# Properties and averaged equations for flows of bubbly liquids

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**Abstract**

Averaged properties of bubbly liquids in the limit of large Reynolds and small Weber numbers are determined as functions of the volume fraction, mean relative velocity, and velocity variance of the bubbles using numerical simulations and a pair interaction theory. The results of simulations are combined with those obtained recently for sheared bubbly liquids [Kang *et al.*, Phys. Fluids **9**, 1540, 1997] and the mixture momentum and continuity equations to propose a complete set of averaged equations and closure relations for the flows of bubbly liquids at large Reynolds and small Weber numbers.

*Keywords*: bubbly flows, averaged equations, potential flow interactions

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# 1 Introduction

The flows of liquid containing bubbles at relatively large Reynolds numbers have been examined by a number of investigators in the past because of their occurrence in many physical and chemical operations. The most significant contributions to our understanding of bubbly liquid flows derive from a series of careful experimental and analytical studies by Professor Leen van Wijngaarden and his colleagues at the University of Twente over the last three decades. It is therefore a great pleasure and indeed an honor to present our recent work on bubbly liquids in a volume dedicated to Professor van Wijngaarden on the occasion of his sixty-fifth birthday and formal retirement from the university.

We shall consider here bubbly flows at large Reynolds number $Re \equiv aV/\nu$ and small Weber number $We \equiv \rho a V^2/\sigma$, $a$ being the bubble radius, $V$ the velocity magnitude of a bubble relative to the mixture, $\sigma$ the interfacial tension, and $\nu$ and $\rho$, respectively, the dynamic viscosity and density of the liquid. In these flows bubbles remain approximately spherical, and the liquid motion induced by the bubbles can be described to a leading order by the potential flow theory. The viscous effects are essentially confined to thin boundary layer regions near the bubble surfaces and to small wakes behind the bubbles and, unlike the case of rigid particles, these viscous effects modify only slightly the fluid velocity field derived from the potential flow approximation as shown by Moore [1,2]. The conditions of small $We$ and large $Re$ are approximately satisfied by about 1 mm diameter bubbles rising through otherwise quiescent water. Because of the relative ease with which the bubble-bubble interactions can be computed, this dual limit can be examined in detail analytically.

We begin with a brief review of the work done to date on the flows satisfying the above dual limit. Unless there are strong variations in the pressure (as is the case in acoustic wave propagation), the volume changes of the bubbles can be neglected, and the interactions among bubbles are primarily governed by the added mass effects. A single massless bubble accelerates in an inviscid fluid as if its mass equals $m/2$, where $m$ is the mass of the liquid displaced by the bubble [3]. This is referred to as the virtual or added mass of the bubble. The momentum change, $m/2$ times the change in the velocity of the bubble is, of course, the change in the liquid momentum as a result of the moving bubble. If there are more bubbles the liquid momentum generated by the motion of bubbles becomes complicated to evaluate, because the flow field induced by one bubble will be distorted by others. In other words, the added mass becomes a function of the configuration of the bubbles. For liquids containing a finite volume fraction $\phi$ of equal-sized bubbles the averaged added mass of a bubble can be expressed as $C_a m/2$, where $C_a$ depends on the volume fraction $\phi$ of the bubbles and the spatial and velocity distributions of the bubbles. Van Wijngaarden [4] was the first to determine rigorously $C_a$ for dilute bubbly liquids, i.e., $\phi \ll 1$. The calculation required a then new technique developed by Batchelor [5] for the renormalization of long-range interactions in Stokes flows. Realizing that $C_a$ will also depend on the way the flow is induced, because of its dependence on the velocity distribution of the bubbles, van Wijngaarden restricted his attention to the special case in which the bubbly liquid at rest is impulsively set into motion by applying equal forces on all the bubbles. For this situation he obtained

$$C_a = 1 + 2.76\phi + O(\phi^2). \tag{1}$$

The bubbles were assumed to be uniformly dispersed in the liquid in this calculation. The fact that the $O(\phi)$ coefficient in Eq. (1) depends on the velocity distribution of the bubbles was further illustrated by Biesheuvel and Spoelstra [6], who considered the case when all the bubbles are accelerated at an equal rate. Their analysis gave the coefficient of the $O(\phi)$ term in the above to be 3.32. In most flows the bubbles are neither uniformly forced nor uniformly accelerated, but it is generally believed that these two special cases provide the two limits on $C_a$ for uniformly dispersed mixtures. This, to some extent, is supported by the investigations of the added mass effects in the suspensions of particles subjected to an oscillatory motion by Felderhof [7] and Sangani, Zhang and Prosperetti [8]. The latter investigators found that the $O(\phi)$ coefficient in this case increased monotonically from 2.76 to 3.32 as the ratio of the particle to the suspending fluid density varied from zero to infinity. They also used numerical simulations to compute $C_a$ for the complete range of values of $\phi$ for the hard-sphere spatial distribution of the oscillating particles, and found the cases of uniformly accelerated and uniformly forced particles to provide two extreme limits of $C_a$. In fact, the difference in $C_a$ for these two extremes is not very significant even when $\phi$ is not small and, as shown in [8], one may use the simple expression

$$C_a = \frac{1 + 2\phi}{1 - \phi}, \tag{2}$$

obtained by Zuber [9] using a cell approximation to estimate $C_a$ in uniform dispersions.

In convective (non-oscillatory) flows the spatial distribution of the bubbles is not necessarily uniform, and therefore one must solve in general a more difficult problem of determining the spatial and velocity distribution of the bubbles for a given imposed flow. For dilute bubbly liquids this requires determination of the spatial and velocity distribution of a pair of interacting bubbles, e.g., by solving for the trajectories of the bubbles initially separated at infinity with a uniform probability. Such calculations have been made for the case of bubbles rising under the influence of gravity by Kok [10,11] and van Wijngaarden [12] for the monodispersed, and by Kumaran and Koch [13] for the polydispersed bubbly liquids. These calculations showed that the equal-sized bubbles tend to undergo repeated collisions with each other with diminishing amplitude, and eventually rise together as a horizontally aligned pair.

The velocity and spatial distribution of bubbles in non-dilute bubbly liquids is most conveniently determined by dynamic simulations. The case of buoyant rise of bubbles by dynamic simulations was examined concurrently by Smereka [14] and Sangani and Didwania [15], in the same year (1993) that van Wijngaarden's work on the pair of bubbles appeared. All these studies, and a more recent study by Yurkovetsky and Brady [16], show that the bubbles align in planes perpendicular to gravity.

The tendency for the formation of these clusters can be explained in terms of potential interactions among bubbles. Pairs of bubbles aligned within about $55^o$ ($\cos^{-1}\sqrt{1/3}$) to the direction of mean bubble motion are repelled by each other due to a Bernoulli effect, while those aligned in a plane perpendicular to that or at an angle greater than $55^o$ with the direction of mean bubble motion are attracted towards each other [17]. This leads to an increased probability of finding bubble pairs that are horizontally aligned. This tendency is diminished when the velocity fluctuations in the bubbly liquids are increased. The steady-state distribution is therefore determined by the magnitude of the rise velocity of the bubbles divided by the root-mean-squared velocity of the bubbles. As shown in [14-16,19] no clusters are seen at sufficiently small value of this ratio, and large clusters in horizontal planes are seen when the ratio is large. Yurkovetsky and Brady [16] referred to this phenomenon as a phase-transition, from a gas-like dispersed state to a condensed state. The added mass coefficient for clustered bubbly liquids is typically much greater than that predicted by the simple expression (2) given by Zuber.

At large but finite Reynolds numbers the magnitude of the velocity fluctuations depends on the rate at which the fluctuations are dissipated by the viscous effects and the rate at which they are created by hydrodynamic interactions. The latter is very small in magnitude for the special case of buoyant rise of equal-size bubbles and, as a consequence, the bubbles tend to form clusters. As suggested by Sangani and Didwania [18], the other flow situations could lead to a greater source of velocity fluctuations, and hence less clustered bubbly liquids. This suggestion is supported by a recent investigation of sheared bubbly liquids by Kang *et al.* [19]. These investigators showed that large velocity fluctuations, and hence a stable dispersion, results when the mean velocity gradient is nonzero. The buoyancy force on the bubbles, and hence the mean rise velocity, was taken to be zero in that investigation.

The above studies were confined to rather special flows of bubbly liquids. More complex flows of bubbly liquids are most conveniently analyzed through averaged equations of bubbly liquids. Derivation of these equations is a subject of numerous investigations (see, for example, [18, 20-24,43]). While the form of the averaged equations is now relatively well established, an outstanding problem that remains to be solved is to prescribe the constitutive relations and closures to be used in these equations. The objective of the present investigation is to provide these relations for the special case of large Reynolds and small Weber number flows.

It is reasonable to suppose that in the large Reynolds number limit the different imposed flows (e.g., the mean shear or the buoyancy induced flows) and the finite viscosity of the liquid are primarily responsible for determining the magnitude of the velocity fluctuations and the mean relative velocity of the bubbles, while the microstructure and the constitutive relations and closures to be used are essentially governed by the inertial interactions among bubbles. Since the inertial interactions preserve the momentum and the kinetic energy of the liquid, we expect the most important nondimensional parameters characterizing the inertial interactions to be the ratio $A$

of the mean relative motion and the root-mean-squared velocity fluctuations of the bubbles and the volume fraction $\phi$ of the bubbles. Note that there is one-to-one correspondence between the steady state momentum and kinetic energy of the liquid to the averaged relative velocity and the velocity variance. The latter set of quantities are not invariants of the potential flow interactions and therefore vary with time but are of more practical significance. The usual averaged equations of motion give added mass, drag coefficient, and other properties in terms of volume fraction only. The present investigation examines the change in microstructure with $A$ and its effect on various properties. Thus we shall use numerical simulations to determine various properties of bubbly liquids as functions of $A$ and $\phi$ and to give averaged equations for describing the flows of bubbly liquids. The limiting case of $A = 0$ was examined in detail by Kang *et al.* [19]. Our results agree with theirs in this limit and extend them to nonzero $A$.

The outline of the article is as follows. In Section 2 we briefly describe the simulation technique and in Section 3 we give the averaged equations for bubbly liquids. Both of these sections are primarily based on previous work by Sangani and Didwania [15,18] and Kang *et al.* [19]. In Section 4 we present the results of dynamic simulations for the pair probability distribution, added mass coefficient, viscous drag coefficient, bubble-phase stress, and the other averaged properties of bubbly liquids as functions of $A$ and $\phi$. The results of simulations are compared with the predictions of a theory for small $\phi$, and analytical expressions that agree well with the dilute theory and the results of numerical simulations for $\phi \leq 0.3$ are given. Also given is an expression for the critical value of $A$ as a function of $\phi$, above which significant clustering in the plane normal to the mean relative velocity can be expected. We combine in Section 5 the results obtained from Kang *et al.* [19] with those obtained in the present investigation and prescribe constitutive relations to be used in modeling the flows of bubbly liquids. The aim of that section is to present a simpler form of averaged equations for the bubble phase and for the mixture that retains the most important features of the dynamics of bubbly liquids. Finally, in Section 6, we discuss how the equations derived here may be modified to account for finite Weber-number flows.

## 2 Simulation technique

In this section we briefly review the dynamic simulation method used in the present study; a more detailed description may be found in Sangani and Didwania [15] and Kang *et al.* [19]. The dynamics of large Reynolds number flows of a liquid containing spherical bubbles are described in terms of a velocity potential $\varphi$ that satisfies the Laplace equation. The velocity of the liquid is decomposed as

$$\mathbf{u}(\mathbf{x}, t) = \overline{\mathbf{u}}(\mathbf{x}, t) + \nabla\varphi, \tag{3}$$

where $\overline{\mathbf{u}}$ is the ensemble-averaged velocity of the gas-liquid mixture. The boundary condition at the surface of a representative bubble $\alpha$ is

$$\mathbf{n} \cdot \nabla\varphi = \mathbf{n} \cdot \mathbf{v}^{\alpha}, \tag{4}$$

where $\mathbf{v}^{\alpha} = \mathbf{w}^{\alpha} - \overline{\mathbf{u}}(\mathbf{x}^{\alpha}, t)$ is the velocity of the bubble relative to the mixture and $\mathbf{n}$ is the unit outward normal from the surface of the bubble, $\mathbf{w}^{\alpha}$ and $\mathbf{x}^{\alpha}$ being the actual velocity and position of the bubble. As shown by Sangani and Didwania [15], the velocity potential is reasonably well described by the point-dipole approximation,

$$\varphi(\mathbf{x}, t) = \mathbf{G} \cdot \mathbf{x} - \sum_{\alpha=1}^{N} a^3 \mathbf{D}^{\alpha} \cdot \nabla S_1(\mathbf{x} - \mathbf{x}^{\alpha}), \tag{5}$$

where $S_1$ is a Green's function for the Laplace equation in a periodic domain [25], $\mathbf{x}^{\alpha}$ the center of bubble $\alpha$, $a$ the radius of the bubble, and $\mathbf{D}^{\alpha}$ the dipole induced by the presence of the bubble. $\mathbf{G}$ represents the induced backflow which maintains $\overline{\mathbf{u}'} = \mathbf{0}$, $\mathbf{u}' = \mathbf{u} - \overline{\mathbf{u}}$ being the deviation from the mean velocity of the gas-liquid mixture. As shown in [15],

$$\mathbf{G} = \frac{3\phi}{N} \sum_{\alpha=1}^{N} \mathbf{D}^{\alpha}. \tag{6}$$

The force balance on the bubbles can be expressed in terms of their impulses defined by

$$\mathbf{I}^{\alpha} = -\rho \int_{S^{\alpha}} \varphi \mathbf{n} dA, \tag{7}$$

where $\rho$ is the density of the liquid and $S^{\alpha}$ the surface of bubble $\alpha$. Note that this definition of impulse differs from that defined in [18,26], where the total impulse is used which equals $\mathbf{I}^{\alpha} - m\overline{\mathbf{u}}$. The sum of impulses over all the bubbles in a unit cell gives the total momentum of the liquid induced by the motion of the bubbles when the back flow is zero. Thus, even though the bubbles themselves are massless, their motion generates additional momentum in the liquid and, in that sense, the impulse of a bubble can be thought of as its virtual momentum. Following Sangani and Didwania [15] and Kang *et al.* [19], the force balance on a bubble $\alpha$, neglecting the mass of the gas inside the bubbles, is given by

$$\frac{d\mathbf{I}^{\alpha}}{dt} = \mathbf{F}_g + \mathbf{F}^{\alpha}_{\overline{\mathbf{u}}} + \mathbf{F}^{\alpha}_p + \mathbf{F}^{\alpha}_v + \mathbf{F}^{\alpha}_c. \tag{8}$$

The forces on the right-hand side arise from the buoyancy, averaged suspension flow, the potential interactions among the bubbles, viscosity, and collisions, respectively. The rest of this section briefly discusses these contributions to the force on a bubble.

The buoyancy force is given by $\mathbf{F}_g = -m\mathbf{g}$, where $m = (4\pi/3)\rho a^3$ is the mass of the liquid displaced by the bubble and $\mathbf{g}$ is the gravitational acceleration.

The force on the bubble due to averaged suspension flow is written as [19]

$$\mathbf{F}^{\alpha}_{\overline{\mathbf{u}}} = m\frac{D\overline{\mathbf{u}}}{Dt} - (\nabla\overline{\mathbf{u}})\cdot\mathbf{I}^{\alpha}, \tag{9}$$

where the derivatives of $\overline{\mathbf{u}}$ are evaluated at the center of bubble $\alpha$ and $D/Dt = (\partial/\partial t + \overline{\mathbf{u}}\cdot\nabla)$ is the derivative following the averaged suspension flow. This expression is consistent with the force on a single bubble derived by Auton, Hunt, and Prud'homme [27], with the forces on bubbles placed in an extensional flow [19], and with the force on bubbles subjected to a small-amplitude oscillatory motion as evaluated by Sangani, Zhang, and Prosperetti [8]. Note that this also includes the lift force on a spherical bubble placed in a weak shear flow, as does the expression by Auton, Hunt, and Prud'homme. In the last case, the overall flow is rotational even though the flow induced by the motion of bubbles, $\mathbf{u}'$, is taken to be irrotational.

The potential interaction force is [18]

$$\mathbf{F}^{\alpha}_{p} = -4\pi\rho a^6 \sum_{\gamma=1}^{N} \mathbf{D}^{\alpha}\mathbf{D}^{\gamma} : \nabla\nabla\nabla S_1(\mathbf{x}^{\gamma} - \mathbf{x}^{\alpha}). \tag{10}$$

In the above, $S_1$ must be first differentiated with respect to $\mathbf{x} - \mathbf{x}^{\gamma}$, and subsequently $\mathbf{x}$ must be taken to equal $\mathbf{x}^{\alpha}$. For $\alpha = \gamma$, the singular part $1/|\mathbf{x} - \mathbf{x}^{\gamma}|$ of $S_1$ must be removed from $S_1$ before differentiating.

The viscous force can be determined by one of the two methods. The first is based on an energy dissipation argument [28] according to which

$$\mathbf{F}^{\alpha}_{v} = -\frac{1}{2}\nabla_{\mathbf{v}^{\alpha}}\dot{E}_{\mathrm{diss}}, \tag{11}$$

where $\dot{E}_{\mathrm{diss}}$ is the rate of energy dissipation per unit cell, which can be evaluated to leading order from the potential flow approximation given by Eq. (5). The other method, which is more convenient and employed in the present study, uses the observation that the viscous force on a bubble evaluated from the velocity gradient of the total dissipation rate must be the same as the force amplitude on a bubble placed in a small-amplitude oscillatory motion examined in Sangani [29] and Sangani, Zhang, and Prosperetti [8].

Finally, a bubble is assumed to bounce back instantaneously upon collision with another bubble in the suspension preserving the total momentum and kinetic energy of the liquid, as has been assumed in other studies [14-16,18,19]. Such collisions occur when a small amount of electrolyte (salt) is present in bubbly liquids [30,31]. The collision impulse is evaluated using the method described in [15].

# 3 Averaged equations for bubbly flows

The bubble-phase continuity and momentum equations are [18,26]

$$\frac{dn}{dt} + n\nabla\cdot\overline{\mathbf{w}} = 0, \tag{12}$$

$$n\frac{d\overline{I_i}}{dt} = -\frac{\partial}{\partial x_j}P_{ij} + n\overline{F_i^b}, \tag{13}$$

where $n$ is the number density of bubbles, $d/dt = (\partial/\partial t + \overline{\mathbf{w}} \cdot \nabla)$ is the derivative following the averaged motion of the bubbles, the quantities with an overbar represent averaged quantities, $P_{ij}$ is the bubble-phase stress tensor, and $\mathbf{F}^b = \mathbf{F}_g + \mathbf{F}_v + \mathbf{F}_{\overline{\mathbf{u}}}$ is the sum of buoyancy, viscous, and mean flow forces. As shown in [18] in detail, the bubble-phase stress tensor represents the transport of momentum due to fluctuations in the velocity of bubbles, collisions, and hydrodynamic interactions. The kinetic part arising from the velocity fluctuations in the bubble motion is

$$P_{ij}^k = n(\overline{I_i v_j} - \overline{I}_i \overline{v}_j). \tag{14}$$

As noted earlier, the impulse of a bubble can be thought of as its virtual momentum and, therefore, this part of the stress is analogous to the pressure in the kinetic theory of dilute (ideal) gases.

The collisional contribution is

$$P_{ij}^c = \frac{n}{2N\triangle t}\sum_{\mathrm{col}}(F_{c,i}^1 - F_{c,i}^2)(x_j^1 - x_j^2), \tag{15}$$

where 1 and 2 represent a pair of colliding bubbles, and $\mathbf{F}_c$'s are the collision impulses acting on the bubbles at the instant of collision, with $\mathbf{F}_c^1 = -\mathbf{F}_c^2$. The summation in the above equation is over all the collisions during a time interval $\triangle t$, and $N$ is the number of bubbles.

The hydrodynamic contribution consists of two parts. The first, arising from the backflow induced by the relative motion of the bubbles, is

$$P_{ij}^M = -\frac{1}{2}\rho G_k G_k \delta_{ij} \tag{16}$$

and will be referred to as the Maxwell stress in analogy with the Maxwell stress in dielectric media. At first sight this expression may appear to differ from that commonly used in the theory of dielectrics or that given in Bulthuis, Prosperetti, and Sangani [26] according to which

$$P_{ij}^M = \rho[G_i G_j - (1/2)\delta_{ij} G_k G_k - 3\phi G_i \overline{D}_j]. \tag{17}$$

However, upon noting that $G_i = 3\phi\overline{D}_i$, we see that the first and third terms on the right-hand side of the above equation cancel each other, and the resulting expression then agrees with that given by Eq. (16). The derivation presented in [26] applies to arbitrary extensional flow, for which $\mathbf{G}$ represents the gradient of averaged velocity potential. In the present study, we treat the force due to mean flow, including pure extensional flows, as a part of $\mathbf{F}_{\overline{\mathbf{u}}}$ and, consequently, $\mathbf{G} = 3\phi\overline{\mathbf{D}}$ is simply the velocity due to backflow generated by the relative motion of the bubbles. Note that Eq. (16) can alternatively be written as

$$P_{ij}^M = -\frac{9}{2}\rho\phi^2 \overline{D}_k \overline{D}_k \delta_{ij}. \tag{18}$$

The contribution from nonzero $\mathbf{G}$ can alternatively be written as a force on the bubble given by

$$F_i^M = -\frac{1}{n}\frac{\partial P_{ij}^M}{\partial x_j} = 3m\overline{D}_k\frac{\partial G_k}{\partial x_i}. \tag{19}$$

This represents the force on a dipole (induced by the bubble) placed in a nonuniform field $\mathbf{G}$. This is analogous to the ponderomotive force in the theory of electrostatics where $\mathbf{G}$ represents the electric field.

The second part of the hydrodynamic stress depends on the detailed spatial and velocity distribution of the bubbles, and can be evaluated in dynamic simulations using [26]

$$P_{ij}^h = -\frac{2\pi n\rho a^6}{N}\sum_{\alpha=1}^{N}\sum_{\gamma=1}^{N} D_m^\alpha D_n^\gamma \Psi_{ijmn}(\mathbf{x}^\alpha - \mathbf{x}^\gamma), \tag{20}$$

where $\Psi_{ijmn}$ is given by

$$\Psi_{ijmn}(\mathbf{x}) = 2\partial_{ijmn}^4 S_2(\mathbf{x}) - \left(\delta_{ij}\partial_{mn}^2 + \delta_{jm}\partial_{in}^2 + \delta_{jn}\partial_{im}^2\right) S_1(\mathbf{x}). \tag{21}$$

$S_2$ and $S_1$ are spatially periodic functions as defined by Hasimoto [32] with $\nabla^2 S_2 = S_1$, and $\partial_i = \partial/\partial x_i$ is a short-hand notation for partial derivatives.

The last term in Eq. (13) can be rewritten as

$$\overline{F}_i^b = -mg_i - 12\pi\mu a C_d \overline{v}_i + m\frac{D\overline{u}_i}{Dt} - \gamma_{ji}\overline{I}_j, \tag{22}$$

where $C_d$ is the viscous drag coefficient and $\gamma_{ji} = \partial\overline{u}_j/\partial x_i$ is the averaged velocity gradient in the gas-liquid mixture. The net impulse $\overline{I}_i$ is related to the added mass coefficient $C_a$ and the mean relative velocity via

$$\overline{I}_i = (m/2)C_a\overline{v}_i. \tag{23}$$

Finally, it can easily be shown that the averaged dipole strength is related to $C_a$ and to the averaged relative velocity by [18,26]:

$$\overline{D}_i = -\frac{1}{3}\left[\frac{1}{2}C_a + 1\right]\overline{v}_i. \tag{24}$$

The stress tensor, $C_a$, and $C_d$ depend on $n$, $\overline{v}_i$ and the velocity variance and, to close the system of equations, we need an equation for determining the velocity variance. The fluctuation energy balance equation has been derived in [18,19,44,45] for some limiting cases. In [18] an equation was derived for $n\overline{I_i v_i}$, the kinetic energy density of the liquid, in the absence of mean shear, while in [19] an equation was derived for the second moment tensor $n\overline{I_i v_j}$ in the absence of mean relative motion. The backflow contribution giving rise to the Maxwell contribution in the stress tensor was not properly included in [18] and therefore it is necessary to rederive the fluctuation energy equation. We shall derive here an equation for the second moments of impulse, $\overline{I_i I_j} - \overline{I}_i\overline{I}_j$, instead of $\overline{I_i v_j}$ or $\overline{v_i v_j}$ used in earlier investigations, as this is easier. The second moments equation is necessary, in general, when large anisotropy in velocity fluctuations is present in the suspensions. Otherwise, a single scalar balance equation obtained by taking the trace of the second moment equation will be adequate in closing the system of averaged equations.

The balance equation for $\overline{I_i I_j} - \overline{I}_i \overline{I}_j$ is written in the same way as for the number density and impulse (cf. Eqs (12)-(13)), and combining the resulting equation with the impulse equation gives

$$\begin{aligned} n\frac{d}{dt}\left[\overline{I_i I_j} - \overline{I}_i\overline{I}_j\right] &= -\frac{\partial}{\partial x_k}\left[n\left(\overline{I_i I_j v_k} - \overline{I_i I_j}\overline{v}_k\right) - \overline{I}_j P^*_{ik} - \overline{I}_i P^*_{jk}\right] \\ &- P^*_{ik}\frac{\partial \overline{I}_j}{\partial x_k} - P^*_{jk}\frac{\partial \overline{I}_i}{\partial x_k} + n\left[\overline{\dot{\psi}_{ij}} - \overline{I}_i(\overline{F}^b_j + \overline{F}^M_j) - \overline{I}_j(\overline{F}^b_i + \overline{F}^M_i)\right] \end{aligned} \tag{25}$$

where $\psi^\alpha_{ij} = I^\alpha_i I^\alpha_j$; the dot in $\dot{\psi}^\alpha$ represents a time derivative following the motion of bubble $\alpha$. For the sake of convenience we have treated the backflow contribution giving rise to the ponderomotive force (cf. Eq.(19)) instead of contributing to the bubble-phase stress. Thus, $P^*_{ij}$ is the total stress minus the Maxwell stress:

$$P^*_{ij} = P_{ij} - P^M_{ij}. \tag{26}$$

For dilute bubbly liquids we can use $I_i \simeq (m/2)v_i$ and then the quantity inside the square brackets in the first term on the right-hand side of Eq. (25) becomes $n(m/2)^2(\overline{\hat{v}_i\hat{v}_j\hat{v}_k})/2$, $\hat{v}_i = v_i - \overline{v}_i$ being the fluctuation velocity. The trace of this quantity is seen to be the same as (except for an additional factor of $m/2$) the heat flux in dilute molecular systems.

We now evaluate separately the contributions to the terms inside the second square brackets on the right-hand side of Eq. (25). The first is a backflow contribution that can be shown to equal

$$\langle\dot{\psi}^M_{ij}\rangle - \overline{I}_i\overline{F}^M_j - \overline{I}_j\overline{F}^M_i = 3m\ \mathrm{sym}\left[(\overline{I_j D_k} - \overline{I}_j\overline{D}_k)\frac{\partial G_i}{\partial x_k}\right], \tag{27}$$

where sym is an operator defined via

$$\mathrm{sym}\ [a_{ij}] = a_{ij} + a_{ji}. \tag{28}$$

The second contribution is from the collisional interaction of bubbles. Using the standard techniques employed in the kinetic theory of dense gases and granular materials, it can be shown that the collisional term is a sum of three terms:

$$n\langle\dot{\psi}^c_{ij}\rangle = S^c_{ij} - \frac{\partial q^c_{ijk}}{\partial x_k} - \gamma_{lk}\tau^c_{ijkl} \tag{29}$$

with

$$S^c_{ij} = \frac{n}{N\triangle t}\sum_{\mathrm{col}}[\triangle\psi^{c,1}_{ij} + \triangle\psi^{c,2}_{ij}], \tag{30}$$

$$q^c_{ijk} = \frac{n}{2N\triangle t}\sum_{\mathrm{col}}[(\triangle\psi^{c,1}_{ij} - \triangle\psi^{c,2}_{ij})(x^1_k - x^2_k)], \tag{31}$$

where, as in Eq. (15), 1 and 2 represent two colliding bubbles, the summation is over all the collisions over a time interval of $\triangle t$, and $\triangle\psi^c_{ij}$ represents the change in $\psi_{ij}$, i.e. in $I_iI_j$, during the collision. Finally, the formula for $\tau^c_{ijkl}$ is obtained by replacing $\psi_{ij}$ in Eq. (31) by $\partial\psi_{ij}/\partial v_l$. $\gamma_{kl} = \partial\overline{u_k}/\partial x_l$ is the gradient of the mean velocity. Physically, the first term on the right-hand side of Eq. (29) represents the source of impulse moments due to collision and the second term represents its flux. The last term in Eq. (29) arises because our phase variables are $\mathbf{I}^\alpha$, or equivalently, $\mathbf{v}^\alpha$, the latter being related to the actual velocity of the bubble by $\mathbf{w}^\alpha - \overline{\mathbf{u}}$. We shall later argue that $\tau^c_{ijkl} \simeq (m/2)(\delta_{il}P^c_{jk} + \delta_{jl}P^c_{ik})$. Thus, the last term in Eq. (29) can be interpreted as proportional to the collision stress times the average velocity gradient of the mixture.

The third is a hydrodynamic contribution that can be derived in a manner similar to that used in [26]:

$$n\langle\dot{\psi}^h_{ij}\rangle = S^h_{ij} - \frac{\partial q^h_{ijk}}{\partial x_k} - \gamma_{lk}\tau^h_{ijkl} \tag{32}$$

with

$$S^h_{ij} = -\frac{3mn}{2N}a^3 \operatorname{sym}\left[\sum_{\alpha=1}^{N}\sum_{\gamma=1}^{N}\left(D^\alpha_m D^\gamma_n I^\alpha_j - D^\gamma_m D^\alpha_n I^\gamma_j\right)\partial^3_{mni}S_1(\mathbf{x}^\alpha - \mathbf{x}^\gamma)\right], \tag{33}$$

$$q^h_{ijk} = -\frac{3mn}{4N}a^3 \operatorname{sym}\left[\sum_{\alpha=1}^{N}\sum_{\gamma=1}^{N} D^\alpha_m D^\gamma_n (I^\alpha_j + I^\gamma_j)\Psi_{mnik}(\mathbf{x}^\alpha - \mathbf{x}^\gamma)\right], \tag{34}$$

where $\boldsymbol{\Psi}$ is defined by Eq. (21). As in the case of collisional contribution, the term $\tau^h_{ijkl}$ can be obtained by replacing $D^\alpha_m D^\gamma_n (I^\alpha_j + I^\gamma_j)$ in Eq. (34) by its derivative with $v^\alpha_l$. Once again, it will be argued that it is proportional to $\delta_{il}P^h_{jk} + \delta_{jl}P^h_{ik}$ so that the last term in Eq. (34) represents the product of the mean velocity gradient with the hydrodynamic stress.

Now we determine contributions from various forces on the bubbles. Buoyancy does not contribute to the fluctuation equation. The contribution from the force due to mixture velocity variations is given by

$$\langle\dot{\psi}^{\overline{\mathbf{u}}}_{ij}\rangle - \overline{F}_{\overline{\mathbf{u}},i}\overline{I}_j - \overline{F}_{\overline{\mathbf{u}},j}\overline{I}_i = -\gamma_{ki}(\overline{I_k I_j} - \overline{I}_k\overline{I}_j) - \gamma_{kj}(\overline{I_k I_i} - \overline{I}_k\overline{I}_i). \tag{35}$$

The mixture velocity gradient $\gamma_{ij}$ can be decomposed into symmetric and asymmetric parts. The symmetric part multiplied with the second moments of impulse will be shown to give a term in the energy equation that approximately equals the work done by the kinetic stress on the mean velocity gradient. The asymmetric part of $\gamma_{ij}$, which gives rise to the lift force, plays the role of altering the distribution of second moments of impulse; its contribution to the impulse variance equation, i.e. to the trace of Eq. (25), is zero.

Finally, the contribution from the viscous forces is

$$n \operatorname{sym}\left[\overline{F_{v,i}I_j} - \overline{F}_{v,i}\overline{I}_j\right]. \tag{36}$$

Collecting various terms, the impulse fluctuation equation is given by

$$n\frac{d}{dt}[\overline{I_i I_j} - \overline{I}_i\overline{I}_j] \quad = \quad S_{ij} - \frac{\partial q_{ijk}}{\partial x_k} - \gamma_{lk}\tau_{ijkl} + \operatorname{sym}\left[-P^*_{ik}\frac{\partial\overline{I}_j}{\partial x_k} - n\gamma_{kj}(\overline{I_i I_k} - \overline{I}_i\overline{I}_k)\right.$$

$$+ \quad 3mn(\overline{I_j D_k} - \overline{I}_j \overline{D}_k)\frac{\partial G_i}{\partial x_k} + n(\overline{F_{v,i} I_j} - \overline{F}_{v,i}\overline{I}_j)\Big], \tag{37}$$

where $S_{ij} = S^h_{ij} + S^c_{ij}$, $\tau_{ijkl} = \tau^h_{ijkl} + \tau^c_{ijkl}$, and

$$q_{ijk} = n\overline{I_i I_j v_k} - n\overline{I_i I_j}\overline{v}_k - \overline{I}_j P^*_{ik} - \overline{I}_i P^*_{jk} + q^c_{ijk} + q^h_{ijk}. \tag{38}$$

As in the case of the momentum equation, various terms appearing in Eq. (37) can be expressed in terms of $n$, $\overline{v}_i$, $\gamma_{ij}$, the velocity moments $T_{ij} \equiv \overline{v_i v_j} - \overline{v}_i \overline{v}_j$, and suitable nondimensional coefficients. The dependence on $\gamma_{ij}$ was examined in detail in Kang *et al.* [19], where the case of zero mean relative motion $\overline{v}_i$ was considered. We shall examine in the next section the dependence on the mean relative velocity.

The impulse moments equation as presented here is indeed quite complex and it would be desirable to make some approximations to make it less unwieldy. The results of numerical simulations in the next section together with the simulations in Kang *et al.* [19] will allow us to achieve this as discussed in Section 5 where we propose a considerably simpler eqaution for determining the moments and hence the bubble-phase temperature. Finally, the above equations for the bubble-phase must be supplemented with the mixture equations, and this will also be considered in Section 5.

# 4 Results

In this section we present results for various averaged quantities appearing in the momentum and fluctuation equations as functions of the mean relative velocity and the velocity variance of the bubbles. The mean velocity gradient $\gamma_{ij}$, viscosity, and buoyancy forces are set to zero. The latter two are not expected to affect the averaged properties in the limit of large Reynolds number, while the effect of mean velocity gradient, as mentioned earlier, was examined in [19].

The dynamic simulations were carried out with 54 bubbles initially placed randomly within a unit cell of a periodic array. Each bubble was given a random impulse chosen from a Gaussian distribution with a prescribed variance (and zero mean), plus a mean impulse along the $x_1$-axis. This determined the total momentum and the kinetic energy of the liquid at time $t = 0$. These quantities are invariants for potential flow interactions. The microstructure, the averaged relative velocity, and velocity variance evolve with time. After about 1000 collisions a steady state was reached. Resulting quantities were subsequently averaged over a time during which at least 3000 collisions occurred. Due to numerical errors in trajectory evaluation and in computations of hydrodynamic interactions, the total kinetic energy varied somewhat - typically by less than 10% over the time interval used in averaging.

## 4.1 Pair-probability distribution

Figure 1 shows simulation results for the pair-probability density for finding two touching bubbles $P(2a, \mu)$. Here, $\mu = \cos\theta$, $\theta$ being the angle made by the line joining the centers of the bubbles with the direction of mean relative velocity. This probability is defined such that $P(aR, \mu) \to n^2$ for widely separated bubbles, $R \to \infty$. In Figure 1, $P$ has been normalized with $n^2\chi$, $\chi$ being the radial distribution function value at contact for the hard-sphere molecular system as given by the Carnahan-Starling [33] approximation:

$$\chi = \frac{1 - \phi/2}{(1 - \phi)^3}. \tag{39}$$

The results shown in Figure 1 correspond to $\phi = 0.05$; the numbers in the figure refer to the value of nondimensional mean relative velocity squared,

$$A = \frac{\overline{v}^2}{T}, \tag{40}$$

with $T$, the bubble-phase temperature, defined by

$$T = \left(\overline{\mathbf{v} \cdot \mathbf{v}} - \overline{v}^2\right)/3. \tag{41}$$

Figure 1 clearly shows that the probability density for bubbles aligned perpendicularly to the mean relative motion, i.e., for $\mu = 0$, increases with increasing $A$, while that for the bubbles aligned along $\mu = 1$ decreases with $A$. This is in agreement with the potential flow theory for two interacting bubbles (see, e.g. van Wijngaarden [12]), which predicts that two bubbles repel each other when their centers are aligned with $\mu > 1/\sqrt{3} = 0.577\ldots$, and attract otherwise.

Dynamic simulation results for volume fractions $\phi = 0.1,\ 0.2$ and $0.3$ at fixed $A$ $(= 1.4 \pm 0.05)$ are presented in Figure 2. We find that the pair probability density normalized by $n^2\chi$ is essentially independent of the volume fraction of the bubbles.

Analytical expressions for $P(aR, \mu)$, and for the other properties of bubbly liquids, can be obtained for dilute bubbly liquids ($\phi \ll 1$) using a theory similar to one employed by Kang *et al.* [19]. The main crux of the theory is that the $N$-bubble probability density for potential flow interactions is known exactly from the investigation of Yurkovetsky and Brady [16], who showed that there exists a Hamiltonian for describing the dynamics of bubbly liquids and that, as a consequence,

$$P_N(\mathcal{C}^N) = Q_N^{-1} \exp\left[-\frac{m\beta}{2}\sum_{\alpha=1}^{N}\left\{\frac{1}{2}\mathbf{I}^\alpha \cdot \mathbf{v}^\alpha - \overline{\mathbf{v}} \cdot \mathbf{I}^\alpha\right\}\right], \tag{42}$$

where $\mathcal{C}^N = \mathbf{x}^1, \mathbf{x}^2, \ldots, \mathbf{x}^N, \mathbf{I}^1, \mathbf{I}^2, \ldots, \mathbf{I}^N$ is the $6N$-dimensional configuration space consisting of the positions and impulses of the bubbles; $\beta$ in the above expression is the inverse of a bubble-phase temperature defined by Yurkovetsky and Brady [16]. As the bubble-phase temperature $T$ defined by Eq. (41) differs from the definition in [16], we shall find below [cf. the discussion after Eq. (44)] a different relation between $\beta$ and $T$. Note also that $\beta$ in [16] equals $m\beta/2$ in the present investigation. $Q_N$ in Eq. (42) is the $N$-bubble partition function. Both $Q_N$ and $\beta$ can be determined, in principle, from the requirements that the probability density for finding a single bubble at a given point in the suspension with any impulse be given by $n$ and that the ratio of the square of the averaged relative velocity to the bubble-phase temperature be given by $A$.

For dilute bubbly liquids, it is more convenient to use one- or two-bubble probability density functions instead of the above, exact, $N$-bubble function. At low volume fractions, these are obtained by simply substituting $N = 1$ and 2 in Eq. (42). Integrating $P_2$ over the impulse subspace and by requiring that the pair probability for two bubbles separated by a large distance must equal $n^2$

$$Q_2 = Q_1^2 = n^{-2}\left(\frac{2\pi}{\beta}\right)^3 e^{A^*}, \tag{43}$$

with

$$A^* = \frac{1}{4}m^2\beta\overline{\mathbf{v}}\cdot\overline{\mathbf{v}}. \tag{44}$$

It will be shown presently that $A^*$ is related to $A$ so that specifying $A$ is equivalent to specifying $A^*$ or $\beta$. It can also be shown that, to leading order in $\phi$, $\beta$ is related to the inverse of impulse variance. To show this, let us first calculate $\overline{I^2}$ using

$$n\overline{I^2} = Q_1^{-1}\int I^2 \exp\left[-\beta I^2/2 + m\overline{\mathbf{v}}\cdot\mathbf{I}\right] d\mathbf{I}. \tag{45}$$

The integration can be carried out by switching to a cylindrical coordinate system. Substituting for $Q_1$ from Eq. (43), and carrying out integration, we obtain

$$\overline{I^2} = \frac{1}{\beta}(3 + A^*). \tag{46}$$

Eqs. (43) and (46) agree with the expressions given in [19] for the special case $A^* = 0$. Upon substituting for $A^*/\beta$ from Eq. (44) into Eq. (46) we obtain

$$\frac{3}{\beta} = \overline{I^2} - \frac{m^2}{4}\overline{\mathbf{v}}\cdot\overline{\mathbf{v}}. \tag{47}$$

We can approximate $A^*$ by writing $\mathbf{I} = m\mathbf{v}/2$ in Eq. (47) and subsequently substituting the result in Eq. (44). This yields, simply,

$$A^* = A[1 + O(\phi)]. \tag{48}$$

Note also that to this, $O(1)$ approximation, $3/\beta = \overline{I^2} - \overline{I}^2$. The $O(\phi)$ correction to the above equation is determined later in this section.

We are now in a position to determine the leading approximation for the pair-probability density $P(aR,\mu)$ in dilute suspensions. By integrating $P_2(\mathcal{C}^2)$ over the impulse space we obtain

$$\begin{aligned} P(a\mathbf{R}) &= \int P_2(\mathcal{C}^2)d\mathbf{I}^\alpha d\mathbf{I}^\gamma \\ &= n^2\left(\frac{\beta}{2\pi}\right)^3 e^{-A^*}\int \exp\left[-m\beta\left(\mathbf{I}^\alpha\cdot\mathbf{v}^\alpha + \mathbf{I}^\gamma\cdot\mathbf{v}^\gamma\right)/4 + m\beta\overline{\mathbf{v}}\cdot(\mathbf{I}^\alpha + \mathbf{I}^\gamma)/2\right] d\mathbf{I}^\alpha d\mathbf{I}^\gamma \end{aligned} \tag{49}$$

where we have used Eqs (42) and (43). To simplify the evaluation of the integrals in Eq. (49) we introduce the transformation

$$\tilde{\mathbf{I}} = (\mathbf{I}^\alpha + \mathbf{I}^\gamma)/2, \qquad \hat{\mathbf{I}} = (\mathbf{I}^\alpha - \mathbf{I}^\gamma)/2, \tag{50}$$

and use $d\mathbf{I}^\alpha d\mathbf{I}^\gamma = 8d\tilde{\mathbf{I}}d\hat{\mathbf{I}}$. The velocities of a pair of bubbles is related to their impulses by [19]

$$v_i^{\alpha,\gamma} = \frac{2}{m}\left[\left(\left(1+\frac{3}{2R^6}\right)\delta_{ij} + \frac{9}{2R^6}k_ik_j\right)I_j^{\alpha,\gamma} + \left(-\frac{3}{2R^3}\delta_{ij} + \frac{9}{2R^3}k_ik_j\right)I_j^{\gamma,\alpha}\right] \tag{51}$$

where $a\mathbf{R} \equiv \mathbf{x}^\gamma - \mathbf{x}^\alpha$ is the separation vector and $\mathbf{k} = \mathbf{R}/R$ is the unit vector along $\mathbf{R}$. The exponential in Eq. (49) can be rewritten using

$$\mathbf{I}^\alpha \cdot \mathbf{v}^\alpha + \mathbf{I}^\gamma \cdot \mathbf{v}^\gamma = \frac{4}{m}\left[\tilde{I}^2 + \hat{I}^2 + \tilde{E}_{ij}\tilde{I}_i\tilde{I}_j + \hat{E}_{ij}\hat{I}_i\hat{I}_j\right] \tag{52}$$

with

$$\tilde{E}_{ij} = \frac{3}{2}\left(-\frac{1}{R^3} + \frac{1}{R^6}\right)\delta_{ij} + \frac{9}{2}\left(\frac{1}{R^3} + \frac{1}{R^6}\right)k_ik_j, \tag{53}$$

$$\hat{E}_{ij} = \frac{3}{2}\left(\frac{1}{R^3} + \frac{1}{R^6}\right)\delta_{ij} + \frac{9}{2}\left(-\frac{1}{R^3} + \frac{1}{R^6}\right)k_ik_j. \tag{54}$$

The resulting integral in Eq. (49) is still difficult to evaluate for arbitrary $R$, and therefore we shall evaluate $P(a\mathbf{R})$ only for large $R$ by keeping terms up to $O(R^{-6})$. The exponent in Eq. (49) is thus approximated to

$$\begin{aligned}\exp\left[-\beta(\tilde{I}^2 + \hat{I}^2) + m\beta\overline{\mathbf{v}}\cdot\tilde{\mathbf{I}}\right] \quad & \left(1 - \beta\tilde{E}_{ij}\tilde{I}_i\tilde{I}_j + \frac{9\beta^2}{8R^6}\left(\tilde{I}^4 - 6(\mathbf{k}\cdot\tilde{\mathbf{I}})^2\tilde{I}^2 + 9(\mathbf{k}\cdot\tilde{\mathbf{I}})^4\right)\right) \times \\ & \left(1 - \beta\hat{E}_{ij}\hat{I}_i\hat{I}_j + \frac{9\beta^2}{8R^6}\left(\hat{I}^4 - 6(\mathbf{k}\cdot\hat{\mathbf{I}})^2\hat{I}^2 + 9(\mathbf{k}\cdot\hat{\mathbf{I}})^4\right)\right).\end{aligned} \tag{55}$$

Substituting now Eq. (55) for the integrand in Eq. (49) and carrying out the integration gives

$$\begin{aligned}P(R,\mu) \quad = \quad & n^2\left[1 + \frac{3A^*}{2R^3}\left(1 - 3\mu^2\right) - \frac{9}{4R^6} + \frac{A^*}{R^6}\left(\frac{87}{80} + \frac{171}{40}\mu^2 + \frac{243}{80}\mu^4\right) + \right. \\ & \left.\frac{A^{*2}}{R^6}\left(\frac{9}{8} - \frac{27}{4}\mu^2 + \frac{81}{8}\mu^4\right)\right].\end{aligned} \tag{56}$$

The terms of $O(R^{-6})$ make negligible contribution to the pair-probability density even at $R = 2$ except for large $A^*$ and for $\mu$ close to unity. Upon neglecting these terms, substituting $A^* = A$, and rearranging Eq. (56) into a Padé approximant yields the following useful expression for the pair-probability density at contact, $R = 2$:

$$P(2a,\mu) = n^2\chi\frac{1 + \frac{1}{16}A(1-3\mu^2)}{1 - \frac{2}{16}A(1-3\mu^2)}. \tag{57}$$

The effect of a finite volume fraction of bubbles is accounted for by multiplying the dilute theory result with the hard-sphere value $\chi$ which partly accounts for the volume exclusion effects in finite

$\phi$ systems. As shown in Figures 1 and 2, the above expression is in excellent agreement with the results of numerical simulations.

Figure 3 shows a snapshot of the bubble positions in a unit cell at the end of the simulation with $A^* = 5.4$ and $\phi = 0.05$. We see that, even though the pair-probability distribution for this case is considerably anisotropic, there is no strong evidence of clustering observed in the simulations of Sangani and Didwania [15] or Smereka [14] with finite viscous and buoyancy forces; apparently, bubble velocity fluctuations are still too high to observe large scale clusters. The dynamic simulation results presented here were obtained by starting with an initially random configuration of bubbles. This initial random configuration sets the limit on the maximum $A$ attainable in the simulations. For $\phi = 0.05$, this corresponds to $A_{max}$ of about 6, and this number decreases with the increasing $\phi$: greater values of $A$ can be attained only if the initial configurations are chosen to be clustered states. Since our interest is mostly in determining the averaged properties of the suspensions that are not highly clustered, we have not carried out simulations with initially clustered bubbly liquids.

### 4.2 Added mass coefficient

Dynamic simulation results for the added mass coefficient $C_a$ are presented in Figure 4. $C_a$ is defined by Eq. (23). As explained in the Introduction, there is a strong dependence of $C_a$ on volume fraction, but we see now also an $A$-dependence. This is, of course, not surprising since the microstructure is a function of $A$. The added mass of a pair of bubbles aligned along their velocities is less than that of a pair of bubbles aligned perpendicularly to it (see, e.g. van Wijngaarden [4]), and we therefore expect $C_a$ to increase with $A$.

To derive an expression for $C_a$ correct to $O(\phi)$, we first notice that the averaged impulse to $O(\phi)$ cannot be determined by simply integrating the added mass increase caused by the presence of a second bubble multiplied by the pair-probability over all possible positions of the second bubble, as this integral is nonabsolutely convergent. Thus, we shall need to renormalize our integral accounting for the effect of the second bubble. In what follows, we use the method outlined in Bulthuis *et al.* [26]. The velocity potential in a suspension containing $N$ bubbles within some domain $\Omega$ is given by the point-dipole approximation:

$$\varphi(\mathbf{x}) = \varphi^{\infty}(\mathbf{x}) - a^3 \sum_{\gamma=1}^{N} \mathbf{D}^{\gamma} \cdot \nabla \frac{1}{|\mathbf{x} - \mathbf{x}^{\gamma}|}, \tag{58}$$

where $\varphi^{\infty}$ is related to the averaged backflow $\mathbf{G} =< \nabla\varphi >$ by (see Bulthuis et al. [26])

$$\nabla\varphi^{\infty}(\mathbf{x}) = \mathbf{G} + a^3 \int_{\Omega} d\mathbf{R} n \overline{\mathbf{D}}(\mathbf{x} - \mathbf{R}) \cdot \nabla\nabla R^{-1}. \tag{59}$$

Note that the above integral, and hence $\varphi^{\infty}$, will approach a constant that depends on the shape of $\partial\Omega$ as the size of $\Omega$ becomes large. The contribution to $\varphi(\mathbf{x})$ from the summation term in Eq.

(58) will cancel the shape-dependent term in $\varphi^\infty$ leading to $\varphi(\mathbf{x})$ being independent of the shape of $\partial\Omega$. Now $\varphi$ can be expanded near the center of bubble $\alpha$ as

$$\varphi(\mathbf{x}) = \varphi_0 + [\mathbf{C}^\alpha + (a/s)^3\mathbf{D}^\alpha] \cdot \mathbf{s} + \dots, \tag{60}$$

where $\mathbf{s} = \mathbf{x} - \mathbf{x}^\alpha$, $\varphi_0$ is a constant, and the dots represent the higher-order harmonics. $\mathbf{C}^\alpha$ is given by

$$\mathbf{C}^\alpha = \nabla\varphi^\infty(\mathbf{x}^\alpha) - a^3 \sum_{\gamma\neq\alpha} \mathbf{D}^\gamma \cdot \nabla\nabla|\mathbf{x}^\alpha - \mathbf{x}^\gamma|^{-1}, \tag{61}$$

where the differentiation is to be carried out with respect to $\mathbf{x}^\alpha - \mathbf{x}^\gamma$. The impulse and velocity of bubble $\alpha$ are related to $\mathbf{C}^\alpha$ and $\mathbf{D}^\alpha$ by [cf. Eqs (4) and (7)]

$$\mathbf{I}^\alpha = -m\left(\mathbf{C}^\alpha + \mathbf{D}^\alpha\right), \tag{62}$$

$$\mathbf{v}^\alpha = \mathbf{C}^\alpha - 2\mathbf{D}^\alpha. \tag{63}$$

Combining the above two expressions and averaging over all the bubbles yields

$$\frac{2}{m}\overline{\mathbf{I}} - \overline{\mathbf{v}} = -3\overline{\mathbf{C}}. \tag{64}$$

$\overline{\mathbf{C}}$ can be evaluated by averaging Eq. (61):

$$\begin{aligned}\overline{\mathbf{C}} \;&=\; \mathbf{G} + a^3\int_\Omega d\mathbf{R}n\overline{\mathbf{D}}(\mathbf{x}-\mathbf{R})\cdot\nabla\nabla R^{-1} - \\ &\frac{1}{n}\int a^3\mathbf{D}(\mathbf{x}^\gamma|\mathbf{x})P_2(\mathbf{x},\mathbf{I}^\alpha,\mathbf{x}^\gamma,\mathbf{I}^\gamma)\cdot\nabla\nabla|\mathbf{x}-\mathbf{x}^\gamma|^{-1}d\mathbf{x}^\gamma d\mathbf{I}^\alpha d\mathbf{I}^\gamma,\end{aligned} \tag{65}$$

with $\mathbf{D}(\mathbf{x}^\gamma|\mathbf{x})$ the dipole strength of a bubble at $\mathbf{x}^\gamma$ given that there is a bubble at $\mathbf{x}$. The first term on the right-hand side of Eq. (65) corresponds to the induced backflow, and it equals $3\phi\overline{\mathbf{D}}$ (cf. Eq. (6)). The integration domain of the second term in Eq. (65) is divided into two parts. The first part, for $R < 2$, is given by

$$na^3\overline{\mathbf{D}}(\mathbf{x})\cdot\int_{R<2}\nabla\nabla R^{-1}dV = \frac{1}{3}na^3\overline{\mathbf{D}}(\mathbf{x})\int_{R=2}\frac{\mathbf{R}}{R}\cdot\nabla R^{-1}dA = -\phi\overline{\mathbf{D}}(\mathbf{x}). \tag{66}$$

The $R > 2$ part will be combined with the last term in Eq. (65) as shown below. To evaluate the latter term we first express the dipole of bubble $\gamma$ in terms of impulses of the bubbles using Eqs (62) and (61):

$$\mathbf{D}^{\alpha,\gamma} = -m^{-1}\mathbf{I}^{\alpha,\gamma} - m^{-1}\mathbf{I}^{\gamma,\alpha}\cdot\nabla\nabla R^{-1} + O(R^{-6}). \tag{67}$$

Note that $\nabla\varphi^\infty$ need not be included, as it is of $O(\phi)$. With the $O(R^{-6})$ terms in Eq. (55) neglected, the last term on the right-hand side of Eq. (65) becomes

$$\begin{aligned}&\frac{1}{mn}\int\left(\tilde{\mathbf{I}} + \tilde{\mathbf{I}}\cdot\nabla\nabla R^{-1}\right)\cdot\nabla\nabla R^{-1}\left(1 + \frac{3}{2}\frac{\beta}{R^3}\tilde{I}^2 - \frac{9}{2}\frac{\beta}{R^3}(\mathbf{k}\cdot\tilde{\mathbf{I}})^2\right)\times \\ &\left(1 - \frac{3}{2}\frac{\beta}{R^3}\hat{I}^2 + \frac{9}{2}\frac{\beta}{R^3}(\mathbf{k}\cdot\hat{\mathbf{I}})^2\right)Q_2^{-1}\exp\left[-\beta(\tilde{I}^2 + \hat{I}^2) + m\beta\overline{\mathbf{v}}\cdot\tilde{\mathbf{I}}\right]d\tilde{\mathbf{I}}d\hat{\mathbf{I}}d\mathbf{R}.\end{aligned} \tag{68}$$

The leading term, of $O(R^{-3})$, which renders the above integral nonabsolutely convergent, can be shown to exactly cancel the $R > 2$ part of the integral in the second term on the right-hand side of Eq. (65). Leaving that part out of Eq. (68) and carrying out the integration gives, for the excess contribution from the last term in Eq. (65),

$$-\phi\overline{\mathbf{v}}\left(\frac{1}{16}+\frac{3}{40}A^*\right). \tag{69}$$

Combining Eq. (64) with Eqs (65), (6), (66) and (69), and comparing the result with Eq. (23) gives

$$C_a = 1 + 3\phi\left(1+\frac{1}{16}+\frac{3}{40}A\right) + O(\phi^2), \tag{70}$$

where we have used Eq. (48) to replace $A^*$ by $A$. The correction factor $3\phi$ in the above expression is familiar from the other added mass calculations referred to in the Introduction: it arises from the backflow induced by the bubbles and from the volume exclusion effect for $R < 2$. The remaining part, arising from the detailed pair interactions of bubbles is small at small $A$ and increases linearly with $A$ in accordance with the results of numerical simulations.

In the Introduction it was mentioned that $C_a$ for spatially uniform suspensions is usually approximated by Eq. (2), which is a function of volume fraction only. A simple formula accounting for the effect of change in microstructure with $A$ is obtained by neglecting the small coefficient 1/16 from Eq. (70) and rearranging the resulting expression in a form similar to Eq. (2):

$$C_a = \frac{1+2\phi+\frac{9}{40}\phi A}{1-\phi}. \tag{71}$$

As shown in Figure 4, the above formula gives very good estimates for $C_a$ at all values of $\phi \leq 0.3$.

### 4.3 Drag coefficient

The drag coefficient relates the averaged viscous drag force on a bubble in the mixture to the averaged relative velocity of the bubbles:

$$\overline{\mathbf{F}}_v = -12\pi\mu a C_d \overline{\mathbf{v}}. \tag{72}$$

Results for $C_d$ obtained from dynamic simulations are shown in Figure 5. $C_d$ is seen to increase both with volume fraction and with $A$. If buoyancy is balanced only by viscous drag this results in an averaged rise velocity that decreases by increasing either the volume fraction or $A$.

An expression for $C_d$ in dilute bubbly liquids is derived by solving for the viscous potential (see [15]) $\nabla^2\varphi^v = 0$, together with the boundary condition $\mathbf{n}\cdot\nabla\varphi^v = -12\mathbf{D}^\alpha\cdot\mathbf{n}$ on the surface of bubble $\alpha$. An expression for $\varphi^v$ can be written in the same manner as for $\varphi$ (cf. Eq. (58)) with $\mathbf{D}$ now replaced by the viscous dipole $\mathbf{D}^v$. The viscous force on the bubble can be evaluated from its viscous dipole by

$$\mathbf{F}_v^\alpha = 4\pi\mu a \mathbf{D}^{v,\alpha}. \tag{73}$$

The viscous potential can be expanded near the center of bubble $\alpha$ in a manner analogous to Eq. (60):

$$\varphi^v(\mathbf{x}) = \varphi_0^v + [\mathbf{C}^{v,\alpha} + (a/s)^3\mathbf{D}^{v,\alpha}] \cdot \mathbf{s} + \ldots . \tag{74}$$

Applying the boundary conditions on $\varphi^v$ yields

$$\mathbf{C}^{v,\alpha} - 2\mathbf{D}^{v,\alpha} = -12\mathbf{D}^{\alpha}. \tag{75}$$

Now $\mathbf{C}^{v,\alpha}$ is given by a viscous potential analogue of Eq. (61). Since $\mathbf{C}^{v,\alpha} = O(\phi)$, we see that, to $O(1)$, $\mathbf{D}^{v,\alpha} = 6\mathbf{D}^{\alpha}$. Substituting this leading estimate of the viscous dipoles in the expression for $\mathbf{C}^{v,\alpha}$ analogous to Eq. (60) yields $\overline{\mathbf{C}}^v = 6\overline{\mathbf{C}} + O(\phi^2)$. On taking an average of Eq. (75) over all the bubbles we obtain

$$6\overline{\mathbf{C}} - 2\overline{\mathbf{D}}^v = -12\overline{\mathbf{D}} = 4\left(C_a/2+1\right)\overline{\mathbf{v}}, \tag{76}$$

where we have used Eq. (24) to evaluate the right-hand side. Upon substituting for $\overline{\mathbf{C}}$ from the added mass calculations, and using the relation between the averaged viscous dipole and $C_d$, we find that

$$C_d = \frac{2}{3}C_a + \frac{1}{3} + O(\phi^2) = 1 + 2\phi\left(1 + \frac{1}{16} + \frac{3}{40}A\right) + O(\phi^2). \tag{77}$$

The correction $2\phi$ arising from the backflow and volume exclusion effects is the same as in the other investigations of viscous drag coefficients in dilute bubbly liquids with uniform spatial distribution [8,12,29]. When the mean relative velocity is large and the pair-probability density is anisotropic the viscous drag coefficient increases, since the drag on a pair aligned perpendicular to the mean motion is greater than in any other direction.

As in the case of $C_a$, the above dilute theory result can be recast into a slightly different form to yield an expression that agrees well with the results of numerical simulations even when $\phi$ is not small, as shown in Figure 5:

$$C_d = \frac{1 + \frac{3}{20}\phi A}{(1-\phi)^2}. \tag{78}$$

The above form of $C_d$ with the term $(1-\phi)^2$ in the denominator is chosen to keep the resulting expression at small $A$ the same as that proposed by Sangani, Zhang, and Prosperetti [8], who calculated $C_d$ for small-amplitude oscillatory flows up to $\phi = 0.5$. Note also that the above expression predicts $C_d$ to continuously increase with increasing $A$, as found in the simulations here and in Sangani and Didwania [15].

An attempt at calculating the viscous drag coefficient for bubbly liquids in which the bubbles form pairs in a plane normal to gravity was also made by van Wijngaarden [12]. He assumed that all the bubbles interact with each other only in the horizontal planes and obtained $C_d = 1 + 1.04\pi a^2 n_a$, where $n_a$ is the number density of bubbles in the horizontal planes. He then related $n_a$ to the volume fraction $\phi$ through a somewhat *ad-hoc* procedure that gave as the final result $C_d = 1 + 1.56\phi + O(\phi^2)$. The present result, Eq. (78), indicates that, in contrast to van Wijngaarden's result, the $O(\phi)$

coefficient for the clustered suspensions is much greater than 2. The main cause of the discrepancy arises from his assumed relationship between $n_a$ and $\phi$, i.e., $n_a = 3\phi/(2\pi a^2)$. As one plane of bubbles contributes $n_a \frac{4}{3}\pi a^3$ per unit area of the plane to the total volume of bubbles in the mixture, this relationship should be $n_a \frac{4}{3}\pi a^3/L = \phi$, where $L$ is the averaged distance between the horizontal planes. This changes van Wijngaarden's final result to $C_d = 1 + 0.78(L/a)\phi$. Apparently van Wijngaarden used $L = 2a$. The numerical simulation results of Sangani and Didwania [15] show, however, that $L$ is much larger than $a$. Thus, the results of numerical simulations and the theory presented here will be consistent with the analysis of van Wijngaarden only if one allows $L$ to increase with $A$.

Eq. (78) has been obtained for high Reynolds number and low Weber number bubbly flows. Experimental data reported by Lammers and Biesheuvel [34] are for bubbly flows with bubbles that are too large for this dual limit to hold. Nevertheless, it is of interest to mention here that they observed that when the bubbly liquid undergoes a transition from a quiescent regime to a so-called agitated regime in which the velocity fluctuations are much greater, the rise velocity of bubbles also increases. This observation is consistent with our calculations that $C_d$ decreases with the increasing velocity fluctuations or decreasing $A$.

## 4.4 Bubble-phase stress tensor

We now present results for the bubble-phase stress tensor, which plays an important role in the momentum balance equation for the bubble phase and in determining the stability of the uniform state of bubbly liquids. The dynamic simulation results will be presented by normalizing the stress tensor with the bubble velocity variance tensor,

$$T_{ij} = \overline{v_i v_j} - \overline{v}_i \overline{v}_j. \tag{79}$$

The ratio $T_{11}/T$ of the velocity variance in the direction of mean relative motion to one-third of the total velocity variance is plotted as a function of $\phi$ and $A$ in Figure 6. Surprisingly, the ratio is very close to unity even when $A$ is not small. Thus, even though a considerable anisotropy in the pair-probability density exists at large $A$, we may treat the velocity fluctuations as nearly isotropic and take

$$T_{ij} = T\delta_{ij}. \tag{80}$$

The kinetic stress [cf. Eq. (14)] is now written as

$$P^k_{ij} = n\left(\overline{I_i v_j} - \overline{I}_i \overline{v}_j\right) = n(m/2)TC^k_{ij}(\phi, A), \tag{81}$$

such that the nondimensional coefficients $C^k_{ij}$ approach unity as $\phi \to 0$ for fixed $A$. The dynamic simulation results for the coefficients $C_k \equiv C^k_{ii}/3$ and $C^k_{11}$ are shown in Figures 7 and 8. From the results it is clear that the two coefficients are essentially the same, and therefore we treat $C^k_{ij}$ as an isotropic tensor, i.e., we write

$$P^k_{ij} = n(m/2)C_k(\phi, A)T\delta_{ij}. \tag{82}$$

The $\phi$-dependence as well as the $A$-dependence of $C_k$ are seen to be weak.

Following [19], the collision stress [cf. Eq. (15)] is written as

$$P^c_{ij} = 4n(m/2)\phi\chi T C^c_{ij}(\phi, A), \tag{83}$$

where $\chi$ is the Carnahan-Starling approximation for the radial distribution function at contact for hard-sphere molecular systems [cf. Eq. (39)]. The dynamic simulation results for $C_c \equiv C^c_{ii}/3$ and $C^c_{11}$ are shown in Figures 9 and 10. Clearly, $C^c_{ij}$ is not isotropic and has a strong $A$-dependence.

To explain these results for $C^c_{ij}$ qualitatively, we note that the collision stress component in the direction of the mean bubble motion, $P^c_{11}$, is most significantly affected by the collisions among pairs of bubbles aligned along the mean relative velocity. The number of such collisions decreases with increasing $A$, and, consequently, one expects $P^c_{11}$ to decrease with $A$. The trace of the collision stress likewise is expected to depend on the averaged value of pair-probability over all orientations of the colliding bubbles. This averaged value is the same as the area under the $P(2a, \mu)$ vs. $\mu$ curve. As seen in Figure 1, this area is greater than unity for most values of $A$. In other words, the total number of collisions increases with $A$ and, as a consequence, one expects $C_c$ to increase with $A$.

Hydrodynamic stress components obtained from dynamic simulations are presented in Figure 11 for $\phi = 0.05$ and in Figure 12 for $\phi = 0.3$. All these stress components have been normalized with $mn\phi T$. In each figure the trace of the Maxwell stress [cf. Eq. (16)] is shown together with the average of the remainder of the hydrodynamic stress components, $P^h_{ii}$ and $P^h_{11}$. The latter two were computed using Eq. (20). These components depend on the detailed spatial and velocity distribution of the bubbles. Also shown in these figures are the predictions for these stress components by Bulthuis *et al.* [26], who considered the special case in which the pair-probability density is uniform for $R > 2$ and the dipoles of all the bubbles are the same. For this case only the volume exclusion effect for $R < 2$ contributes to the hydrodynamic stress, and

$$P^h_{ij} = (9/10)\rho\phi\overline{D}_m\overline{D}_n(\delta_{ij}\delta_{mn} + \delta_{im}\delta_{jn} + \delta_{in}\delta_{jm}). \tag{84}$$

According to this expression the hydrodynamic stress components are positive and, when normalized by $\rho T$ as in Figures 11-12, increase with $A$ (since $\overline{v}$, and, hence, $\overline{D}$, increase with increasing $A$ at fixed $T$). The results of numerical simulations show this trend for small $A$, but at large $A$ (at least for $\phi = 0.05$) the hydrodynamic stress is seen to become negative, indicating that the effect of anisotropy in the pair-probability density and detailed bubble-bubble interactions is opposite to the volume exclusion effect.

Also shown in Figure 11-12 is the Maxwell contribution to the trace of stress as given by Eqs (18) and (24). It is seen that the total hydrodynamic stress is dominated by the Maxwell stress $P^M_{ij}$, thus, we may approximate the total hydrodynamic stress by simply the Maxwell stress.

To obtain approximate formulas for the parameters introduced in Eqs (82) and (83), we first derive dilute-theory results for $C_k$ and $C_c$.

To estimate $C_k$ we need to evaluate $\overline{\mathbf{I}\cdot\mathbf{v}}$. Once again, simply substituting Eq. (51), multiplying with the pair-probability density $P_2(\mathcal{C}^2)$, and integrating over phase space leads to nonabsolutely convergent integrals. A proper renormalization is found by writing, from Eqs (62) and (63),

$$m^{-1}\overline{\mathbf{I}\cdot\mathbf{v}} = -\overline{C^2} + \overline{\mathbf{C}\cdot\mathbf{D}} + 2\overline{D^2}. \tag{85}$$

The first two terms on the right-hand side can be evaluated directly as shown below, but the last term would require renormalization. To avoid its evaluation we shall use the following identity for $\overline{I^2}$ obtained from, once again, Eqs (62) and (63):

$$m^{-2}\overline{I^2} = \overline{C^2} + 2\overline{\mathbf{C}\cdot\mathbf{D}} + \overline{D^2}. \tag{86}$$

Now $\overline{I^2}$ can be expressed readily in terms of $\overline{v}$ and $A^*$ by using Eqs (44) and (46):

$$\overline{I^2} = \frac{m^2}{4}\overline{v}^2(1+3/A^*). \tag{87}$$

Thus we can use Eq. (86) to eliminate $\overline{D^2}$ from Eq. (85). We remark in passing that the velocity variance is obtained similarly from

$$\overline{v^2} = \overline{C^2} - 4\overline{\mathbf{C}\cdot\mathbf{D}} + 4\overline{D^2}, \tag{88}$$

which we need below to obtain the value of $C_k$.

The term $\overline{C^2}$ in Eq. (85) is readily calculated by noticing that, by Eqs (61) and(59), it equals

$$n\overline{C^2} = a^6\int P_2(\mathbf{x},\mathbf{I}^\alpha,\mathbf{x}+\mathbf{R},\mathbf{I}^\gamma)D^\alpha_m D^\gamma_n \partial^2_{mi}R^{-1}\partial^2_{ni}R^{-1}d\mathbf{R}d\mathbf{I}^\alpha d\mathbf{I}^\gamma + O(\phi^2). \tag{89}$$

In writing the above expression use has been made of the fact that $\mathbf{G}$ is of $O(\phi)$. Now, since the integrand in the above is already $O(R^{-6})$, we can simply substitute $\mathbf{D}^{\alpha,\gamma} = -m^{-1}\mathbf{I}^{\alpha,\gamma}$ and use a limiting form of $P_2(\mathcal{C}^2)$ for $R\to\infty$. The integration is now trivial, and leads to

$$\overline{C^2} = \frac{1}{4}\phi m^{-2}\overline{I^2} + O(\phi^2), \tag{90}$$

where Eq. (46) is used to express $\beta$ in $\overline{I^2}$.

In a similar manner, combining Eqs (61) and (59) with Eq. (66) and using $\mathbf{G} = 3\phi\overline{\mathbf{D}}$, we obtain

$$\begin{aligned}\overline{\mathbf{C}\cdot\mathbf{D}}(\mathbf{x}) \quad &= \quad 2\phi\overline{D}^2(\mathbf{x}) + \frac{1}{n}\int_{R>2} a^3\Big[D^\alpha_i D^\gamma_j \partial^2_{ij}R^{-1}P_1(\mathbf{x},\mathbf{I}^\alpha)P_1(\mathbf{x}+\mathbf{R},\mathbf{I}^\gamma) - \\ &\qquad D^\alpha_i D^\gamma_j \partial^2_{ij}R^{-1}P_2(\mathbf{x},\mathbf{I}^\alpha,\mathbf{x}+\mathbf{R},\mathbf{I}^\gamma))\Big]\, d\mathbf{R}d\mathbf{I}^\alpha d\mathbf{I}^\gamma.\end{aligned} \tag{91}$$

Integration yields

$$\overline{\mathbf{C}\cdot\mathbf{D}} = \frac{1}{2}\phi\overline{v}^2 + \frac{\phi}{4}\overline{v}^2\left(-\frac{3}{8} + \frac{1}{4}A^* + \frac{3}{20}A^{*2}\right)/A^*, \tag{92}$$

where use has been made of Eq. (44).

Eqs (86), (90) and (92) can be substituted into Eq. (85) to give

$$m^{-1}\overline{\mathbf{I}\cdot\mathbf{v}} = \frac{1}{2}m\overline{v}^2\left(1-\frac{15}{4}\phi-\frac{9}{40}\phi A^*\right)+\frac{m\overline{v}^2}{2A^*}\left(3-\frac{9}{16}\phi\right). \tag{93}$$

The relation between $A^*$ and $A$, in which the results are to be presented, is also readily obtained using Eq. (88):

$$A^* = A\left(1+\frac{3}{16}\phi-\frac{37}{16}\phi A-\frac{3}{20}\phi A^2\right). \tag{94}$$

Upon substituting Eq. (94) in Eq. (93) and comparing the result with Eq. (82), we finally obtain

$$C_k = 1-\frac{3}{8}\phi+O(\phi^2). \tag{95}$$

This expression agrees exactly with the result of Kang *et al.* [19] for $A=0$, where no renormalization was needed. More importantly, this shows that $C_k$ is independent of $A$ in agreement with the results of numerical simulations (cf. Figure 7). In Figure 7, the solid lines represent the expression given by Kang *et al.* [19], obtained by fitting the results of numerical simulations with $A=0$:

$$C_k = 1-0.35\phi-0.42\phi^2. \tag{96}$$

We see that this expression applies to non-dilute bubbly liquids even when $A$ is nonzero.

The collision stress given by Eq. (15) can alternatively be expressed as [19]

$$P^c_{ij}(\mathbf{x}) = -4a^3\int_{\mathbf{g}'\cdot\mathbf{k}>0} F_{c,i}k_j\mathbf{g}'\cdot\mathbf{k}P_2(\mathcal{C}^2)d\mathbf{k}d\mathbf{I}^\alpha d\mathbf{I}^\gamma \tag{97}$$

with $\mathbf{g}'=\mathbf{v}^\alpha-\mathbf{v}^\gamma$ the relative velocity between the two colliding bubbles just before contact and $\mathbf{k}$ a unit vector along $\mathbf{x}^\gamma-\mathbf{x}^\alpha$, bubble $\alpha$ being placed at $\mathbf{x}$, i.e., $\mathbf{x}^\alpha=\mathbf{x}$. The collision force is (see [15] and [19])

$$\mathbf{F}_c = -m_a(\mathbf{g}'\cdot\mathbf{k})\mathbf{k}, \tag{98}$$

where $m_a=(m/2)(1-3R^{-3}+6R^{-6})^{-1}|_{R=2}\simeq 0.7m$ is the added mass at contact. Carrying out the integrations in Eq. (97), and comparing the result with Eq. (83) gives

$$C_c = 1-\frac{1}{R^6}\left(\frac{9}{4}-\frac{3}{2}A-\frac{9}{10}A^2\right)\Big|_{R=2}, \tag{99}$$

where we have replaced $A^*$ by $A$. We take this opportunity to report that there was a typographical error in Kang *et al.* [19] in the result for $C_c$ at $A=0$; the coefficient in front of $R^{-6}$ in Eq. (55) of that study should be changed from 9/8 to 9/4, and the resulting expression then agrees with the present result, Eq. (99), at $A=0$.

It is found that only a small correction for larger volume fractions is needed to fit the dynamic simulation data shown in Figure 9:

$$C_c = \left[1-\frac{1}{64}\left(\frac{9}{4}-\frac{3}{2}A-\frac{9}{10}A^2\right)\right](1-0.42\phi). \tag{100}$$

Thus the $A-$dependence of $C_c$ observed in simulations is well predicted by the theory for $\phi \ll 1$.

The numerical simulation results for $C_{11}^c$ can be adequately represented by means of

$$C_{11}^c = \left[1 - \frac{1}{64}\left(\frac{9}{4} - 0.1A + 0.01A^2\right)\right](1 - 0.42\phi); \tag{101}$$

Thus the collision stress is given by

$$P_{ij}^c = 4n(m/2)T\phi\chi\left(\frac{1}{2}(C_{ii}^c - C_{11}^c)\delta_{ij} + \frac{1}{2}(3C_{11}^c - C_{ii}^c)e_i e_j\right), \tag{102}$$

with the coefficients $C_{11}^c$ and $C_{ii}^c$ given by Eqs (100) and (101), and $e_i$ a unit vector in the direction of the mean bubble motion.

Figure 13 shows the trace of the total bubble-phase stress as a function of $A$ for various values of $\phi$. At small $A$ the kinetic and collision stresses dominate, and they are always positive. The hydrodynamic stress, on the other hand, is always negative and increases in magnitude with $A$, thus we expect that the total stress will become negative at sufficiently large $A$ for a given $\phi$. We extrapolated the results shown in Figure 13 to determine $A_{cr}$ at which this stress will become zero. The results are shown in Figure 14. One would expect that clustering would be very significant for $A > A_{cr}$ and therefore the results shown in this figure may be used as a rough criterion for estimating the magnitude of velocity fluctuations needed for preventing clusters in bubbly liquids. The solid line shown in Figure 14 corresponds to the prediction of a simple theory outlined below.

Since we have seen that $P_{ij}^h$ is much smaller in magnitude compared with the Maxwell stress, we set it to zero. The total stress is then given by

$$\begin{aligned} P \equiv P_{kk}/3 &= (m/2)n[C_k T(1 + 4\phi\chi(C_c/C_k)) - 9\phi\overline{D}^2] \\ &= (m/2)nT\left[C_k(1 + 4\phi\chi(C_c/C_k)) - \phi A((C_a/2) + 1)^2\right], \end{aligned} \tag{103}$$

where we have related the averaged dipole to the averaged velocity via Eq. (24). Equating the total stress to zero, we obtain

$$A_{cr} = \frac{C_k[1 + 4\phi\chi(C_c/C_k)]}{\phi[(C_a/2) + 1]^2}. \tag{104}$$

Since $C_a$ and $C_c$ are functions of both $\phi$ and $A_{cr}$, we must solve for $A_{cr}$ in the above by trial and error. An explicit expression is obtained if we neglect the dependence of these coefficients on $A_{cr}$. The upper curve in Figure 14 was generated by taking $C_c = C_k$ and using Eqs (2) and (96) to estimate $C_a$ and $C_k$, respectively. We see that this simple procedure gives fairly accurate estimates of $A_{cr}$; the error at $\phi = 0.05$ is within the confidence range of extrapolation of the numerical results shown in Figure 13. Evidently, the increase in $C_a$ with $A$ is partially compensated by the increase in $C_c$ with $A$.

Note that according to this expression $A_{cr} \to 4/(9\phi)$ as $\phi \to 0$, indicating that small velocity fluctuations will be adequate in preventing clustering in very dilute bubbly liquids. Finally, the above analytical result can be easily extended to $\phi > 0.3$. For example, at $\phi = 0.5$ it predicts

$A_{cr} \simeq 2$, indicating that $A_{cr}$ is nearly constant for $\phi > 0.25$. The increase in $C_a$ at higher $\phi$ is offset by the increase in the collision stress, or $\chi$.

The stability criterion for the uniform state of bubbly liquids derived from the continuity and momentum equations for the bubble-phase depends on the compressibility $dP/d\phi$ of the bubble-phase. Although we shall not undertake the complete stability analysis in the present study, it is of some interest to determine the value $A'_{\mathrm{cr}}$ at which the isothermal (constant $T$) compressibility becomes zero. This can be done readily be differenting Eq. (103) at constant $T$ and setting the derivative to zero. The result for $A'_{\mathrm{cr}}$ thus obtained is represented by the lower curve in Figure 14. We see that $A'_{\mathrm{cr}}$ approximately equals $A_{\mathrm{cr}}/2$ for the complete range of $\phi$. Considering that we have not seen complete phase-transition from the random gas-like dispersed state to a clustered state in any of our simulations, we see that the uniform state is stable even when the isothermal compressibility becomes negative. Thus, a complete analysis including the energy equation may be needed for predicting the exact phase-transition point.

### 4.5 Terms in the impulse fluctuation equation

The impulse fluctuation equation (37) is expressed in terms of the impulse variance, whereas we shall be usually interested in the bubble phase temperature. We therefore define

$$\overline{I^2} - \overline{I}^2 = 3C_I(m/2)^2 T. \tag{105}$$

Note that, in the light of the observed isotropy of bubble velocity fluctuations (cf. Figure 6), we restrict our attention to the trace of $\overline{I_i I_j}$ (the impulse variance tensor has indeed found to be nearly isotropic in the simulation results). Upon substituting Eq. (94) in Eq. (87) it is seen that $C_I$ for dilute bubbly liquids is given by

$$C_I = 1 - \frac{3}{16}\phi(1 - A), \tag{106}$$

where we have used Eqs (24) and (70). This dilute-theory result is seen to hold also for higher volume fractions in Figure 15. Notice that the ordinate scale varies only from about 0.94 to 1.06 for the results shown in the figure, and hence the scatter in the data should be interpreted as rather small.

The energy-flux $q_{ijk}$ given by Eq. (38) is written as

$$q_{ijk} = n\left(\frac{m}{2}\right)^2 \left[f_1 \overline{v}_i \overline{v}_j \overline{v}_k + f_2 T \delta_{ij} \overline{v}_k + f_3 T\left(\delta_{ik}\overline{v}_j + \delta_{jk}\overline{v}_i\right)\right]. \tag{107}$$

Dynamic simulation results for $f_1, f_2$ and $f_3$ are shown in Figures 16–18. We see that $f_2 \simeq f_3$. It is easy to show that $f_1, f_2$ and $f_3$ are $O(\phi)$. Therefore the results of numerical simulations were fitted as follows:

$$q_{ijk} = \left(\frac{m}{2}\right)^2 n\phi\left[(\overline{v}_i\delta_{jk} + \overline{v}_j\delta_{ik} + \overline{v}_k\delta_{ij})T\left(\frac{3}{2} - \frac{A^2}{18}\right) + \overline{v}_i\overline{v}_j\overline{v}_k\left(\frac{4}{3} - \frac{5}{A}\right)\right]. \tag{108}$$

The solid lines in Figures 16-18 compare the above fitted expressions to the results of numerical simulations.

The trace of the fluctuation impulse flux is given by

$$q_{iik} = \left(\frac{m}{2}\right)^2 n\phi\overline{v}_k \left[\frac{5}{2}T + \left\{\frac{4}{3} - \frac{5A}{18}\right\}\overline{v}^2\right]. \tag{109}$$

The source term $S_{ij}$ in the energy equation was also evaluated in dynamic simulations. Our simulations showed that its magnitude was always very small and that it fluctuated around zero for all $A$ and $\phi$. Thus, we may set $S_{ij}$ to zero for the case of mean relative motion in the absence of imposed velocity gradients. It should be noted that this quantity does play a very important role in the energy equation of sheared suspensions, as shown in [19]. That its value must be zero for the case of mean relative motion examined here can also be concluded from the fact that at steady state all the other terms in the energy equation vanish for inviscid interactions in homogeneous suspensions.

The calculation of $\tau^c_{ijkl}$ amounts to determining the change in $\partial(I^\alpha_i I^\alpha_j)/\partial v^\alpha_l$ during collisions of bubble $\alpha$ with the other bubbles in the suspension. As shown in Kang *et al.* [19] such quantities, which appear in the calculation of the collision stress, may be evaluated reasonably accurately by treating the bubbles as if they were rigid particles of mass $(m/2)C_k$. Their calculations required the derivative of $I^\alpha_i v^\alpha_j$ and a reasonable approximation for our purpose can be made by multiplying their result by $(m/2)C_I/C_k$. Thus a reasonable approximation to $\tau^c_{ijkl}$ is

$$\tau^c_{ijkl} = (m/2)(C_I/C_k)[\delta_{il}P^c_{jk} + \delta_{jl}P^c_{ik}] \tag{110}$$

Likewise, we also relate $\tau^h_{ijkl}$ to the hydrodynamic stress.

The ponderomotive term requires the calculation of $C_{DI}$ defined by

$$\overline{I_i D_j} - \overline{I}_i \overline{D}_j = -(m/4)C_{DI}T_{ij}. \tag{111}$$

By using Eqs (62) and (63) it is easy to show that $C_{DI} = (2/3)C_k + C_I$.

Only the viscous dissipation term, $\overline{\mathbf{F}_v \cdot \mathbf{I}} - \overline{\mathbf{F}_v} \cdot \overline{\mathbf{I}}$, in Eq. (37) remains to be calculated. The finite viscosity affects the fluctuation equation in two ways. The first one corresponds to giving rise to the viscous dissipation term in the fluctuation equation. To leading order this can be determined by evaluating the above term for the spatial and velocity configurations given by the potential flow interactions, as has been done for the other terms in the averaged equations in the present study. This term will give a sink term in the impulse variance equation as given by $36\pi\mu a n R^I_{diss}\overline{I}^2$, where $R^I_{diss}$, as a function of $\phi$ and $A$, will be evaluated below. The second effect of finite viscosity arises from the fact that the viscous forces acting on the bubbles will alter slightly the bubble trajectories and velocity distribution. The viscous, as well as potential forces acting on the bubbles, will be modified as a consequence, and this will lead to a term that can be treated as a part of $S_{ij}$ (note that it will then depend on the viscosity of the liquid). The source of energy fluctuations arising from such interactions has been evaluated for the case of solids sedimenting through a viscous gas by Koch [35] for dilute, and by Koch and Sangani [36] for non-dilute, gas-solid suspensions. The balance of the source of energy fluctuations produced by hydrodynamic interactions and the sink of energy with the viscous dissipation determines the magnitude of $T/\overline{v}^2$ in homogeneous, monodisperse sedimenting suspensions. Since the dynamic simulations of buoyant rise of bubbles by Smereka [14] and Sangani and Didwania [15] have shown that velocity fluctuations continue to decrease with increasing time when the viscosity is finite, we conclude that the source of energy fluctuations arising from hydrodynamic interactions with small viscosity in the absence of mean shear must be very small in magnitude. Thus, we shall determine here only the sink term arising from finite viscosity of the liquid.

The energy dissipation term is written as

$$n\left(\overline{F_{v,i}I_i} - \overline{F}_{v,i}\overline{I}_i\right) = -36\pi\mu a n(m/2)TR^I_{\mathrm{diss}}(\phi, A). \tag{112}$$

The simulation results for $R^I_{\mathrm{diss}}$ are shown in Figure 19. A dilute-theory result for $R^I_{\mathrm{diss}}$ is straightforwardly obtained by calculating $\overline{\mathbf{F}_v \cdot \mathbf{I}}$ using Eq. (73) and the discussion thereafter. The result reads

$$R^I_{\mathrm{diss}} = 1 - \frac{3}{16}\phi + \frac{7}{48}\phi A. \tag{113}$$

This dilute-theory result is shown in Figure 19 to represent data up to $\phi = 0.3$ well, so no higher-order volume-fraction correction is needed.

# 5 Simplified equations

With all the terms in the averaged equations calculated for the case of nonzero mean relative motion in the present study, and for the case of nonzero velocity gradient in Kang *et al.* [19], we are now in a position to propose the complete set of averaged equations for flows of bubbly liquids at large Reynolds and small Weber numbers. The continuity and momentum equations for the bubble phase are given by Eqs (12) and (13). The bubble-phase stress calculated in the present study can now be combined with that in Kang *et al.* [19]. It was shown there that the rheology of the bubble phase is non-Newtonian, exhibiting shear thickening and nonzero normal stress differences. To explain these simulation results and multiple steady states of sheared bubbly liquids it was necessary to solve for the second moments of the velocity distribution. Significant normal-stress differences were found only when the Reynolds number based on the shear rate was small (say less than 50) and when the volume fraction is less than say 0.05. Outside this range, however, a much simpler Newtonian stress tensor was found to be adequate. For the sake of simplicity therefore we shall adopt that expression and write

$$P^*_{ij} = (\rho/2)\phi T(C_k + 4\phi\chi C_c)\delta_{ij} - [\kappa - (2/3)\mu_s]\, e^p_{kk}\delta_{ij} - 2\mu_s e^p_{ij}, \tag{114}$$

where $\kappa$ and $\mu_s$ are the bulk and shear viscosities of the bubble phase. Note that these are *not* the viscosities of the gas inside the bubble; instead, as shown in [19], their values will be close to that of a dense monatomic gas whose molecules have a mass equal to $C_k(\phi)m/2$, as in the case of viscosities of granular materials (Jenkins and Savage [37], Babic [38], Sangani *et al.* [39]). Using the analogy with the kinetic theory of dense gases and granular materials, we write (see Babic [38], Sangani *et al.* [39])

$$\mu_s = \frac{16}{5\pi^{1/2}} C_k(\rho/2) a T^{1/2} \phi^2 \chi \left[ 1 + \frac{\pi}{12} \left( 1 + \frac{5}{8\phi\chi} \right)^2 \right], \tag{115}$$

$$\kappa = \frac{16}{3\pi^{1/2}} C_k(\rho/2) a T^{1/2} \phi^2 \chi. \tag{116}$$

The particle strain rate $e^p_{ij}$ to be used in Eq. (114) is defined as

$$e^p_{ij} = (\gamma^p_{ij} + \gamma^p_{ji})/2, \qquad \gamma^p_{ij} = \frac{\partial}{\partial x_j}(\overline{u}_i + C_a \overline{v}_i). \tag{117}$$

It should be noted that Kang *et al.* [19] considered the case of simple shear flows with no mean force on the bubbles and therefore in their analysis the mean relative velocity $\overline{v}_i$ was zero, and the bubble phase strain rate $e^p_{ij}$ was equal to the mixture strain rate $e_{ij}$. In general the two are, of course, not equal, and one must substitute $e^p_{ij}$ for $e_{ij}$ in [19] since one expects the transfer of momentum in the bubble phase with nonzero $e^p_{ij}$ to occur even when $e_{ij} = 0$. A closer examination of the fluctuation equation derived in Section 3 suggested that the work done by the stress $P^*_{ij}$ must be multiplied by $e^p_{ij}$ and not by the gradient of $\overline{w}_i = \overline{u}_i + \overline{v}_i$.

Although the expression for the stress tensor proposed here is Newtonian, it must be kept in mind that $T$ in our system, as in the granular flows, depends on the shear rate unlike molecular systems, where $T$ is independent of the shear rate. A higher shear rate will lead to larger velocity fluctuations and hence greater $T$. Thus the relation proposed here allows for the most important feature — the shear thickening behavior — of the bubble-phase rheology found in Kang *et al.* [19].

The fluctuation energy equation is

$$\frac{3}{2}(\rho/2)\phi \frac{dT}{dt} = -\frac{\partial Q_j}{\partial x_j} - P^*_{ij} e^p_{ij} - 36\pi\mu a n (T - \alpha \overline{v}^2) - 3(\rho/2)\phi T_{ij} \frac{\partial G_i}{\partial x_j}. \tag{118}$$

In writing the above equation we have set $C_k$, $C_I$, $C_{DI}$, and $R^I_{\text{diss}}$ all to unity. Our computations in Section 4 showed that all these coefficients related to the fluctuations in impulse or velocity moments were not very different from unity unlike $C_a$ and $C_d$ appearing in the average momentum equation for which the long-range nature of interactions are significant and the coefficients differ from unity significantly. We estimate that setting all these coefficients to unity would result in an overall estimate of $T$ that will differ by less than 15% for $\phi < 0.3$ from that obtained by keeping these coefficients.

The term on the left-hand side of Eq. (118) can be interpreted as the rate of change of kinetic energy per unit volume of the suspension due to fluctuation motion of the bubbles. The first term on the right-hand side represents the flux of energy. Supplementing the expression obtained in Section 4 with the Fourier's conduction law, we obtain

$$Q_j = -k\frac{\partial T}{\partial x_j} + (\rho/2)\phi^2\overline{v}_j\left[\frac{5}{4}T + \left\{\frac{2}{3} - \frac{5\overline{v}^2}{36T}\right\}\overline{v}^2\right] \tag{119}$$

with the conductivity $k$ given by

$$k = \frac{8}{\pi^{1/2}}(\rho/2)aT^{1/2}\phi^2\chi\left[1 + \frac{25\pi}{512\phi^2\chi^2}\right], \tag{120}$$

The above expression, and the coefficients of viscosities, are taken from the granular flow literature (see, e.g. [37-39]) by substituting the added mass $m/2$ of the bubbles for the mass of the particles in granular flows.

The second term on the right-hand side of Eq. (118) represents the rate of work done (per unit volume) by the bubble-phase stress (minus the Maxwell stress) on the bubble-phase rate of strain $e^p_{ij}$. (Note that our definition of the stress is the negative of the stress $\sigma_{ij}$ used in the usual fluid mechanics literature.) This term then represents the conversion of mechanical work into internal or fluctuation energy.

The third term on the right-hand side represents the effect of viscosity on the fluctuation equation. Here, we have added an *ad-hoc* $\alpha\overline{v}^2$ term that is supposed to represent the source of fluctuation energy arising from the viscous effects as discussed earlier. We have not evaluated $\alpha$ because, as argued earlier, it is expected to be small in magnitude for pure potential flow interactions at small Weber numbers. Note that at steady state, in the absence of imposed shear, and for homogeneous, buoyancy driven flow, $\alpha = T/\overline{v}^2 = A^{-1}$. The fact that the dynamic simulations for this condition shows formation of large clusters indicate that $\alpha < A^{-1}_{cr}$. As discussed later it may be possible, however, to choose $\alpha$ from the experiments conducted at finite Weber numbers for which the clusters are not observed.

The last term in Eq. (118) represents the rate of work done by the force induced by the backflow. Recall that $G_i = 3\phi\overline{D}_i$. Apparently, it also equals the work done by the kinetic part of the stress on the backflow gradient, although this may just be an artifact of taking $C_{DI} = C_k$. Unlike all the other terms introduced so far this term requires all the second moments $T_{ij} = \overline{v_i v_j} - \overline{v}_i\overline{v}_j$ of the velocity distribution. Recognizing that the kinetic stress can be evaluated by using the coefficients of viscosity for ideal gases ($\phi \to 0$) we estimate $P^k_{ij} = (\rho/2)\phi T_{ij}$ from (cf. Eq.(114))

$$P^k_{ij} = (\rho/2)\phi T\delta_{ij} - 2\mu_s(0)[e^p_{ij} - (1/3)e^p_{kk}\delta_{ij}] \tag{121}$$

where $\mu_s(0)$ is the shear viscosity obtained by taking the limit $\phi \to 0$ in Eq. (115).

Finally, it should be noted that the lift force and the $S_{ij}$ term do not contribute to the energy equation. Both of these do affect the individual components of the second moments of velocity but not to the fluctuation energy. Note that $S_{ii} = 0$ is a consequence of our assumption that the collision between bubbles conserve the kinetic energy of the liquid.

Thus, we see that the average equations for the bubble-phase are very similar to those for the rapid granular and high Stokes gas-solid suspensions [38,39]. The bubbles can be treated as if their mass equals $m/2$ as far as the energy equation and the rheology is concerned, and as if their mass equals $C_a m/2$ as far as their net momentum is concerned. The mean relative velocity of the bubbles in addition creates a negative Maxwell pressure, and this can play an important role in the stability of bubbly liquids when the temperature fluctuations are not large. In addition, we also see that additional terms in the energy flux vector arises when the relative velocity of the bubbles is nonzero. Another important difference is that the behavior of bubbly liquids depends additionally on the mixture velocity $\overline{u}_i$ through the force $\mathbf{F}_{\overline{\mathbf{u}}}$ in the momentum equation and through $e_{ij}$ that appears in the rheology of the bubble phase. Thus, in addition to the above continuity, momentum, and fluctuation energy equations for the bubble phase, we also need to solve, in general, the appropriate mixture equations. These, we present next.

The continuity and the momentum equations for the gas-liquid mixture are derived in [18,20]:

$$\nabla \cdot \overline{\mathbf{u}} = 0, \tag{122}$$

$$\rho \left[ \frac{\partial}{\partial t}(1-\phi)\overline{u_i}^L + \frac{\partial}{\partial x_j}(1-\phi)\overline{u_i}^L \overline{u_j}^L \right] = -\frac{\partial \overline{p}}{\partial x_i} + \rho(1-\phi)g_i - \frac{\partial}{\partial x_j}\hat{\Sigma}_{ij}. \tag{123}$$

Here $\overline{u_i}^L = \overline{u}_i - \phi\overline{v}_i/(1-\phi)$ is the averaged liquid velocity, $\overline{p}$ is the averaged mixture pressure plus one-third of the trace of $\Sigma_{ij}$, and $\hat{\Sigma}_{ij} \equiv \Sigma_{ij} - (1/3)\delta_{ij}\Sigma_{kk}$ is the deviatoric part of the mixture stress $\Sigma_{ij}$ defined by

$$\Sigma_{ij} = \Sigma_{ij}^R + \Sigma_{ij}^S + \Sigma_{ij}^v \tag{124}$$

with the Reynolds, interfacial, and viscous stresses defined as, respectively,

$$\Sigma_{ij}^R = \rho\langle\Theta(u_i - \overline{u_i}^L)(u_j - \overline{u_j}^L)\rangle, \qquad \Sigma_{ij}^S = an\langle\int_{S^\alpha}(p^L - p^G)n_i n_j dA\rangle, \tag{125}$$

$$\Sigma_{ij}^v = -\langle\Theta\tau_{v,ij}\rangle - na\langle\int_{S^\alpha}\tau_{v,ik}n_k n_j dA\rangle. \tag{126}$$

In the above expressions $\Theta(\mathbf{x})$ is the liquid-indicator function whose value is unity when $\mathbf{x}$ is inside the liquid and zero otherwise, $\tau_{v,ij}$ is the viscous stress in liquid, i.e. total stress plus $p^L\delta_{ij}$, $p^L$ being the pressure determined from the inviscid equations, $n_i$ is the unit outer normal on the surface of a bubble, and $n$ is the number density of the bubbles. Note that the interfacial stress equals the number density of bubbles times the (inviscid) stresslet induced by the motion of bubbles.

For large Reynolds number flows the Reynolds and interfacial stresses will dominate the total deviatoric stress in most regions of the flows. The viscous stress, however, will become important in the regions where the volume fraction of the bubbles is small, e.g. near the wall. Thus, we must determine the complete $\phi$ dependence of $\Sigma_{ij}^R$ and $\Sigma_{ij}^S$ using an inviscid flow theory while it will suffice to determine the viscous stress for dilute bubbly liquids. As shown in Appendices A and B the total stress can be evaluated from

$$\Sigma_{ij} = P^*_{ij} + (\rho/2)\phi\overline{v}_i\overline{v}_j \left[C_a + \phi\left\{(C_a+2)(2+\phi(C_a+2)) - 2(1-\phi)^{-1}\right\}\right] - 2\mu(1+5\phi/3)e_{ij} + \text{ I.T.}, \tag{127}$$

where I.T. stands for the isotropic terms. Their evaluation is not necessary since we are only interested in the deviatoric part of the mixture stress. The mixture stress is seen to consist of the bubble-phase stress (note that the Maxwell stress is isotropic and therefore $P^*_{ij}$ can be replaced by total $P_{ij}$ without any consequences) and an extra part that depends on the mean relative velocity of the bubbles. The leading term in this extra part, i.e. $(\rho/2)\phi\overline{v}_i\overline{v}_j$, agrees with the expressions given by Biesheuvel and van Wijngaarden [20] and Sangani and Didwania [18], among others, who considered dilute bubbly liquids with all the bubbles rising with the same velocity. As noted first in [20], the mixture stress is anisotropic even in the absence of an imposed velocity gradient. We also see that in the large Reynolds number limit the liquid viscosity is modified by a factor of $1+5\phi/3$. This factor is different from the more familiar Einstein factor $1+5\phi/2$ which applies to the effective viscosity of dilute suspensions of rigid spheres in the zero Reynolds number limit. Note also that, because $\Sigma_{ij}$ is directly related to the bubble-phase stress, the mixture will also exhibit a shear thickening behavior – larger shear rates will increase the velocity fluctuations, and this in turn will lead to greater mixture viscosity.

Finally, it should be noted that the effective pressure $\overline{p}$ must be treated as an unknown in addition to $\overline{\mathbf{u}}$ unlike the stress $P$ in the bubble-phase equation where it was treated as a function of $\phi$, $T$, and $e_{ij}$. The effective pressure defined here is the averaged mixture pressure plus the trace of $\Sigma_{ij}$; the detailed relation between the two is unnecessary since in most applications, e.g. in determining the pressure drop across the length of a pipe in which the bubbly liquid is flowing, it is $\overline{p}$ that is important and not the ensemble-averaged pressure.

# 6 Concluding remarks

We have presented a complete set of averaged equations for the gas-liquid mixture and the bubble-phase in this study for the special case of large Reynolds, small Weber number flows. Considerable simplifications that occur in the equations describing the flow induced by the relative motion of the bubbles make it possible to fully analyze the effect of bubble interactions at finite $\phi$ on the closure relations in these flows. From a practical point of view, however, the small Weber number limit is somewhat restrictive: the bubbles in most ordinary flows quickly coalesce to become 2-3 mm in diameter, and then the deformation of the bubbles and the resulting wake and form drag effects become important. Also, the Weber number based on the root-mean-squared velocity may become large in some instances even though say the Weber number based on imposed shear is small, as shown by numerical simulations in Kang *et al.* [19]. To extend the range of application of the equations derived here to larger bubbles, an approach that seems promising at present is to rely on experiments at relatively small to moderate Weber numbers to determine the finite Weber number corrections to some of the terms in the average equations derived here. For example, the added mass and viscous drag coefficients may be multiplied by the corresponding correction factors for single ellipsoidal bubble determined by Moore [2], as has been done recently by Lammers and Biesheuvel [34]. The viscous term in the energy equation may be multiplied by a correction factor given by Kang *et al.* [19] (cf. their Eq. (93) with $We_v$ there replaced by the $We_T = \rho a T/\sigma$, the Weber number based on the magnitude of velocity fluctuations) so that $T$ does not increase in an unbounded manner with the increasing shear rate. Likewise the constant $\alpha$ introduced in the energy equation (cf. Eq. (118)) should be determined as a function of Weber number and $\phi$ from the recent measurements of void fraction wave speeds in a bubble column by Lammers and Biesheuvel [34]. The numerical simulations of bubbly liquids at finite Weber and Reynolds number through direct numerical solution of Navier-Stokes equations of motion as done, for example, by Tryggvason and his colleagues [40–42], although limited at present to relatively small Reynolds numbers, also offer a possibility of determining $\alpha$ in finite Weber number flows. We believe that this approach of combining the averaged equations based on detailed calculations with small $We$ with a few modifications determined from available experimental data provide the most promising approach to developing a reliable set of averaged equations for describing flows of bubbly liquids. Our future work will be concerned with investigating in detail the consequences of these equations to the stability and other experimentally well-examined bubbly liquid flows.

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## Appendix A: Reynolds and interfacial stresses

In addition to the bubble-phase stress tensor $P_{ij}$, which occurs in the averaged momentum equation for the bubbles, the stress tensor $\Sigma_{ij}$ in the momentum equation of the mixture also needs to be evaluated. Biesheuvel and van Wijngaarden [20] and Sangani and Didwania [18] determined $\Sigma_{ij}$ for dilute bubbly liquids. In this Appendix, $\Sigma_{ij}$ is related to the bubble-phase stress $P_{ij}$, for which results for arbitrary volume fractions were presented in Section 4. Since we are mainly interested in determining the mixture and bubble velocities, and not the mixture pressure, only the deviatoric part of $\Sigma_{ij}$ is important.

The Reynolds stress contribution to $\Sigma_{ij}$ is expressed in terms of the fluid velocity fluctuations about the averaged liquid velocity $\overline{u}_i^L$ by Eq. (125). To relate this Reynolds stress to bubble-phase stress components we first eliminate the averaged liquid velocity from this definition using $\overline{u}_i^L = \overline{u}_i - \frac{\phi}{1-\phi}\overline{v}_i$. Substituting this, together with $u_i' = u_i - \overline{u}_i$ in Eq. (125), gives for the Reynolds stress

$$\Sigma_{ij}^R = \rho\langle\Theta u_i' u_j'\rangle - \rho\frac{\phi^2}{1-\phi}\overline{v}_i\overline{v}_j. \tag{128}$$

Here we have used that $\overline{\Theta u_i'} = -\phi\overline{v}_i$, which follows from the requirement that the average of $u_i'$ in the mixture is zero. The hydrodynamic interaction stress in Bulthuis *et al.* [26] is formulated in terms of the fluid velocity relative to the average backflow induced by the bubble motion, $G_i$. We therefore substitute $u_i' = G_i + u_i^*$ in Eq. (128):

$$\Sigma_{ij}^R = \rho\langle\Theta u_i^* u_j^*\rangle - \rho\left[(1-\phi)G_iG_j + \phi(G_i\overline{v}_j + G_j\overline{v}_i) + \frac{\phi^2}{1-\phi}\overline{v}_i\overline{v}_j\right]. \tag{129}$$

The only unknown on the right-hand side is the first term, but that can be related to the bubble-phase stress tensor by using Eq. (63) in Bulthuis *et al.* [26], i.e., by using

$$-\rho^{-1}P_{ij}^h = \overline{\Theta T_{ij}} + n\langle\int_{S^\alpha} n_k s_j T_{ik} dA\rangle, \tag{130}$$

with $S^\alpha$ the surface of a test bubble $\alpha$, $T_{ij} = u_i^* u_j^* - \frac{1}{2}\delta_{ij}u_k^* u_k^*$, and $n_k = s_k/a$ the outward normal vector at the bubble surface. Substituting this into the above expression for the Reynolds stress gives

$$\Sigma_{ij}^R = P_{ij}^h - \rho n\langle\int_{S^\alpha} n_k s_j T_{ik} dA\rangle - \rho\left[G_iG_j(1-\phi) + \phi(G_i\overline{v}_j + G_j\overline{v}_i) + \frac{\phi^2}{1-\phi}\overline{v}_i\overline{v}_j\right] + \text{I.T.}, \tag{131}$$

where I.T. refers to isotropic terms. The integral over the surface of a bubble is combined with the interfacial stress below.

The interfacial stress is given by

$$\Sigma_{ij}^S = n\langle\int_{S^\alpha} p^L n_i s_j dA\rangle + \text{I.T.}\,. \tag{132}$$

In Eq. (132) $p^L$ is the pressure in the liquid just outside a test bubble. We shall assume that the flow is incompressible and irrotational. Then $p^L$ follows (to a constant) from the Bernoulli equation,

$$-\frac{p^L}{\rho} = \frac{\partial \varphi}{\partial t} + \frac{1}{2}u^2 = \left(\frac{\partial \varphi}{\partial t}\right)_{\mathbf{s}} - w_i^\alpha u_i + \frac{1}{2}u^2. \tag{133}$$

Here $\mathbf{s}$ is a coordinate system that moves with bubble $\alpha$ (having velocity $\mathbf{w}^\alpha$). The contribution of the partial time derivative of $\varphi$ to $\Sigma_{ij}^S$ plays an important part in the propagation of sound waves through bubbly flows, as the bubble radius is then a function of time [20]. Here, however, the bubbles are incompressible, for which case the contribution of $\partial\varphi/\partial t$ to $\Sigma_{ij}^S$ can be shown to be unimportant. Thus, neglecting the time derivative, and substituting Eq. (133) into Eq. (132),

$$\Sigma_{ij}^S = \rho n \langle \int_{S^\alpha} \left( w_k^\alpha u_k - \frac{1}{2}u^2 \right) n_i s_j dA \rangle + \text{I.T.}. \tag{134}$$

Again we write the velocity in the mixture as $\mathbf{u} = \overline{\mathbf{u}} + \mathbf{G} + \mathbf{u}^*$ to give, with $\mathbf{G}^t \equiv \mathbf{G} + \overline{\mathbf{u}}$,

$$\Sigma_{ij}^S - \rho n \langle \int_{S^\alpha} n_k s_j T_{ik} dA \rangle = \rho n \langle \int_{S^\alpha} (v_k^\alpha - G_k)\left(u_k^* n_i s_j - u_i^* n_k s_j\right) dA \rangle + \text{ I.T.}. \tag{135}$$

The integral on the right-hand side can be evaluated using the point-dipole approximation Eq. (60):

$$\rho \int_{S^\alpha} (n_i u_k^* - n_k u_i^*) s_j dA = \delta_{kj}(I_i^\alpha - mG_i) - \delta_{ij}(I_k^\alpha - mG_k). \tag{136}$$

Combining now the two stress contributions we have

$$\Sigma_{ij}^R + \Sigma_{ij}^S = P_{ij}^h + n < I_i^* v_j^* > -\rho[(1-\phi)G_i G_j + \phi(G_i \overline{v}_j + G_j \overline{v}_i) + \phi^2 \overline{v}_i \overline{v}_j/(1-\phi)] + \text{ I.T.}, \tag{137}$$

where $I_j^{*,\alpha} = I_j^\alpha - mG_j$. and $v_j^{*,\alpha} = v_j^\alpha - G_j$.

The above can be corrected for the collision of the bubbles by assuming that very short-range forces act on the bubbles during the collision process which alter only the pressure distribution in the gap region between the two colliding bubbles. Let $p^{\text{col}}$ be the excess pressure during the short collision process. This gives an additional contribution to the interfacial stress as given by

$$2n \int_{S^\alpha} p^{\text{col}} n_i s_j dA = -2nan_j^{\text{col}} F_{c,i}, \tag{138}$$

with $F_{c,i}$ the total collision force on one of the bubbles and $n_j^{\text{col}}$ the normal on the surface of that bubble at the point of contact with the other bubble. The factor 2 arises from the fact that the collision involves two bubbles. This collision contribution is, after averaging Eq. (138),

$$\Sigma_{ij}^c = -2na\langle n_j^{\text{col}} F_{c,i} \rangle = -\frac{2an}{N\Delta t}\sum n_j^{\text{col}} F_{c,i} = P_{ij}^c. \tag{139}$$

Thus, the contribution of collisions to the stress in the mixture equation is the same as that to the stress in the momentum equation for the bubbles.

Combining now the above stress with Eq. (137), using $\overline{I}_i = (m/2)C_a \overline{v}_i$, and $G_i = 3\phi\overline{D}_i = -(C_a/2+1)\overline{v}_i$, and simplifying we obtain the result given in the main text (cf. (Eq. (127)).

## Appendix B: Viscous stress $\Sigma_{ij}^v$

As mentioned in the main text, the viscous stress will be important in large Reynolds number flows only in the regions where the volume fraction of the bubbles is small. We shall therefore evaluate the leading order effect of the bubbles on stress at large Reynolds numbers. The viscous stress at a point in the liquid is given by $\tau_{v,ij} = -p^v\delta_{ij} + 2\mu e_{ij}$. (We shall use a slightly different notation in this Appendix and refer to the local strain rate as $e_{ij}$ and the average strain rate as $E_{ij}$ even though we used $e_{ij}$ in the main text to denote the average strain rate of the mixture.) $p^v$ is the correction to the inviscid pressure arising from the small viscous effects. We expect this pressure and $\tau_v$ to be linear in the velocity of the bubbles and to the imposed velocity gradient $E_{ij}$ unlike the inviscid pressure which was bilinear in the velocity of the bubbles. Thus, the average viscous stress, being a second-order tensor, will be independent of the mean velocity of the bubbles in an isotropic or dilute suspension, and we only need to consider the effect of finite $E_{ij}$. Now, the viscous pressure, and hence the stress, can be evaluated in the manner outlined in [8,29] for the small-amplitude oscillatory flows induced by an imposed time-dependent strain rate, $E_{ij}e^{i\omega t}$, in the same way as we have used it to determine the viscous drag forces on the bubbles. Alternatively, we can use dissipation arguments to first evaluate the total viscous dissipation and then equate it to $-\Sigma_{ij}^v E_{ij}$ to determine $\Sigma_{ij}^v$. Both methods yield exactly the same result although we shall present here only the analysis using the latter.

Let the velocity in the liquid be given by $u_i = E_{ij}x_j + u_i^*$, where $u_i^*$ is the disturbance velocity field due to the presence of the bubble at origin. It is to show that

$$u_i^*(\mathbf{r}) = \frac{a^5}{3r^5}E_{ij}\left[x_j\delta_{ik} + x_i\delta_{jk} - 5\frac{x_ix_jx_k}{r^2}\right], \tag{140}$$

with $\mathbf{x} = \mathbf{r}/r$. Writing $e_{ij} = E_{ij} + e_{ij}^*$, the dissipation rate per unit volume of the mixture is given by

$$\dot{E}_{\text{diss}} = 2\mu\langle\Theta e_{ij}e_{ij}\rangle = 2\mu[(1-\phi)E_{ij}E_{ij} + E_{ij}\langle\Theta e_{ij}^*\rangle + \langle\Theta e_{ij}^*e_{ij}^*\rangle]. \tag{141}$$

The second term on the right-hand side is zero and the third term can be evaluated by converting it to a volume integral outside the test bubble and using a divergence theorem which converts the volume integral in the liquid to an area integral on the surface of the bubble:

$$\langle\Theta e_{ij}^*e_{ij}^*\rangle = -\frac{n}{2}\langle\int_{S^\alpha}\frac{\partial u^{*2}}{\partial r}dA\rangle, \tag{142}$$

where $n$ is the number density of the bubbles. The above integral can be shown to equal $32\pi a^3/9$. Combining this result with Eq. (141) we have

$$\dot{E}_{\text{diss}} = -\Sigma_{ij}^v E_{ij} = \mu(1 + 5\phi/3)E_{ij}E_{ij}, \tag{143}$$

or, since $E_{ij}$ is arbitrary, $\Sigma_{ij}^v = -2\mu(1 + 5\phi/3)E_{ij}$.

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## Figure captions

**Fig. 1.**
$P(2a, \mu)$, the pair probability density for two bubbles in contact with the line joining their centers at an angle $\cos^{-1}\mu$ with the direction of the mean bubble flow, as a function of $\mu$ and $A$. Markers indicate simulation results and lines represent Eq. (57). Numbers refer to values of $A$. $\chi$ is the radial distribution function value at contact for the hard-sphere molecular systems.

**Fig. 2.**
$P(2a, \mu)$ as a function of $\mu$ and volume fraction $\phi$ for $A = 1.4 \pm 0.05$. Markers indicate simulation results, line represents Eq. (57). $\square$, volume fraction $\phi = 0.1$; $\times$, $\phi = 0.2$; $+$, $\phi = 0.3$.

**Fig. 3.**
Side view (a) and top view (b) of bubble positions at the end of a simulation with $A = 5.3$ and $\phi = 0.05$. The coordinates have been scaled with the bubble radius.

**Fig. 4.**
Added mass coefficient $C_a$ as a function of $A$ for different volume fractions $\phi$. Markers indicate simulation results and lines represent Eq. (71).

**Fig. 5.**
Drag coefficient $C_d$ as a function of $A$ for different volume fractions $\phi$. Markers indicate simulation results and lines represent Eq. (78).

**Fig. 6.**
The ratio of variance in the bubble velocity along its mean motion and $T$ as a function of $A$ for different volume fractions $\phi$. Simulation results. $\square$, $\phi = 0.05$; $\times$, $\phi = 0.1$; $+$, $\phi = 0.15$; $\diamond$, $\phi = 0.2$; $*$, $\phi = 0.3$.

**Fig. 7.**
$C_k$, the kinetic stress coefficient, as a function of $A$ and $\phi$. Refer Fig. 5 for legends; lines represent Eq. (96).

**Fig. 8.**
$C_{11}^k$ as a function of $A$ and $\phi$. Refer Fig. 5 for legends; lines represent Eq. (96).

**Fig. 9.**
$C_c$, the collision stress coefficient, as a function of $A$ and $\phi$. Simulation results: $\square$, $\phi = 0.05$; $*$, $\phi = 0.3$. Lines represent Eq. (100).

**Fig. 10.**
$C^c_{11}$ as a function of $A$ and $\phi$. Simulation results: $\Box$, $\phi = 0.05$; $*$, $\phi = 0.3$; lines represent curve fits (cf. Eq. (101)).

**Fig. 11.**
Hydrodynamic interaction stress tensor components normalized by $\rho T$. Markers indicate simulation results for $\phi = 0.05$; $\Box$, trace $P^M_{ii}$ of the Maxwell stress; $+$ trace $P^h_{ii}$ of the hydrodynamic stress without Maxwell stress; $\times$, normal component in the direction of mean bubble motion $P^h_{11}$ of the hydrodynamic stress without the Maxwell stress. Line through the squares is the result for the Maxwell stress obtained from Eq. (18) and Eq. (24); the other lines refer to the predictions of $P^h_{11}$ and $P^h_{ii}$ by Bulthuis *et al.* [32] (cf. their equation (47)) for the case when the dipoles of all the bubbles are equal and spatial distribution is uniform; the lower line is $P^h_{11}$, the upper line is $P^h_{ii}$.

**Fig. 12.**
Same as in Fig. 11, but for $\phi = 0.3$.

**Fig. 13.**
The trace of total bubble-phase stress as a function of $A$. $\Box$, $\phi = 0.05$; $+$, $\phi = 0.1$; $\times$, $\phi = 0.15$.

**Fig. 14.**
Values of $A$ above which the total bubble-phase stress trace becomes zero ($A_{cr}$) or the isothermal compressibility becomes zero ($A'_{cr}$). The symbols represent the values of $A_{cr}$ obtained by extrapolating the simulation results such as those given in Fig. 13 while the curves are predictions for $A_{cr}$ and $A'_{cr}$.

**Fig. 15.**
$C_I$ as a function of $A$ for different volume fractions. Markers indicate simulation results, and lines represent Eq. (106). $+$, $\phi = 0.15$; $\Diamond$, $\phi = 0.2$; $*$, $\phi = 0.3$.

**Fig. 16.**
$f_1$ as a function of $A$ and $\phi$. $\Box$, $\phi = 0.05$; $\times$, $\phi = 0.15$; $+$, $\phi = 0.3$.

**Fig. 17.**
$f_2$ as a function of $A$ and $\phi$. Refer Fig. 16 for legends.

**Fig. 18.**
$f_3$ as a function of $A$ and $\phi$. Refer Fig. 16 for legends.

**Fig. 19.**

$R^I_{diss}$ as a function of $A$ and $\phi$. Markers are simulation results, lines are Eq. (113). $\square$, $\phi = 0.05$; $\times$, $\phi = 0.15$; $+$, $\phi = 0.3$.