

# Effective equations for wave propagation in bubbly liquids

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We derive a system of effective equations for wave propagation in a bubbly liquid. Starting from a microscopic description, we obtain the effective equations by using Foldy's approximation in a nonlinear setting. We discuss in detail the range of validity of the effective equations as well as some of their properties.

## 1. Introduction

Wave propagation in a liquid containing gas bubbles is a complex phenomenon that has been studied theoretically both in the linear small-amplitude regime and in the weakly nonlinear regime, including effects of temperature, surface tension, viscosity, etc. (d'Agostino & Brennen 1983; Batchelor 1969; Drew & Cheng 1982; Drumheller & Bedford 1979; Hsieh 1982; Van Wijngaarden 1968, 1972; Wallis 1969; and additional references therein). A good deal of the theory of waves in a bubbly liquid can be deduced from a set of nonlinear differential equations that were proposed by Van Wijngaarden (1968, 1972).

These equations were written down on the basis of physical reasoning. It is not clear how they arise from the equations that describe the microscopic motion of the liquid and the gas bubbles. The purpose of this paper is to show that the equations of Van Wijngaarden can be obtained from the microscopic equations in a specific asymptotic limit that we describe in detail. From this analysis one gets a clear idea of the range of validity of Van Wijngaarden's equations.

Let us review briefly Van Wijngaarden's equations. The macroscopic state of the gas-bubble-liquid mixture is described by its density  $\rho(t, \mathbf{x})$ , pressure  $p(t, \mathbf{x})$ , velocity  $\mathbf{u}(t, \mathbf{x})$ , gas volume fraction  $\beta(t, \mathbf{x})$  and bubble radius  $R(t, \mathbf{x})$  for some time  $t > 0$  and  $\mathbf{x}$  in three-dimensional space  $\mathbb{R}^3$ . The bubble radius field  $R(t, \mathbf{x})$  is a continuum variable and specifies some average bubble radius for bubbles in the neighbourhood of a point  $\mathbf{x}$ . The equations of Van Wijngaarden (1972) are

$$\rho_t + \nabla \cdot (\rho \mathbf{u}) = 0, \quad (1.1)$$

$$\rho(\mathbf{u}_t + \mathbf{u} \cdot \nabla \mathbf{u}) + \nabla p = 0, \quad (1.2)$$

$$\rho = \rho_l(1 - \beta), \quad (1.3)$$

$$\frac{p_g \beta}{1 - \beta} = \text{constant}, \quad (1.4)$$

$$p_g R^3 = \text{constant}, \quad (1.5)$$

$$p_g - p = \rho_l \left\{ R R_{tt} + \frac{3}{2} R_t^2 \right\}. \quad (1.6)$$

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Initial conditions are given, for example, for  $\rho$ ,  $\mathbf{u}$ ,  $p$ ,  $R$  and  $R_t$ . The volume fraction  $\beta$  and the gas pressure  $p_g$  are determined by (1.4) and (1.5) once the constants on the right are prescribed. The liquid density  $\rho_l$  is related to  $p$  by an equation of state for the liquid, or is simply taken as constant if the compressibility of the liquid is negligible.

We find in §§2–4 that (1.1)–(1.6) (in the somewhat simplified form ((5.1)–(5.4) given below) can be derived by an adaptation of Foldy's method (Foldy 1945; Carstensen & Foldy 1947). The essential point in this method is to argue that the pressure and velocity fields felt by each bubble are the macroscopic ones; each bubble does not feel the local fields of the other bubbles. Obviously this requires small gas-bubble volume fraction. Let  $n$  be the number of bubbles per unit volume,  $R_0$  a typical bubble radius and  $\lambda$  a typical wavelength of a disturbance in the mixture, with  $\lambda \gg R_0$ . Then we will show that (1.1)–(1.6) are valid if  $n\lambda^2 R_0$  is of order one. Note that the volume fraction  $\frac{4}{3}\pi R_0^3 n = \frac{4}{3}\pi(R_0/\lambda)^2 (n\lambda^2 R_0)$  is then small.

It is useful to note why (1.1)–(1.6) are reasonably good for sound propagation in a bubbly liquid (Van Wijngaarden 1972). Equations (1.1) and (1.2) are the usual conservation laws for mass and momentum of the mixture. Equation (1.3) defines the macroscopic density as the liquid density  $\rho_l$  times the liquid volume fraction. The term  $\rho_g \beta$  could be added to account for the mass density of the gas, but the ratio  $\rho_g/\rho_l$  is negligibly small for typical values of  $\rho_g$  and  $\rho_l$ . Equation (1.4) is a consequence of the assumption that the mass of the gas per unit mass of the liquid is the constant  $\rho_g \beta/\rho_l(1-\beta)$ . This is valid when the gas and the liquid move with the same velocity. The isothermal equation of state in the gas (1.5) can be used to eliminate  $\rho_g$  in the mass ratio, and this gives (1.4). Equation (1.6) is Rayleigh's equation (Plesset & Prosperetti 1977; Keller & Miksis 1980; Prosperetti 1983) for radial bubble oscillations of a single bubble, with  $p$  being the pressure far away from the bubble. Note that in the macroscopic description the bubble radius is a field variable  $R(t, \mathbf{x})$ , but (1.6) involves only time derivatives. In §4 we explain how (1.6) arises from a Foldy approximation in the continuum limit. The presence of (1.6) in the system (1.1)–(1.6) indicates that typical interbubble distances must be large compared with typical bubble radii, i.e. small gas-bubble volume fraction.

The plan of this paper is as follows. In §2 we introduce the microscopic equations of motion, including a description of the bubble geometry, and a number of assumptions about the physical conditions. The appropriate scaling of the microscopic equation is introduced in §3, and under this scaling the non-dimensionalized macroscopic equations (4.1)–(4.4) are derived in §4. In dimensional form the equations are (5.1)–(5.4). The effective sound speed, the resonant frequency and an energy function for (4.1)–(4.4) are derived and analysed in §5. In §6 the addition of surface tension, viscosity and heat conduction is outlined.

## 2. The microscopic problem

We consider wave propagation through a liquid with gas bubbles dispersed in it, for example water with air bubbles. Let  $\rho$ ,  $\mathbf{u}$ ,  $p$  denote the fluid density, velocity and pressure. Whenever necessary the subscript  $l$  or  $g$  will be added to denote the property of liquid or gas respectively.

We make a number of assumptions about the physical characteristics of the fluid motion. First, since we are interested in wave propagation rather than in bulk motion, we assume that the bubble centres do not move. Secondly, we assume that the bubbles are spherical with a uniform internal pressure distribution. The first assumption is

consistent with the scaling in §3. The drift velocity of the bubble centre is of the order of the velocity far away from the bubble (as in (3.7)) and of higher order than the velocity of the bubble surface (as in (3.12)). The pressure will be uniform because the inertia of the gas is negligible. The sphericity assumption is self-consistent with the approximate solution found in §4, since in that solution the wavelength is much larger than the bubble radius and the bubble feels only a uniform pressure fluctuation. The sphericity could also be justified on the basis of surface tension.

However, as a third simplifying assumption we do not explicitly include surface tension, viscosity or heat conduction. This assumption is removed in §6. The fourth assumption is that the liquid is nearly incompressible with constant density and sound speed and that the flow is irrotational. Scaling assumptions will be introduced at the end of this section and in §3.

Suppose there are  $N$  bubbles with centres  $\mathbf{x}_1, \dots, \mathbf{x}_N$  and radii  $R_1(t), \dots, R_N(t)$  in the region  $\Omega$ . The corresponding equations of motion in the liquid region

$$\{\mathbf{x} : |\mathbf{x} - \mathbf{x}_j| > R_j \text{ for all } j\}$$

are

$$\frac{1}{\rho_l c_l^2} (\rho_l + \mathbf{u} \cdot \nabla p) + \nabla \cdot \mathbf{u} = 0, \tag{2.1}$$

$$\rho_l (\mathbf{u}_t + \mathbf{u} \cdot \nabla \mathbf{u}) + \nabla p = 0, \tag{2.2}$$

with  $\rho_l$  and  $c_l$  taken to be constant and with  $\nabla \times \mathbf{u} = 0$ . The boundary conditions on the bubble surfaces  $\{|\mathbf{x} - \mathbf{x}_j| = R_j\}$  are continuity of pressure and normal velocity, i.e.

$$\frac{\partial}{\partial t} R_j = \mathbf{u} \cdot \hat{\mathbf{n}}, \tag{2.3}$$

$$p = p_g \tag{2.4}$$

at  $|\mathbf{x} - \mathbf{x}_j| = R_j$  for  $j = 1, \dots, N$ , in which  $\hat{\mathbf{n}}$  is the normal to the bubble surface. The equation of state for the gas in the  $j$ th bubble is

$$p_g = \kappa \left( \frac{M_j}{\frac{4}{3}\pi R_j^3} \right)^\gamma, \quad j = 1, \dots, N, \tag{2.5}$$

in which  $M_j$  is the mass of the  $j$ th bubble (which is constant in time) and the parenthetical term in (2.5) is the gas density. Equations (2.1)–(2.5) should be complemented by initial conditions for  $p$ ,  $\mathbf{u}$  and  $R_j$ , as well as specification of the constants  $\rho_l$ ,  $c_l$ ,  $\mathbf{x}_j$ ,  $M_j$ ,  $\kappa$ ,  $\gamma$ .

We are interested in the limit of an infinite number of bubbles; so we make one further assumption that the bubble configuration tends to a continuum. This is formulated as follows.

For each  $N$  let the bubble-centre configuration be  $\{\mathbf{x}_1^N, \dots, \mathbf{x}_N^N\}$  and define

$$\frac{\theta^N(A)}{N} = \frac{\text{number of points } \mathbf{x}_j^N \text{ in a set } A}{N}. \tag{2.6}$$

Then there is a function  $\theta(\mathbf{x})$ , the continuum bubble-centre density, which is positive in  $\Omega$  and zero outside, such that as  $N$  tends to infinity

$$\frac{1}{N} \theta^N(A) \rightarrow \int_A \theta(\mathbf{x}) \, d\mathbf{x}, \tag{2.7}$$

for all subsets  $A$  in space. Another equivalent but more useful way of stating (2.7) is that as  $N$  tends to infinity

$$\frac{1}{N} \sum_{j=1}^N \phi(x_j^N) \rightarrow \int \phi(x) \theta(x) dx \tag{2.8}$$

for each continuous function  $\phi$ .

### 3. Scaling

Let  $\lambda$  denote the wavelength of a disturbance propagating in the bubbly liquid, let  $V$  be the volume of the region  $\Omega$  containing the bubbles and let  $R_0$  be a typical bubble radius. The dimensionless inter-bubble-centre distance  $\epsilon$ , dimensionless bubble radius  $\delta$  and gas volume fraction  $\beta$  are defined by

$$\epsilon = \frac{1}{\lambda} \left( \frac{V}{N} \right)^{\frac{1}{3}}, \tag{3.1}$$

$$\delta = \frac{R_0}{\lambda}, \tag{3.2}$$

$$\beta = \frac{1}{V} \frac{4}{3} \pi R_0^3 N = \frac{4}{3} \pi \left( \frac{\delta}{\epsilon} \right)^3. \tag{3.3}$$

An additional parameter, which enters in §4, is

$$\chi \equiv NV^{-1} \lambda^3 \delta = \frac{\delta}{\epsilon^3}. \tag{3.4}$$

The first scaling assumption is that  $\epsilon$  and  $\delta$  are very small and that  $\chi$  is of order one (relative to  $\epsilon$  and  $\delta$ ). Then (3.4) implies that

$$\delta = O(\epsilon^3); \tag{3.5}$$

this in turn implies that the volume fraction  $\beta$  is very small, i.e.  $\beta = O(\delta^2)$ .

The density will be scaled relative to the liquid density  $\rho_l$ . The pressure will be scaled relative to some reference equilibrium pressure  $p_0$ , such as the atmospheric pressure. The typical bubble mass  $M_0$  is related to  $p_0$  and  $R_0$  by

$$p_0 = \kappa \left[ \frac{M_0}{\frac{4}{3} \pi R_0^3} \right]^\gamma. \tag{3.6}$$

We scale velocities relative to a reference speed  $\bar{c}$ , which should be thought of as the effective sound speed of the bubbly liquid. Its value will be derived *a posteriori* in §5. Velocities are taken to be small compared with  $\bar{c}$ , so that convective effects are of higher order. All this leads to the following scaling in which the dimensionless variables are primed:

$$\left. \begin{aligned} x &= \lambda x', & t &= \frac{1}{f} t', & \rho &= \rho_l \rho', & R &= R_0 R' = \lambda \delta R', \\ p &= p_0 p', & \mathbf{u} &= \bar{c} \delta^2 \mathbf{u}' = R_0 f \delta \mathbf{u}', & \theta &= V^{-1} \theta'. \end{aligned} \right\} \tag{3.7}$$

Here  $f$  is a reference frequency such that  $\lambda f = \bar{c}$ . The function  $\theta'$  is the ratio of the local number density of bubbles ( $N\theta$ ) to the global number density of bubbles ( $N/V$ ), and is one for a uniform mixture.

The definitions (3.7) implicitly assert a second scaling assumption of this paper: the dimensionless variables in (3.7) are assumed to have order-one magnitude in the

continuum limit of small  $\delta$ . However, the scaling is non-uniform, since  $u'$  turns out to be of order  $\delta^{-1}$  in the neighbourhood of the bubble surfaces (cf. (3.12)). This boundary-layer behaviour cannot be anticipated by the general scaling (3.7).

Next define dimensionless parameters  $\zeta$  and  $C$  by

$$\zeta = \frac{1}{\delta^2} \frac{p_0}{\rho_\ell \bar{c}^2}, \tag{3.8}$$

$$C^2 = \left(\frac{c_\ell}{\bar{c}}\right)^2. \tag{3.9}$$

The parameters  $\zeta$  and  $C$  may be quite large in practice. However, as a further scaling assumption we require  $\zeta$  and  $C$  to be of order one relative to  $\delta$ .

Using (3.7) we obtain the following dimensionless form of the microscopic equations (2.1)–(2.6). In the region  $\{\mathbf{x}' : |\mathbf{x}' - \mathbf{x}'_j{}^N| > R'_j{}^N \text{ for all } j\}$  we have

$$\zeta C^{-2} (p'_\ell + \delta^2 \mathbf{u}' \cdot \nabla' p') + \nabla' \cdot \mathbf{u}' = 0, \tag{3.10}$$

$$\mathbf{u}'_\ell + \delta^2 \mathbf{u}' \cdot \nabla' \mathbf{u}' + \zeta \nabla' p' = 0. \tag{3.11}$$

On the  $j$ th bubble surface  $|\mathbf{x}' - \mathbf{x}'_j{}^N| = R'_j{}^N$  there are boundary conditions

$$\mathbf{u}' \cdot \mathbf{n} = \delta^{-1} \frac{\partial}{\partial t'} R'_j{}^N, \tag{3.12}$$

$$p' = F'_j{}^N(R'_j{}^N), \tag{3.13}$$

and the gas pressure and bubble configuration are described by

$$p'_g = F'_j{}^N(R'_j{}^N) = \left(\frac{M'_j{}^N}{(R'_j{}^N)^3}\right)^\gamma, \tag{3.14}$$

$$\frac{V}{\lambda^3} \frac{1}{N} (\text{number of } \mathbf{x}'_j{}^N \text{ in a set } A') \rightarrow \int_{A'} \theta'(\mathbf{x}') d\mathbf{x}'. \tag{3.15}$$

These equations are complemented by initial conditions

$$p'(0, \mathbf{x}') = \tilde{p}(\mathbf{x}'), \quad \mathbf{u}'(0, \mathbf{x}') = \tilde{\mathbf{u}}(\mathbf{x}'), \tag{3.16}$$

$$R'_j{}^N(0) = \tilde{R}(\mathbf{x}'_j{}^N), \quad \dot{R}'_j{}^N(0) = \tilde{R}'_\ell(\mathbf{x}'_j{}^N), \tag{3.17}$$

$$M'_j{}^N = \tilde{M}(\mathbf{x}'_j{}^N), \tag{3.18}$$

in which  $\nabla \times \tilde{\mathbf{u}} = 0$  and  $\tilde{R}$  and  $\tilde{M}$  are smooth functions defined in  $\Omega$ . Equation (3.18) and  $\nabla \times \mathbf{u}' = 0$  hold for all time. The initial data  $\tilde{p}$ ,  $\tilde{\mathbf{u}}$  are for the ambient fluid away from the bubbles. The actual spatial dependence of the initial data will be assumed to be consistent with the expansion described in §4. In particular initially  $u'$  must satisfy (3.12) with radial velocity given by (3.17). Following (2.8), (3.15) may be written as

$$\frac{1}{N} \sum_{j=1}^N \phi(\mathbf{x}'_j{}^N) \rightarrow \frac{\lambda^3}{V} \int \phi(\mathbf{x}) \theta'(\mathbf{x}') d\mathbf{x}' \tag{3.19}$$

for any continuous  $\phi$ .

Before continuing, we make several remarks about the parameter  $\zeta$ . Using the expressions (5.9) and (5.11) for the effective sound speed  $\bar{c}$  and resonant frequency  $\omega_0$ , which will be derived in §5, we find that

$$\zeta = \frac{4\pi^2}{3\gamma} \left(\frac{\omega_0}{\omega}\right)^2, \tag{3.20}$$

$$C^2 = \left(1 - \frac{4\pi}{3\gamma} \frac{\delta}{\epsilon^3} \zeta^{-1}\right)^{-1} = \left[1 - \frac{\delta}{\pi\epsilon^3} \left(\frac{\omega}{\omega_0}\right)^2\right]^{-1}. \tag{3.21}$$

Thus  $\zeta$  is essentially the square of the ratio of the resonant frequency to the reference frequency. The relations in (3.21) show that upper limits on  $\zeta^{-1}$  or  $\omega/\omega_0$  are implicit in our scaling assumptions.

The assumption that  $\zeta$  be of order one relative to  $\delta$  may be understood in two different ways. First it can be thought of as a relation

$$\frac{\rho_g}{\rho_l} = O(\delta^2) \tag{3.22}$$

between the geometric parameter  $\delta$  and the physical parameters  $\rho_g$  and  $\rho_l$ . To derive (3.22), note that  $\rho_g \bar{c}^2 \approx p_0$ , so that

$$\zeta = \frac{1}{\delta^2} \frac{p_0}{\rho_l \bar{c}^2} \approx \frac{\rho_g}{\delta^2 \rho_l}.$$

Alternatively the order-one size of  $\zeta$  could be a statement about the size of the pressure variations in the liquid. If these variations are of size  $\Delta p_0$  rather than  $p_0$ , then we should change the pressure scaling in (3.7) to  $p = p_0 + \Delta p_0 p'$ . The dimensionless parameter  $\zeta$  would then be replaced by  $\xi = (1/\delta^2) \Delta p_0 / \rho_l \bar{c}^2$ . If  $\rho_l$  and  $\bar{c}$  are fixed, this is the statement that

$$\Delta p_0 = O(\delta^2). \tag{3.23}$$

On the other hand, after taking the limit  $\delta \rightarrow 0$  with  $\zeta$  fixed, as will be done in §4, we may consider  $\zeta$  to be large. The effective equations (4.1)–(4.4) for average quantities  $\bar{p}$ ,  $\bar{u}$ ,  $R$  imply that, for  $\zeta$  large,  $p_g(R) \approx \bar{p}$ ,  $\bar{u}$  is size  $\zeta$ , and  $\bar{p}$ ,  $\bar{u}$  obey the acoustic equations for the liquid alone. Thus the effective equations are uniformly valid in this limit of pure liquid.

#### 4. Derivation of effective equations

The main result of this paper is a systematic derivation of effective equations ((4.1)–(4.7) below) from the scaled equations (3.10)–(3.18), in the limit  $\delta$  going to zero. In this section primes are dropped from (3.10)–(3.18). We shall show formally the following proposition.

Suppose that the  $u$ ,  $p$  and  $R_j^N$  satisfy (3.10)–(3.18). Let  $\delta \rightarrow 0$  and  $N \rightarrow \infty$ , with  $\chi$ ,  $\zeta$  and  $C$  held constant. Then  $u$  and  $p$  converge to  $\bar{u}$  and  $\bar{p}$ , and  $R_j^N$  converges to the continuum radius field  $R$  (in the sense that  $R_j^N - R(x_j^N) \rightarrow 0$  for all  $j$ ). Furthermore, the limits  $\bar{u}$ ,  $\bar{p}$  and  $R$  satisfy equations

$$\zeta C^{-2} \bar{p}_t + \nabla \cdot \bar{u} - (\frac{4}{3} \pi R^3 \theta \chi)_t = 0, \tag{4.1}$$

$$\bar{u}_t + \zeta \nabla \bar{p} = 0, \tag{4.2}$$

$$R R_{tt} + \frac{3}{2} R_t^2 = \zeta (F(R) - \bar{p}), \tag{4.3}$$

with the equation of state

$$F(R) = \left( \frac{M}{R^3} \right)^\gamma \tag{4.4}$$

and initial conditions

$$\bar{p}(0, \mathbf{x}) = \tilde{p}(\mathbf{x}), \tag{4.5}$$

$$\bar{u}(0, \mathbf{x}) = \tilde{u}(\mathbf{x}), \tag{4.6}$$

$$R(0, \mathbf{x}) = \tilde{R}(\mathbf{x}), \quad R_t(0, \mathbf{x}) = \tilde{R}_t(\mathbf{x}), \tag{4.7}$$

and with  $\theta$  and  $M = \tilde{M}$  defined by (3.15) and (3.18) (with primes dropped).

In this section the limit will be formally derived by analysing (3.10)–(3.18).

First we rewrite the equations using the velocity potential  $\phi$  satisfying

$$\mathbf{u} = \nabla\phi. \tag{4.8}$$

Equations (3.10) and (3.11) become

$$\zeta p + \phi_t + \frac{1}{2}\delta^2(\nabla\phi)^2 = 0, \tag{4.9}$$

$$\phi_{tt} - C^2 \Delta\phi + 2\delta^2 \nabla\phi \cdot \nabla\phi_t + \delta^4(\nabla\phi \nabla\phi) : (\nabla\nabla\phi) = 0 \tag{4.10}$$

for  $|\mathbf{x} - \mathbf{x}_j^N| > \delta R_j^N$  for all  $j$ . The boundary conditions are

$$\frac{\partial\phi}{\partial n} = \frac{1}{\delta} \frac{\partial}{\partial t} R_j^N, \tag{4.11}$$

$$\phi_t + \frac{1}{2}\delta^2(\nabla\phi)^2 = -\zeta F_j^N(R_j^N) \tag{4.12}$$

on  $|\mathbf{x} - \mathbf{x}_j^N| = \delta R_j^N(t)$ . The initial conditions are

$$\phi(0, \mathbf{x}) = \check{\phi}(\mathbf{x}), \quad \text{with} \quad \nabla\check{\phi} = \check{\mathbf{u}}(\mathbf{x}), \tag{4.13}$$

$$\phi_t(0, \mathbf{x}) = -\zeta\check{p}(\mathbf{x}) - \frac{1}{2}\delta^2(\nabla\check{\phi})^2. \tag{4.14}$$

Let  $G(t, \mathbf{x})$  be the Green function for the wave operator, i.e.

$$G_{tt} - C^2 \Delta G = \delta(t) \delta(\mathbf{x}) \quad (t > 0, \mathbf{x} \in \mathbb{R}^3), \tag{4.15}$$

$$G(t, \mathbf{x}) \equiv 0 \quad (t < 0).$$

We use  $\delta(\cdot)$  to denote the delta function only in (4.15). Let the  $j$ th gas bubble be denoted by

$$H_j^N(t) = \{\mathbf{x} \in \mathbb{R}^3 \text{ such that } |\mathbf{x} - \mathbf{x}_j^N| \leq \delta R_j^N\} \tag{4.16}$$

and let the liquid region be denoted by

$$\Omega^N(t) = \mathbb{R}^3 - \bigcup_{j=1}^N H_j^N(t). \tag{4.17}$$

Using Green's theorem, we may write (4.10)–(4.14) in integral equation form. For  $t > 0$  and  $\mathbf{x} \in \Omega^N(t)$ , we have

$$\begin{aligned} \phi_t(t, \mathbf{x}) = & -\delta^2 \int_0^t d\tau \int_{\Omega^N(\tau)} d\mathbf{y} G_t(t-\tau, \mathbf{x}-\mathbf{y}) [2\nabla\phi(\tau, \mathbf{y}) \cdot \nabla\phi_\tau(\tau, \mathbf{y}) + \delta^2(\nabla\phi \nabla\phi) : (\nabla\nabla\phi)] \\ & - C^2 \sum_{j=1}^N \int_0^t d\tau \int_{\partial H_j^N(\tau)} ds [G_t(t-\tau, \mathbf{x}-\mathbf{y}) \hat{\mathbf{n}} \cdot \nabla\phi(\tau, \mathbf{y}) \\ & \quad + \phi_\tau(\tau, \mathbf{y}) \hat{\mathbf{n}} \cdot \nabla G(t-\tau, \mathbf{x}-\mathbf{y})] \\ & + \int_{\Omega^N(0)} d\mathbf{y} [G_t(t, \mathbf{x}-\mathbf{y}) \phi_t(0, \mathbf{y}) + C^2 \nabla G(t, \mathbf{x}-\mathbf{y}) \cdot \nabla\phi(0, \mathbf{y})]. \end{aligned} \tag{4.18}$$

Here  $\partial H_j^N$  denotes the boundary of  $H_j^N$  on which the boundary conditions (4.11)–(4.12) hold.

We may write (4.18) as

$$\phi_t = \mathbf{N}(\phi) + \sum_{j=1}^N \mathbf{F}_j^N(\phi) + \mathbf{L}\phi_0. \tag{4.19}$$

Here  $\mathbf{L}$  is the linear operator corresponding to the last term in (4.18). It maps the field  $\phi$  at time  $t = 0$ , which is  $\phi_0$ , to the one at time  $t$  with  $\phi$  evolving according to

the wave equation  $\phi_{tt} - C^2 \Delta \phi = 0$  without boundary conditions.  $N$  is the nonlinear operator corresponding to the first integral on the right of (4.18) and  $F_j^N$  is the nonlinear operator associated with the sum over  $j$  on the right of (4.18). The operators  $F_j^N$  are nonlinear because the radii  $R_j^N(t)$  of the spheres  $H_j^N(t)$  depend on the solution via (4.11) and (4.12).

In Foldy's method, expressed somewhat differently here, (4.19) is solved approximately as follows. For  $\delta$  small and  $N$  large with  $\chi = NV^{-1}\lambda^3\delta = O(1)$ , we shall show that the field  $\phi$  will tend to a limit  $\bar{\phi}$ , the continuum limit. It is then sufficient to calculate  $F_j^N(\bar{\phi})$  for each sphere separately and to evaluate the limit of the sum over  $N$  in (4.19). This is what is meant by saying that each bubble feels only the average pressure and velocity fields around it and not the local fields of the other bubbles.

We proceed to implement this. First it is clear that the nonlinear term  $N(\phi)$  will make no contribution in the limit because it is formally of order  $\delta^2$ . Now consider a single bubble which may be centred at the origin and let  $\mathbf{z} = \mathbf{x}/\delta$ , so that the sphere has radius  $R(t)$  on the  $\mathbf{z}$ -scale. To leading order in  $\delta$  the potential  $\phi$  outside the bubble satisfies

$$\Delta_{\mathbf{z}} \phi = 0 \quad (|\mathbf{z}| > R), \tag{4.20}$$

along with the boundary conditions

$$\hat{\mathbf{n}} \cdot \nabla_{\mathbf{z}} \phi = \frac{\partial R}{\partial t} \tag{4.21}$$

and

$$\frac{1}{2}(\nabla_{\mathbf{z}} \phi)^2 + \phi_t = -\zeta F(R) \quad \text{on } |\mathbf{z}| = R. \tag{4.22}$$

Far away from the bubble, for  $|\mathbf{z}|$  large, we expect that  $\phi$  behaves like  $\bar{\phi}$ , the continuum potential field. Clearly the solution is

$$\phi = \frac{-R_t R^2}{|\mathbf{z}|} + \bar{\phi}, \tag{4.23}$$

which satisfies (4.20), (4.21) and the large- $|\mathbf{z}|$  condition. Condition (4.22) leads to Rayleigh's equation (4.3).

With the  $\phi$  determined locally about a bubble in the above way, we look at a typical integrand in  $F_j^N$ ,

$$I_j^N(\mathbf{x}, t, \tau) = \int dS [G_t(t-\tau, \mathbf{x}-\mathbf{y}) \frac{\partial \phi(\tau, \mathbf{y})}{\partial n} + \phi_\tau(\tau, \mathbf{y}) \hat{\mathbf{n}} \cdot \nabla G(t-\tau, \mathbf{x}-\mathbf{y})], \tag{4.24}$$

where the integral is over the surface of the  $j$ th bubble, and  $\mathbf{x}$  is away from the other bubbles. Note that  $F_j^N = \int I_j^N d\tau$ . We may replace  $\partial \phi / \partial n$  in (4.24) by  $\delta^{-1} \partial R_j^N / \partial t$  in view of (4.11). We may also evaluate  $G$  at  $\mathbf{y} = \mathbf{x}_j^N$ , making an error of order  $\delta$ . Since  $\phi_\tau$  by (4.22) is of order one, we see that

$$I_j^N(\mathbf{x}, t, \tau) = 4\pi \delta (R_j^N(\tau))^2 G_t(t-\tau, \mathbf{x}-\mathbf{x}_j^N) \frac{\partial}{\partial t} R_j^N(\tau) \tag{4.25}$$

plus terms of order  $\delta^2$ .

Let  $R(t, \mathbf{x})$  satisfy (4.3). Then  $R_j^N(t)$  is close to the continuum field  $R(t, \mathbf{x}_j^N)$  in the sense that

$$\frac{1}{N} \sum_{j=1}^N (R_j^N(t) - R(t, \mathbf{x}_j^N))^2 \rightarrow 0 \quad \text{as } N \rightarrow \infty. \tag{4.26}$$



The precise nature of this convergence will not be checked in detail. Using (4.26) in (4.25) allows us to replace  $R_j^N(\tau)$  by  $R(\tau, \mathbf{x}_j^N)$ . If we insert this expression for  $I_j^N(\mathbf{x}, t, \tau)$  in the sum in (4.18) and ignore terms that are of order  $\delta^2$ , we obtain

$$\begin{aligned} \bar{\phi}_t(t, \mathbf{x}) = & -(4\pi N\delta) C^2 \lim_{N \rightarrow \infty} \frac{1}{N} \sum_{j=1}^N \int_0^t d\tau G_t(t-\tau, \mathbf{x}-\mathbf{x}_j^N) R^2(\tau, \mathbf{x}_j^N) R_t(\tau, \mathbf{x}_j^N) \\ & + \int_{\mathbf{R}^3} d\mathbf{y} [G_t(t, \mathbf{x}-\mathbf{y}) (-\zeta\tilde{p}(\mathbf{y})) + C^2 \nabla G(t, \mathbf{x}-\mathbf{y}) \cdot \tilde{\mathbf{u}}(\mathbf{y})]. \end{aligned} \quad (4.27)$$

Using (3.4) and (3.19) on the right-hand side of (4.27), we arrive at the following integral equation for the continuum field  $\bar{\phi}$ :

$$\begin{aligned} \bar{\phi}_t(t, \mathbf{x}) = & -\chi C^2 \int_0^t d\tau \int_{\mathbf{R}^3} d\mathbf{y} G_t(t-\tau, \mathbf{x}-\mathbf{y}) (\frac{4}{3}\pi R^3(\tau, \mathbf{y}) \theta(\mathbf{y}))_t \\ & + \int_{\mathbf{R}^3} d\mathbf{y} [G_t(t, \mathbf{x}-\mathbf{y}) (-\zeta\tilde{p}(\mathbf{y})) + C^2 \nabla G(t, \mathbf{x}-\mathbf{y}) \cdot \tilde{\mathbf{u}}(\mathbf{y})]. \end{aligned} \quad (4.28)$$

The integral equation (4.28) is equivalent to

$$C^{-2}\bar{\phi}_{tt} - \nabla^2\bar{\phi} + (\frac{4}{3}\pi R^3\theta\chi)_t = 0, \quad (4.29)$$

$$\nabla\bar{\phi}(0, \mathbf{x}) = \tilde{\mathbf{u}}(\mathbf{x}), \quad \bar{\phi}_t(0, \mathbf{x}) = -\zeta\tilde{p}(\mathbf{x}), \quad (4.30)$$

which is equivalent to (4.1), (4.2), (4.5) and (4.6), with  $\nabla\bar{\phi} = \mathbf{u}$  and  $R$  solving (4.3), (4.4) and (4.7). This finishes the demonstration of the main proposition.

### 5. Properties of the effective equations

The system of effective equations (4.1)–(4.4) has in dimensional variables the form

$$\frac{1}{\rho_\ell c_\ell^2} p_t + \nabla \cdot \mathbf{u} - (\frac{4}{3}\pi R^3 n)_t = 0, \quad (5.1)$$

$$\rho_\ell \mathbf{u}_t + \nabla p = 0, \quad (5.2)$$

$$p_g = \kappa \left( \frac{M}{\frac{4}{3}\pi R^3} \right)^\gamma, \quad (5.3)$$

$$RR_{tt} + \frac{3}{2}R_t^2 = \frac{1}{\rho_\ell} (p_g - p). \quad (5.4)$$

Here we assume that  $\rho_\ell, c_\ell, M, \kappa$  and  $n = N\theta$  (the number of bubble centres per unit volume) are constants. Initial values are given for  $p, \mathbf{u}, R$  and  $R_t$ .

In this section we shall compare (5.1)–(5.4) with Van Wijngaarden's equations, compute the effective sound speed and resonant frequency, and derive an energy function for (5.1)–(5.4). The expression for sound speed and resonant frequency are well known; the energy function is new.

Van Wijngaarden's equations (1.1)–(1.6) reduce to (5.1)–(5.4) if (i) the volume fraction is assumed to be small, (ii) the velocity fluctuations are assumed to be small and (iii) the liquid is assumed to be nearly incompressible, which is the familiar approximation used in acoustics. The last condition is implemented by replacing  $d\rho_\ell/dt$  by  $c_\ell^{-2} dp/dt$  and then treating  $\rho_\ell$  and the liquid sound speed  $c_\ell$  as constants. In addition we replace the isothermal equation of state (1.5) by the more general equation (5.3) and identify the volume fraction as  $\beta = \frac{4}{3}\pi R^3 n$ .

To obtain the effective sound speed and resonant frequency, we linearize (5.1)–(5.4) about  $\mathbf{u} = 0$ ,  $p = p_0$  and  $R = R_0$ , such that

$$p_0 = \kappa \left( \frac{M_0}{\frac{4}{3}\pi R_0^3} \right)^\gamma, \quad (5.5)$$

to obtain 
$$\frac{1}{\rho_\ell c_\ell^2} p_t + \nabla \cdot \mathbf{u} - 4\pi R_0^2 n R_t = 0, \quad (5.6)$$

$$\rho_\ell \mathbf{u}_t + \nabla p = 0, \quad (5.7)$$

$$R_{tt} + \omega_0^2 R = -\frac{1}{R_0 \rho_\ell} p. \quad (5.8)$$

Here 
$$\omega_0 = \left( \frac{3\gamma p_0}{R_0^2 \rho_\ell} \right)^{\frac{1}{2}} \quad (5.9)$$

is the natural frequency of radial bubble oscillations. The dispersion relation for (5.6)–(5.8) is

$$\frac{1}{\bar{c}^2} = \left( \frac{k}{\omega} \right)^2 = \frac{1}{c_\ell^2} + \frac{3\beta_0}{R_0^2(\omega_0^2 - \omega^2)}, \quad (5.10)$$

with  $\beta_0 = \frac{4}{3}\rho R_0^3 n$ . In (5.10)  $\bar{c}$  is the effective phase velocity of infinitesimal disturbances.

In our derivation of the effective equations (5.1)–(5.4) there is no condition that the disturbance frequency  $\omega$  be less than the resonance frequency  $\omega_0$ . However, the derivation is valid only if the solution of (5.1)–(5.4) is bounded. Note that if dissipative effects were included, as in §6, the solution would be bounded through resonance.

When  $\omega$  is small compared with  $\omega_0$ , i.e. the low-frequency case, (5.10) simplifies to

$$\frac{1}{\bar{c}^2} = \frac{1}{c_\ell^2} + \frac{\beta_0 \rho_\ell}{\gamma p_0}, \quad (5.11)$$

which is the well-known formula for the effective sound speed  $\bar{c}$  (Van Wijngaarden 1972).

With the values  $p_0 = 10^6$  dyn/cm<sup>2</sup> (atmospheric pressure),  $\rho_\ell = 1$  g/cm<sup>3</sup> for water,  $R_0 = 10^{-1}$  cm and  $\gamma = 1$ , the resonant bubble frequency  $\omega_0 = 1.7 \times 10^4$  rad/s (2750 Hz). If  $\omega$  is less than 1000 Hz, say, and  $\beta_0$  is of order  $10^{-2}$ , then  $c_\ell^{-2}$  is negligible ( $c_\ell = 1400$  m/s) in (5.11) and  $\bar{c}$  is about 100 m/s. This is even smaller than the sound speed in the gas ( $c_g \sim 330$  m/s). Only for very small bubble volume fraction, say of order 0.01% ( $\beta_0 = 10^{-4}$ ) or smaller, does  $\bar{c}$  of (5.11) differ from that of the simpler formula

$$\bar{c}^2 = \frac{\gamma p_0}{\beta_0 \rho_\ell}, \quad (5.12)$$

at low frequencies.

This striking behaviour of the effective sound speed of the mixture as a function of bubble volume fraction has been confirmed experimentally by Silberman (1957), and makes bubbly liquids an interesting medium. For example, layers of bubbly liquids can be used to reflect or isolate sound fields (Domenico 1982). This behaviour is also interesting mathematically, because the bubbly liquid is a two-component composite medium with singular behaviour at small volume fraction: a very small volume of bubbles changes the effective properties of the mixture drastically.

The energy density for the system (5.1)–(5.4) is given by

$$E = \frac{1}{2}\rho_l u^2 + \frac{1}{2} \frac{1}{\rho_l c_l^2} p^2 + 2\pi n \rho_l R^3 R_t^2 + \frac{n}{\gamma - 1} p_g \left(\frac{4}{3}\pi R^3\right). \tag{5.13}$$

With this definition, we have the energy-conservation equation

$$\frac{\partial}{\partial t} E + \nabla \cdot (p\mathbf{u}) = 0. \tag{5.14}$$

For suitable conditions at infinity, this equation implies the conservation of energy:

$$\frac{d}{dt} \int E(t, \mathbf{x}) d\mathbf{x} = 0. \tag{5.15}$$

The various terms in the energy density (5.3) have the following physical interpretation. The first two terms  $\frac{1}{2}\rho_l u^2 + \frac{1}{2}(\rho_l c_l^2)^{-1} p^2$  are the energy density for a linearized, isentropic, compressible flow without bubbles. The last term in (5.13),  $n(\gamma - 1)^{-1} p_g \frac{4}{3}\pi R^3$ , is the energy of the gas bubbles with equation of state (5.3). This is seen by noting that

$$\frac{d}{dt} \frac{n}{\gamma - 1} p_g \left(\frac{4}{3}\pi R^3\right) = -p_g \frac{d}{dt} \left(n \frac{4}{3}\pi R^3\right), \tag{5.16}$$

which has the form  $dE = -p_g dV$ , with  $V = n \frac{4}{3}\pi R^3$  being the total gas-bubble volume.

The term  $n 2\pi \rho_l R^3 R_t^2$  is the number of bubbles per unit volume times the kinetic energy in an incompressible flow outside a radially oscillating sphere of radius  $R(t)$ . The flow outside the sphere has the form

$$v(t, \mathbf{x}) = \frac{R^2 R_t}{|\mathbf{x}|^2} \quad (|\mathbf{x}| > R(t)), \tag{5.17}$$

so the kinetic energy of the fluid induced by the radial oscillations of the bubble is

$$\begin{aligned} \int_{|\mathbf{x}| > R} \frac{1}{2}\rho_l v^2 d\mathbf{x} &= 4\pi \int_R^\infty \frac{1}{2}\rho_l (R^2 R_t)^2 r^{-2} dr \\ &= 2\pi \rho_l R^3 R_t^2. \end{aligned} \tag{5.18}$$

Equations (5.1)–(5.4) can be put in variational form by introducing the potential  $\phi$  defined by

$$\mathbf{u} = \nabla\phi, \quad \phi_t = -\frac{1}{\rho_l} p. \tag{5.19}$$

The Lagrangian density is given by

$$L(\phi, R, \phi_t, R_t) = \frac{1}{2}\rho_l \left( (\nabla\phi)^2 - \frac{1}{c_l^2} (\phi_t)^2 \right) - n \rho_l 2\pi R^3 R_t^2 - n \rho_l \frac{4}{3}\pi R^3 \phi_t + \frac{n}{\gamma - 1} \frac{\kappa M^\gamma}{\left(\frac{4}{3}\pi R^3\right)^{\gamma - 1}}. \tag{5.20}$$

It is also interesting to note that (5.1)–(5.4) is a Hamiltonian system.

Since the energy  $E$  is strictly positive and  $\int E d\mathbf{x}$  is conserved, finite-energy solutions of (5.1)–(5.4) are stable. In the case of plane waves, when all quantities in (5.1)–(5.4) depend on one space coordinate  $x_1$  and the velocity is  $\mathbf{u} = (u_1, 0, 0)$ , one can show much more. If, at  $t = 0$ ,  $p$ ,  $u_1$  and  $R$  are smooth (with continuous first derivatives) and  $R$  is positive for all  $x_1$ , then there is a smooth bounded solution of (5.1)–(5.4) with  $R > 0$  for all  $t$ . This shows that there is no bubble cavitation or shock-wave formation.

The proof of this result is rather technical, and is given elsewhere (Caflisch 1984). It relies on a representation of (5.1)–(5.4) as a nonlinear system in characteristic form. If

$$f = (\rho_l c_l^2)^{-1} p + c_l^{-1} u_1, \quad g = (\rho_l c_l^2)^{-1} p - c_l^{-1} u_1, \quad D = RR_t \tag{5.21}$$

then (5.1)–(5.4) become

$$\left. \begin{aligned} \frac{\partial}{\partial t} f + c_l \frac{\partial}{\partial x_1} f &= n 4\pi R D, \\ \frac{\partial}{\partial t} g - c_l \frac{\partial}{\partial x_1} g &= n 4\pi R D, \\ \frac{\partial}{\partial t} R &= R^{-1} D, \\ \frac{\partial}{\partial t} D &= -\frac{1}{2} R^{-2} D^2 + \frac{\kappa}{\rho_l} \left(\frac{M}{\frac{2}{3}\pi}\right)^\gamma R^{-3\gamma} - \frac{1}{2} \rho_l c_l^2 (f + g). \end{aligned} \right\} \tag{5.22}$$

Besides its usefulness for mathematical analysis, the form (5.22) is particularly amenable to numerical solution.

### 6. Effects of surface tension, viscosity and heat conduction

In §5 we neglected these effects in order to focus attention on the basic aspects of the continuum limit in the Foldy approximation of §4. The result of §4 can be extended easily to handle effects of surface tension, viscosity and heat conduction. These extensions are summarized in this section. The effect of radiation damping due to the compressibility of the liquid, which is a higher-order dissipative mechanism in the present scaling, has not been included.

The inclusion of surface-tension, viscosity and heat-conduction effects in the Foldy approximation of §5 is easy, because the local fields around each bubble do not interact directly. Moreover, these effects are most important in a small neighbourhood of each bubble only. Therefore including these effects merely changes the Rayleigh–Plesset equation (1.6) or (4.3) in the same way as Plesset & Prosperetti (1977).

Let  $\sigma$  denote the coefficient of surface tension and let  $W$  be the dimensionless Weber number

$$W = \frac{p_0 R_0}{\sigma}. \tag{6.1}$$

The Weber number  $W$  for air bubbles in water at atmospheric pressure is of order one only for bubbles of micron size and smaller. Let  $\nu_l$  denote the kinematic viscosity of the liquid, and  $Re$  the Reynolds number based on the bubble radius, i.e.

$$Re = \frac{\delta \bar{c} R_0}{\nu_l} = f \frac{R_0^2}{\nu_l}, \tag{6.2}$$

in which  $\delta^2 \bar{c}$  is the reference velocity.

The inclusion of surface tension and viscosity in the Foldy approximation leads again to (4.1)–(4.4) (or (5.1)–(5.4)), but with two additional terms in (4.4), so that it becomes

$$RR_{tt} + \frac{3}{2} R_t^2 + \frac{4R_t}{Re R} + \frac{2\zeta}{WR} = \zeta [p_g(R) - \bar{p}]. \tag{6.3}$$

In dimensional variables (6.3) is

$$\rho_l(RR_{tt} + \frac{3}{2}R_t^2) + \frac{4\mu_l R_t}{R} + \frac{2\sigma}{R} = p_g - \bar{p}. \quad (6.4)$$

Heat-conduction effects can be included by using the results of Miksis & Ting (1984). Their results hold when the ratio of thermal-diffusion length to the radius is small, i.e.

$$\sigma_T = \frac{l_T}{R_0} = \left( \frac{D_g}{\omega R_0^2} \right)^{\frac{1}{2}} \ll 1. \quad (6.5)$$

Here  $D_g$  is the reference thermal diffusivity of the gas. The principal effect is confined in a thin thermal layer inside the bubble.

With  $\sigma_T$  as the small expansion parameter, Miksis & Ting constructed an approximate solution and arrived at an integral relationship for the bubble radius and the pressure in the gas phase. The equation in dimensionless form is

$$[p_g(t)]^{1/\gamma} [R(t)]^3 = 1 + \frac{3\sigma_T}{2\pi(\pi)^{\frac{1}{2}}} \int_0^{s^{\frac{1}{2}}} \{[\tilde{p}_g(s - \xi^2)]^{(1-\gamma)/\gamma} - 1\} d\xi, \quad (6.6)$$

where  $s = 16\pi^2 \int_0^t [R(t)]^4 p_g(t) dt$  and  $\tilde{p}_g(s) = p_g(t)$ . Coupling (6.6) with the Rayleigh-Plesset equation (6.3) gives us a closed system that relates  $R(t)$  and  $p_g(t)$  for a single bubble in an unbounded liquid region with a far-field pressure  $\bar{p}(t)$ .

## 7. Conclusions

We have derived a system of effective equations for wave propagation in a bubbly liquid. If surface tension and dissipative effects are neglected the equations are (5.1)–(5.4) (or (4.1)–(4.4) in dimensionless form). They are nearly identical with the well-known system (1.1)–(1.6), but differ from (1.1)–(1.6) by the omission of convective terms and terms of size  $\beta^2$ . These omitted terms are negligible in the regime we have considered.

If surface tension, viscosity and heat conduction are included, the equations are (5.1)–(5.3), (6.4) and (6.6). The heat-conduction term in these equations differs from that of previous studies (e.g. Prosperetti 1977). It comes from the analysis by Miksis & Ting (1984) of heat-conduction effects for the oscillation of a single bubble as described in §6.

Our main contribution in this paper is to derive these equations systematically and thereby to find their range of validity. We have found this system to be an accurate approximation of the microscopic equations for waves in a bubbly liquid if  $\epsilon \ll 1$ ,  $\delta \ll 1$ ,  $\chi = \delta/\epsilon^3 = O(1)$  and if  $\zeta$  and  $C$  are independent of  $\delta$  and  $\epsilon$ , in which  $\epsilon = \lambda^{-1}(V/N)^{\frac{1}{3}}$  is the scaled interbubble distance,  $\delta = R_0/\lambda$  is the scaled bubble radius,  $N$  is total bubble number in volume  $V$ , and  $\zeta$  and  $C$  are defined by (3.8) and (3.9). In addition we have assumed that the fluid velocity is small enough to neglect convection.

In addition we have derived in §4 a new conserved-energy function and the well-known sound speed and resonant frequency for the effective equations.

Since  $\delta/\epsilon^3 = \chi = O(1)$ , the volume fraction  $\beta = \frac{4}{3}\pi(\delta/\epsilon)^3$  is very small. At larger volume fractions the Foldy method is not applicable since the local pressure fields around different bubbles interact directly. In a subsequent paper (Caffisch *et al.* 1984) we shall present a linear analysis of wave propagation in liquids with finite bubble

volume fraction, as well as a theory which is valid uniformly in bubble volume fraction. We show that the bubble deformations are predominantly non-spherical and calculate the effective sound speed.

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### Appendix. Geometric assumptions and the Foldy approximation

The analysis of §4 shows how the passage from (3.10)–(3.18) to (4.1)–(4.7) is effected via the Foldy approximation. Of course, it is clear that there are many technical steps to be filled out. One point however is essential in the method and must be faced. How do we know that in this asymptotic limit ( $\delta \rightarrow 0$ ,  $\epsilon \rightarrow 0$ ,  $\delta \sim \epsilon^3$ ) bubbles do not feel the local potential fields of other bubbles? From (4.23) we see that the local field of each bubble has the form of a monopole

$$\delta \frac{(R_j^N)^2 \partial R_j^N / \partial t}{|x - x_j^N|}. \quad (\text{A } 1)$$

The field that the  $j$ th bubble feels due to the local fields of the other bubbles is, to principal order,

$$\delta \sum_{i \neq j} \frac{R^2(t, x_j^N) \partial R(t, x_j^N) / \partial t}{|x_j^N - x_i^N|}, \quad (\text{A } 2)$$

where we replace  $R_j^N(t)$  by  $R(t, x_j^N)$  as in (4.26). We would like to have some control on (A 2) uniformly on  $i = 1, 2, \dots, N$ . Suppose we assume that

$$\max_{i=1, \dots, N} \left| \frac{1}{N} \sum_{\substack{j=1 \\ j \neq i}}^N \frac{\psi(x_j^N)}{|x_i^N - x_j^N|} - \int \frac{\psi(y) \theta(y) dy}{|x_j^N - y|} \right| \rightarrow 0 \quad (\text{A } 3)$$

as  $N \rightarrow \infty$  for every function  $\psi(y)$  that is smooth. Note that we have used the scaling  $N\delta \sim 1$  and  $\chi \sim 1$  here. Note also that (A 3) is not implied by (2.7) or (2.8); it is an additional assumption. Now with (A 3) one can show in similar but simpler problems (Hrushlov & Marchenko 1974; Papanicolaou & Varadhan 1980) that indeed the Foldy approximation is valid. We expect that the same is true for the present problem.

Condition (A 3) (or the weaker one given by (1.8) in Papanicolaou & Varadhan 1980) is needed because, although the bubble centres may tend to a continuum configuration with a smooth density, some bubble centres may come too close to each other, and this would interfere with the Foldy approximation. With (A 3) we gain some control on how close bubbles can come to each other without interfering with the Foldy approximation. Of course there are simpler conditions one can impose, such as

$$\min_{i \neq j} |x_i^N - x_j^N| \geq \alpha N^{-\frac{1}{3}}, \quad (\text{A } 4)$$

for some  $\alpha > 0$  independent of  $N$ . This is valid, for example, in a periodic configuration of bubble centres. Configurations of independent identically distributed  $\{x_j^N\}$  cannot satisfy (A 4), but they can satisfy (A 3) with probability arbitrarily close to one.

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