

## Thermal attenuation and dispersion of sound in a periodic emulsion

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We investigate the attenuation and dispersion of sound waves in suspensions and emulsions caused by the thermal-transport process. They combine to constitute the effective compressibility of the system. We begin with an attempt to justify the Isakovich formula for calculating the effective compressibility. The formula is then rewritten in terms of the interfacial heat flux. Isakovich's analysis is simply an independent-particle approximation. It is the purpose of this paper to consider the effect of interparticle interactions. The effective compressibility is calculated for an array of spherical particles or droplets centered at the points of a periodic lattice, immersed in a fluid of different species. Ewald's method of fast-convergent lattice sums in electrostatics is extended to a technique for the heat-conduction problem in a periodic emulsion. The computation for cubic lattices reveals that the interparticle interactions act to reduce, in the lower-frequency range, both the attenuation coefficient and the departure of the sound velocity from its high-frequency limit. The striking feature is that a drastic change in attenuation occurs when the thermal conductivity of the particle is substantially larger than that of the ambient fluid.

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### I. INTRODUCTION

Sound waves propagating through a fluid medium entail attenuation due to the existence of viscous dissipation and thermal conduction. When the systems are mixtures of two or more materials, the increased attenuation and dispersion of sound are observed. These phenomena are of great importance in connection with the acoustic-wave propagation through fog, ocean or ocean bottom, and ultrasonic measurements of biological membranes and biomolecules, etc.

A theoretical treatment of the viscous-dissipation loss of sound in a suspension dates back to Sewell [1], who placed the restriction that the suspended particle is rigid and immovable. This restriction was relaxed by Lamb [2] to allow for the free motion of a particle. Epstein [3] extended the theory to include elastic particles.

As for the thermal-conduction loss, Zener and co-workers [4] developed a theory in the context of polycrystals (see Ref. [5]). Isakovich [6] also pointed out the significance of heat exchange between particles and the medium in suspensions and emulsions. He reasoned that compression or dilatation of volume during the passage of a sound wave brings about a temperature jump at the droplet-fluid interface, causing a heat flow from one component to another. Such a heat-exchange process leads to internal entropy production, which results in enhanced attenuation and dispersion of sound. A comprehensive theoretical framework that includes both viscous dissipation and thermal conduction was given independently by Epstein and Carhart [7]. Chow [8] generalized the framework to include the surface tension and larger-amplitude oscillations of particles. Allegra and Hawley [9] carried through a further analysis to be applicable to isotropic

solids. They gave a thorough review of previous experimental as well as theoretical studies.

The relative importance of heat and momentum transfer in sound propagation depends on the material constants of systems. However, some theoretical arguments and elaborate experimental tests clarified that the thermal-transport process can be the major factor in the sound attenuation in many systems such as air containing water droplets, aqueous suspensions of polystyrene spheres, and various kinds of emulsions of one liquid in another [8–12]. On this ground, we focus our attention on the heat-transfer mechanism.

Several attempts have been made to extend the above-mentioned work. Among them are studies to include the dispersion of sound velocity [13], to establish a theory applicable in a wider frequency range [14], and to take account of phase transitions, by accommodating interfacial mass transfer as well [15–17]. Despite this long history of the subject, very little is known about the effect of particle-to-particle or droplet-to-droplet interactions upon sound attenuation and dispersion in three-dimensional suspensions and emulsions.

The objective of the present investigation is first to provide the formulation of the problem of the excess attenuation and dispersion of sound in an emulsion due to heat transfer, and second, to elucidate the effect of the mutual interactions of particles. In keeping with this, we neglect the viscosity of fluid. In other words, we leave the shear wave out of consideration. Besides, we postulate that the wavelength of the compressional wave is much longer than that of the thermal wave and the interparticle distances as well as the particle size. This means that the pressure fluctuation is regarded as uniform over the region under investigation.

To be more specific, we explain the heat-exchange mechanism by the passage of sound in some detail. The basic laws of fluid mechanics express the conservation of mass, momentum, and energy. The corresponding partial differential equations combine to yield, after linearization, the following system of equations [14]:

$$\beta \frac{\partial^2 p}{\partial t^2} - \nabla \cdot \left[ \frac{1}{\rho_0} \nabla p \right] - \alpha \frac{\partial^2 T}{\partial t^2} = 0, \quad (1.1)$$

$$\frac{\partial T}{\partial t} - \frac{1}{\rho_0 c_p} \nabla \cdot (\kappa \nabla T) - \frac{T_0 \alpha}{\rho_0 c_p} \frac{\partial p}{\partial t} = 0. \quad (1.2)$$

In the above,  $p$  denotes the pressure fluctuation,  $T$  the temperature fluctuation, and  $T_0$  and  $\rho_0$  stand for the values of temperature and density in equilibrium, respectively. The constant  $\alpha = -(1/\rho)(\partial\rho/\partial T)_p$  is the coefficient of thermal expansion,  $\beta = (1/\rho)(\partial\rho/\partial p)_T$  the isothermal compressibility,  $\kappa$  the thermal conductivity, and  $c_p$  the specific heat at constant pressure. For derivation of (1.2), it suffices to recall the thermodynamic relation

$$Tds = c_p dT - \frac{T\alpha}{\rho} dp, \quad (1.3)$$

where  $s$  denotes the entropy per unit mass. In the absence of  $\alpha$ , (1.1) and (1.2) express the propagation of compressional and thermal waves, respectively. It follows that  $\alpha$  plays the key role of linking the thermal wave to the compressional wave. Komura *et al.* [14] solved both (1.1) and (1.2) directly for a one-dimensional periodic heterogeneous medium with no restriction placed on the frequency range. Such a coupled equation system may be considered to be equivalent to those employed by Epstein and Carhart and Allegra and Hawley. They dealt with the problem of a single spherical particle under the stipulation that the wavelength of sound is much longer than that of the thermal wave as well as the particle size. But it seems to be a hard task to search for the solution of (1.1) and (1.2) for many-particle systems in three dimensions.

Temkin and Dobbins [13] devised an intriguing approach that relies on coupled equations for one-dimensional variables derived by taking a certain average in each component (see Ref. [18]). Though the sound attenuation deduced by them agrees with that of Ref. [7] at low frequencies, it exhibits discrepancies at high frequencies. Moreover, it is not straightforward to incorporate the particle-to-particle interactions into their formalism.

For these reasons, the most appropriate approach for our purpose is to come back to Isakovich's idea. Its key point is to skip the compressional-wave equation (1.1) and thereby to reduce the problem to the solution of the heat-conduction problem (1.2) only. This is made possible because of the long wavelength of the pressure wave, under which the interplay of compressional and thermal waves becomes weak [7,14]. The effective compressibility, or the complex wave number, is then accessible owing to linearity and causality of the system under investigation. A formula for calculating the dispersion relation of sound velocity and the coefficient of sound attenuation

was derived by Isakovich in an ingenious but rather intuitive manner.

In Sec. II, we give a theoretical justification for Isakovich's approach, starting from the entropy production formula in thermodynamics and fluid mechanics. Thereafter his formula is cast into an alternative form which is quite illuminating, admitting a lucid physical interpretation. It is then apparent that Isakovich's analysis neglects the interparticle interactions through thermal waves and replaces the many-particle problem by the isolated-sphere problem. The interparticle interactions are the main subjects of this paper. Our formalism, presented in Sec. II, gives rise to a by-product even under the isolated-sphere approximation: for emulsions with very different thermometric conductivity (the sphere's being much smaller), a frequency region shows up where the attenuation coefficient does not depend on the frequency.

In Sec. III, we highlight the interparticle interactions. As a step towards it, we turn our attention to dilute emulsions of spherical particles arranged in a periodic lattice. To construct a solution of (1.2), we resort to a singular perturbation technique called the method of matched asymptotic expansions. In manipulating the matching conditions, a fast-convergent method of lattice sums devised by Ewald [19] is extended to a new technique applicable to the heat-conduction problem. Numerical computation is performed for three kinds of cubic lattices, namely, simple cubic, body-centered, and face-centered cubic lattices. It will be shown that the importance of particle-interaction effect depends sensitively upon the material properties, especially the thermal conductivity. The final section (Sec. IV) is devoted to a summary and conclusions.

## II. DERIVATION OF THE FORMULA FOR THE ATTENUATION AND DISPERSION OF SOUND

In this section, we shall derive a formula for calculating the complex wave number whose real part gives the sound speed and whose imaginary part is the excess attenuation coefficient. We begin with the rederivation of the Isakovich formula [6].

### A. Isakovich formula

Isakovich proposed his formula via an intuitive argument on the generalized compressibility in the presence of the heat conduction, using the relation  $\delta\rho/\rho = \beta\delta p - \alpha\delta T$ . It is informative to approach it from the opposite side, namely, the energy dissipation or the sound attenuation.

The rate of energy dissipation  $\dot{E}_{\text{mech}}^{(T)}$ , attributable to heat conduction, is given by

$$\dot{E}_{\text{mech}}^{(T)} = -T_0 \int \frac{1}{T} \nabla \cdot (\kappa \nabla T) dV. \quad (2.1)$$

Let us divide the temperature field into the equilibrium temperature  $T_0$  and fluctuating part  $T$ , which is periodic in time. Substituting  $T \rightarrow T_0 + T$  into (2.1) and taking

only terms up to second order in the fluctuation, we are left with, after the time average over one period,

$$\overline{\dot{E}_{\text{mech}}^{(T)}} = \frac{1}{T_0} \int_V \overline{T \nabla \cdot (\kappa \nabla T)} dV, \quad (2.2)$$

where the overbar signifies the time average. Substitution from (1.2) then gives

$$\overline{\dot{E}_{\text{mech}}^{(T)}} = - \int_V \alpha T \frac{\partial p}{\partial t} dV. \quad (2.3)$$

We send a monochromatic wave into the medium and put the pressure and temperature perturbations to be  $p = \text{Re}(Pe^{-i\omega t})$  and  $T = \text{Re}(Te^{-i\omega t})$ , where  $\omega$ , being a real number, is the angular frequency, and the symbol Re means the real part of the following expression. Presuming no confusion, we use  $T$  both for the total temperature perturbation and for its amplitude indistinguishably. Then (2.3) is reduced to

$$|\overline{\dot{E}_{\text{mech}}^{(T)}}| = -\frac{\omega}{2} \int_V \alpha \text{Im}(TP^*) dV, \quad (2.4)$$

where the symbol Im means the imaginary part and the asterisk indicates the complex conjugate.

Let us define  $c$  as the sound speed for the heterogeneous medium under consideration. The mean total energy would be expressed as

$$\bar{E} = \frac{1}{c^2 \langle \rho_0 \rangle} \int_V \overline{p^2} dV = \frac{1}{2c^2 \langle \rho_0 \rangle} \int_V PP^* dV, \quad (2.5)$$

where  $\langle \dots \rangle$  stands for the spatial average. We do not give here a mathematical proof for this. The reader may take this as a plausibility argument but could accept it from the sound energy density,  $e$

$$e = \frac{1}{2} \rho_0 v^2 + \frac{1}{2\rho_0} \left[ \frac{\partial p}{\partial \rho} \right]_s \rho^2 \quad (2.6)$$

and

$$\bar{E} = \int_V \frac{1}{\rho_0} \left[ \frac{\partial p}{\partial \rho} \right]_s \overline{\rho^2} dV, \quad (2.7)$$

where use has been made of the virial theorem of mechanics. In the above,  $\mathbf{v}$  is the fluid velocity and  $\rho$  is the density fluctuation [20].

The attenuation coefficient  $\delta$  is defined by

$$\delta = |\overline{\dot{E}_{\text{mech}}^{(T)}}| / (2c\bar{E}). \quad (2.8)$$

Introducing (2.4) and (2.7) into (2.8), we find that

$$\delta = -\frac{1}{2} c \omega \langle \rho_0 \rangle \text{Im} \langle \alpha T / P \rangle. \quad (2.9)$$

Here the assumption that the pressure may be thought of as uniform is to be remembered.

Now we are in a position to introduce a complex wave number  $k(\omega) = k_r + ik_i$ , whose real part is related to the bulk sound speed  $c$  through  $c = \omega/k_r$  and whose imaginary part is the attenuation coefficient  $\delta = k_i$ . Then, (2.9) reads

$$\text{Im}[k^2(\omega)] = -\omega^2 \langle \rho_0 \rangle \text{Im} \langle \alpha T / P \rangle, \quad (2.10)$$

giving

$$\left[ \frac{k(\omega)}{\omega} \right]^2 = a_1 - \langle \rho_0 \rangle \left\langle \frac{\alpha T}{P} \right\rangle, \quad (2.11)$$

where  $a_1$  is a real function of  $\omega$ . The quantity  $[k(\omega)/\omega]^2$  plays the role of susceptibility relating the response  $\rho$  to the stimulus  $p$ . Owing to the linearity and causality of the system, the Kramers-Kronig relationship guarantees that once the  $\omega$ -dependent imaginary part of the susceptibility is available, the real part is uniquely determined without going into details of the dynamics [21–23]. However, because of lack of the knowledge of functional form of (2.10), we have no alternative but to appeal to the limiting behavior of  $k(\omega)$  as  $\omega \rightarrow \infty$ . In the high-frequency limit, the internal process becomes adiabatic, so that the temperature is given by

$$T = \left[ \frac{\partial T}{\partial p} \right]_s p = \frac{T_0 \alpha}{\rho_0 c_p} p, \quad (2.12)$$

and that the limiting sound velocity  $c_{LL}$  is made simply from the volume average of the adiabatic compressibility  $\beta/\gamma$ :

$$c_{LL} = 1 / \{ \langle \rho_0 \rangle \langle \beta/\gamma \rangle \}^{1/2}, \quad (2.13)$$

where  $\gamma$  is the specific-heat ratio. By comparing (2.11) with the limiting expression  $k = \omega/c_{LL}$ , we obtain the form of  $a_1$  and thus we reach the Isakovich formula

$$k(\omega) = \omega \{ \langle \rho_0 \rangle \langle \xi / P \rangle \}^{1/2}. \quad (2.14)$$

Here  $\xi$  is the so-called condensation defined by

$$\xi = \beta P - \alpha T, \quad (2.15)$$

and use has been made of the thermodynamic relation  $\beta(1 - 1/\gamma) = T_0 \alpha^2 / (\rho_0 c_p)$ .

### B. Alternative form of the Isakovich formula

The physical meaning of the Isakovich formula is made clear if we transform (2.14) into another form derived in what follows. For a monochromatic wave proportional to  $e^{-i\omega t}$ , (1.2) is written as

$$-i\omega T = \frac{1}{\rho_0 c_p} \nabla \cdot (\kappa \nabla T) - \frac{i\omega T_0 \alpha}{\rho_0 c_p} P. \quad (2.16)$$

Insertion of (2.16) into (2.14) with (2.15) yields

$$k^2 = \frac{\omega^2 \langle \rho_0 \rangle}{V} \int_V \left[ \left[ \beta - \frac{T_0 \alpha^2}{\rho_0 c_p} \right] - \frac{i\alpha}{\omega \rho_0 c_p P} \nabla \cdot (\kappa \nabla T) \right] dV, \quad (2.17)$$

where  $V$  is the total volume of the system.

We try to transform the last term of (2.17) into the surface integrals at the interfaces. For this, we restrict our attention to emulsions in which suspended particles or droplets are composed of an identical substance with common material constants, and specify the suspended particles by the subscript 1 and the suspending fluid by the subscript 2. The proper boundary conditions on the surface  $S^{(i)}$  of the  $i$ th particle are

$$T_1 = T_2, \quad (\kappa_1 \nabla T_1) \cdot \mathbf{n} = (\kappa_2 \nabla T_2) \cdot \mathbf{n}, \quad (2.18)$$

where  $\mathbf{n}$  is the unit vector along the outward normal to the particle. In addition, we neglect the contribution from the surface integral at the boundary receding to infinity by assuming the sufficiently rapid decay of the temperature field with distances. Then (2.17) becomes, with the help of Gauss's theorem,

$$k = \omega \left\{ \langle \rho_0 \rangle \left[ \left\langle \frac{\beta}{\gamma} \right\rangle - \frac{i}{\omega P} \left( \frac{\alpha_2}{\rho_2 c_{p_2}} - \frac{\alpha_1}{\rho_1 c_{p_1}} \right) \right] \times \frac{1}{V} \sum_i q_i \right\}^{1/2}, \quad (2.19)$$

where

$$q_i = - \int_{S^{(i)}} (\kappa \nabla T) \cdot \mathbf{n} \, dS, \quad (2.20)$$

and the summation is taken over all particles. The expression (2.19) implies that the thermal attenuation and dispersion are featured by the heat-transfer rate  $q_i$  across the interfaces. Note that the boundary conditions (2.18) pertain to the systems that exhibit no phase transitions. With phase transition, due to the latent-heat generation inside the interface, (2.18) must be replaced by another one and (2.19) ceases to be valid [15,16].

The Isakovich formula can be derived from hydrodynamical principles under certain approximations. We will not go into detail but will give an outline of our derivation. We start with Epstein-Carhart's formalism [7], where the pressure and the temperature fields are expressed as linear combinations of two scalar fields  $\varphi$  and  $\Phi$ . To be more specific, the sum  $\varphi + \Phi = \bar{\varphi}$  is the scalar velocity potential so that the fluid velocity  $\mathbf{v}$  is expressed as  $\mathbf{v} = \nabla \bar{\varphi}$ . Let  $T$  be the fluctuating part of the temperature field. Following Epstein and Carhart, we may safely separate the  $T$  field as  $T = T_\varphi + T_\Phi$ , where

$$T_\varphi = -i\omega[(\gamma-1)/\alpha q^2]\varphi, \quad T_\Phi = -(\rho_0 c_p / \alpha \kappa)\Phi,$$

with  $q^2 = \gamma / \rho_0 \beta$ . The latter satisfies the thermal diffusion equation, namely,

$$\frac{\partial T_\Phi}{\partial t} - \frac{1}{\rho_0 c_p} \nabla \cdot (\kappa \nabla T_\Phi) = 0.$$

Then we obtain

$$\overline{\dot{E}_{\text{mech}}^{(T)}} = \frac{1}{T_0} \int_V \overline{T_\varphi \nabla \cdot (\kappa \nabla T_\Phi)} \, dV. \quad (2.21)$$

To arrive at the expression (2.21), we have neglected the Stokes-Kirchhoff dissipation with a minor renormalization arising from the reflection waves in the  $p$  field, namely,

$$\int_V \overline{T_\varphi \nabla \cdot (\kappa \nabla T_\varphi)} \, dV$$

has been neglected. This is justified as long as the volume fraction of the suspended particles is not too small. It is

noted that

$$\int_V \overline{T_\Phi \nabla \cdot (\kappa \nabla T_\Phi)} \, dV = 0,$$

for  $T_\Phi$  and  $\nabla^2 T_\Phi$  are entirely out of phase. Omitting  $(k/K)^2$ , with  $K = (1+i)(\rho_0 c_p \omega / 2\kappa)^{1/2}$ , we obtain

$$p = -i\omega \rho_0 (\varphi + \Phi) = \frac{\rho_0 \alpha q^2}{\gamma - 1} T_\varphi + \text{const} T_\Phi.$$

Then (2.21) is written as

$$\overline{\dot{E}_{\text{mech}}^{(T)}} = \int_V \frac{\alpha}{\rho_0 c_p} \overline{p \nabla \cdot (\kappa \nabla T_\Phi)} \, dV,$$

where we have used the thermodynamic identity

$$\frac{\alpha q^2}{\gamma - 1} = \frac{c_p}{\alpha T_0}.$$

Hence we arrive at

$$\overline{\dot{E}_{\text{mech}}^{(T)}} \approx \frac{1}{2} \left[ \frac{\alpha_2}{\rho_2 c_{p_2}} - \frac{\alpha_1}{\rho_1 c_{p_1}} \right] \text{Re} \left[ P^* \sum_i q_i \right]. \quad (2.22)$$

Starting from (2.22) in place of (2.3), we are able to reach (2.19) again.

Equation (2.22) is helpful in understanding the heat-exchange mechanism. In response to the incident pressure wave, the temperature difference

$$\left[ \frac{\alpha_2}{\rho_2 c_{p_2}} - \frac{\alpha_1}{\rho_1 c_{p_1}} \right] T_0 P e^{-i\omega t} \quad (2.23)$$

is established between the two phases with no time lag. It is followed by the heat current  $q_i$  from the higher temperature phase to the lower one. The amount of work done associated with this relaxation process is furnished by (2.22).

### C. Dilute emulsion of independent spherical particles

It is worthwhile to revisit the problem of a very dilute emulsion of spherical droplets or particles. With the phrase "very dilute," we mean that mutual interactions among the particles may be disregarded. The result is included in Ref. [6], and we make a few explanatory comments on it.

We consider a spherical particle or droplet of radius  $a$  embedded in a fluid of different sort and choose polar coordinates  $(r, \theta, \phi)$  with the origin taken at the center of the sphere. Then the boundary conditions reduce to

$$T_1 = T_2, \quad \kappa_1 \frac{\partial T_1}{\partial r} = \kappa_2 \frac{\partial T_2}{\partial r} \quad \text{at } r = a. \quad (2.24)$$

Subject to (2.24), the solution of (2.16) takes the form

$$T_1 = \left\{ \frac{T_0 \alpha_1}{\rho_1 c_{p_1}} + \frac{A_1}{r} \sinh[(1-i)n_1 r] \right\} P e^{-i\omega t} \quad \text{for } r \leq a, \quad (2.25a)$$

$$T_2 = \left\{ \frac{T_0 \alpha_2}{\rho_2 c_{p_2}} + \frac{A_2}{r} e^{-[(1-i)n_2 r]} \right\} P e^{-i\omega t}$$

for  $r \geq a$ , (2.25b)

where  $n_i = (\omega \rho_i c_{p_i} / 2\kappa_i)^{1/2}$  for  $i=1,2$  is the wavelength of the thermal wave, and  $A_1$  and  $A_2$  are the constants com-

plying with (2.24). Insertion of (2.25) into (2.14) or (2.19) yields

$$k(\omega) = \frac{\omega}{c_{LL}} \left[ 1 + \frac{3i\epsilon T_0 \langle \rho_0 \rangle c_{LL}^2}{a^2 \omega} \hat{A} \right]^{1/2}, \quad (2.26a)$$

where  $\epsilon$  is the volume fraction of the suspended material characterized by the subscript 1 and

$$\hat{A} = \left[ \frac{\alpha_2}{\rho_2 c_{p_2}} - \frac{\alpha_1}{\rho_1 c_{p_1}} \right]^2 \times \frac{\kappa_1 \kappa_2 [(1-i)n_2 a + 1] \{ (1-i)n_1 a - \tanh[(1-i)n_1 a] \}}{\kappa_1 \{ (1-i)n_1 a - \tanh[(1-i)n_1 a] \} + \kappa_2 \tanh[(1-i)n_1 a] [(1-i)n_2 a + 1]}. \quad (2.26b)$$

Here the definition of  $c_{LL}$  given by (2.13) is to be understood.

For  $\epsilon \ll 1$ , the limiting forms of this expression for both cases where the thermal wavelength is much greater and much less than the size of the suspended particles are easily written out. At very low frequencies ( $n_1 a \ll 1$  and  $n_2 a \ll 1$ ),

$$c = \frac{\omega}{k_r} \sim c_{LL} \left[ 1 - \frac{\epsilon}{2} T_0 c_{LL}^2 \langle \rho_0 \rangle \rho_1 c_{p_1} \times \left[ \frac{\alpha_2}{\rho_2 c_{p_2}} - \frac{\alpha_1}{\rho_1 c_{p_1}} \right]^2 \right], \quad (2.27a)$$

$$\delta \sim \frac{\epsilon}{6\kappa_1} T_0 c_{LL} \langle \rho_0 \rangle \rho_1^2 c_{p_1}^2 \left[ \frac{1}{5} + \frac{\kappa_1}{\kappa_2} \right] \times \left[ \frac{\alpha_2}{\rho_2 c_{p_2}} - \frac{\alpha_1}{\rho_1 c_{p_1}} \right]^2 a^2 \omega^2. \quad (2.27b)$$

At very high frequencies ( $n_1 a \gg 1$  and  $n_2 a \gg 1$ ),

$$c \sim c_{LL} \left[ 1 - \frac{3\epsilon}{2a\sqrt{2\omega}} T_0 c_{LL}^2 \langle \rho_0 \rangle \left[ \frac{\alpha_2}{\rho_2 c_{p_2}} - \frac{\alpha_1}{\rho_1 c_{p_1}} \right]^2 \times \frac{1}{(\kappa_1 \rho_1 c_{p_1})^{-1/2} + (\kappa_2 \rho_2 c_{p_2})^{-1/2}} \right], \quad (2.28a)$$

$$\delta \sim \frac{3\epsilon\sqrt{\omega}}{2\sqrt{2}a} T_0 c_{LL} \langle \rho_0 \rangle \left[ \frac{\alpha_2}{\rho_2 c_{p_2}} - \frac{\alpha_1}{\rho_1 c_{p_1}} \right]^2 \times \frac{1}{(\kappa_1 \rho_1 c_{p_1})^{-1/2} + (\kappa_2 \rho_2 c_{p_2})^{-1/2}}. \quad (2.28b)$$

At low frequencies, the attenuation coefficient  $\delta$  given by (2.27b) is proportional to  $\omega^2$ , which happens to agree with the Stokes-Kirchhoff formula in the homogeneous media. But their physical mechanisms are different from each other and these effects are additive. The source of (2.27b) is traced to the nonstationary term in (2.16). The forms of (2.28a) and (2.28b) are inherent in the heterogeneous media. At high frequencies,  $\delta$  is proportional to  $\omega^{1/2}$ , because only a small fraction of the volume confined to the thermal boundary layer with thickness  $(\omega \rho c_p / \kappa)^{-1/2}$  takes part in the loss mechanism. From the standpoint of

(2.22), we may say that the heat flux  $q$  is proportional to  $\omega^{1/2}$  because the temperature variation is confined to the skin depth of thermal clothes. It is not difficult to realize that the dependence of (2.28) on frequency is in accord with that associated with sound reflection from a solid wall [24,25].

It deserves emphasis that a novel feature shows up when the thermometric conductivity  $\chi = \kappa / \rho c_p$  of the suspended substance is much smaller than that of the ambient fluid. Manipulation of (2.26) uncovers that for  $n_1 a \gg 1$  and  $n_2 a \ll 1$  under the constraint  $\kappa_1(n_1 a) \gg \kappa_2$ ,

$$c \sim c_{LL} \left[ 1 - \frac{3\epsilon n_2}{2a\omega} T_0 c_{LL}^2 \langle \rho_0 \rangle \kappa_2 \left[ \frac{\alpha_2}{\rho_2 c_{p_2}} - \frac{\alpha_1}{\rho_1 c_{p_1}} \right]^2 \times \left[ 1 + \frac{\kappa_2}{2\kappa_1(n_1 a)(n_2 a)} \right] \right], \quad (2.29a)$$

$$\delta \sim \frac{3\epsilon}{2a^2} T_0 c_{LL} \langle \rho_0 \rangle \kappa_2 \left[ \frac{\alpha_2}{\rho_2 c_{p_2}} - \frac{\alpha_1}{\rho_1 c_{p_1}} \right]^2. \quad (2.29b)$$

The examination of the temperature field (2.25) in this frequency range indicates that the boundary conditions (2.18) imposed at the interface are virtually equivalent to the fixed temperature condition ( $T_1 = T_2 = [T_0 \alpha_1 / \rho_1 c_{p_1}] P e^{-i\omega t}$  at  $r = a$ ) on account of the relative largeness of the specific heat of the particle. In this case, the heat flux  $q$  is proportional to  $\omega^0$ , so is the attenuation coefficient. We are reminded that the same frequency dependence is touched on in Ref. [20]. This particular behavior is intrinsic to spherical particles or, more precisely, curved interfaces. For flat interfaces, heat flux independent of  $\omega$  is in no way compatible with the fixed boundary condition. It is interesting to note that the systems undergoing the first-order phase transition within the interface show the same behavior [16].

To illustrate the significance of the heat-exchange mechanism, we show in Fig. 1 the excess attenuation per wavelength  $\lambda = 2\pi/k$  and the sound velocity normalized by  $c_{LL}$  simultaneously as functions of the normalized frequency  $n_2 a$  for an emulsion of 20% by volume toluene in water. For comparison, the attenuation per wavelength for pure water, based on the formula of Stokes-Kirchoff, is shown as a dashed line. The material constants employed here and hereafter are taken from the tables in Ref. [9], except for air, and the values at 20°C, which are

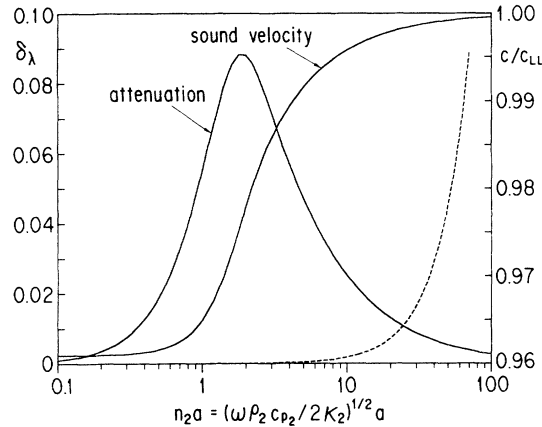


FIG. 1. Excess attenuation per wavelength  $\delta_\lambda (=2\pi\delta/k)$  and normalized sound velocity  $c/c_{LL}$  as functions of  $n_2a = (\omega\rho_2c_{p_2}/2\kappa_2)^{1/2}a$ , the ratio of droplet radius  $a$  to skin depth of thermal wave in the suspending fluid, for an emulsion of 20% by volume toluene in water at 20°C. Here  $c_{LL}$  is the high-frequency limit of the sound velocity and is given by (2.13). The values are calculated from (2.26a) and (2.26b). The dashed line displays the attenuation per wavelength for pure water based on the formula of Stokes-Kirchhoff.

adopted, are listed in Table I. For air, the physical parameters at 22°C are chosen from Ref. [7]. It is apparent that the sound attenuation is dominated by the interfacial heat transfer over a wide frequency range, but that this is not the case when the frequency becomes very large. The transition between them was clearly exemplified by Komura *et al.* [14], dealing with a one-dimensional medium.

Figure 2(a) presents the frequency dependence of the attenuation coefficients for suspensions of 0.1% by volume polystyrene in water (the solid line) and in air (the dashed line). Figure 2(b) is the corresponding graph for the deficiency  $(c_{LL} - c)/c_{LL}$  of sound speed. The heat conduction is more effective in air than in water, mainly because the magnitude of  $\alpha/\rho c_p$  of air is much larger than that of water. More remarkable is that for the air suspension, an intermediate frequency region is identifiable where the frequency dependence is the same as that predicted by (2.29). To see this, it is enough to refer to the difference of values of the thermometric conductivity.

However, all of the results mentioned above forget the influence of the neighboring particles or droplets on heat-transport process. At low frequencies the tempera-

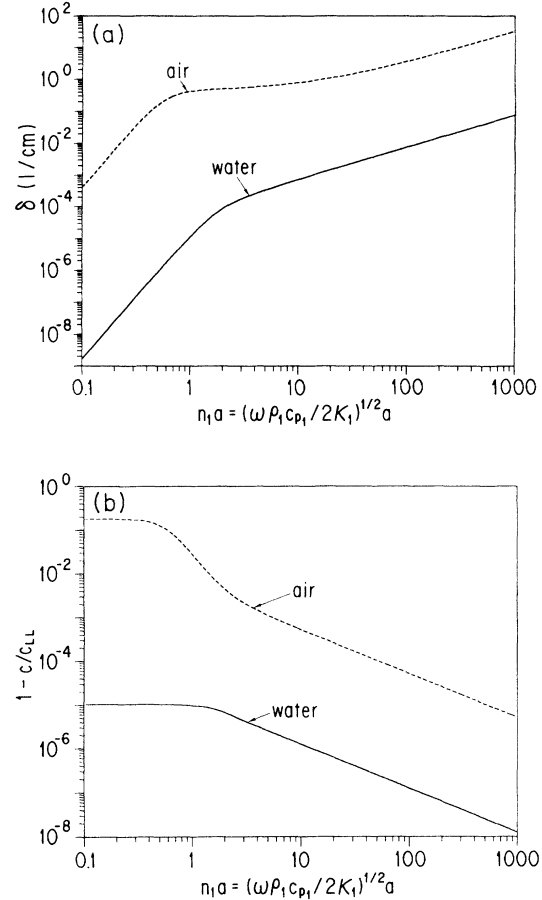


FIG. 2. Excess attenuation  $\delta$  (a) and deficiency  $1 - c/c_{LL}$  of sound velocity  $c$  (b) as functions of  $n_1a = (\omega\rho_1c_{p_1}/2\kappa_1)^{1/2}a$ , the ratio of particle radius  $a$  to skin depth of thermal wave in the particle, for an emulsion of 0.1% by volume polystyrene in water (solid line) and in air (dashed line) at 20°C. Here  $c_{LL}$  is the high-frequency limit of the sound velocity and is given by (2.13). The values are calculated from (2.26a) and (2.26b).

ture changes gradually over a thick layer surrounding the interface. As a result, each droplet is dressed in a thick clothes of such boundary layer of temperature penetration that may contain adjacent droplets. The heat-exchange process may be significantly different from Isakovich's case. In the next section, we inquire into the modifications brought about when particle-to-particle interactions are called into play.

TABLE I. Physical constants of substances employed in our calculation. The values, except for the last column (air), are calculated at  $T = 20^\circ\text{C}$ , using the information listed in the tables of Allegra and Hawley [9]. The values for air, given at  $T = 22^\circ\text{C}$ , are chosen from the table of Epstein and Carhart [7].

	Water	Toluene	Polystyrene	Air (22°C)
$\rho$ (g/cm <sup>3</sup> )	0.9964	0.8656	1.055	$1.17 \times 10^{-3}$
$\kappa$ (10 <sup>4</sup> erg/cm sec °C)	5.87	1.6	1.15	0.243
$c_p$ (10 <sup>7</sup> erg/g °C)	4.181	1.666	1.19	1.00
$c$ (10 <sup>5</sup> cm/sec)	1.483	1.360	2.38	0.344
$\alpha$ (10 <sup>-4</sup> /°C)	2.042	10.71	2.6	36.6

### III. PERIODIC EMULSION

The question of interparticle interactions with respect to the thermal-transport process was addressed recently by Onuki [16]. He dealt with sound attenuation in the critical regime of a phase-separating fluid system and commented upon the interaction effect for a uniform distribution of droplets. But in several respects, his treatment is insufficient. In this section, we consider spatially periodic emulsions and give a more comprehensive analysis on the attenuation and dispersion of sound. Though limited in application, this situation has some relevance to ordered colloidal dispersions, which consist of electrically interacting charged Brownian particles arranged in a lattice.

A distinguishing advantage inherent in a periodic system is the availability of a powerful mathematical tool for lattice sums exploited by Ewald [19] (see also Ref. [26]). This technique has been applied to a variety of solid-fluid or solid-solid systems, such as the sedimentation problem for spheres in a lattice [27,28], the effective viscosity of a periodic suspension [29,30], and the effective conductivity of composite materials [31,32]. Here we would like to point out that although there has been an abundance of studies concerning periodic systems, the calculations of lattice sums so far implemented have been limited to systems with very long-range interactions, such as arrays of Coulomb charges and Stokeslets. However, the thickness of the thermal boundary layer plays an important role in the physical mechanism of heat transfer and so the variation of its thickness cannot be ruled out. Our intention is to accomplish an extension of Ewald's technique to lattices of spherical droplets with nonstationary interfacial heat transfer. This procedure is then tied up with a singular perturbation method.

#### A. Formulation of the problem: matched asymptotic expansions

Consider a set of identical spherical particles or droplets of radius  $a$ , which form an infinite three-dimensional regular lattice embedded in another fluid. Coordinates of sphere centers are

$$\mathbf{r}_l = l_1 \mathbf{a}^{(1)} + l_2 \mathbf{a}^{(2)} + l_3 \mathbf{a}^{(3)} \quad (l_1, l_2, l_3 = 0, \pm 1, \pm 2, \dots), \quad (3.1)$$

where  $\mathbf{a}^{(1)}$ ,  $\mathbf{a}^{(2)}$ , and  $\mathbf{a}^{(3)}$  are the basic lattice vectors that constitute the basis for a unit cell having volume  $\tau_0 = \mathbf{a}^{(1)} \cdot (\mathbf{a}^{(2)} \times \mathbf{a}^{(3)})$ . The volume fraction of suspended substance is  $\epsilon = 4\pi a^3 / 3\tau_0$ , which is assumed to be small. We are reminded that the wavelength  $2\pi/k$  of the pressure wave is much longer than the lattice parameter  $h$  (particle-to-particle distance) as well as the particle radius, so that the pressure may be taken as uniform. Upon the thermal wave, we impose a condition that the thickness  $1/n$  with  $n = (\omega\rho c_p / 2\kappa)^{1/2}$  of the thermal boundary layer is much larger than the particle size, but it may be comparable with the lattice parameter. Also, the condition that  $n_1 \sim n_2$  or  $\chi_1 \sim \chi_2$  is necessary for our expansion scheme to be valid.

With these conditions, mutual interaction between the particles need be considered. The above conditions are simply expressed by

$$a \ll 1/n_1 \sim 1/n_2 \sim h \ll 2\pi/k. \quad (3.2)$$

The equation to be solved both inside and outside the sphere is provided by (1.2) or (2.16). The boundary conditions (2.24) apply to the interface of the sphere centered at the origin. The coexistence of conflicting symmetries, namely, the spherical symmetry associated with the particle shape and the symmetry associated with the arrangement of the particles, renders the problem difficult. A way to circumvent this difficulty is to take advantage of a singular perturbation technique called the method of matched asymptotic expansions, an explanation of which is given in the following.

We concentrate on a specific particle and the surrounding unit cell bounded by the midplanes between that particle and the nearest-neighbor ones. The Cartesian coordinates  $\mathbf{r}' = (x'_1, x'_2, x'_3)$  or polar coordinates  $(r', \theta, \phi)$  are introduced such that the origin coincides with the center of the sphere. Here as a tentative notation we distinguish the dimensional coordinates by primed variables. The inner region consists of both the interior of the particle and its immediate neighborhood [ $r' \sim O(a)$ ]. The outside of it [ $r' \sim O(1/n_2)$ ] is referred to as the outer region. Accordingly, we introduce the inner variable  $\mathbf{r} = \mathbf{r}'/a$  and the outer one  $\bar{\mathbf{r}} = n_2 \mathbf{r}'$ , with which we nondimensionalize (2.16) in each region. As in Sec. II B, the temperature field is decomposed into the incident wave and the diffraction component  $T'_i$  and thus expressed, corresponding to the pressure perturbations  $p = \text{Re}[Pe^{-i\omega t}]$ , as

$$T_i = \text{Re}[\hat{T}_i Pe^{-i\omega t}], \quad (3.3a)$$

with

$$\hat{T}_i = T'_i + T_0 \alpha_i / (\rho_i c_{p_i}). \quad (3.3b)$$

Then, in the inner region, the diffraction component obeys

$$\Delta T'_1 = -2i\hat{n}^2 \Omega T'_1 \quad \text{for } r < 1, \quad (3.4a)$$

$$\Delta T'_2 = -2i\Omega T'_2 \quad \text{for } 1 < r \leq R_c/a. \quad (3.4b)$$

In the outer region, it takes the form

$$\tilde{\Delta} \tilde{T}'_2 = -2i\tilde{T}'_2 \quad \text{for } \bar{r} \geq n_2 R_c, \quad (3.5)$$

where  $\Omega = (n_2 a)^2$ ,  $\hat{n} = n_1/n_2$ , and  $\Delta$  is the three-dimensional Laplacian. As is seen from (3.2),  $\Omega \ll 1$  and  $\hat{n} \sim O(1)$ . There is a substantial region of overlap between the inner and outer regions and  $R_c$  is a typical radius characterizing it. We seek the solution of (3.4) and (3.5) supplemented with (2.24) to yield the temperature distribution in the inner and outer regions separately, in the form of series expansions in a small parameter  $\Omega^{1/2}$ . The resulting fields are then matched to each other.

In the inner region, where the nonstationary effect acts as a small perturbation, we proceed by postulating the following forms of the temperature fields:

$$T_1' = T_1^{(0)} + \Omega T_1^{(1)} + \Omega^{3/2} T_1^{(3/2)} + \Omega^2 T_1^{(2)} + \Omega^{5/2} T_1^{(5/2)} + \Omega^3 T_1^{(3)} + \Omega^{7/2} T_1^{(7/2)} + \dots, \quad (3.6a)$$

$$T_2' = T_2^{(0)} + \Omega T_2^{(1)} + \Omega^{3/2} T_2^{(3/2)} + \Omega^2 T_2^{(2)} + \Omega^{5/2} T_2^{(5/2)} + \Omega^3 T_2^{(3)} + \Omega^{7/2} T_2^{(7/2)} + \dots. \quad (3.6b)$$

In the outer region, where the nonstationary effect balances with the heat conduction, the expansion is of the form

$$\tilde{T}_2' = \Omega^{3/2} \tilde{T}_2^{(3/2)} + \Omega^2 \tilde{T}_2^{(2)} + \Omega^{5/2} \tilde{T}_2^{(5/2)} + \Omega^3 \tilde{T}_2^{(3)} + \Omega^{7/2} \tilde{T}_2^{(7/2)} + \Omega^4 \tilde{T}_2^{(4)} + \dots. \quad (3.7)$$

It is straightforward to obtain the first few terms of the inner expansions. If we suppose that (3.7) is true, there exists no field in the outer region up to  $O(\Omega)$ , which supplies heat to the inner region. Therefore, we have

$$T_1^{(0)} = T_0 \left[ \frac{\alpha_2}{\rho_2 c_{p_2}} - \frac{\alpha_1}{\rho_1 c_{p_1}} \right] \equiv \Theta, \quad (3.8a)$$

$$T_2^{(0)} = 0, \quad (3.8b)$$

and

$$T_1^{(1)} = -\frac{\kappa_2}{2\kappa_1} A \left[ r^2 - \left[ \frac{2\kappa_1}{\kappa_2} + 1 \right] \right], \quad (3.9a)$$

$$T_2^{(1)} = \frac{A}{r}, \quad (3.9b)$$

where

$$A = \frac{2i\kappa_1}{3\kappa_2} \hat{n}^2 \Theta. \quad (3.10)$$

Notice, as shown by (2.23),  $\Theta$  is the initial temperature discrepancy across the interface that drives the resulting heat current.

### B. Periodic fundamental solution

The crucial step is the construction of the periodic fundamental solution  $\tilde{T}_2^{(3/2)}$  of (3.5) and the deduction of the matching condition on the inner solution. Below, we describe it at some length.

The solution (3.9b) requires that

$$\tilde{T}_2^{(3/2)} \sim \frac{A}{\tilde{r}} \quad \text{as } \tilde{r} \rightarrow 0, \quad (3.11)$$

which represents the temperature field produced by a point source of strength  $-4\pi A$  at  $\tilde{r}=0$ . If account is taken of the periodicity of the system, the solution of (3.5) subject to (3.11) is shown to be reducible to the solution of

$$\tilde{\Delta} \tilde{T}_2^{(3/2)} + 2i\tilde{T}_2^{(3/2)} = -4\pi A \sum_l \delta^3(\tilde{\mathbf{r}} - \tilde{\mathbf{r}}_l), \quad (3.12)$$

where the summation is taken over all integers  $l = (l_1, l_2, l_3) \in \mathbb{Z}^3$ . As usual, we write down the formal solution of (3.12) in the form of a Fourier series

$$\tilde{T}_2^{(3/2)} = \frac{A}{\pi \tilde{\tau}_0} \sum_m \frac{e^{-2\pi i(\mathbf{k}_m \cdot \tilde{\mathbf{r}})}}{k_m^2 - \frac{i}{2\pi^2}}, \quad (3.13)$$

where  $\tilde{\tau}_0$  is the normalized volume of unit cell ( $n_2^3 \tau_0$ ) and  $\mathbf{m} \in \mathbb{Z}^3$ . Furthermore,

$$\mathbf{k}_m = m_1 \tilde{\mathbf{b}}^{(1)} + m_2 \tilde{\mathbf{b}}^{(2)} + m_3 \tilde{\mathbf{b}}^{(3)} \quad (m_1, m_2, m_3 = 0, \pm 1, \pm 2, \dots) \quad (3.14)$$

is the wave number normalized by  $n_2$  with  $(\tilde{\mathbf{b}}^{(1)}, \tilde{\mathbf{b}}^{(2)}, \tilde{\mathbf{b}}^{(3)})$  being the reciprocal basic lattice vectors, which are given by

$$\begin{aligned} \tilde{\mathbf{b}}^{(1)} &= [\tilde{\mathbf{a}}^{(2)} \times \tilde{\mathbf{a}}^{(3)}] / \tilde{\tau}_0, & \tilde{\mathbf{b}}^{(2)} &= [\tilde{\mathbf{a}}^{(3)} \times \tilde{\mathbf{a}}^{(1)}] / \tilde{\tau}_0, \\ \tilde{\mathbf{b}}^{(3)} &= [\tilde{\mathbf{a}}^{(1)} \times \tilde{\mathbf{a}}^{(2)}] / \tilde{\tau}_0. \end{aligned} \quad (3.15)$$

Now we turn to derivation of the functional form of (3.13) as  $\tilde{r} \rightarrow 0$ . In connection with this, Ewald [19] gave a useful argument in the context of electromagnetic waves and we refashion it for our purpose.

We start with transformation of (3.13) into an integral representation:

$$\tilde{T}_2^{(3/2)} = \frac{A}{\tilde{\tau}_0} \sum_m \lim_{\epsilon \rightarrow 0^+} \int_0^\infty e^{-\pi[k_m^2 - (i/2\pi^2) + \epsilon]\xi - 2\pi i(\mathbf{k}_m \cdot \tilde{\mathbf{r}})} d\xi. \quad (3.16)$$

A small positive number  $\epsilon$  is introduced for correctness. Here we divide the domain of integration into two parts; one is from 0 to  $\eta$  and the other from  $\eta$  to  $\infty$ . By applying to the former part the theta transformation formula

$$\sum_m e^{-\pi k_m^2 \xi - 2\pi i(\mathbf{k}_m \cdot \tilde{\mathbf{r}})} = \frac{\tilde{\tau}_0}{\xi^{3/2}} \sum_l e^{-\pi(\tilde{\mathbf{r}} - \tilde{\mathbf{r}}_l)^2 / \xi}, \quad (3.17)$$

we have

$$\begin{aligned} \tilde{T}_2^{(3/2)} &= A \left[ \frac{2}{\sqrt{\pi}} \sum_l \int_{\sqrt{\pi/\eta}}^\infty e^{-(\tilde{\mathbf{r}} - \tilde{\mathbf{r}}_l)^2 \xi^2 + (i/2\pi^2)\xi} d\xi \right. \\ &\quad \left. + \frac{\eta}{\tilde{\tau}_0} \sum_m e^{-2\pi i(\mathbf{k}_m \cdot \tilde{\mathbf{r}})} \right. \\ &\quad \left. \times \lim_{\epsilon \rightarrow 0^+} \int_1^\infty e^{-\pi[k_m^2 - (i/2\pi^2) + \epsilon]\eta \xi} d\xi \right]. \end{aligned} \quad (3.18)$$

In the first term of (3.18), we convert the integration variable  $\xi$  into  $\mu$  defined by

$$\mu = \tilde{R}_l \xi + \frac{1-i}{2\xi} \quad \text{or} \quad \xi = \frac{\mu \pm [\mu^2 - 2(1-i)\tilde{R}_l]^{1/2}}{2\tilde{R}_l},$$

with  $\tilde{R}_l = |\tilde{\mathbf{r}} - \tilde{\mathbf{r}}_l|$ . Thereby that term is split into two terms, to one of which we again apply transformation of the integration variable

$$v^2 = \mu^2 - 2(1-i)\tilde{R}_l.$$

In the resulting expression, there still remains an undetermined sign (plus or minus). With one sign chosen to be compatible with (3.18), we get



$$\begin{aligned} \tilde{T}_2^{(3/2)} = A \left[ \frac{1}{2} \sum_l \left\{ \frac{e^{(1-i)|\tilde{r}-\tilde{r}_l|}}{|\tilde{r}-\tilde{r}_l|} \operatorname{erfc} \left[ |\tilde{r}-\tilde{r}_l| \left( \frac{\pi}{\eta} \right)^{1/2} + \frac{1-i}{2} \left( \frac{\eta}{\pi} \right)^{1/2} \right] \right. \right. \\ \left. \left. + \frac{e^{-(1-i)|\tilde{r}-\tilde{r}_l|}}{|\tilde{r}-\tilde{r}_l|} \operatorname{erfc} \left[ |\tilde{r}-\tilde{r}_l| \left( \frac{\pi}{\eta} \right)^{1/2} - \frac{1-i}{2} \left( \frac{\eta}{\pi} \right)^{1/2} \right] \right\} + \frac{1}{\pi \tilde{r}_0} \sum_m \frac{e^{-2\pi i(\mathbf{k}_m \cdot \tilde{\mathbf{r}}) - \pi[k_m^2 - (i/2\pi^2)]\eta}}{k_m^2 - \frac{i}{2\pi^2}} \right], \end{aligned} \quad (3.19)$$

where

$$\operatorname{erfc}(z) = \frac{2}{\sqrt{\pi}} \int_z^\infty e^{-w^2} dw. \quad (3.20)$$

It should be noted that (3.20) represents a line integral in the complex plane rather than the real integral. The contour of integration is prescribed to guarantee the convergence of (3.20).

We are now prepared to deduce the behavior of the fundamental periodic solution as  $\tilde{r} \rightarrow 0$ . To this end, we use Hobson's theorem [33], which is expressed, if we limit ourselves to cubic lattices, as

$$\begin{aligned} \tilde{T}_2^{(3/2)} = \tilde{T}_s + \sum_{n=0, n \neq 1}^\infty \sum_{m=0}^{m \leq n/2} (a_{nm}^{(0)} + a_{nm}^{(2)} \tilde{r}^2 + \dots \\ + a_{nm}^{(2k)} \tilde{r}^{2k} + \dots) \\ \times Y_{2n}^{4m}(\tilde{x}_1, \tilde{x}_2, \tilde{x}_3), \end{aligned} \quad (3.21)$$

where  $\tilde{T}_s$  is a particular solution including all of the singular part and is given, for example, by

$$\tilde{T}_s = \frac{A}{2\tilde{r}} [e^{(1-i)\tilde{r}} + e^{-(1-i)\tilde{r}}], \quad (3.22)$$

and  $Y_n^m$  is the solid spherical harmonic

$$Y_n^m(x_1, x_2, x_3) = r^n P_n^m(\cos\theta) \cos m\phi, \quad (3.23)$$

with  $P_n^m(\cos\theta)$  being the associated Legendre function of degree  $n$  and rank  $m$ . The coefficients  $a_{nm}^{(2k)}$  are determined by

$$a_{nm}^{(0)} = c_{nm} Y_{2n}^{4m} \left[ \frac{\partial}{\partial \tilde{x}_1}, \frac{\partial}{\partial \tilde{x}_2}, \frac{\partial}{\partial \tilde{x}_3} \right] \left[ \tilde{T}_2^{(3/2)} - \tilde{T}_s \right] \Big|_{\tilde{r}=0}, \quad (3.24a)$$

$$\begin{aligned} a_{nm}^{(2k)} = \frac{c_{nm}}{2^k k! (4n+3)(4n+5) \cdots (4n+2k+1)} \\ \times \tilde{\nabla}^{2k} Y_{2n}^{4m} \left[ \frac{\partial}{\partial \tilde{x}_1}, \frac{\partial}{\partial \tilde{x}_2}, \frac{\partial}{\partial \tilde{x}_3} \right] \left[ \tilde{T}_2^{(3/2)} - \tilde{T}_s \right] \Big|_{\tilde{r}=0} \\ (k \geq 1), \end{aligned} \quad (3.24b)$$

with

$$c_{nm} = \frac{\epsilon_m 2^{2n} (2n)! (2n-4m)!}{(4n)! (2n+4m)!}, \quad \epsilon_m = \begin{cases} 1 & \text{for } m=0 \\ 2 & \text{for } m>0. \end{cases} \quad (3.24c)$$

Substitution from (3.19) thus gives rise to

$$\begin{aligned} \tilde{T}_2^{(3/2)} \sim A \left[ \frac{1}{\tilde{r}} - (1-i+b) - i\tilde{r} + \frac{i}{3} (1-i+b) \tilde{r}^2 - \frac{\tilde{r}^3}{6} \right. \\ \left. + \frac{1-i+b}{30} \tilde{r}^4 + a_{20}^{(0)} Y_4^0(\tilde{x}_1, \tilde{x}_2, \tilde{x}_3) \right. \\ \left. + a_{21}^{(0)} Y_4^1(\tilde{x}_1, \tilde{x}_2, \tilde{x}_3) + \dots \right] \text{ as } \tilde{r} \rightarrow 0. \end{aligned} \quad (3.25)$$

Here the parameter  $b$  is the crucial quantity in our analysis. This parameter represents the particle-to-particle or droplet-to-droplet interactions and is given by

$$\begin{aligned} b = \frac{2}{\sqrt{\eta}} e^{i\eta/2\pi} - (1-i) \operatorname{erfc} \left[ \frac{1-i}{2} \left( \frac{\eta}{\pi} \right)^{1/2} \right] - \frac{2\pi i}{\tilde{r}_0} e^{i\eta/2\pi} \\ - \sum_{l \neq 0}' \left\{ \frac{e^{(1-i)\tilde{r}_l}}{2\tilde{r}_l} \operatorname{erfc} \left[ \tilde{r}_l \left( \frac{\pi}{\eta} \right)^{1/2} + \frac{1-i}{2} \left( \frac{\eta}{\pi} \right)^{1/2} \right] \right. \\ \left. + \frac{e^{-(1-i)\tilde{r}_l}}{2\tilde{r}_l} \operatorname{erfc} \left[ \tilde{r}_l \left( \frac{\pi}{\eta} \right)^{1/2} - \frac{1-i}{2} \left( \frac{\eta}{\pi} \right)^{1/2} \right] \right\} - \sum_{m \neq 0}' \frac{e^{-\pi[k_m^2 - (i/2\pi^2)]\eta}}{\pi \tilde{r}_0 \left[ k_m^2 - \frac{i}{2\pi^2} \right]}. \end{aligned} \quad (3.26)$$

The prime on the sums means that the index  $l = \mathbf{m} = \mathbf{0}$  is to be removed. It is noticeable that all other particles simultaneously take part in the heat-conduction process. The limiting form (3.25) thus obtained in turn furnishes a matching condition on the solution at higher orders.

### C. Formula for attenuation and dispersion of sound

The construction of higher-order inner and outer solutions is implemented with no difficulty. We have repeated the procedure up to  $O(\Omega^{7/2})$  for the inner solution and up to  $O(\Omega^4)$  for the outer solution. We omit details and write down the necessary expressions of the resulting field over the entire space.

In the inner region,

$$\hat{T}_1 = \frac{T_0 \alpha_1}{\rho_1 c_{p_1}} + \Theta - \frac{\kappa_2}{2\kappa_1} A \left[ r^2 - \left( \frac{2\kappa_1}{\kappa_2} + 1 \right) \right] \Omega - A(1-i+b)\Omega^{3/2} + \left\{ \frac{i\kappa_2}{\kappa_1} \hat{n}^2 A \left[ \frac{r^4}{20} - \frac{1}{6} \left( \frac{2\kappa_1}{\kappa_2} + 1 \right) r^2 \right] + A_1^{(2)} \right\} \Omega^2 + \left[ \frac{iA}{3} \hat{n}^2 (1-i+b)r^2 + A_1^{(5/2)} \right] \Omega^{5/2} + \dots \quad \text{for } r < 1, \quad (3.27a)$$

$$\hat{T}_2 = \frac{T_0 \alpha_2}{\rho_2 c_{p_2}} + \frac{A}{r} \Omega - A(1-i+b)\Omega^{3/2} + \left[ -iAr + \frac{B}{r} \right] \Omega^2 + \left[ \frac{iA}{3} (1-i+b)r^2 + A_2^{(5/2)} + \frac{C}{r} \right] \Omega^{5/2} + \dots \quad \text{for } 1 \leq r \leq R_c/a. \quad (3.27b)$$

In the outer region,

$$\hat{T}_2 = \frac{T_0 \alpha_2}{\rho_2 c_{p_2}} + \frac{[A\Omega^{3/2} + B\Omega^{5/2} + C\Omega^3 + D\Omega^{7/2} + E\Omega^4 + \dots]}{\pi \bar{\tau}_0} \times \sum_m \frac{e^{-2\pi i(\mathbf{k}_m \cdot \bar{\mathbf{r}})}}{k_m^2 - \frac{i}{2\pi^2}} \quad \text{for } \bar{\mathbf{r}} \geq n_2 R_c, \quad (3.28)$$

where the definitions of  $\Theta$  and  $A$  are given by (3.8a) and (3.10), respectively, and other constants are

$$B = -iA \left[ 1 - \frac{2}{3} \hat{n}^2 \left( \frac{\kappa_1}{\kappa_2} + \frac{1}{5} \right) \right], \quad (3.29a)$$

$$C = \frac{2iA}{3} (1-i+b) \left[ 1 - \frac{\kappa_1}{\kappa_2} \hat{n}^2 \right], \quad (3.29b)$$

$$D = A \left\{ -\frac{1}{2} + \frac{4\kappa_1}{3\kappa_2} \hat{n}^2 - \frac{2}{9} \hat{n}^4 \left[ 2 \left( \frac{\kappa_1}{\kappa_2} \right)^2 + \frac{4\kappa_1}{5\kappa_2} + \frac{4}{35} \right] \right\} - iB, \quad (3.29c)$$

$$E = A(1-i+b) \left[ \frac{2}{15} - \frac{2\kappa_1}{3\kappa_2} \hat{n}^2 + \frac{4\kappa_1}{9\kappa_2} \hat{n}^4 \left( \frac{\kappa_1}{\kappa_2} + \frac{1}{5} \right) \right] + \frac{2iB}{3} (1-i+b) \left[ 1 - \frac{\kappa_1}{\kappa_2} \hat{n}^2 \right] - iC, \quad (3.29d)$$

and

$$A_1^{(2)} = -iA \left[ 2 - \frac{1}{3} \hat{n}^2 \left( \frac{2\kappa_1}{\kappa_2} + \frac{7}{5} + \frac{7\kappa_2}{20\kappa_1} \right) \right], \quad (3.29e)$$

$$A_1^{(5/2)} = \frac{iA}{3} (1-i+b) \left[ 3 - \hat{n}^2 \left[ 1 + \frac{2\kappa_1}{\kappa_2} \right] \right] - B(1-i+b), \quad (3.29f)$$

$$A_2^{(5/2)} = -B(1-i+b), \quad (3.29g)$$

with  $\hat{n} = n_1/n_2$ .

The last remaining task is to evaluate the volume average of the temperature field, and thus to obtain the effective compressibility or the complex wave number of the two-phase system as prescribed by (2.14), along with (2.15). In so doing, the formula

$$\frac{1}{\pi \bar{\tau}_0} \int_{\bar{\tau}_0} \sum_m \frac{e^{-2\pi i(\mathbf{k}_m \cdot \bar{\mathbf{r}})}}{k_m^2 - \frac{i}{2\pi^2}} d\bar{\mathbf{r}} = 2\pi i \quad (3.30)$$

is of great help. After some manipulations, we eventually arrive at the desired formula:

$$k + i\delta = \frac{\omega}{c_{LL}} \left[ 1 + \epsilon c_{LL}^2 \langle \rho_0 \rangle \rho_1 c_{p_1} T_0 \left( \frac{\alpha_2}{\rho_2 c_{p_2}} - \frac{\alpha_1}{\rho_1 c_{p_1}} \right)^2 \right. \\ \times \left. \left[ 1 + \frac{2i}{3} \left( \frac{\kappa_1}{\kappa_2} + \frac{1}{5} \right) \hat{n}^2 \Omega - \frac{2i\kappa_1}{3\kappa_2} (1-i+b) \hat{n}^2 \Omega^{3/2} + \frac{4\kappa_1}{3\kappa_2} \left[ 1 - \frac{1}{3} \left( \frac{\kappa_1}{\kappa_2} + \frac{2}{5} + \frac{2\kappa_2}{35\kappa_1} \right) \hat{n}^2 \right] \hat{n}^2 \Omega^2 \right. \right. \\ \left. \left. - \frac{4\kappa_1}{3\kappa_2} (1-i+b) \left[ 1 - \frac{2}{3} \left( \frac{\kappa_1}{\kappa_2} + \frac{1}{5} \right) \hat{n}^2 \right] \hat{n}^2 \Omega^{5/2} + \dots \right] \right]^{1/2}. \quad (3.31)$$

The same result is obtained if use is made of the formulas (2.19) and (2.20), which serve as a consistency check. The parameter  $b$  is defined by (3.26). It is emphasized here that  $b$  is a universal quantity representing the effect of interparticle interactions in the periodic-emulsion system. In the absence of  $b$ , (3.31) agrees with the behavior of (2.26) at low frequencies, as it should be. Furthermore, in view of (3.28), (3.31) is contributed by monopole temperature field only and in line with this fact, the diffracted or scattered compressional wave comprises an assembly of monopole sounds. What we have done is to work out the correction to the strength of point heat source due to surrounding particles.

A salient feature of our treatment is that the parameter  $b$  is not a constant but that it has a weak dependence on the frequency  $\omega$ . In the limit of  $\omega \rightarrow 0$ , it will restore the known values. Before proceeding to the discussion of numerical results, we examine the behavior of (3.31) in this limit in order to get some insight.

We define the lattice parameter  $h$  characterizing the distance between the nearest-neighbor particles, along with the basic vectors. For three types of cubic lattices we have the following.

(i) Simple cubic lattice (sc):

$$\left. \begin{matrix} \mathbf{a}^{(1)} \\ \mathbf{a}^{(2)} \\ \mathbf{a}^{(3)} \end{matrix} \right\} = h \left\{ \begin{matrix} (1,0,0) \\ (0,1,0) \\ (0,0,1) \end{matrix} \right\}, \quad \left. \begin{matrix} \mathbf{b}^{(1)} \\ \mathbf{b}^{(2)} \\ \mathbf{b}^{(3)} \end{matrix} \right\} = 1/h \left\{ \begin{matrix} (1,0,0) \\ (0,1,0) \\ (0,0,1) \end{matrix} \right\}. \quad (3.32a)$$

(ii) Body-centered cubic lattice (bcc):

$$\left. \begin{matrix} \mathbf{a}^{(1)} \\ \mathbf{a}^{(2)} \\ \mathbf{a}^{(3)} \end{matrix} \right\} = \frac{h}{2} \left\{ \begin{matrix} (1,1,-1) \\ (-1,1,1) \\ (1,-1,1) \end{matrix} \right\}, \quad \left. \begin{matrix} \mathbf{b}^{(1)} \\ \mathbf{b}^{(2)} \\ \mathbf{b}^{(3)} \end{matrix} \right\} = 1/h \left\{ \begin{matrix} (1,1,0) \\ (0,1,1) \\ (1,0,1) \end{matrix} \right\}. \quad (3.32b)$$

(iii) Face-centered cubic lattice (fcc):

$$\left. \begin{matrix} \mathbf{a}^{(1)} \\ \mathbf{a}^{(2)} \\ \mathbf{a}^{(3)} \end{matrix} \right\} = \frac{h}{2} \left\{ \begin{matrix} (1,1,0) \\ (0,1,1) \\ (1,0,1) \end{matrix} \right\}, \quad \left. \begin{matrix} \mathbf{b}^{(1)} \\ \mathbf{b}^{(2)} \\ \mathbf{b}^{(3)} \end{matrix} \right\} = 1/h \left\{ \begin{matrix} (1,1,-1) \\ (-1,1,1) \\ (1,-1,1) \end{matrix} \right\}. \quad (3.32c)$$

When the frequency is so low that the thermal skin depth around each particle contains so many particles, that is, for  $N(n_1 h) \ll 1$  and  $N(n_2 h) \ll 1$  with  $N$  being a very large number, (3.31) is reduced to

$$k + i\delta \approx \frac{\omega}{c_{LL}} \left\{ 1 + \epsilon c_{LL}^2 \langle \rho_0 \rangle \rho_1 c_{p_1} T_0 \left[ \frac{\alpha_2}{\rho_2 c_{p_2}} - \frac{\alpha_1}{\rho_1 c_{p_1}} \right]^2 \right. \\ \left. \times \left[ 1 - \frac{\kappa_1}{\kappa_2} \hat{n}^2 \epsilon + \frac{2i}{3} \left[ \frac{1}{5} + \frac{\kappa_1}{\kappa_2} (1 - d \epsilon^{1/3}) \right] + 3\epsilon \frac{\kappa_1}{\kappa_2} \left[ 1 - \frac{2}{3} \left[ \frac{\kappa_1}{\kappa_2} + \frac{1}{5} \right] \hat{n}^2 \right] \right] \hat{n}^2 \Omega + \dots \right\}^{1/2}, \quad (3.33a)$$

$$d = \begin{cases} 1.760119 & \text{for sc} \\ 1.791858 & \text{for bcc} \\ 1.791747 & \text{for fcc} \end{cases}. \quad (3.33b)$$

Here  $d$  is a constant reflecting the very long-range interactions among particles. The same values were obtained in different problems by several authors [27,28]. Furthermore, if  $\epsilon \ll 1$  in addition to the above assumptions, (3.33) may be separated into the real and imaginary parts, giving

$$c \approx c_{LL} \left\{ 1 - \frac{\epsilon}{2} c_{LL}^2 \langle \rho_0 \rangle \rho_1 c_{p_1} T_0 \left[ \frac{\alpha_2}{\rho_2 c_{p_2}} - \frac{\alpha_1}{\rho_1 c_{p_1}} \right]^2 \right. \\ \left. \times \left[ 1 - \epsilon \frac{\rho_1 c_{p_1}}{\rho_2 c_{p_2}} \right] + \dots \right\}, \quad (3.34a)$$

$$\delta \approx \frac{\epsilon}{3} c_{LL} \langle \rho_0 \rangle \rho_1 c_{p_1} T_0 \left[ \frac{\alpha_2}{\rho_2 c_{p_2}} - \frac{\alpha_1}{\rho_1 c_{p_1}} \right]^2 \\ \times \left\{ \frac{1}{5} + \frac{\kappa_1}{\kappa_2} (1 - d \epsilon^{1/3}) \right. \\ \left. + 3\epsilon \frac{\kappa_1}{\kappa_2} \left[ 1 - \frac{2}{3} \left[ \frac{\kappa_1}{\kappa_2} + \frac{1}{5} \right] \hat{n}^2 \right] \right\} \omega (n_1 a)^2 + \dots \quad (3.34b)$$

We observe from (3.34a) that the effective adiabatic compressibility for a two-phase state takes a larger value than the simply averaged value. We may say that the medium becomes softened on account of heat exchange. Equation (3.34b) states that the interparticle interactions act to diminish the attenuation coefficient and that the main correction term is essentially proportional to  $\epsilon^{1/3}$ . The magnitude of the correction terms for the three types of cubic lattices does not differ very much from each other. The reduction is more effective for bcc and fcc than for sc. We would like to stress that the ratio  $\kappa_1/\kappa_2$  is vital to the action of the interparticle interactions. The larger  $\kappa_1/\kappa_2$  is, the more influential the contribution from the surrounding particles is.

## D. Results

As noted before, the interaction parameter  $b$ , the most important quantity given by (3.26), varies with frequency. A choice of the integration contour in the complex plane is at our disposal. We have picked out two different contours, say, a straight line parallel to the real axis, and a contour composed of a straight line parallel to the imaginary axis and the real axis itself. Furthermore, a return is

made to the original formula (3.18). All of the results produced by them agree within sufficiently many digits. To bring out the features of the particle-to-particle interactions, we depict the real and imaginary parts of  $(n_2h)b$  as a function of  $n_2h$  in Figs. 3(a) and 3(b) separately. In view of (3.31), the real part of  $b$  is tied primarily to the attenuation coefficient and the imaginary part primarily to the dispersion of the sound velocity. It is worthwhile to note the limiting behavior of  $b$  as  $n_2h \rightarrow 0$ :

$$(n_2h)b = -\frac{2\pi i}{(n_2h)^2 \hat{\tau}_0} + c_1 h - (1-i)n_2 h - 2i \left[ \frac{c_2}{h} \right] (n_2h)^2 + O((n_2h)^3), \quad (3.35a)$$

with

$$c_1 h = \begin{cases} 2.837297 & \text{for sc} \\ 3.639233 & \text{for bcc} \\ 4.584862 & \text{for fcc,} \end{cases} \quad (3.35b)$$

$\hat{\tau}_0 = \tau_0/h^3$ , and  $c_2/h$  is a constant of the array.

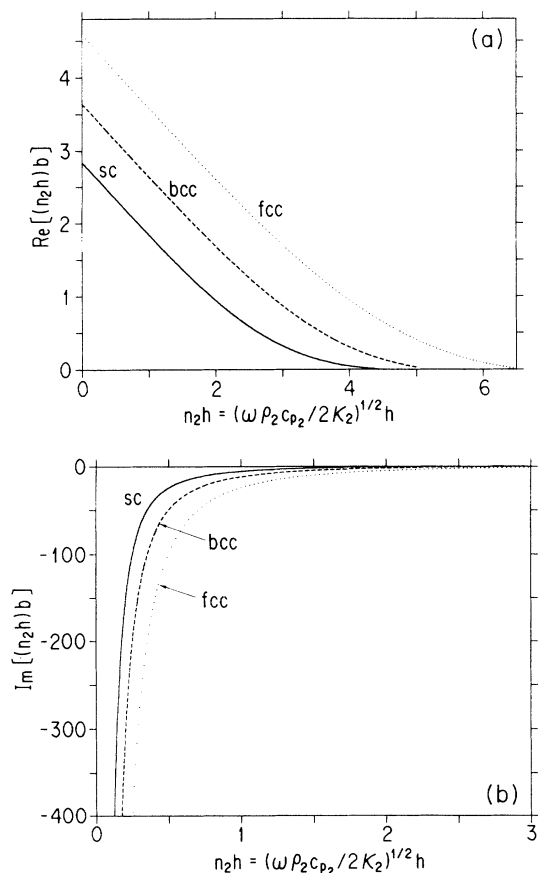


FIG. 3. The real part (a) and the imaginary part (b) of  $(n_2h)b$ , with  $b$  being the interaction parameter provided by (3.26), as functions of  $n_2h = (\omega\rho_2c_{p_2}/2\kappa_2)^{1/2}h$ , the ratio of lattice parameter  $h$  to skin depth of thermal wave in the suspending fluid. The solid line displays the result for the simple cubic (sc) array, the dashed line for the body-centered cubic (bcc) array, and the dotted line for the face-centered cubic (fcc) array.

The interaction parameter  $(n_2h)b$  exhibited in Figs. 3(a) and 3(b) is found to be given by (3.35) at very low frequencies and tends to zero as  $n_2h \rightarrow \infty$ . This is consistent with our intuition that at high frequencies, the thermal skin depth is much thinner than the particle-to-particle distances, and hence the interparticle interactions become irrelevant. The attenuation and the dispersion of sound are calculated for the real materials with the parameter values listed in Table I.

As the first example, we consider suspensions of polystyrene spheres in toluene in which the ratio  $\kappa_1/\kappa_2$  of thermal conductivity does not differ very much from unity. Figure 4(a) shows the attenuation per wavelength as a function of  $n_2a$  for typical values of volume fraction  $\epsilon$ . The simple cubic array is assumed. Figure 4(b) is the cor-

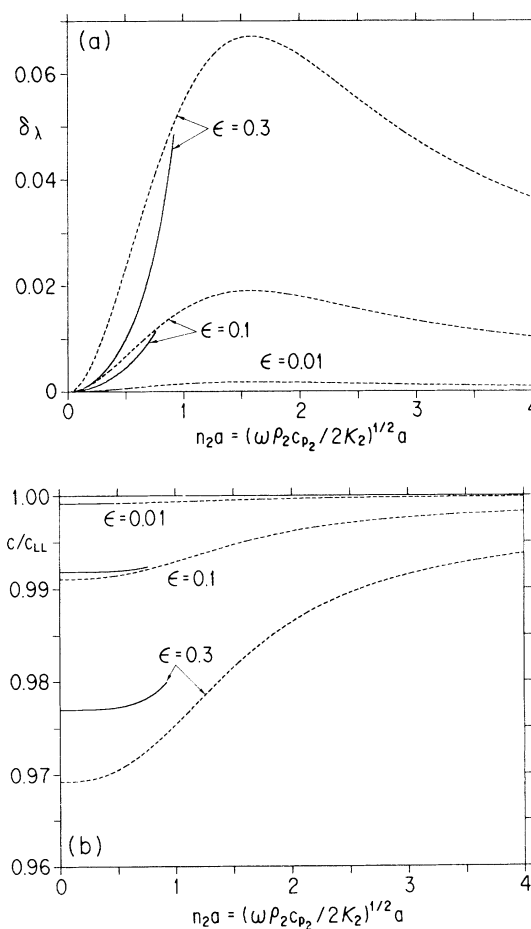


FIG. 4. Excess attenuation per wavelength  $\delta_\lambda (=2\pi\delta/k)$  (a) and normalized sound velocity  $c/c_{LL}$  (b) as functions of  $n_2a = (\omega\rho_2c_{p_2}/2\kappa_2)^{1/2}a$ , the ratio of particle radius  $a$  to skin depth of thermal wave in the suspending fluid, for a simple cubic (sc) lattice of polystyrene spheres suspended in toluene at 20°C with typical values of volume fraction  $\epsilon$  (solid lines). Here  $c_{LL}$  is the high-frequency limit of the sound velocity and is given by (2.13). The values of  $\epsilon$  are indicated on each line. The results are obtained from (3.31) and plotted on a linear-linear scale. The dashed lines display Isakovich's results, which ignore particle-to-particle interactions, as given by (2.26a) and (2.26b).

responding graph for the sound velocity normalized by  $c_{LL}$ . The results for a periodic suspension are drawn by solid lines. For comparison, we show the results obtained from Isakovitch's theory for independent particles by dashed lines. At first glance, we see that the attenuation coefficient grows, roughly speaking, in proportion to the volume fraction. The same is true of the deviation of the sound speed from the limiting value  $c_{LL}$  as  $\omega \rightarrow \infty$ . The particle-to-particle interactions come into play at low frequencies, say,  $n_1 a \sim n_2 a < 1$ . The interactions necessarily act to reduce the attenuation coefficient. The deviation of the sound speed from  $c_{LL}$  is reduced by the interactions. The interactions become quite influential for a volume fraction such as  $\epsilon = 0.3$ . It is noteworthy that for  $\epsilon = 0.3$ , the interactions bring in a drastic modification of Isakovitch's values for both attenuation and dispersion.

Figures 5–7 extend Fig. 4(a) to a wider frequency range and are plotted against  $\log(n_2 a)$ . Figure 5 is drawn for the case of  $\epsilon = 0.01$ , Fig. 6 for  $\epsilon = 0.1$ , and Fig. 7 for  $\epsilon = 0.3$ . In order to illustrate an advantage of our scheme that takes the variation of thickness of the thermal boundary layer into consideration, we show, by dotted lines, the results of very-low-frequency behavior supplied by (3.33). It is conspicuous from Fig. 7 that the attenuation is substantially modified by the interparticle interactions. Besides, the same figure clarifies that inclusion of the variation of the boundary-layer thickness is indispensable for producing satisfactory results. But this is not the case with dilute suspensions as shown by Fig. 5 ( $\epsilon = 0.01$ ) and Fig. 6 ( $\epsilon = 0.1$ ).

The correction from the interactions depends slightly on the structure of the cubic lattice. We make a comparison between sc, bcc, and fcc, though the dependence is of little practical importance. Figures 8(a) and 8(b) display

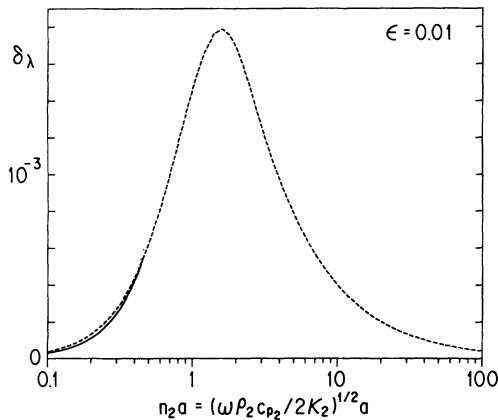


FIG. 5. Excess attenuation per wavelength  $\delta_\lambda (= 2\pi\delta/k)$  as a function of  $n_2 a$ , the ratio of particle radius  $a$  to skin depth of thermal wave in the suspending fluid, for a simple cubic (sc) lattice of polystyrene spheres suspended in toluene at 20°C with volume fraction  $\epsilon = 0.01$  (solid line). The result is obtained from (3.31) in a wider frequency range and plotted on a semilog scale. The dashed line displays Isakovitch's result for independent particles, as given by (2.26a) and (2.26b). The dotted line shows the very-low-frequency behavior (3.33a) and (3.33b) obtained by ignoring frequency-dependence of skin depth of thermal wave.

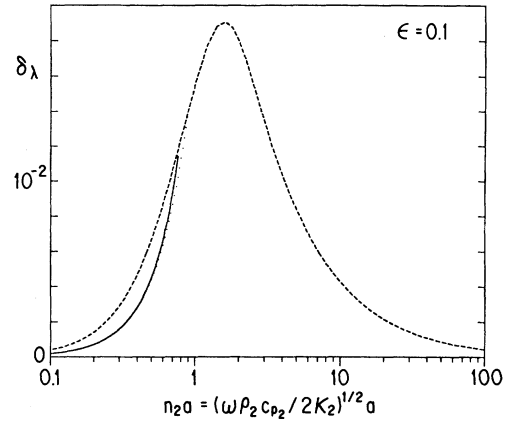


FIG. 6. As Fig. 5, but for  $\epsilon = 0.1$ .

the attenuation per wavelength and the normalized sound velocity, respectively, for  $\epsilon = 0.3$ . The difference between bcc represented by a broken line and fcc marked with crosses is hard to discern. As for the reduction in attenuation, we admit a larger influence for bcc and fcc than for sc. In contrast, regarding the sound velocity, sc undergoes a larger influence than bcc and fcc.

In the above, our argument has been directed exclusively toward suspensions composed of two substances with comparable thermal conductivity. Expressions (3.31) and (3.33) suggest that the interaction is fairly sensitive to the ratio  $\kappa_1/\kappa_2$  of the thermal conductivity. Subsequently, we shed light on this aspect.

In Figs. 9(a) and 9(b) we present the attenuation per wavelength and the normalized sound velocity for an aqueous suspension of polystyrene spheres of 30% by volume arranged in a sc lattice. The characteristic of the ambient water is its large thermal conductivity. Therefore,  $\kappa_1/\kappa_2$  takes a small value in this system. We learn from Fig. 9(a) that the neighboring particles do not intervene so much in the heat-exchange process despite the rather high concentration.

The opposite situation is demonstrated by Figs. 10(a) and 10(b), where the sound attenuation and velocity are calculated for a 5% emulsion of water globules in toluene. This is an example of large  $\kappa_1/\kappa_2$  in comparison

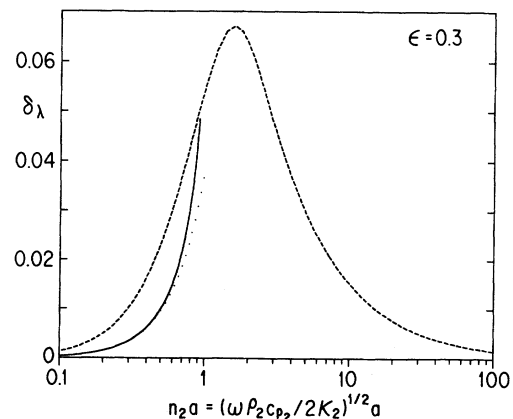


FIG. 7. As Fig. 5, but for  $\epsilon = 0.3$ .

with unity. Although this example seems to be somewhat artificial, it discloses rich details. In a usual sense, this example appears to belong to a dilute emulsion. But Fig. 10(a) implies that the independent-particle approximation breaks down. Therefore, we have to be careful in identifying a small volume fraction of suspended particles with a dilute emulsion. The substantial thermal effect in such a dilute emulsion originates from two sources, which can be seen in (3.34b). First, there is a correction proportional to  $\epsilon^{1/3}$ , which acts to suppress the attenuation. The dependence of  $\epsilon^{1/3}$  is attributed to the ordered structure of dispersed particles. The second correction term, which is proportional to  $\epsilon$ , is negative in sign in this case, and thus contributes to reinforce the action of reduction. It should be borne in mind that these cooperative effects result from the largeness of  $\kappa_1/\kappa_2$ . In a higher-frequency region, we notice that the solid line deflects downward from the dotted line as opposed to the previous examples.

This fact may reflect the undue truncation of our expansions, so it is necessary to implement the higher-order expansions in order to obtain satisfactory values at higher frequencies.

We conceive the mechanism of the reduction of attenuation in the following way. Under a sound wave in a heterogeneous medium, adiabatic temperature variations, different in different substances, immediately follow in response to compression or expansion of the substances. Then the heat exchange arises between an emulsion particle and the ambient medium. The strength of the heat flow is proportional to the temperature gradient. The heat flow produces the entropy production or the energy dissipation in proportion to the volume integral of the square of the temperature gradient. When the temperature of the particles happens to be raised more than that of the ambient medium, the heat flows out from the particles. Viewed globally, it implies that the neighboring par-

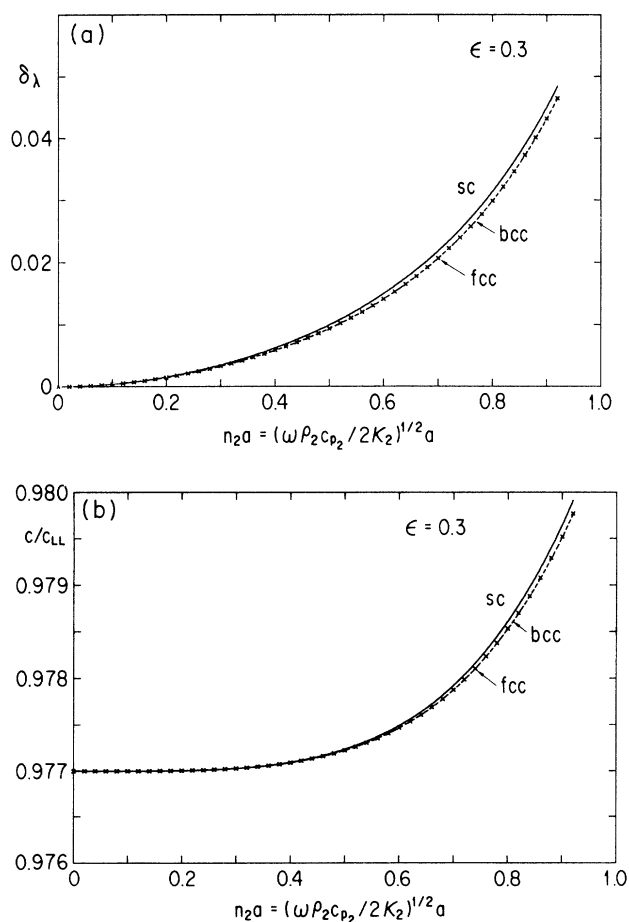


FIG. 8. Excess attenuation per wavelength  $\delta_\lambda (=2\pi\delta/k)$  (a) and normalized sound velocity  $c/c_{LL}$  (b) as functions of  $n_2 a$ , the ratio of particle radius  $a$  to skin depth of thermal wave in the suspending fluid, for suspensions of 30% by volume polystyrene in toluene at 20°C, as calculated from (3.31). Here  $c_{LL}$  is the high-frequency limit of the sound velocity and is given by (2.13). The solid line is for the simple cubic (sc) array, the dashed line is for the body-centered cubic (bcc) array, and crosses are for the face-centered cubic (fcc) array. The discrepancy between bcc and fcc is not discernible.

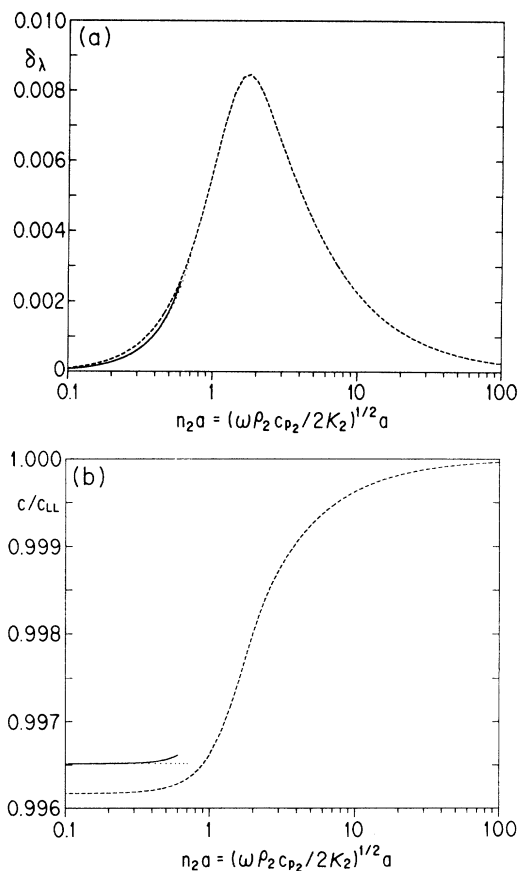


FIG. 9. Excess attenuation per wavelength  $\delta_\lambda (=2\pi\delta/k)$  (a) and normalized sound velocity  $c/c_{LL}$  (b) as functions of  $n_2 a$ , the ratio of particle radius  $a$  to skin depth of thermal wave in the suspending fluid, for an aqueous suspension of a simple cubic (sc) lattice of toluene globules at 20°C with volume fraction  $\epsilon=0.3$  (solid line). Here  $c_{LL}$  is the high-frequency limit of the sound velocity and is given by (2.13). The result is obtained from (3.31). The dashed line displays Isakovitch's result for independent particles, as given by (2.26a) and (2.26b). The dotted line shows the very-low-frequency behavior (3.33a) and (3.33b) obtained by ignoring frequency dependence of skin depth of thermal wave.

ticles supply each particle with heat. The opposite is true when the particle temperature is lower. It follows that for given density variations of the particles and the medium, the magnitude of temperature gradient in the interacting-particle system is smaller than that in the independent-particle system. As a result, the sound attenuation is smaller in the interacting-particle system than in the independent-particle system.

The many-particle effect is less remarkable in the sound velocity than in the attenuation. In a heterogeneous system, Isakovitch obtained a certain decrease of the sound speed  $c$  from  $c_{LL}$ , that is, the proper spatial average of the adiabatic compressibility. Our analysis takes into consideration the many-particle effect neglected in Isakovitch's theory. The decrease of  $c$  from  $c_{LL}$  is reduced by our many-particle effect. Since the reduction is small, we skip the physical explanation for this reduction.

It is emphasized here that the correction due to the many-particle effect in the water-in-toluene emulsion is very large even in the dilute case of  $\epsilon=0.05$ , while in the toluene-in-water emulsion, the effect is small even in the dense case of  $\epsilon=0.3$ . We note that  $\hat{n}$  is not very much

different from unity. In these examples,  $\kappa_{\text{water}}$  is much larger than  $\kappa_{\text{toluene}}$  and therefore the droplet-to-droplet interactions are mainly controlled by  $\kappa_1$ , the heat conductivity of the droplet.

The clue to comprehending the sensitive dependence of the correction on  $\kappa_1/\kappa_2$  lies in the expressions (2.19) or (2.22). The energy dissipation or the complex wave number is corrected by the heat conduction through the product of the temperature discrepancy (2.23) or (3.8a) caused adiabatically and the resulting interfacial heat flux  $q_i$ . Plainly, the adiabatic temperature variation has no connection with the heat-conduction process as evidenced by (3.8a). The heat flux is determined by the solution of the coupled equations (3.4a) and (3.4b) complying with the boundary condition (2.18). But in our formulation, the temperature gradient  $\partial T_1^{(k)}/\partial r$  at the order of  $\Omega^k$  is determined solely by the distribution of  $T_1^{(k-1)}$  and is thus irrelevant to the temperature distribution  $T_2^{(k)}$  of the same order in the suspending fluid. In fact, (3.4a) and the regularity condition yield

$$\frac{\partial T_1^{(k)}}{\partial r} = -\frac{2i\hat{n}^2\Omega}{r^2} \int_0^r \xi^2 T_1^{(k-1)}(\xi) d\xi. \quad (3.36)$$

Because of (3.8a),  $\partial T_1^{(1)}/\partial r$  has no dependence on  $\kappa_1$  or  $\kappa_2$ , so the interfacial heat flux  $\kappa_1 \partial T_1^{(1)}/\partial r$  is proportional to  $\kappa_1$ . This is the reason why the heat source  $A$  in (3.12) is proportional to  $\kappa_1$ . All of the suspended particles are then endowed with the same heat source. They coherently excite the locally uniform temperature field  $T_1^{(3/2)} = T_2^{(3/2)} = -A(1-i+b)$  of  $O(\Omega^{3/2})$ . In consequence,  $\partial T_1^{(5/2)}/\partial r|_{r=1} = 2iA(1-i+b)\hat{n}^2/3$ . The parameter  $b$ , which measures the influence of the interparticle interactions, is coupled in  $\kappa_1 \partial T_1^{(5/2)}/\partial r$  with  $\kappa_1 A$ , the product of the thermal conductivity  $\kappa_1$ , and the strength of the heat source. The basic mechanism expressed by temperature gradient (3.36) at the interface, which appears to be irrespective of that of the suspending fluid, together with the temperature discrepancy  $\Theta$  driven adiabatically, gives rise to the selective importance of  $\kappa_1$  in manifesting the mutual interactions between the suspended particles. It is to be understood that the uneven roles of  $\kappa_1$  and  $\kappa_2$  are peculiar to the shape of the minority phase.

#### IV. SUMMARY AND CONCLUSION

We have investigated, in this paper, the coefficient of sound attenuation and the dispersion relation of the sound velocity in suspensions and emulsions. Otherwise stated, we have worked out the effective compressibility of heterogeneous media. Following several considerations, we have disregarded the viscous dissipation and spotlighted the process of the interfacial heat transfer. A great simplification has been achieved by taking advantage of Isakovitch's framework.

We provide a theoretical justification of the Isakovitch formula, which produces the attenuation coefficient and the sound velocity at the same time. For this, we start with the entropy production formula rather than the density variation  $\delta\rho$  or the compressibility. After the

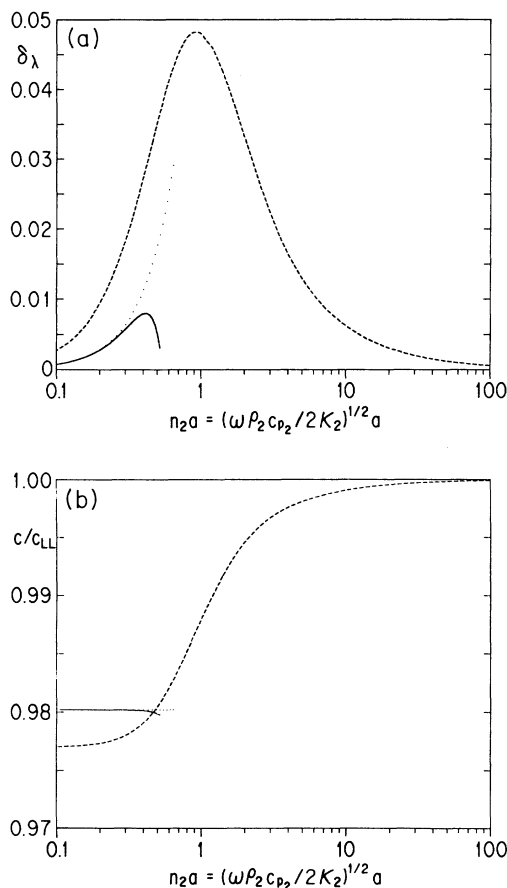


FIG. 10. Excess attenuation per wavelength  $\delta_\lambda (=2\pi\delta/k)$  (a) and normalized sound velocity  $c/c_{LL}$  (b) as functions of  $n_2a$  for a simple cubic (sc) lattice of water globules suspended in toluene at 20°C with volume fraction  $\epsilon=0.05$  (solid line). The dashed line displays Isakovitch's result for independent particles. The dotted line shows the very-low-frequency behavior (3.33a) and (3.33b).

rederivation, we rewrite his formula in terms of the interfacial heat flux and the adiabatically driven temperature difference between two phases. This version helps to realize the underlying physical mechanism of heat transfer in the presence of the compressional wave.

The problem of a single spherical droplet immersed in a fluid of different species, which was originally solved by Isakovich, is revisited. As shown by him, the attenuation coefficient  $\delta$  behaves as  $\omega^2$  at very low frequencies [ $(n_1 a)^2 \ll 1$ ], while it behaves as  $\omega^{1/2}$  at very high frequencies [ $(n_1 a)^2 \gg 1$ ]. In addition, we uncover the existence of a new scaling regime for emulsions with the material property of  $\chi_1 \ll \chi_2$ . What we have found is that  $\delta$  is proportional to  $\omega^0$  and  $(1 - c/c_{LL})$  is proportional to  $\omega^{-1}$  at frequencies satisfying  $n_1 a \gg 1$  and  $n_2 a \ll 1$  with the condition  $n_1 a \gg \kappa_2/\kappa_1$  being necessary. A suspension of polystyrene spheres in air exemplifies it. It is worth emphasizing that this behavior has much in common with the low-frequency behavior in mixture undergoing the first-order phase transition in the thin interface [16]. In both cases, the heat-conduction process is virtually subjected, at frequencies in question, to the boundary condition that the temperature is fixed at the interface.

The second half of this paper is concerned with the elucidation of the effect of multiparticle interactions upon the thermal attenuation process. We have dealt with a lattice of spherical particles or droplets embedded in a medium of another liquid. Isakovich's framework, which requires the functional forms of the temperature distribution both inside and outside the particle, has much in its favor. They are derived in the form of expansions in powers of  $\Omega^{1/2}$ , using the method of matched asymptotic expansions. The correction coming from the multiparticle interactions is then obtainable through the fast-convergent lattice sum of the contour integrals in the complex plane. Here we accomplish an extension of Ewald's method to the heat-conduction problem. The numerical calculation is implemented for the typical cubic lattices, namely, sc, bcc, and fcc lattices. To enlighten the sensitive dependence of the interactions upon the ratio  $\kappa_1/\kappa_2$  of the thermal conductivity, we pick up, as examples, an aqueous polystyrene lattice ( $\kappa_1 < \kappa_2$ ), a suspension of polystyrene particles in toluene ( $\kappa_1 \sim \kappa_2$ ), and an emulsion of water in toluene ( $\kappa_1 > \kappa_2$ ). Our results are summarized as follows.

(i) In general, the interparticle interactions act to suppress both the sound attenuation and the deviation of the sound velocity from the high-frequency limit.

(ii) At such high frequencies that the thermal skin depth is much thinner than the particle-to-particle distances, the effect of the neighboring particles is inconse-

quential.

(iii) In contrast, at very low frequencies, there is a significant correction, depending upon the value of  $\kappa_1/\kappa_2$ , to the attenuation coefficient whose dominant term is proportional to  $\epsilon^{1/3}$  with  $\epsilon$  being the volume fraction of the minority phase. The reduction in attenuation is more effective for bcc and fcc than for sc. In accordance with it, the deviation of the sound velocity is relaxed, but sc is more susceptible to it than bcc and fcc, as opposed to the attenuation. In either event, the difference is not large enough to be of practical importance.

(iv) For moderate volume fractions, the consideration of the skin-depth variation with frequency achieves an improvement in quantitative accuracy.

(v) The most relevant physical parameter prompting the interparticle interactions is the thermal conductivity  $\kappa_1$  of the suspended substance. When  $\kappa_1/\kappa_2$  is large compared with unity, the word "dilute," with respect to  $\epsilon$ , no longer makes sense. Even if the concentration of water is a few percent in toluene, the attenuation curve exhibits a marked difference with Isakovich's result. The monopole temperature field or the point source emanating from each particle participates in the attenuation and the modification of sound speed. This fact accounts for the relative importance of  $\kappa_1$ .

Here, a question is raised as to the contribution from the viscous force. As  $\epsilon$  and/or  $\kappa_1/\kappa_2$  are increased, the attenuation goes through a considerable reduction from Isakovich's result. If so, we cannot dismiss the possibility that the viscous dissipation and the thermal conduction exchange roles. In reality, there is no doubt that an increase of  $\epsilon$  causes an increase of the force exerted on the particles and therefore an increase of the contribution from the viscous dissipation. At present, it does not seem to be straightforward to incorporate the viscous dissipation into the formula (2.14).

Our choice of periodic emulsions of spherical particles is not sufficient to have much practical bearing. A recent experiment revealed that for aqueous suspensions of polystyrene spheres, the occurrence of aggregation phenomena, on the contrary, promotes the attenuation in the low-frequency range [12]. It implies that the sound attenuation may be quite sensitive to the distribution of particles. Furthermore, for emulsions, the minority phase cannot always be reckoned to be of spherical shape. These and other factors call for individual treatment. For this, the alternative formula (2.19) and (2.20) will be helpful.

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