{n}dS-\tau\mathbf{U}^\nu\right)\cdot\nabla_y\nabla_y\frac{1}{r}. \tag{A7}$$

Recalling equation (2) for the kinetic energy, we see that in order to calculate the kinetic energy of the bubbly flow, we shall need the integrals $\int_{\partial\Omega^\nu}\phi\mathbf{n}dS$. Applying the divergence theorem and placing the origin at the center of the bubble, we find

$$\frac{1}{a}\int_{\partial\Omega^\nu}\phi\mathbf{x}dS=\int_{\partial\Omega^\nu}\phi\mathbf{n}dS=\int_{V_f}\nabla\phi dV.$$

To determine the above quantity, we derive a Faxén-type law, which relates the integral $\int_{\partial\Omega^\nu}\phi\mathbf{n}dS$ to the analogous integral $\int_{\partial\Omega^\nu}\phi'\mathbf{n}dS$ in the ν bubble's absence (the fact being denoted by the prime), known from the potential theory to be equal to $\tau\nabla_y\phi'(\mathbf{R}^\nu)$ (cf. Kellogg[29]), and the dipole of that bubble. As a first step, we rewrite (A1) in the form appropriate for a point on the surface of the bubble,

$$\phi(\mathbf{x})-2\phi'(\mathbf{x})=\frac{1}{2\pi a}\int_{\partial\Omega^{\nu_y}}\left(\mathbf{F}\frac{1}{r}+\phi\frac{(\mathbf{x}-\mathbf{y})}{r^3}\right)\cdot\mathbf{y}dS_y.$$

Now multiply both sides of the above expression by $\mathbf{x}$ and integrate the product over the surface of the sphere with respect to the variable $\mathbf{x}$ to give

$$\int_{\partial\Omega^{\nu_x}}\mathbf{x}[\phi(\mathbf{x})-2\phi'(\mathbf{x})]dS_x$$
$$=\frac{1}{2\pi a}\int_{\partial\Omega^{\nu_x}}\int_{\partial\Omega^{\nu_y}}\mathbf{x}\left[\left(\mathbf{F}\frac{1}{r}+\phi\frac{(\mathbf{x}-\mathbf{y})}{r^3}\right)\cdot\mathbf{y}\right]dS_ydS_x. \tag{A8}$$

The evaluation of the right-hand side of (A8) yields

$$\int_{\partial\Omega^{\nu_x}}\mathbf{x}[\phi(\mathbf{x})-2\phi'(\mathbf{x})]dS_x$$
$$=-\frac{8}{9}\pi a^4\mathbf{U}-\frac{1}{3}\int_{\partial\Omega^{\nu_y}}\mathbf{y}\phi(\mathbf{y})dS_y,$$

or just

$$\frac{2}{3}\overline{\mathbf{x}\phi(\mathbf{x})}-\overline{\mathbf{x}\phi'(\mathbf{x})}=-\frac{a^2}{9}\mathbf{U}, \tag{A9}$$

where

$$\overline{\mathbf{x}\phi(\mathbf{x})}=\frac{1}{4\pi a^2}\int_{\partial\Omega^{\nu_x}}\mathbf{x}\phi(\mathbf{x})dS_x.$$

We can now introduce the following quantity:

$$\overline{\nabla\phi(\mathbf{x})}=\frac{1}{\tau}\int_{V_f}\nabla\phi(\mathbf{x})dV,$$

and, by virtue of the divergence theorem,

$$\overline{\mathbf{x}\phi(\mathbf{x})}=\frac{a^2}{3}\overline{\nabla\phi(\mathbf{x})}.$$

Now (52) can be put in the form

$$\frac{2}{3}\overline{\nabla\phi(\mathbf{x})}-\overline{\nabla\phi'(\mathbf{x})}=-\frac{\mathbf{U}}{3}. \tag{A10}$$

We remark here briefly that a Faxén-type relation, analogous to (A10), holds for the quantity

$$\overline{\phi(\mathbf{x})}=\frac{1}{4\pi a^2}\int_{\partial\Omega^{\nu_x}}\phi(\mathbf{x})dS_x,$$

and it reads as

$$\overline{\phi(\mathbf{x})}-\overline{\phi'(\mathbf{x})}=\frac{q}{4\pi a},$$

which, upon recalling the expression for the charge (A5), reduces to the simple form

$$\overline{\phi(\mathbf{x})}-\overline{\phi'(\mathbf{x})}=0.$$

For bubble μ, in the absence of the gradient of any external potential, i.e., when the fluid's motion is wholly due to the motion of the bubbles, using (A7), we obtain

$$\nabla\phi'(\mathbf{R}^\mu)=-\frac{a^3}{3}\sum_{\nu\neq\mu}(\overline{\nabla\phi(\mathbf{R}^\nu)}-\mathbf{U}^\nu)\cdot\nabla_y\nabla_y\frac{1}{r_{\mu\nu}},$$

where $r_{\mu\nu}=|\mathbf{R}^\mu-\mathbf{R}^\nu|$. Combining the above result with the Faxén law (A9) and recalling that $\overline{\nabla\phi'(\mathbf{x})}=\nabla\phi'(\mathbf{x})$ (cf. Kellogg[29]), we have

$$\overline{\nabla\phi(\mathbf{R}^\mu)}=-\frac{\mathbf{U}^\mu}{2}-\frac{a^3}{2}\sum_{\nu\neq\mu}(\overline{\nabla\phi(\mathbf{R}^\nu)}-\mathbf{U}^\nu)\cdot\nabla_y\nabla_y\frac{1}{r_{\mu\nu}}. \tag{A11}$$

Now we find it convenient to put (A11) in matrix form,

$$\overline{\nabla\phi(\mathbf{R})}=\mathbf{M}_1\cdot\overline{\nabla\phi(\mathbf{R})}+\mathbf{M}_2\cdot\mathbf{U},$$

where $\overline{\nabla\phi(\mathbf{R})}$ and $\mathbf{U}$ are vectors defined as

$$\overline{\nabla\phi(\mathbf{R})}=[\overline{\nabla\phi(\mathbf{R}^1)},\overline{\nabla\phi(\mathbf{R}^2)},\ldots,\overline{\nabla\phi(\mathbf{R}^N)}]^T$$

and

$$\mathbf{U}=[\mathbf{U}^1,\mathbf{U}^2,\ldots,\mathbf{U}^N]^T,$$

whereas $\mathbf{M}_1$ and $\mathbf{M}_2$ are matrices of the following form:

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$$\mathbf{M}_1 = -\frac{a^3}{2}\begin{pmatrix} 0 & \nabla_y\nabla_y r_{12}^{-1} & \nabla_y\nabla_y r_{13}^{-1} & \cdots & \nabla_y\nabla_y r_{1N}^{-1} \\ \nabla_y\nabla_y r_{21}^{-1} & 0 & \nabla_y\nabla_y r_{23}^{-1} & \cdots & \nabla_y\nabla_y r_{2N}^{-1} \\ \cdots & \cdots & \cdots & \cdots & \cdots \\ \nabla_y\nabla_y r_{N1}^{-1} & \nabla_y\nabla_y r_{N2}^{-1} & \nabla_y\nabla_y r_{N3}^{-1} & \cdots & 0 \end{pmatrix}, \tag{A12}$$

$$\mathbf{M}_2 = -\frac{a^3}{2}\begin{pmatrix} a^{-3}\mathbf{I} & -\nabla_y\nabla_y r_{12}^{-1} & -\nabla_y\nabla_y r_{13}^{-1} & \cdots & -\nabla_y\nabla_y r_{1N}^{-1} \\ -\nabla_y\nabla_y r_{21}^{-1} & a^{-3}\mathbf{I} & -\nabla_y\nabla_y r_{23}^{-1} & \cdots & -\nabla_y\nabla_y r_{2N}^{-1} \\ \cdots & \cdots & \cdots & \cdots & \cdots \\ -\nabla_y\nabla_y r_{N1}^{-1} & -\nabla_y\nabla_y r_{N2}^{-1} & -\nabla_y\nabla_y r_{N3}^{-1} & \cdots & a^{-3}\mathbf{I} \end{pmatrix}. \tag{A13}$$

We are now in a position to deduce a formula that will establish a direct relation between the vectors $\overline{\nabla\phi(\mathbf{R})}$ and $\mathbf{U}$,

$$\overline{\nabla\phi(\mathbf{R})} = (\mathbf{I}-\mathbf{M}_1)^{-1}\cdot\mathbf{M}_2\cdot\mathbf{U}.$$

We shall define as the *mass matrix* the following matrix of tensors:

$$\mathbf{M} \equiv -(\mathbf{I}-\mathbf{M}_1)^{-1}\cdot\mathbf{M}_2 .$$

The reason why it is appropriate for the matrix $\mathbf{M}$ to be called mass matrix will become clear in the course of the following argument. Once again, recall the expression (2) for the total kinetic energy of the fluid in our problem together with the relation

$$\sum_\nu \mathbf{U}^\nu \cdot \int_{\partial\Omega^\nu} \phi\mathbf{n}^\nu dS = \tau\mathbf{U}\cdot\overline{\nabla\phi} = -\tau\mathbf{U}\cdot\mathbf{M}\cdot\mathbf{U}.$$

Then the expression for the total kinetic energy can be rewritten as

$$\mathcal{T} = \tfrac{1}{2}\rho\tau\mathbf{U}\cdot\mathbf{M}\cdot\mathbf{U}, \tag{A14}$$

and it is now obvious that, indeed, the matrix $\mathbf{M}$ plays here the role of virtual, or added mass of the entire system, normalized by a factor of $\rho\tau$.

2. Extension to infinite systems

To model the infinite medium, we apply periodic boundary conditions to a cell of volume V_c containing N particles. We are no longer in a position to argue that the velocity at infinity is zero. Rather, the whole medium is now set into motion, i.e., there is a bulk flow; the kinetic energy (per unit cell) of this flow is

$$\mathcal{T}_{bf} = \frac{\mathbf{P}_t\cdot\mathbf{P}_t}{2\rho V_c(1-f)},$$

and there is a corresponding velocity,

$$\mathbf{U}_{bf} = \frac{\mathbf{P}_t}{\rho V_c(1-f)},$$

where f is the void fraction.

Returning to the potential flow problem, the bulk flow corresponds to a continuous dipole distribution throughout the medium. We must account for this distribution in our derivation of an expression for the gradient of the potential. Bonnecaze and Brady[27] have addressed this problem; proceeding along the lines of Secs. IV and V of their paper, we rewrite the equation for the potential gradient in a form similar to (A4), recalling the expression for the charge (A5) and accounting for an infinite number of bubbles [cf. Eq. (35), Bonnecaze and Brady[27]],

$$\nabla\phi(\mathbf{x}) - \nabla\phi^E(\mathbf{x}) = -\frac{n\langle\mathbf{S}\rangle}{3} - \frac{1}{4\pi}\sum_\nu \mathbf{S}^\nu\cdot\nabla_y\nabla_y\frac{1}{r} + \frac{1}{4\pi}\int_{V-V_\epsilon} n\langle\mathbf{S}\rangle\cdot\nabla_y\nabla_y\frac{1}{r}dV,$$

where $n\langle\mathbf{S}\rangle$ is the average particle dipole density, related to the motion of the entire liquid as a whole, V is the volume enclosed by S^∞, a distant boundary surface, which surrounds $\mathbf{x}$, and V_ϵ is the volume of a sphere of radius ϵ surrounding $\mathbf{x}$.

The cell is periodically replicated to fill all space; the sums over the infinite particles are replaced by double sums over the N particles in the lth cell of the system and over all the replicated cells. To make the resulting double sums converge rapidly, we use the Ewald summation expression (A2.4) from Bonnecaze and Brady,[27]

$$\frac{n\langle\mathbf{S}\rangle}{3} + \frac{1}{4\pi}\sum_\nu \mathbf{S}^\nu\cdot\nabla_y\nabla_y\frac{1}{r} - \frac{1}{4\pi}\int_{V-V_\epsilon} n\langle\mathbf{S}\rangle\cdot\nabla_y\nabla_y\frac{1}{r}dV = \frac{\mathbf{S}^{\mu_0}}{4\pi}\frac{40}{3}\xi^3\pi^{-\frac{1}{2}} + \frac{1}{4\pi}\sum_l \sum_{\substack{\nu \\ \nu_l\neq\mu_0}} \mathbf{S}^{\nu_l}\cdot\mathbf{\Delta}^{(r)} + \frac{1}{4\pi}\frac{1}{V_c}\sum_{\substack{k \\ k\neq 0}} \sum_\nu \mathbf{S}^{\nu_l}\cdot\mathbf{\Delta}^{(k)},$$

where

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$$\mathbf{\Delta}^{(r)}=\left[(8\xi^7r^4-36\xi^5r^2+16\xi^3+4\xi r^{-2})\frac{e^{-\xi^2r^2}}{\sqrt{\pi}}+\frac{2\ \mathrm{erfc}(\xi r)}{r^3}\right]\frac{(\mathbf{R}^{\mu 0}-\mathbf{R}^{\nu l})(\mathbf{R}^{\mu 0}-\mathbf{R}^{\nu l})}{r^2}+\left[(4\xi^5r^3-12\xi^3r+2\xi r^{-1})\frac{e^{-\xi^2r^2}}{\sqrt{\pi}}+\frac{\mathrm{erfc}(\xi r)}{r^2}\right]\times\left[\frac{(\mathbf{R}^{\mu 0}-\mathbf{R}^{\nu l})(\mathbf{R}^{\mu 0}-\mathbf{R}^{\nu l})}{r^3}-\frac{\mathbf{I}}{r}\right],$$

$$\mathbf{\Delta}^{(k)}=\frac{4\pi}{k^2}\left[1+\frac{1}{4}\left(\frac{k}{\xi}\right)^2+\frac{1}{8}\left(\frac{k}{\xi}\right)^4\right]e^{-k^2/4\xi^2}\times\cos[\mathbf{k}\cdot(\mathbf{R}^{\mu}-\mathbf{R}^{\nu})]\mathbf{kk}.$$

In the expressions above, l is the cell index number, ν is the particle index number, $\mathbf{R}^{\mu 0}$ is the position of the particle μ in the zeroth cell, $\mathbf{R}^{\nu l}$ is the position of the particle ν in the lth cell, $\mathbf{k}$ is the periodic cell reciprocal or wave vector, k is its magnitude, $r=|\mathbf{R}^{\mu 0}-\mathbf{R}^{\nu l}|$, and $\mathbf{R}^{\mu}-\mathbf{R}^{\nu}$ is the position vector difference between bubbles μ and ν within a cell. The arbitrary number ξ regulates the speed of convergence for the sums and is typically chosen to be $1/V_c^{1/3}$. Each sum converges exponentially fast.

It is now straightforward to rewrite matrices $\mathbf{M}_1$ and $\mathbf{M}_2$ of (A12) and (A13) in terms of the Ewald sums. All of the analysis of the previous section will apply to the case of an infinite number of bubbles, except the total kinetic energy as given by (A14) should now be thought of as that of an elementary cell rather than all fluid [the same obviously applies to the total momentum as given by (6)].

[1] G. K. Batchelor, "Developments in microhydrodynamics," *Theoretical and Applied Mechanics*, edited by W. Koiter (North-Holland, Amsterdam, 1976), p. 33.

[2] J. M. Delhaye and J. L. Achard, "On the averaging operators introduced in two-phase flow modeling," *Proceedings of the 1st OECD/NEA Specialist Meeting on Transient Two-Phase Flow*, Toronto, 1976, Vol. 1, p. 5.

[3] E. J. Hinch, "An averaged-equation approach to particle interactions in a fluid suspension," J. Fluid Mech. **83**, 695 (1977).

[4] A. V. Voinov and A. G. Petrov, "On the stress tensor in a fluid containing dispersed particles," J. Appl. Math. Mech. **41**, 368 (1977).

[5] S. Banerjee and A. M. C. Chan, "Separated flow model I. Analysis of the averaged and local instantaneous formulations," J. Multiphase Flow **6**, 1 (1980).

[6] D. A. Drew, "Mathematical modeling of two-phase flow," Annu. Rev. Fluid Mech. **15**, 261 (1983).

[7] A. Biesheuvel and L. van Wijngaarden, "Two-phase flow equations for a dilute dispersion of gas bubbles in liquid," J. Fluid Mech. **148**, 301 (1984).

[8] C. Pauchon and S. Banerjee, "Interphase momentum interaction effects in the averaged multifield model. Part I: Void propagation in bubbly flows," Int. J. Multiphase Flow **12**, 559 (1986).

[9] J. A. Geurst, "Virtual mass in two-phase bubbly flow," Physica A **129**, 233 (1985).

[10] J. A. Geurst, "Variational principles and two-fluid hydrodynamics of bubbly liquid/gas mixtures," Physica A **135**, 455 (1986).

[11] C. Pauchon and P. Smereka, "Momentum interactions in dispersed flow: An averaging and a variational approach," Int. J. Multiphase Flow **18**, 65 (1992).

[12] P. Smereka, "On the motion of bubbles in a periodic box," J. Fluid Mech. **254**, 79 (1993).

[13] A. S. Sangani and A. K. Didwania, "Dispersed phase stress tensor in flows of bubbly liquids at large Reynolds numbers," J. Fluid Mech. **248**, 27 (1993).

[14] A. S. Sangani and A. K. Didwania, "Dynamic simulations of flows of bubbly liquids at large Reynolds numbers," J. Fluid Mech. **250**, 307 (1993).

[15] A. Biesheuvel and W. C. M. Gorissen, "Void fraction disturbances in a uniform bubbly liquid," Int. J. Multiphase Flow **16**, 211 (1990).

[16] L. van Wijngaarden and C. Kapteyn, "Concentration waves in dilute bubble/liquid mixtures," J. Fluid Mech. **212**, 111 (1990).

[17] D. W. Moore, "The boundary layer on a spherical gas bubble," J. Fluid Mech. **16**, 161 (1963).

[18] D. W. Moore, "The velocity of rise of distorted gas bubbles in a liquid of small viscosity," J. Fluid Mech. **23**, 749 (1965).

[19] J. B. W. Kok, "Dynamics of gas bubbles moving through liquid. Part I. Theory," Eur. J. Mech. B Fluids **12**, No. 4, 515 (1993).

[20] J. B. W. Kok, "Dynamics of gas bubbles moving through liquid. Part II. Experiment," Eur. J. Mech. B Fluids **12**, No. 4, 541 (1993).

[21] G. K. Batchelor, *An Introduction to Fluid Dynamics* (Cambridge University Press, Cambridge, 1967).

[22] H. Lamb, *Hydrodynamics*, 6th ed. (Cambridge University Press, Cambridge, 1932) (Dover edition 1945).

[23] E. J. Hinch and L. C. Nitsche, "Nonlinear drift interactions between fluctuating colloidal particles: Oscillatory and stochastic motions," J. Fluid Mech. **256**, 343 (1993).

[24] L. D. Landau and E. M. Lifshitz, *Statistical Physics* (Pergamon Press, Oxford, 1969).

[25] T. L. Hill, *Statistical Mechanics* (McGraw–Hill, New York, 1956, Dover edition 1987).

[26] M. P. Allen and D. J. Tildesley, *Computer Simulation of Liquids* (Clarendon Press, Oxford, 1987).

[27] R. T. Bonnecaze and J. F. Brady, "A method for determining the effective conductivity of dispersions of particles," Proc. R. Soc. London Ser. A **430**, 285 (1990).

[28] R. T. Bonnecaze and J. F. Brady, "The effective conductivity of random suspensions of spherical particles," Proc. R. Soc. London Ser. A **432**, 445 (1991).

[29] O. D. Kellogg, *Foundations of Potential Theory* (Dover, New York, 1953).

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