An extended coupled phase theory for the sound propagation in polydisperse concentrated suspensions of rigid particles

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An extension of the classical coupled phase theory is proposed to account for hydrodynamic interactions between neighboring rigid particles, which are essential to describe properly the sound propagation in concentrated suspensions. Rigorous ensemble-averaged equations are derived for each phase and simplified in the case of acoustical wave propagation. Then, closure is achieved by introducing a self-consistent scheme originally developed by Buyevich and Shchelchkova [Prog. Aerosp. Sci. **18**, 121–151 (1978)] for incompressible flows, to model the transfer terms between the two phases. This provides an alternative to the effective medium self-consistent theory developed by Spelt *et al.* [J. Fluid Mech. **430**, 51–86 (2001)] in which the suspension is considered as a whole. Here, a significantly simpler formulation is obtained in the long wavelength regime. Predictions of this self-consistent theory are compared with the classical coupled phase theory and with experimental data measuring the attenuation in concentrated suspensions of silica in water. Our calculation is shown to give a good description of the attenuation variation with volume fraction. This theory is also extended to the case of polydisperse suspensions. Finally, the link between the self-consistent theory and the different orders of the multiple scattering theory is clarified. [DOI: 10.1121/1.2723648]

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I. INTRODUCTION

The propagation of sound waves through dilute suspensions of different natures has been the subject of many studies since the pioneering article of Sewell[1] in 1910 who considered immovable rigid particles suspended in a gas. Several phenomena can be involved in the attenuation and dispersion of sound, depending on the particles nature and the wave frequency. In hydrosols, the acoustic damping is mainly induced by the visco-inertial terms (Lamb[2]).

To study the influence of particles on the sound propagation in suspensions, two methods have been principally developed. First, the scattering theory, also called ECAH theory based on the work of Epstein and Carhart[3] and Allegra and Hawley.[4] In this model, a spherical particle is considered and the waves propagating inside and outside the particle are decomposed into three modes: compressional, shear, and thermal ones. Potentials are expressed in terms of Bessel functions series satisfying the boundary conditions at the particle surface. Second, there is the coupled phase theory[5,6] based on the two-phase hydrodynamic equations. The primary advantage of the scattering theory is to be valid over the whole frequency range, although some difficulties arise with the series truncation due to the nonuniform convergence of the Bessel series.[7] The coupled phase theory gives a good framework to incorporate phenomena that would be difficult to include in the scattering theory such as mass transfers or chemical reactions. Moreover, it also leads to an explicit dispersion equation that is simpler to interpret physically and calculate, which can be useful when dealing with the inverse problem.

These theories agree well with experimental data in dilute suspensions, but they both neglect some parts, of the "multiple scattering" whose importance increases with concentration. The coupled phase theory inherently integrates "multiple scattering" but only at first order (cf. Sec. II C). Thus, it neglects interactions that occur when the viscous or/and thermal boundary layer of neighboring particles overlap one another. The ECAH theory was originally considering a simple superposition of each particle contribution and was therefore not considering "multiple scattering."

To incorporate the "reverberant multiple scattering" (in the sense of geometrical redirection of energy), the multiple scattering theory[8–13] has been introduced into the ECAH theory.[14]

To account for "dissipative multiple scattering," that is to say overlapping of thermal waves in the case of emulsions, Hemar *et al.*[15] have introduced a so-called "core-shell model:" the particle is surrounded by a cell of pure fluid,

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where the presence of the particle is rather unlikely, which is itself embedded in an effective medium. Such models show a good agreement with experiments, but they require the introduction of several unknown parameters. The first one is the radius of the pure medium cell b. Many investigators choose $b=a/\alpha_d^{1/3}$ (where a is the particle radius and α_d is the volume fraction occupied by the particles), although this value is not appropriate for randomly distributed spheres. It would be more adequate for systems in which the distance between particles is almost uniform (cf. Refs. 16 and 17). Then, the effective properties of the medium must also be introduced. In the work by Hemar *et al.* and in the article by Hipp *et al.*,[18] the volume averaged parameters are introduced, but one could also choose a different set of effective parameters. The primary advantage of the present study is that the effective properties are introduced in a consistent way. One can note that the multiple scattering theory and the "core-shell model" have been merged in the article by McClements and Hermann.[19] The results of this model have then been compared in detail with the classical coupled phase theory by Evans and Attenborough.[6]

Finally, and to the authors' knowledge, the only articles dealing with the overlapping of visco-inertial potentials are the articles by Dukhin and Goetz[20] and by Spelt *et al.*[17] The first one is the so-called cell model,[21,22] which is based on empirical grounds. Although it provides a relatively good estimation of viscous interactions between neighboring particles, there is no rigorous justification of this procedure. Moreover, it does not integrate the intrinsic (bulk) losses in the formulation. The second one, as the present work, uses the self-consistent approximation, but the suspension is considered as a whole. Its advantage is to give a theory that is valid whatever the frequency of the incident wave and the nature of the particle. The present work is limited to the long wavelength regime (LWR) but provides a simpler expression to describe the attenuation and dispersion in suspensions of rigid particles. This work also differs in the expression of the closure relations that are not limited to plane waves. These considerations will be developed in Sec. II D 4.

In this paper, we will first derive the ensemble averaged conservation equations and simplify them in the case of acoustical wave propagation. Then we will introduce the self-consistent scheme originally developed by Buyevich[23,24] for an incompressible flow and we will discuss the link between the closure assumptions introduced here in the effective medium theory (EMT), and the one introduced in the multiple scattering theory (MST). Once the semi-analytical dispersion equation is established, the results are compared to the experimental data of Hipp *et al.*[25] Finally, the equations will be extended to the case of polydisperse solutions.

II. THEORY

In the coupled phase theory, averaged conservation equations are written down separately for each phase. These equations are coupled by the transfer terms between the two phases. In the case of rigid particles in a liquid matrix, the thermal transfer and intrinsic absorption can be neglected as they are both proportional to γ_c-1, where γ_c is the specific heat ratio of the continuous phase that is almost equal to one in liquids. Thus, only the mass and momentum conservation equations are required. In this section, we will consider a monodisperse suspension of spherical rigid particles isotropically distributed.

A. Ensemble-averaged equations

The ensemble-averaged equations are calculated from the local constitutive equations of the continuous and dispersed phases by using a configurational average.[24]

If the interfaces between the two phases have no mass, one can introduce the generalized functions for density and momentum based on the "fine-grained" definition in each phase under the form

$$\begin{bmatrix} \rho' \\ \rho' \boldsymbol{v}' \end{bmatrix} = \sum_{k=c,d} \chi_k \begin{bmatrix} \rho_k' \\ \rho_k' \boldsymbol{v}_k' \end{bmatrix},$$

where c,d denote respectively the continuous and dispersed phase, ρ_k' and $\boldsymbol{v}_k'$ are the "fine grained" density and velocity, and χ_k is the phase function defined by

$$\chi_k(\boldsymbol{x},t) = \begin{cases} 1 & \text{if } \boldsymbol{x} \text{ is in phase } k \text{ at time } t, \\ 0 & \text{otherwise,} \end{cases} \tag{1}$$

with of course $\chi_c=1-\chi_d$.

If interface forces due to the surface tension are neglected and no external force field is considered, the generalized mass and momentum conservation equations can be written under the form

$$\frac{\partial \rho'}{\partial t} + \operatorname{div}(\rho' \boldsymbol{v}') = 0, \tag{2}$$

$$\frac{\partial}{\partial t}(\rho' \boldsymbol{v}') + \operatorname{div}(\rho' \boldsymbol{v}' \otimes \boldsymbol{v}') = \operatorname{div}(\boldsymbol{\Pi}'), \tag{3}$$

where $\boldsymbol{\Pi}'$ is the generalized stress tensor:

$$\boldsymbol{\Pi}' = \sum_{k=c,d} \chi_k \boldsymbol{\Pi}_k',$$

Π_k' being the stress tensor in the kth phase.

To obtain separate averaged equations for each phase, Eqs. (2) and (3) are multiplied by the phase function and then averaged via the configurational average:

$$\langle G'(\boldsymbol{x},t) \rangle = \int G'(\boldsymbol{x},t|\boldsymbol{x}_1,\ldots,\boldsymbol{x}_N) p(t,\boldsymbol{x}_1,\ldots,\boldsymbol{x}_N) d\boldsymbol{x}_1 \cdots d\boldsymbol{x}_N;$$

where $p(t,\boldsymbol{x}_1,\cdots,\boldsymbol{x}_N)dx_1 \cdots dx_N$ is the probability of finding the first particle center in the vicinity of $\boldsymbol{x}_1$ at t, while at the same time the second particle is in the vicinity of $\boldsymbol{x}_2$ and so forth. Considering indistinguishable particles, this expression can be rewritten as the probability $p(t,C_N)dC_N$ of finding the N particles in the vicinity of $C_N=(\boldsymbol{x}_1,\ldots,\boldsymbol{x}_N)$, regardless of their order.

The left hand side of Eqs. (2) and (3) are of the form

$$\frac{\partial G'}{\partial t} + \mathrm{div}(G'\boldsymbol{v}').$$

Providing that the fluctuations $G''=G'-\langle G'\rangle$ of the local field G' relative to the mean field $\langle G'\rangle$ are neglected and as long as no phase change occurs, the previously described average yields[26]

$$\left\langle \chi_k \left[\frac{\partial G'}{\partial t} + \mathrm{div}(G'\boldsymbol{v}') \right] \right\rangle = \frac{\partial}{\partial t}(\alpha_k G_k) + \mathrm{div}(\alpha_k G_k v_k), \tag{4}$$

with $\alpha_k=\langle\chi_k\rangle$ the mean volume fraction occupied by phase k, and G_k the phasic average of the variable G':

$$G_k = \langle \chi_k G'\rangle/\alpha_k. \tag{5}$$

The next fundamental step is to express $\langle\chi_c\mathrm{div}(\boldsymbol{\Pi}')\rangle$ and $\langle\chi_d\mathrm{div}(\boldsymbol{\Pi}')\rangle$ in terms of quantities that we will be able to calculate for a test particle in order to achieve closure. Our derivation is based on the theory developed by Buyevich,[24,27] here modified to account for the compressibility of the continuous phase as required for sound propagation. From $\chi_c = 1-\chi_d$, we easily obtain

$$\langle\chi_c\mathrm{div}(\boldsymbol{\Pi}')\rangle = \mathrm{div}\langle\boldsymbol{\Pi}'\rangle - \langle\chi_d\mathrm{div}(\boldsymbol{\Pi}')\rangle \quad \text{with } \langle\boldsymbol{\Pi}'\rangle = \langle\chi_c\boldsymbol{\Pi}'\rangle + \langle\chi_d\boldsymbol{\Pi}'\rangle. \tag{6}$$

In the continuous phase, the "fine-grained" stress tensor expression is the one of a Newtonian fluid, and thus

$$\langle\chi_c\boldsymbol{\Pi}'\rangle = -\alpha_c p_c \boldsymbol{I} + 2\mu_c\langle\chi_c\boldsymbol{D}'\rangle + \lambda_c\langle\chi_c\mathrm{div}(\boldsymbol{v}')\rangle\boldsymbol{I}, \tag{7}$$

where p_c is the phasic average of the local pressure, $\boldsymbol{D}'$ is the strain rate tensor, $\boldsymbol{I}$ is the unit tensor, and μ_c and $\zeta_c=\lambda_c+2\mu_c/3$ are respectively the shear and bulk viscosities of the continuous phase.

Owing to the rigidity of the particles, the strain rates $\boldsymbol{D}'$ and the volume variation $\mathrm{div}(\boldsymbol{v}')$ vanish inside the particle, and thus we obtain

$$\langle\chi_c\boldsymbol{D}'\rangle = \langle\boldsymbol{D}'\rangle - \langle\chi_d\boldsymbol{D}'\rangle = \langle\boldsymbol{D}'\rangle \equiv \boldsymbol{D}, \tag{8}$$

$$\langle\chi_c\mathrm{div}(\boldsymbol{v}')\rangle = \mathrm{div}\langle\boldsymbol{v}'\rangle - \langle\chi_d\mathrm{div}(\boldsymbol{v}')\rangle = \mathrm{div}\langle\boldsymbol{v}'\rangle = \mathrm{div}(\boldsymbol{v}), \tag{9}$$

with $\boldsymbol{v}=\alpha_c\boldsymbol{v_c}+\alpha_d\boldsymbol{v_d}$ and $\boldsymbol{D}=1/2(\nabla\boldsymbol{v}+\nabla^t\boldsymbol{v})$.

From Eqs. (7)–(9), we get

$$\langle\boldsymbol{\Pi}'\rangle = -\alpha_c p_c\boldsymbol{I} + 2\mu_c\boldsymbol{D} + \lambda_c\mathrm{div}(\boldsymbol{v})\boldsymbol{I} + \langle\chi_d\boldsymbol{\Pi}'\rangle. \tag{10}$$

Finally, by taking into acount (4), (6), and (10) in Eqs. (2) and (3), the following system of mass and momentum conservation stands:

$$\frac{\partial}{\partial t}(\alpha_c\rho_c) + \mathrm{div}(\alpha_c\rho_c\boldsymbol{v_c}) = 0, \tag{11}$$

$$\frac{\partial}{\partial t}(\alpha_d\rho_d) + \mathrm{div}(\alpha_d\rho_d\boldsymbol{v_d}) = 0, \tag{12}$$

$$\frac{\partial}{\partial t}(\alpha_c\rho_c\boldsymbol{v_c}) + \mathrm{div}(\alpha_c\rho_c\boldsymbol{v_c}\otimes\boldsymbol{v_c}) = \mathrm{div}(\boldsymbol{\Pi}) - \boldsymbol{F}, \tag{13}$$

$$\frac{\partial}{\partial t}(\alpha_d\rho_d\boldsymbol{v_d}) + \mathrm{div}(\alpha_d\rho_d\boldsymbol{v_d}\otimes\boldsymbol{v_d}) = \boldsymbol{F}, \tag{14}$$

with the expressions of the effective stress tensor $\boldsymbol{\Pi}=\langle\boldsymbol{\Pi}'\rangle$ given by Eq. (10) and the effective force $\boldsymbol{F}$ given by

$$\boldsymbol{F} = \langle\chi_d\mathrm{div}(\boldsymbol{\Pi}')\rangle. \tag{15}$$

B. The test particle problem

Thus, only the quantities $\langle\chi_d\boldsymbol{\Pi}'\rangle$ and $\langle\chi_d\mathrm{div}(\boldsymbol{\Pi}')\rangle$ remain to be expressed in terms of the averaged fields to achieve closure. To address this issue, the link between these expressions and the so-called test particle problem must be established. Let us introduce some notations that will be useful in this problem.

First, the conditional averages with one or two (or more) particles positions being known are defined by

$$\langle G'\rangle_{\boldsymbol{x}'}(\boldsymbol{x},t) = \int G'(\boldsymbol{x},t|C_N)p(t,C_{N-1}|\boldsymbol{x}')\,dC_{N-1},$$

$$\langle G'\rangle_{\boldsymbol{x}',\boldsymbol{x}''}(\boldsymbol{x},t) = \int G'p(t,C_{N-2}|\boldsymbol{x}',\boldsymbol{x}'')\,dC_{N-2}.$$

Then, we can introduce the unconditional probability density $p(t,\boldsymbol{x}')$ of finding one of the N sphere centers in $\boldsymbol{x}'$ at t, and $p(t,\boldsymbol{x}';\boldsymbol{x}'')$ the same probability but conditioned by the presence of another sphere center in $\boldsymbol{x}''$:

$$p(t,\boldsymbol{x}') = \sum_{j=1}^{N}\int\cdots\int p(t,C_N)_{x^j=\boldsymbol{x}'}\prod_{i\neq j}d\boldsymbol{x}^i,$$

$$p(t,\boldsymbol{x}';\boldsymbol{x}'') = \sum_{j\neq k}\int\cdots\int p(t,C_N)_{x^j=\boldsymbol{x}',x^k=\boldsymbol{x}''}\prod_{i\neq j,k}d\boldsymbol{x}^i.$$

We can note that $p(t,\boldsymbol{x}')$ is nothing but the mean concentration number of particles by volume, which will be noted $n(t,\boldsymbol{x}')$ in the rest of this paper.

From the above definitions, Buyevich and Shchelchkova[24] establish the link between quantities averaged over the dispersed phase and integrals over a test particle surface or volume:

$$\langle\chi_d G'\rangle(\boldsymbol{x},t) = \int_{|\boldsymbol{x}-\boldsymbol{x}'|\leq a} n(t,\boldsymbol{x}')\langle G'\rangle_{\boldsymbol{x}'}(\boldsymbol{x},t)\,d\boldsymbol{x}'. \tag{16}$$

With this equation and providing that the macroscopic scale L (that is to say, the wavelength λ in acoustics) is much larger than the radius a of the particle, they obtain the following formula:

$$\alpha_d(t,\boldsymbol{x}) = \langle\chi_d\rangle(t,\boldsymbol{x}) \approx 4/3\,\pi a^3 n(t,\boldsymbol{x}), \tag{17}$$

$$\boldsymbol{F} = \langle\chi_d\mathrm{div}(\boldsymbol{\Pi}')\rangle \approx \frac{3\alpha_d}{4\pi a^3}\oint\langle\boldsymbol{\Pi}'\rangle_x\cdot\mathbf{n}\,dS, \tag{18}$$

where $\mathbf{n}$ is the normal vector. Equation (18) clearly shows that $\boldsymbol{F}$ is nothing but the force applied on a test sphere by a fictitious medium, whose properties significantly differ from

the pure ambient fluid, as they include the influence of the other distributed spheres.

Finally, with a complex reasoning that we will not reproduce here, they prove that if no external torque acts on the particle, and the inertial terms due to the particle rotation can be neglected (assumptions well satisfied for acoustical waves), $\langle \chi_d \mathbf{\Pi}' \rangle$ can be expressed by the following surface integral over the test sphere surface:

$$\langle \chi_d \mathbf{\Pi}' \rangle \approx \frac{3\alpha_d}{4\pi a^3} \oint^{(s)} \boldsymbol{a} \otimes (\boldsymbol{n} \cdot \langle \mathbf{\Pi}' \rangle_{\boldsymbol{x}}) \, dS, \tag{19}$$

where the superscript (s) means that only the symmetric part of the tensor appearing in the integrand is considered.

C. The self-consistent closure scheme

The link between constitutive equations and the test particle problem is now established via (18) and (19). To compute these expressions, one should first determine the boundary conditions and secondly derive a set of equations for the conditionally averaged fields. The first issue can be solved by the following considerations:

- As no phase changes occur, the conditionally averaged velocity is equal to the velocity of the test sphere on the particle surface ($r=a$).
- Far from the test particle (when $r \to \infty$), the perturbation of the fields induced by the presence of the heterogeneity vanishes so that the conditionally averaged fields asymptotically coincide with the unconditionally averaged fields:

$$\langle G' \rangle_{\boldsymbol{x}'} \to \langle G' \rangle \quad \text{when } r \to \infty . \tag{20}$$

To address the second issue, the same equations can be derived for the conditionally averaged field as for the averaged field, but, this time, constitutive equations are expressed in terms of the averaged field with two particle positions being known:

$$\boldsymbol{F}_{\boldsymbol{x}'} = \int_{\boldsymbol{x}''} \int_{|\boldsymbol{x}-\boldsymbol{x}'| \le a} n(t,\boldsymbol{x}',\boldsymbol{x}'') \langle \mathrm{div}(\mathbf{\Pi}') \rangle_{\boldsymbol{x}',\boldsymbol{x}''} \, d\boldsymbol{x}' \, d\boldsymbol{x}'',$$

$$\langle \chi_d \mathbf{\Pi}' \rangle_{\boldsymbol{x}'} = \int_{\boldsymbol{x}''} \int_{|\boldsymbol{x}-\boldsymbol{x}'| \le a} n(t,\boldsymbol{x}',\boldsymbol{x}'') \langle (\mathbf{\Pi}') \rangle_{\boldsymbol{x}',\boldsymbol{x}''} \, d\boldsymbol{x}' \, d\boldsymbol{x}''.$$

Of course, one could also calculate the averaged equations with the position of two particles being known and so on. In such a way, one would obtain an infinite hierarchy of mutually dependent equations conditioned by the position of an increasing number of particles. So the problem arises of an efficient truncation or closure of this hierarchy. A truncation at the first level would result in the calculation of the constitutive equations when the particles are embedded in the pure ambient fluid. Here, mutual interactions of two or more spheres are completely left out. This approximation is usually used in dilute mixtures and corresponds to the classical version of the coupled phase theory. However, even at this level, a part of the multiple scattering is included because the particles are excited by the mean field (cf. Fig. 1).

At the next level, binary interactions of pairs of spheres are accounted for, while ternary, quadruple, and higher order loops are neglected. To achieve closure at this level, one should calculate the constitutive equations when two particles positions are known, which is not an easy matter.

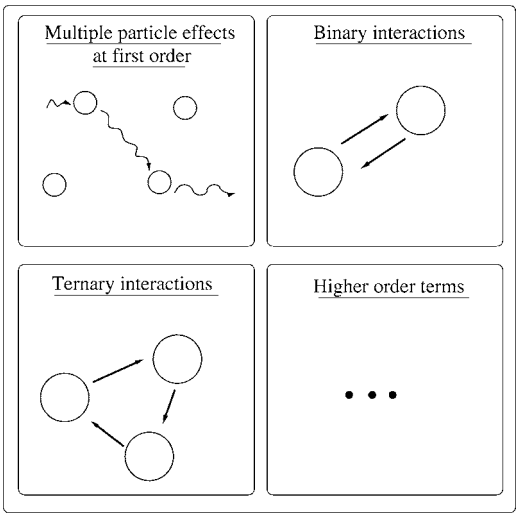

FIG. 1. Different orders of multiple scattering.

Of course, one could theoretically truncate this hierarchy at any order to integrate higher order loops, but the complexity of the calculation would greatly increase with the order. Anyway, this procedure would result in a polynomial expansion with respect to the particle concentration α_d and will thus be limited to relatively dilute mixtures. Moreover, nothing ensures that mutual interaction between n particles are dominant over interactions between $n+1$ particles when the concentration increases.

To overcome these limitations, Buyevich proposed a self-consistent scheme.[16,24] The starting point of this procedure is that, anyway, the resolution of the previous mutually dependent equations would result in an infinite polynomial series for the effective mixture properties. Instead of truncating the hierarchy at a certain level, the particles are supposed to be embedded in this *final* effective medium. Thanks to this procedure and by introducing a plausible form for integrals (18) and (19), the effective properties will be computed with an iterative scheme. All orders of interaction will thus be included in this formulation. Moreover, the correlations of particles in position can also be incorporated according to the choice of the expression of the conditional volume fraction.

D. Application to the propagation of an acoustic wave

1. Linearized equations

We will now adapt the previous system to the propagation of an acoustic wave. In this case, Eqs. (11)–(15) can be linearized. If we denote the equilibrium state with a subscript "*o*", the following equations stand:

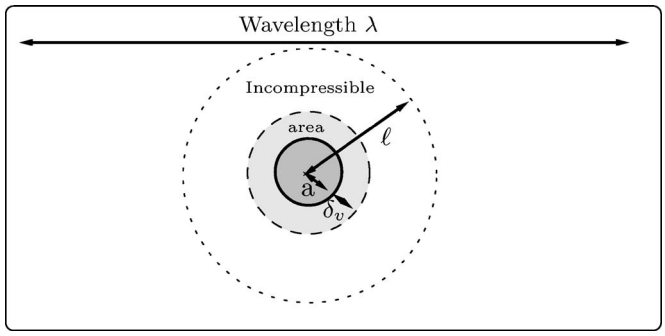

FIG. 2. The mesoscopic scale.

mass conservation:

$$\rho_{co}\left(\boxed{\frac{\partial \alpha_c}{\partial t}}+\alpha_{co}\mathrm{div}(\boldsymbol{v_c})\right)+\boxed{\alpha_{co}\frac{\partial \rho_c}{\partial t}}=0, \tag{21}$$

$$\boxed{\frac{\partial \alpha_d}{\partial t}}+\alpha_{do}\mathrm{div}(\boldsymbol{v_d})=0, \tag{22}$$

$$\alpha_d=1-\alpha_c; \tag{23}$$

momentum conservation:

$$\alpha_{co}\rho_{co}\frac{\partial \boldsymbol{v_c}}{\partial t}=-\nabla(\alpha_c p_c)+\mu_c\Delta \mathbf{v}+\boxed{(\lambda_c+\mu_c)\boldsymbol{\nabla}\mathrm{div}(\boldsymbol{v})} +\mathrm{div}\langle\chi_d\boldsymbol{\Pi}'\rangle-\boldsymbol{F}, \tag{24}$$

$$\alpha_{do}\rho_{do}\frac{\partial \boldsymbol{v_d}}{\partial t}=\boldsymbol{F}. \tag{25}$$

In these expressions, all terms linked to the compressibility of the continuous phase have been outlined.

2. The long wavelength regime (LWR)

To perform the explicit calculation of constitutive equations, we will consider the LWR. In this case, we can introduce a mesoscopic scale l around the test particle where the compressibility of the continuous phase can be neglected and such as, when $r\to l$, the perturbation induced by the test particle vanishes (cf. Fig. 2):

$$a+\delta_v \ll l \ll \lambda,$$

where λ is the acoustic wavelength and $\delta_v=\sqrt{2\mu_c/\omega\rho_c}$ is the thickness of the viscous boundary layer. This inequality is always satisfied in the cases treated in this paper. At distances smaller than l (cf. Fig. 2), all (outlined) compressible terms can be neglected and, after Fourier transform, we obtain

$$\mathrm{div}(\boldsymbol{v_c})=\mathrm{div}(\boldsymbol{v_d})=0, \tag{26}$$

$$-\alpha_{co}\rho_{co}(i\omega)\boldsymbol{v_c}=-\nabla(\alpha_c p_c)+\mu_c\Delta\boldsymbol{v}+\mathrm{div}\langle\chi_d\boldsymbol{\Pi}'\rangle-\boldsymbol{F}, \tag{27}$$

$$-\alpha_{do}\rho_{do}(i\omega)\boldsymbol{v_d}=\boldsymbol{F}. \tag{28}$$

The same equations can be derived for the conditionally averaged field, but, this time, the conditional volume fraction at equilibrium $\alpha_{do,\boldsymbol{x}'}$ replaces the unconditional one:

$$\mathrm{div}(\alpha_{co,\boldsymbol{x}'}\boldsymbol{v_{c,x}}')=\mathrm{div}(\alpha_{do,\boldsymbol{x}'}\boldsymbol{v_{d,x}}')=0, \tag{29}$$

$$-\alpha_{co,\boldsymbol{x}'}\rho_{co}(i\omega)\boldsymbol{v_{c,x}}'=-\nabla(\alpha_{c,\boldsymbol{x}'}p_{c,\boldsymbol{x}'})+\mu_c\Delta\boldsymbol{v}_{\boldsymbol{x}'} +div\langle\chi_d\boldsymbol{\Pi}'\rangle_{\boldsymbol{x}'}-\boldsymbol{F}_{\boldsymbol{x}'}, \tag{30}$$

$$-\alpha_{do,\boldsymbol{x}'}\rho_{do}(i\omega)\boldsymbol{v_{d,x}}'=\boldsymbol{F}_{\boldsymbol{x}'}. \tag{31}$$

3. Correlations of particles in position

The difference between $\alpha_{do,\boldsymbol{x}'}$ and α_{do} stems from the correlations of particles in position, that is to say, the perturbation of the particle repartition induced by the presence of a test sphere in $\boldsymbol{x}'$. The simplest approximation consists in neglecting this difference:

$$\alpha_{do,\boldsymbol{x}'}=\alpha_{do}, \tag{32}$$

and thus ignoring the non-overlapping property of the spheres. For the sake of simplicity, we will adopt this hypothesis and we will precisely discuss its validity in Sec. III. Of course, more elaborate expressions[16] can be derived to describe properly the repartition of particles within groups of several spheres (cf. the Kirkwood and Percus-Yevick models as reviewed in the book by Croxton[28]) and therefore include the correlations of position.

4. The self-consistent condition

Even if the equations have been simplified in the incompressible region, we still have to deal with the entire hierarchy of equations, and we therefore need to close the system. For that purpose, we will use the condition expressed in the Sec. II C: the particles will be supposed to be embedded in the *final* effective medium (made of the whole series expansion). Let us apply this condition to our case.

First, Eqs. (29)–(31) can be rewritten in a convective reference frame related to the velocity of the test particle center, that is to say $\boldsymbol{v_d}|_{r=0}$:

$$\mathrm{div}(\boldsymbol{V_{c,x}}')=\mathrm{div}(\boldsymbol{V_{d,x}}')=0, \tag{33}$$

$$-\alpha_{co}\rho_{co}(i\omega)\boldsymbol{V}_{\boldsymbol{c,x}'}=-\nabla(\alpha_{c,\boldsymbol{x}'}p_{c,\boldsymbol{x}'})+\mu_c\Delta\boldsymbol{V}_{\boldsymbol{x}'} +\mathrm{div}\langle\chi_d\boldsymbol{\Pi}'\rangle_{\boldsymbol{x}'}-\boldsymbol{F}_{\boldsymbol{x}'}-\alpha_{co}\rho_{co}\nabla\Psi, \tag{34}$$

$$-\alpha_{do}\rho_{do}(i\omega)\boldsymbol{V}_{\boldsymbol{d,x}'}=\boldsymbol{F}_{\boldsymbol{x}'}-\alpha_{do}\rho_{do}\nabla\Psi, \tag{35}$$

with

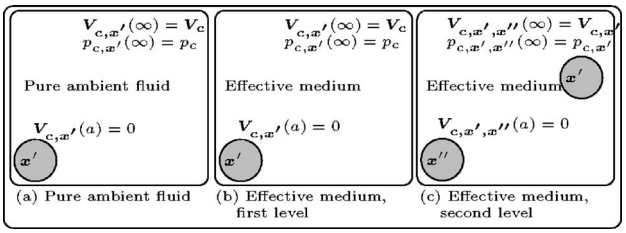

FIG. 3. Calculation of closure terms at different levels of the hierarchy.

$$\{\boldsymbol{V}_{\boldsymbol{d},\boldsymbol{x}'}, \boldsymbol{V}_{\boldsymbol{c},\boldsymbol{x}'}\} = \{\boldsymbol{v}_{\boldsymbol{d},\boldsymbol{x}'}, \boldsymbol{v}_{\boldsymbol{c},\boldsymbol{x}'}\} - \boldsymbol{v}_{\boldsymbol{d}}|_{r=0},$$

$$\Psi = -i\omega \boldsymbol{r} \cdot \boldsymbol{v}_{\boldsymbol{d}}|_{r=0}. \tag{36}$$

The terms proportional to $\nabla\Psi$ appearing in these equations are due to the change of reference frame. The boundary conditions can also be rewritten:

$$\boldsymbol{V}_{\boldsymbol{c},\boldsymbol{x}'} \to 0 \quad \text{when } r \to a, \tag{37}$$

$$\boldsymbol{V}_{\boldsymbol{c},\boldsymbol{x}'} \to \boldsymbol{V}_{\boldsymbol{c}} \quad \text{and} \quad p_{c,\boldsymbol{x}'} \to p_c \quad \text{when } r \to \infty. \tag{38}$$

Then, integrands (18) and (19) can be calculated for a particle embedded in a pure incompressible fluid, with boundary conditions (37) and (38) [cf. Fig. 3(a)]. The first integrand (18) corresponds to the classical calculation of the force applied on a moving sphere embedded in an unsteady nonuniform velocity field, sometimes called the Basset-Boussinesq-Oseen force.[29] The calculation of the second integrand (19) is less usual and can be found in some papers by Buyevich:[29,23]

$$\boldsymbol{F} = m_1(\boldsymbol{V}_{\boldsymbol{c}} - \boldsymbol{V}_{\boldsymbol{d}}) + m_2 \Delta \boldsymbol{V}_{\boldsymbol{c}} + m_3 \nabla \Psi, \tag{39}$$

$$\mathrm{div}\langle \chi_d \boldsymbol{\Pi}' \rangle = -\nabla(\alpha_d p_c) + m_0 \Delta \boldsymbol{V}_{\boldsymbol{c}}, \tag{40}$$

where m_0, m_1, m_2, and m_3 depend on the properties of the pure ambient fluid (μ_c, ρ_{co}) and on frequency ω. In these expressions, $m_1(\boldsymbol{V}_{\boldsymbol{c}} - \boldsymbol{V}_{\boldsymbol{d}})$ corresponds to the sum of the Stokes drag, the Basset hereditary, and the total inertial forces; $m_2 \Delta \boldsymbol{V}_{\boldsymbol{c}}$ is the Oseen correction due to the nonuniformity of the ambient fluid velocity; and $m_3 \nabla \Psi$ comes from the change of reference frame.

In the effective medium, the same relations stand:

$$\boldsymbol{F} = m_1^*(\boldsymbol{V}_{\boldsymbol{c}} - \boldsymbol{V}_{\boldsymbol{d}}) + m_2^* \Delta \boldsymbol{V}_{\boldsymbol{c}} + m_3^* \nabla \Psi, \tag{41}$$

$$\mathrm{div}\langle \chi_d \boldsymbol{\Pi}' \rangle = -\nabla(\alpha_d p_c) + m_0^* \Delta \boldsymbol{V}_{\boldsymbol{c}}, \tag{42}$$

but m_0^*, m_1^*, m_2^*, and m_3^* depend on the effective properties of the surrounding fluid (μ_{eff}, ρ_{eff1}, ρ_{eff2}), where ρ_{eff1} and ρ_{eff2} are some effective volume fractions, respectively linked to the inertial phenomena and the change of frame of reference, and μ_{eff} is the effective viscosity of the suspension.

Here arises the problem of determining these effective properties and this is the fundamental point in which our theory differs from the so-called "core shell model." These effective densities and viscosity will be calculated in a consistent way instead of being empirically introduced.

The expressions of $\boldsymbol{F}$ and $\mathrm{div}\langle \chi_d \boldsymbol{\Pi}' \rangle$ come from the calculation of integrands (18) and (19) in the *final* effective medium with boundary conditions (37) and (38) as illustrated by Fig. 3(b). At the next level of the hierarchy, the particles are also embedded in the *final* effective medium and the boundary conditions become [cf. Fig. 3(c)]

$$\boldsymbol{V}_{\boldsymbol{c},\boldsymbol{x}',\boldsymbol{x}''} \to 0 \quad \text{when } r \to a, \tag{43}$$

$$\boldsymbol{V}_{\boldsymbol{c},\boldsymbol{x}',\boldsymbol{x}''} \to \boldsymbol{V}_{\boldsymbol{c},\boldsymbol{x}'} \quad \text{and } p_{c,\boldsymbol{x}',\boldsymbol{x}''} \to p_{c,\boldsymbol{x}'} \quad \text{when } r \to \infty. \tag{44}$$

This is exactly the same problem, but the boundary conditions are expressed in terms of the conditionally averaged fields instead of the averaged fields. So $\boldsymbol{F}_{\boldsymbol{x}'}$ and $\mathrm{div}\langle \chi_d \boldsymbol{\Pi}' \rangle$ will be related to $\boldsymbol{V}_{\boldsymbol{c},\boldsymbol{x}'}, \boldsymbol{V}_{\boldsymbol{d},\boldsymbol{x}'}$, and $\nabla\Psi$ with *exactly the same coefficients* m_0^*, m_1^*, m_2^*, and m_3^*:

$$\boldsymbol{F}_{\boldsymbol{x}'} = m_1^*(\boldsymbol{V}_{\boldsymbol{c},\boldsymbol{x}'} - \boldsymbol{V}_{\boldsymbol{d},\boldsymbol{x}'}) + m_2^* \Delta \boldsymbol{V}_{\boldsymbol{c},\boldsymbol{x}'} + m_3^* \nabla \Psi, \tag{45}$$

$$\mathrm{div}\langle \chi_d \boldsymbol{\Pi}' \rangle_{\boldsymbol{x}'} = -\nabla(\alpha_{d,\boldsymbol{x}'} p_{c,\boldsymbol{x}'}) + m_0^* \Delta \boldsymbol{V}_{\boldsymbol{c},\boldsymbol{x}'}. \tag{46}$$

It is the equality of these coefficients at every order of the hierarchy that expresses the self-consistent condition. With this condition, there is no need to truncate the hierarchy at a finite order because the system is already closed as we will see.

To determine the expressions of the effective parameters in a consistent way, previous equations must be combined properly to obtain a final system of equations in the effective medium similar to the equations that would stand in the pure medium, that is to say,

$$\mathrm{div}(\boldsymbol{V}_{\boldsymbol{c},\boldsymbol{x}'}) = 0, \tag{47}$$

$$-\rho_{\mathrm{eff1}}(i\omega)\boldsymbol{V}_{\boldsymbol{c},\boldsymbol{x}'} = -\nabla p_{c,\boldsymbol{x}'} + \mu_{\mathrm{eff}} \Delta \boldsymbol{V}_{\boldsymbol{c},\boldsymbol{x}'} - \rho_{\mathrm{eff2}} \nabla \Psi. \tag{48}$$

If we replace relations (45) and (46) in Eqs. (33)–(35), we obtain together with Eq. (48) a set of 11 equations. On the other hand, we have 11 unknown parameters: the six components of velocities $\boldsymbol{V}_{\boldsymbol{d},\boldsymbol{x}'}$ and $\boldsymbol{V}_{\boldsymbol{c},\boldsymbol{x}'}$, the three effective parameters ρ_{eff1}, ρ_{eff2}, and μ_{eff}, the volume fraction $\alpha_{d,\boldsymbol{x}'}$, and the pressure $p_{c,\boldsymbol{x}'}$. Therefore, the effective properties can be expressed in terms of the coefficients m_k^* (for more details, cf. the original derivation by Buyevich[23]):

$$\rho_{\mathrm{eff1}} = \alpha_{co}\rho_{co} + \frac{\alpha_{do}\rho_{do} m_1^*}{m_1^* - i\omega\alpha_{do}\rho_{do}}, \tag{49}$$

$$\mu_{\mathrm{eff}} = \alpha_{co}\mu_c + m_0^* + \frac{\alpha_{do}\rho_{do} i\omega m_2^* + \alpha_{do}\mu_c(m_1^* - i\omega m_2^* \rho_{\mathrm{eff1}}/\mu_{\mathrm{eff}})}{m_1^* - i\omega\alpha_{do}\rho_{do}}, \tag{50}$$

$$\rho_{\mathrm{eff2}} = \alpha_{co}\rho_{co} + \alpha_{do}\rho_{do}\frac{m_1^* - m_3^* i\omega}{m_1^* - i\omega\alpha_{do}\rho_{do}}. \tag{51}$$

Now Eqs. (47) and (48) can be solved[29,23] with boundary conditions (37) and (38) to calculate integrands (18) and (19). We will obtain the same expressions as in the case of the pure fluid, but the effective properties (μ_{eff}, ρ_{eff1}, ρ_{eff2}) will stand instead of the pure fluid properties (μ_c, ρ_{co}):

$$m_0^* = \frac{5\alpha_{do}\mu_{\text{eff}}\exp(\beta)}{2(1+\beta)}, \tag{52}$$

$$m_1^* = \frac{9\alpha_{do}}{2a^2}(1+\beta+\beta^2/3)\mu_{\text{eff}}, \tag{53}$$

$$m_2^* = \frac{9\alpha_{do}}{2\beta^2}(\exp(\beta)-(1+\beta+\beta^2/3))\mu_{\text{eff}}, \tag{54}$$

$$m_3^* = \alpha_{do}\rho_{\text{eff2}}, \tag{55}$$

with

$$\beta^2 = -(i\omega)\rho_{\text{eff1}}a^2/\mu_{\text{eff}}. \tag{56}$$

Thus, coefficients m_k^* are expressed in terms of the effective properties and the system is closed. There only remains to solve numerically the self-consistent system formed by Eqs. (49)–(55) in the complex plane. This can be achieved by a simple iterative procedure, but some more elaborate schemes such as the so-called "Globally Convergent Newton's Method" can also be used.

We can notice that in the steady regime, we simply obtain

$$\rho_{\text{eff1}} = \rho_{\text{eff2}} = \alpha_{co}\rho_{co} + \alpha_{do}\rho_{do} \equiv \rho, \quad \mu_{\text{eff}} = \mu_c/(1-5/2\alpha_{do}).$$

This simple case illustrates the strength of the self-consistent scheme. A truncation of the hierarchy at the first order would have given the well-known Einstein formula:

$$\mu_{\text{eff}} = \mu_c(1+5/2\alpha_{do}) + O(\alpha_{do}^2).$$

It can be simply obtained from the expression (50) of μ_{eff} by replacing the coefficients m_k^* by their expression in the pure fluid m_k and by taking the asymptotic limit when $\omega=0$.

A truncation at order n would have given a formula of the form

$$\mu_{\text{eff}} = 1+5/2\alpha_{do} + \sum_{i=2}^{n} K_i\alpha_{do}^i + O(\alpha_{do}^{n+1}).$$

Thus all these formulas are limited to $\alpha_{do} \ll 1$. With the self-consistent theory, we directly obtain the whole series expression.

To conclude this part we would like to point out some differences with other models. First, we can note that the above steady effective properties are commonly used in the "core shell" model. Thus, the evolution of these parameters with frequency is neglected, contrary to the present study. Then, one of the differences with the model proposed by Spelt *et al.*[17] is that, in our theory, the relation between the closure terms and the averaged fields (expressed by coefficients m_k^*) is deduced from the pure fluid expressions. In the article by Spelt *et al.*, the authors say that each closure relation can be expressed in terms of any of the averaged fields as these field are also related to each other through algebraic equations that depend on the frequency and the effective wave number. This is a correct argument but only for plane waves because more complicated relations stand between averaged fields when dealing with spherical or more complicated wavefronts.

E. Dispersion equation for a plane acoustic wave

Now, we will derive the dispersion equation for a plane wave such as $G=G_o+\tilde{G}e^{i(k_* x-\omega t)}$, where $\tilde{G}$ is the amplitude of the wave, G_o is the equilibrium state, and k_* is the complex effective wave number. In this case, from Eqs. (21)–(25) and the expression of closure relations (41) and (42) we get the final system:

mass conservation:

$$-i\omega(\rho_{co}\tilde{\alpha}_c + \alpha_{co}\tilde{\rho}_c) + ik_*\alpha_{co}\rho_{co}\tilde{v}_c = 0, \tag{57}$$

$$i\omega\tilde{\alpha}_c + ik_*\alpha_{do}\tilde{v}_d = 0; \tag{58}$$

momentum conservation:

$$i\omega\alpha_{co}\rho_{co}\tilde{v}_c - ik_*\tilde{p}_c = k_*^2(\lambda_c+2\mu_c)(\alpha_{co}\tilde{v}_c + \alpha_{do}\tilde{v}_d) + k_*^2 m_0^*\tilde{v}_c + [m_1^*(\tilde{v}_c-\tilde{v}_d) - k_*^2 m_2^*\tilde{v}_c - i\omega m_3^*\tilde{v}_d], \tag{59}$$

$$-i\alpha_{do}\rho_{do}\omega\tilde{v}_d = m_1^*(\tilde{v}_c-\tilde{v}_d) - k_*^2 m_2^*\tilde{v}_c - i\omega m_3^*\tilde{v}_d; \tag{60}$$

state equation:

$$\tilde{p}c = c_{co}^2\tilde{\rho}_c, \tag{61}$$

where c_{co} is the sound velocity at rest in the continuous phase and parameters m_k^* can be numerically calculated from expressions (49)–(55) as mentioned earlier. This system is therefore a linear system of five equations, with five unknowns, $\tilde{\alpha}_c$, $\tilde{\rho}_c$, $\tilde{v}_c$, $\tilde{v}_d$, and $\tilde{p}_c$, and is consequently well posed. If we introduce the following parameters,

$$M_k^* = \frac{m_k^*}{\alpha_{do}\rho_{do}}, \quad d_r = \frac{\alpha_{do}\rho_{do}}{\alpha_{co}\rho_{co}}, \quad \text{and} \quad r = \frac{\rho_{co}}{\rho_{do}},$$

we get from (60)

$$\tilde{v}_d = [h_v - k_*^2 h_c]\tilde{v}_c \tag{62}$$

with

$$h_v = \frac{M_1^*}{M_1^* + i\omega(M_3^*-1)} \quad \text{and} \quad h_c = \frac{M_2^*}{M_1^* + i\omega(M_3^*-1)}.$$

Finally, by combining the conservation equations, we obtain the following bicubic equation, which can easily be solved to calculate the effective wave number:

$$Ak_*^4 + Bk_*^2 + C = 0, \tag{63}$$

$$A = d_r h_c\left[\frac{(\lambda_c+2\mu_c)}{\rho_{do}i\omega} + \frac{rc_{co}^2}{\alpha_{co}\omega^2}\right], \tag{64}$$

$$B = -d_r\left[h_c + \frac{M_0^* + (\lambda_c+2\mu_c)(\alpha_{co}+\alpha_{do}h_v)}{i\omega}\right] - \frac{c_{co}^2}{\alpha_{co}\omega^2}[1+d_r r h_v], \tag{65}$$

$$C = 1 + d_r h_v. \tag{66}$$

This calculation can be simplified because $h_v/k_*^2 h_c \gg 1$. This can be proved either by calculating it numerically or by noticing that this ratio is of the form of an effective acoustic Reynolds number, which is therefore large compared to unity. Thus, all terms proportional to h_c in the preceding equations can be neglected and we finally obtain

$$\left(\frac{k_*}{\omega} c_{co}\right)^2 = I(\omega) V(\omega),$$

$$I(\omega) = \left[1 - \frac{\alpha_{co} d_r i\omega}{c_{co}^2 (1 + d_r r h_v)} \times \left[M_0^* + \frac{(\lambda_c + 2\mu_c)(\alpha_{co} + \alpha_{do} h_v)}{\alpha_{do} \rho_{do}}\right]\right]^{-1},$$

$$V(\omega) = \left[1 + d_r \frac{(\alpha_{co} - r) h_v - \alpha_{co} r}{1 + d_r r h_v}\right].$$

In this expression $I(w)$ corresponds to the intrinsic (bulk) losses in the medium and $V(\omega)$ to the visco-inertial interactions between the two phases.

III. COMPARISON WITH EXPERIMENTS AND OTHER THEORIES

In this section, we will first compare the effective medium theory (EMT) with the multiple scattering theory (MST) from a theoretical point of view. Then, we will compare the predictions of our theory with the experimental data of Hipp *et al.*[25] and also with the "classical coupled phase theory" in which the calculation of the closure terms is based on the pure ambient fluid parameters instead of the effective ones.

A. Theoretical comparison with the multiple scattering theory

The hierarchy appearing in the EMT is similar to the hierarchy that also appears in the MST [see Ref. 11, Eq. (2.13)]. However, in the EMT the hierarchy is a succession of mutually dependent equations governing the conditionally averaged fields, whereas in the MST the hierarchy concerns the exciting field. This is one of the fundamental points in which these two theories differ.

Before delving into this crucial problem, let us clarify the terminology used here. The exciting field ($G_j'^E$) acting on the jth particle is the sum of the original field that would exist in the absence of particles (G'^0), and the wave scattered by every particle ($G_k'^S$) except the jth:

$$G_j'^E(\boldsymbol{x}, t|\boldsymbol{x_1}, \ldots, \boldsymbol{x_N}) = G'^0(\boldsymbol{x}, t) + \sum_{k \neq j} G_k'^S(\boldsymbol{x}, t|\boldsymbol{x_1}, \ldots, \boldsymbol{x_N}). \tag{67}$$

Note that this deterministic formulation is exact and that only the last scattering event of the particle j is omitted. Thus $G_k'^S$ may involve previous scattering by the jth particle. Otherwise, resonant scattering between a cluster of particles, i.e., loops, would be neglected.

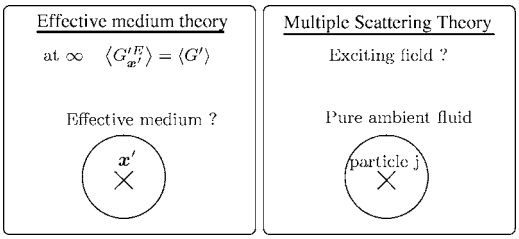

FIG. 4. Comparison between the self-consistent theory and the multiple scattering theory.

In the linear regime, the wave scattered by the jth particle can be related to the exciting field acting on it by the introduction of a linear operator T_j so that

$$G_j'^E(\boldsymbol{x}, t|\boldsymbol{x_1}, \ldots, \boldsymbol{x_N}) = G'^0(\boldsymbol{x}, t) + \sum_{k \neq j} T_k G_k'^E(\boldsymbol{x}, t|\boldsymbol{x_1}, \ldots, \boldsymbol{x_N}). \tag{68}$$

The introduction of this operator is a crucial step in the MST and it means that this theory is limited to the linear regime whereas the EMT is not. We can also note that the operator T_j is computed by using the properties of the pure ambient fluid.

The following relation stands between the total field (G'), the exciting field acting on the jth particle ($G_j'^E$), and the wave scattered by the same particle ($G_j'^S$):

$$G'(\boldsymbol{x}, t|\boldsymbol{x}_1, \ldots, \boldsymbol{x}_N) = G_j'^E(\boldsymbol{x}, t|\boldsymbol{x}_1, \ldots, \boldsymbol{x}_N) + G_j'^S(\boldsymbol{x}, t|\boldsymbol{x_1}, \ldots, \boldsymbol{x_N}). \tag{69}$$

Now we can clarify the previous assertion. In the EMT, a test particle is considered in $\boldsymbol{x}'$, and the value of the averaged exciting field acting on it is known far from the particle as the influence of the test particle vanishes:

$$\langle G_{\boldsymbol{x}'}'^E \rangle_{\boldsymbol{x}'} \to \langle G' \rangle \quad \text{when } r \to \infty \quad \text{as } \langle G_{\boldsymbol{x}'}'^S \rangle_{\boldsymbol{x}'} \to 0, \tag{70}$$

where $G_{\boldsymbol{x}'}'^E$ and $G_{\boldsymbol{x}'}'^S$ are respectively the exciting field acting on the particle located in $\boldsymbol{x}'$ and the wave scattered by this particle. This relation corresponds to the boundary condition (20). The fundamental problem here is thus to determine the expression of the effective medium surrounding the particle to perform integrands (18) and (19).

In the multiple scattering theory, we suppose that all particles are embedded in the pure ambient fluid (by introducing the operator T) and the problem, in this case, is to determine the form of the exciting field. These two approaches can be summarized by Fig. 4. We will now show that, at the lowest level, these two theories are equivalent. For that purpose, let us recall the assumptions implicitly made by Foldy[8] (and listed by Waterman and Truell[11]), when he identifies the average of the exciting field with the average of the total field to achieve closure. The starting point of his derivation is Eq. (68).

(1) Its first assumption is that the exciting field acting on the jth particle is the total field that would exist if this particle was not there:

$$G_j'^E(\boldsymbol{x},t|\boldsymbol{x}_1,\ldots,\boldsymbol{x}_N) = G'(\boldsymbol{x},t|\boldsymbol{x}_1,\ldots,\boldsymbol{x}_{j-1},\boldsymbol{x}_{j+1},\ldots\boldsymbol{x}_N),$$

Here, Foldy neglects all mutual interactions of the particles (loops) as the jth particle cannot influence the other particles that produce the exciting field.

(2) Then he assumes that the probability density conditioned by the position of one particle $p(t,\boldsymbol{x}_1,\ldots,\boldsymbol{x}_{j-1},\boldsymbol{x}_{j+1},\ldots,\boldsymbol{x}_N|\boldsymbol{x}_j)$ is equal to the unconditional probability: $p(t,\boldsymbol{x}_1,\ldots,\boldsymbol{x}_{j-1},\boldsymbol{x}_{j+1},\ldots,\boldsymbol{x}_N)$. This assumption leads to the statistical independence so that the correlations of particles in position are neglected and thus the particles can overlap one another.
(3) His third hypothesis is not restrictive; he assumes that the contributions of a single particle on the mean field can be neglected. This will be valid whenever the number N of particles appearing in the statistical average process is large enough, as the contribution of a single particle is of the order $1/N$.

Of course the first and second hypotheses are linked: to account properly for interactions of two or more particles, one should determine the correlations in position. But they are also undoubtedly distinct. One could, for example, estimate the interactions of pairs with an inaccurate distribution of the particles, for example by supposing that they can overlap one another. This would lead to an expression that would not be valid for too concentrated solutions but that would nevertheless incorporate pair interactions.

Assumptions 1–3 lead to the simple relation:

$$\langle G_j'^E(\boldsymbol{x},t)\rangle_j = \langle G'(\boldsymbol{x},t)\rangle. \tag{71}$$

So, the average of the exciting field acting on the jth particle is equal to the mean field, and the particle is embedded in the pure ambient fluid. This situation is therefore equivalent to the first level of the EMT hierarchy. However, even at this level, there is still a fundamental difference between these two theories. In the coupled phase theory, averaged equations are derived with respect to the volume fraction occupied by each phase. In the derivation of Foldy, however, the particles are supposed to be pointlike. Thus the decrease of the volume fraction occupied by the continuous phase due to the increase of the number of particles is not accounted for. The difference between these two theories can be neglected when the number of particles is large but the corresponding volume fraction is small. However, this difference becomes important when the particles occupy a large volume fraction: in the coupled phase theory when $\alpha_{do}\to 1$, only the intrinsic absorption in the dispersed phase remains whereas in the theory of Foldy the effects are maximum.

To account for interactions of pairs of particles, Lax introduced the so-called "quasi-crystalline approximation"[10] according to which the averaged exciting field acting on the jth particle when the position of two particles is known (j,k) is approximately equal to the averaged exciting field acting on the jth particle when only the position of the latter is known

$$\langle G_j'^E(\boldsymbol{x},t)\rangle_{jk} \approx \langle G_j'^E\rangle_j. \tag{72}$$

This approximation comes within the scope of a more general frame in which the averaged exciting field with n positions of particles being known is supposed to be approximately equal to the averaged exciting field with $n-1$ known positions.

To conclude this part, we can underline the fact that such a procedure will always result in a polynomial development with respect to the particle concentration and will thus be limited to relatively dilute mixtures whereas the EMT is not.

B. Comparison with the classical coupled phase theory and experimental data

Figure 5 compares the results obtained with our theory with the experimental data of Hipp *et al.*[14] and also with the "classical coupled phase theory." In this figure, we can see how much the predictions are improved by the introduction of the effective parameters instead of the pure fluid values, for low frequencies, when the interactions between the particles are strong because of the overlapping of boundary layers. However, when the frequency increases, some differences between the predictions and the experiments appear. They may be explained by the following considerations.

When the correlations of particles in position are considered, the more we approach the test particle, the more unlikely is the presence of another particle because they cannot overlap one another. So, it means that the conditional volume fraction $\alpha_{do,\boldsymbol{x}'}\to 0$ when $r\to a$. On the opposite, far from the test particle, the modification of the particle distribution induced by the presence of the test sphere vanishes so that $\alpha_{do,\boldsymbol{x}'}\to\alpha_{do}$ when $r\to\infty$. The transition between these two regimes appears approximately when $r\approx 2a$, which is the characteristic length that can be introduced because of the non-overlapping property of the particles. On the other hand, we can notice that the boundary layer thickness δ_v is inversely proportional to the square root of the frequency, so that when the frequency increases, this thickness decreases.

In our model, the correlations of particles in position are not considered and thus the effective properties do not depend on the distance from the particle surface. As long as $\delta_v \gg a$, approximating the effective medium surrounding the particle by homogeneous parameters based on the approximation $\alpha_{do,\boldsymbol{x}'}(r)=\alpha_{do}$ is accurate. But when $\delta v\approx a$, the variations of the conditional volume fraction with the distance r from the particle center cannot be neglected anymore. When $\delta_v \ll a$, the parameters of the effective medium in the boundary layer are even very close to the pure fluid values as almost no particles are present in this region. Consequently, the approximation that consists in taking the pure fluid parameters (ρ_{co},μ_c) to calculate the closure terms should give better results in this frequency range. To verify the validity of this hypothesis, we have plotted the attenuation curves for larger particles (cf. Fig. 6). In this case the characteristic frequency coresponding to $\delta_v=a$ is equal to $f_c=11$ MHz, whereas for Fig. 5 it was equal to 101 MHz.

On the above curves, we can observe the transition between the two asymptotic limits, around the characteristic frequency f_c, therefore corroborating our hypothesis. Our

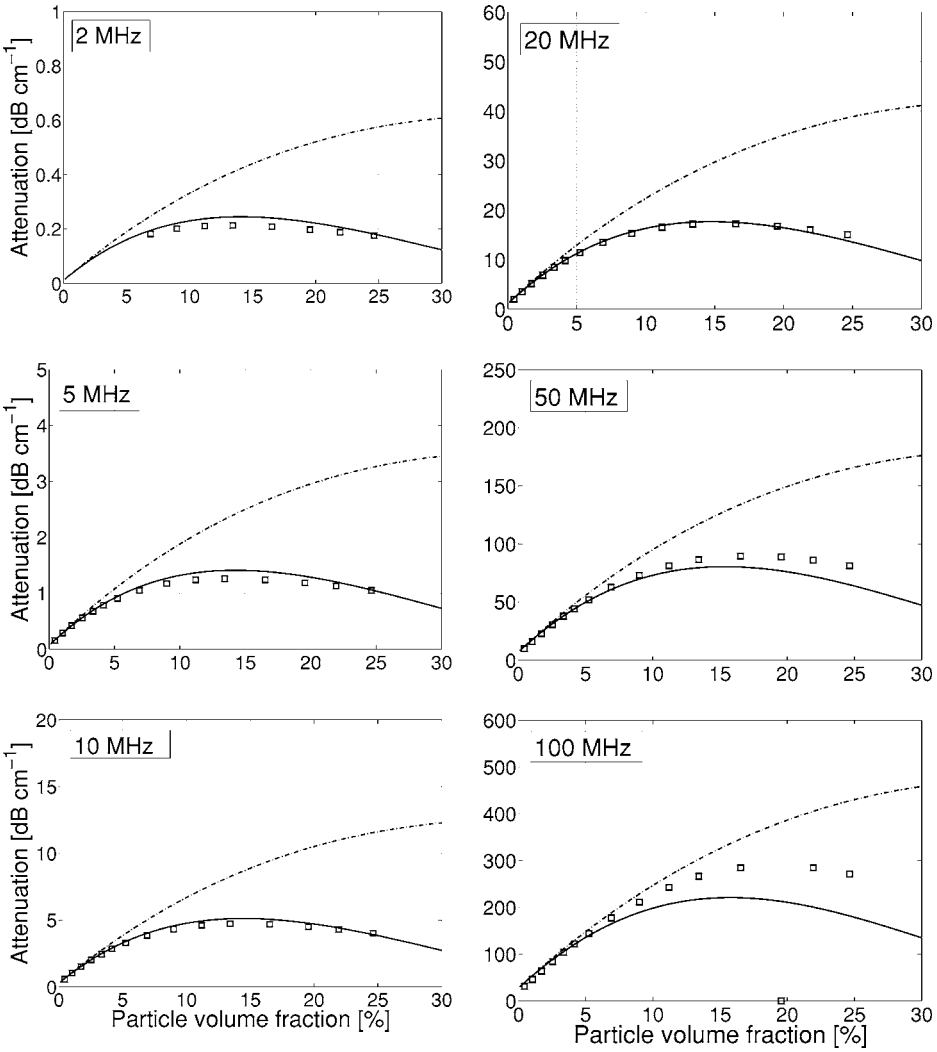

FIG. 5. Attenuation as a function of the volume fraction at various frequencies for silica particles of 56-nm radius in water. The solid lines correspond to our theory (—), the broken line to the classical coupled phase theory (- · -), and the symbols to the experimental data.

analysis shows how important the correlations in position are in the calculation of the effective parameters in acoustics. Thus, their integration in the calculation of coefficients m_k^* should give the smooth transition between these two limiting cases.

To conclude this section, we also plotted the curves with and without the intrinsic (bulk) losses, which are often neglected in the coupled phase models (cf. Fig. 7). These effects prove to be important for volume fraction up to 6% and must therefore be included correctly in formulation.

IV. EXTENSION TO THE POLYDISPERSE CASE

In this section, we will extend previous equations to polydisperse suspensions by using the same procedure as the one introduced by Gubaidullin and Nigmatulin[30] in their treatment of polydisperse aerosols. When a polydisperse suspension is considered, the probability $p(t,\boldsymbol{x})$ of finding any particle in x at t is replaced by the probability $p(t,\boldsymbol{x},a)$ of finding a particle of radius a in x at t. In this case, the properties of the dispersed phase also depend on the particule radius. So instead of introducing directly the phasic average over the whole dispersed phase $\langle \chi_d G' \rangle / \alpha_d$, we will split it into two steps:

- first, an average over all particles with the same radius $G_p(a) = \langle \chi_a G' \rangle / \alpha_p(a)$, where

$$\chi_a(\boldsymbol{x},t) = \begin{cases} 1 & \text{if } \boldsymbol{x} \text{ is in a particle of size } a \text{ at time } t, \\ 0 & \text{otherwise,} \end{cases}$$

  and $\alpha_p(a) = \langle \chi_a(\boldsymbol{x},t) \rangle$ is the volume fraction occupied by particles of radius a,
- and second, an average $\langle \rangle_a$ over all the particles sizes:

$$G_d = \frac{1}{\alpha_d} \langle G_p(a) \rangle_a = \frac{1}{\alpha_d} \int_{a_{\min}}^{a_{\max}} \alpha_p(a) G_p(a)\, da.$$

We can now derive the equations in the polydisperse case. The equations of the continuous phase will remain the same, but the momentum conservation equation of the dispersed phase will be derived for each particle size so that the set of Eqs. (57)–(61) becomes

mass conservation:

$$-i\omega(\rho_{co}\tilde{\alpha}_c + \alpha_{co}\tilde{\rho}_c) + ik_*\alpha_{co}\rho_{co}\tilde{v}_c = 0,$$

$$i\omega\tilde{\alpha}_c + ik_*\alpha_{do}\langle \tilde{v}_p \rangle_a = 0;$$

momentum conservation:

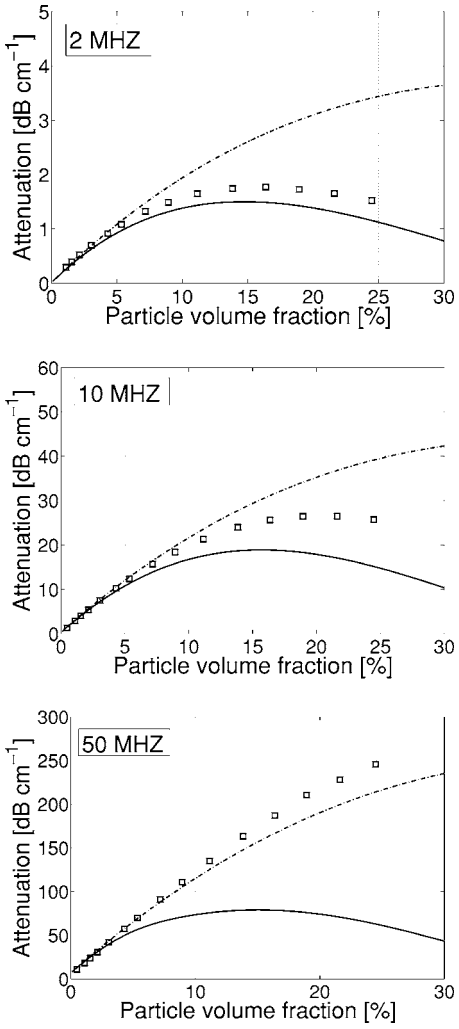

FIG. 6. Attenuation as a function of the volume fraction at various frequencies for silica particles of 164.5-nm radius in water. The solid lines correspond to our theory (—), the broken line to the classical coupled phase theory (- · -), and the symbols to the experimental data.

$$i\omega\alpha_{co}\rho_{co}\tilde{v}_c - ik_*\tilde{p}_c$$
$$= k_*^2(\lambda_c + 2\mu_c)(\alpha_{co}\tilde{v}_c + \alpha_{do}\langle\tilde{v}_p\rangle_a) + k_*^2\langle m_0^*\rangle_a\tilde{v}_c$$
$$+ \langle m_1^*(\tilde{v}_c - \tilde{v}_p)\rangle_a - k_*^2\langle m_2^*\rangle_a\tilde{v}_c - i\omega\langle m_3^*\tilde{v}_p\rangle_a,$$
$$- i\alpha_{do}\rho_{do}\omega\tilde{v}_p = m_1^*(\tilde{v}_c - \tilde{v}_p) - k_*^2 m_2^*\tilde{v}_c - i\omega m_3^*\tilde{v}_p;$$

state equation:

$$\tilde{p}_c = c_{co}^2\tilde{\rho}_c.$$

In these equations, the coefficients m_k^* depend on the particle size and must therefore be calculated for each radius a.

Then we can express the velocity of the dispersed phase $\tilde{v}_p$ in terms of the velocity of the continuous $\tilde{v}_c$ phase, which is independent of the particle radius:

$$\tilde{v}_p = [h_v(a) - k_*^2 h_c(a)]\tilde{v}_c.$$

In this way, we can extract the continuous phase properties from the average $\langle\rangle_a$. If we now combine the conservation equations, we finally obtain the same dispersion equation as in the monodisperse case but with respectively $\langle h_v\rangle_a$, $\langle h_c\rangle_a$, and $\langle M_0^*\rangle_a$ instead of h_v, h_c, and M_0^*. Thus, only the average

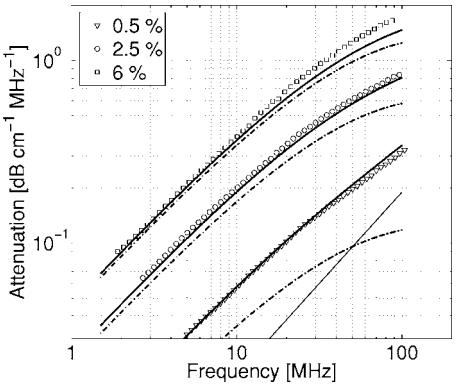

FIG. 7. Attenuation as a function of the frequency at different volume fractions for silica particles of 56-nm radius in water. The solid lines correspond to the complete theory (—), the broken line to the theory without intrinsic losses (- · -), the symbols to the experimental data, and the thin straight line to the intrinsic losses in water.

of these three parameters must be calculated to extend the validity of our dispersion relation to the polydisperse case.

V. CONCLUSION

The coupled phase theory has been improved to consider polydisperse suspensions and viscous interactions with the use of an effective medium, self-consistent theory. Our derivation is based on Buyevich's incompressible hydrodynamic model extended here to acoustical waves propagation. This theory turns out in practice to be very effective, as it amounts finally to a dispersion relation, which can be used directly for measurements or simulations. When compared with experiments, this model provides an accurate description of the attenuation at low frequencies, for concentrated suspensions for which interactions between particles are strong. In particular, the self-consistent approach has the ability to take properly into account interactions at all orders, such as the overlapping of viscous boundaries layers and the loops in the sense of the multiple scattering theory (MST) of waves. The link between the MST and the effective medium theory (EMT) has also been clarified.

Finally, our derivation could be extended to higher frequencies (or equivalently larger particles) and higher volume fractions by taking into account the correlations of particles in position, which would affect the effective properties of the medium. Its validity could even be enlarged to the high frequency regime by including the compressibility of the liquid when calculating the closure terms.[31,32]

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