On the influence of spatial correlations on sound propagation in concentrated solutions of rigid particles

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In a previous paper [J. Acoust. Soc. Am. **121**, 3386–3387 (2007)], a self-consistent effective medium theory has been used to account for hydrodynamic interactions between neighboring rigid particles, which considerably affect the sound propagation in concentrated solutions. However, spatial correlations were completely left out in this model. They correspond to the fact that the presence of one particle at a given position locally affects the location of the other ones. In the present work, the importance of such correlations is demonstrated within a certain frequency range and particle concentration. For that purpose, spatial correlations are integrated in our two-phase formulation by using a closure scheme similar to the one introduced by Spelt *et al.* ["Attenuation of sound in concentrated suspensions theory and experiments," J. Fluid Mech. **430**, 51–86 (2001)]. Then, the effect is shown through a careful comparison of the results obtained with this model, the ones obtained with different self-consistent approximations and the experiments performed by Hipp *et al.* ["Acoustical characterization of concentrated suspensions and emulsions. 2. Experimental validation," Langmuir, **18**, 391–404 (2002)]. With the present formulation, an excellent agreement is reached for all frequencies (within the limit of the long wavelength regime) and for concentrations up to 30% without any adjustable parameter.
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I. INTRODUCTION

A precise prediction of the attenuation and dispersion of acoustical waves induced by the presence of particles in suspensions of different natures would be of great interest for acoustic spectroscopy.[1,2] Although the propagation in dilute suspensions is now well described (see Ref. 3 for bubbles, the ECAH theory,[4,5] and the coupled phase theory[6] for emulsions and the models of Gubaidullin and Nigmatulin,[7] Gumerov *et al.*,[8] and Duraiswami and Prosperetti[9] for aerosols), there remain some difficulties in concentrated suspensions as the interactions between neighboring particles must be taken into account. For that purpose, different methods have been used: first, numerical methods which are generally based on the so called "multipole expansion" (see Refs. 10 and 11 for the Helmholtz equation, and Refs. 12–15 for the Stokes and Brinkman equations). Although this numerical treatment of the problem is required for the study of particular configurations (when the particles are not homogeneously distributed), it does not take advantage of the average homogeneous distribution of the particles for randomly distributed spheres. That is why, a statistical treatment of the equations is interesting in this case. First, a hierarchy of mutually dependent averaged equations can be derived (see Ref. 16 for the multiple scattering theory developed for the Helmholtz equation and Ref. 17 for the two-phase Navier–Stokes equations). Then arises the problem of the efficient closure of this hierarchy. In many papers, this hierarchy is truncated at a certain order (generally at first or second order[18–20]). At first order, mutual interactions between neighboring particles are completely left out. At second order, only mutual interactions between two particles are taken into account. This truncation of the hierarchy cannot be used for concentrated suspensions because in this case, mutual interactions between N particles cannot be neglected compared to mutual interactions between $N+1$ particles. In order to avoid this truncation, self-consistent effective medium theories have been widely used in many branches of physics (see Ref. 21 for acoustical waves in bubbly liquids, Ref. 22 for elastic waves in composites, and Refs. 23–25 for two-phase flow, etc.). These methods consist of calculating the constitutive equations by considering a test particle surrounded by an effective medium whose properties are determined in a consistent way. To take into account spatial correlations (that is to say the modification of the particles location due to the presence of the test sphere), different approximations have been introduced (see Refs. 25–27 for a comparison of the different

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ones). Some take into account the continuous variation of the conditional volume fraction with the distance from the test particle and others approximate this variation by a step function and therefore reduce to "core shell models." A core-shell approximation (originally introduced by Dodd *et al.*[28]) has been successfully used by Spelt *et al.*[29] to compute the propagation of acoustical waves in suspensions of different natures. In their work, the suspension is considered as a whole and the plane acoustical wave impinging a test particle is decomposed according to the ECAH theory[4,5] into compressional, thermal, and viscous modes. Therefore, this model can be used for a large range of frequencies and different kinds of suspensions.

In the following, the same closure scheme (self-consistent approximation with core-shell approximation) is introduced to compute the exchange terms in our two-phase formulation. However, simpler equations are obtained by considering only the scattering mechanisms and pole orders necessary for the study of rigid particle in the long wavelength regime (LWR). Moreover, vectorial expressions of the closure terms (force and stresslet) are obtained and the acceleration of the particles is taken into account. These expressions can therefore be used for other purposes than plane acoustical waves. We must note that for a plane acoustical wave propagating in suspensions of rigid particles in the LWR, the two models should correspond.

The present model is used to study the influence of spatial correlations on the sound propagation in solutions of rigid particles. In the first section, we recall the linearized two-phase equations obtained in a previous paper[30] to describe the sound propagation in solutions of rigid particles. Then, we derive the equations for the conditionally averaged fields which should be solved to take into account spatial correlations and, in particular, the continuous variation of the conditional volume fraction with the distance from the test particle. To perform the explicit calculation of the dispersion equation, a simplification of this problem is used: the conditional volume fraction is approximated by a step function. In this way, a "core-shell" model is obtained but with a "core radius" related to the particle volume fraction and radius by a complex function calculated from Percus–Yevick (PY) theory for hard spheres.[31,32] The results are finally compared to the experiments of Hipp *et al.*[33] performed in solutions of silica particles for different frequencies, particle sizes, and concentration and an excellent agreement is reached.

II. COUPLED PHASE THEORY

A. Linearized ensemble averaged equations and the hierarchy of balance equations

In a precedent paper,[30] ensemble averaged equations have been derived to describe the propagation of acoustical waves in homogeneous solutions of monodisperse rigid particles. They were obtained from local-instant balance equations in each phase by the introduction of the phasic function:

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

which allows to broaden the validity of local balance equations to every position and time, and by the use of a statistical average (noted $\langle\,\rangle$ hereafter) such as

$$\langle G'(\mathbf{x},t)\rangle = \int G'(\mathbf{x},t|C_N)p(t,C_N)dC_N,$$

where $p(t,C_N)dC_N$ is the probability of finding the N particles in the vicinity of $C_N=(\mathbf{x}_1,\ldots,\mathbf{x_N})$, regardless of their order and $G'=\Sigma_k\chi_k G'_k$ is a local function generalized to every position and time by the use of the phasic function.

Once linearized, the balance equations stand under the following form.

Mass conservation,

$$\rho_{co}\left(\frac{\partial\alpha_c}{\partial t}+\alpha_{co}\,\mathrm{div}(\mathbf{v_c})\right)+\alpha_{co}\frac{\partial\rho_c}{\partial t}=0, \tag{1}$$

$$\rho_{do}\left(\frac{\partial\alpha_d}{\partial t}+\alpha_{do}\,\mathrm{div}(\mathbf{v_d})\right)+\alpha_{do}\frac{\partial\rho_d}{\partial t}=0, \tag{2}$$

$$\alpha_d = 1-\alpha_c. \tag{3}$$

Momentum conservation,

$$\alpha_{co}\rho_{co}\frac{\partial\mathbf{v_c}}{\partial t} = -\nabla(\alpha_c p_c)+\mu_c\Delta\mathbf{v}+(\lambda_c+\mu_c)\nabla\ \mathrm{div}(\mathbf{v}) + \mathrm{div}\,\mathbf{S}-\mathbf{F}, \tag{4}$$

$$\alpha_{do}\rho_{do}\frac{\partial\mathbf{v_d}}{\partial t}=\mathbf{F}. \tag{5}$$

Equations of state,

$$\rho_c = p_c/c_{co}^2, \tag{6}$$

$$\rho_d = p_c/c_{do}^2. \tag{7}$$

In these equations, the subscripts o, c, and d denote, respectively, the equilibrium state, and the continuous and the dispersed phases, $\alpha_k=\langle\chi_k\rangle$ is the volume fraction of phase k while $\rho_k=\langle\chi_k\rho'\rangle$ is its mean density and $\mathbf{v_k}=\langle\chi_k\rho'\mathbf{v}'\rangle/\langle\chi_k\rho'\rangle$ its mean velocity. Finally, p_c, μ_c, and $\zeta_c=\lambda_c+2\mu_c/3$ are, respectively, the pressure, the shear, and the bulk viscosities of the continuous phase, c_{ko} the sound speed in phase k, $\mathbf{v}=\alpha_{co}\mathbf{v_c}+\alpha_{do}\mathbf{v_d}$ the average velocity of the suspension, $\mathbf{F}=\langle\chi_d\,\mathrm{div}(\mathbf{\Pi}')\rangle$ the interphase force, and $\mathbf{S}=\langle\chi_d\mathbf{\Pi}'\rangle$ the average of the local generalized stress tensor $\mathbf{\Pi}'=\Sigma_{k=c,d}\chi_k\Pi'_k$. We can note that in Eq. (2) (mass conservation for the dispersed phase), the compressibility of the particles has been taken into account in order to give a correct prediction of both attenuation and dispersion for concentrated suspensions of silica nanoparticle. Even if these particles are 35 times less compressible than water (ratio 2, 2 for density, 4 for the sound speed, and therefore 35 for the compressibility $\xi=1/\rho c^2$), their departure from a perfectly rigid behavior might have an influence on the effective sound speed in concentrated suspensions. However, this modification does not affect the attenuation curves and they will only induce a global shift of the effective sound speed as we are far from the particle resonances in the frequency range considered here.

To achieve closure, the interphase force $\mathbf{F}$ and the stresslet $\mathbf{S}$ must be expressed in terms of the averaged fields. The first step is to establish the link between these expressions and the so-called test particle problem. This particular issue was addressed by Buyevich and Shchelchkova:[24]

$$\mathbf{F} = \langle \chi_d \,\mathrm{div}(\mathbf{\Pi}') \rangle \approx \frac{3\alpha_d}{4\pi a^3} \oint \langle \mathbf{\Pi}' \rangle_{\mathbf{x}} \,.\, \mathbf{n} dS, \tag{8}$$

$$\mathbf{S} = \langle \chi_d \mathbf{\Pi}' \rangle \approx \frac{3\alpha_d}{4\pi a^3} \oint^{(s)} \mathbf{a} \otimes (\mathbf{n} \,.\, \langle \mathbf{\Pi}' \rangle_{\mathbf{x}}) dS, \tag{9}$$

where a is the radius of the particle, $\mathbf{n}$ the normal vector, and $\langle\ \rangle_{\mathbf{x}}$ the statistical average conditioned by the knowledge of the position of one particle in $\mathbf{x}$:

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

With these expressions, the interphase force and stresslet are related to the conditionally averaged fields. One could therefore decide to derive the balance equations for the conditionally averaged fields. The same equations would be obtained, but now the conditional force $\mathbf{F_x}$ and stresslet $\mathbf{S_x}$ would depend on the averaged fields with the positions of two particles being known and so on. In this way, an infinite hierarchy of interdependent equations, similar to the cluster expansion which appears in statistical physics, would be disclosed. It was rigorously established by Hinch[17] in 1977.

Now arises the problem of the efficient closure of this hierarchy. The first idea consists of truncating this hierarchy at a certain order. At first order, the constitutive Eqs. (8) and (9) are calculated for a sphere embedded in the pure continuous phase. In this case, interactions between particles are completely left out. This is the approximation classically used in two-phase models.[6,34] At second order, the conditional force $\mathbf{F_x}$ and stresslet $\mathbf{S_x}$ are calculated for a cluster of two spheres lying in the pure continuous phase. In this case, binary interactions of pairs of sphere are taken into account while ternary or higher order interactions are completely left out. One could of course calculate these expressions for higher order clusters but this method is limited. First, because the calculation of the constitutive equations becomes more and more difficult when the size of the cluster increases. Second, because interactions between $N+1$ particles are no more negligible compared to interactions between N particles in very concentrated solutions, and thus the whole hierarchy must be considered.

It is the merit of the pioneering work of Lundgren[23] and Buyevich *et al.*[24,35] to have proposed an alternative *self-consistent effective medium theory* that takes into account interactions at all order within a certain approximation. These self-consistent schemes have then been extended to even more concentrated medium, when spatial correlations must be considered. For this purpose, different approximations have been introduced and they are compared in a paper by Sangani and Yao.[26,27] In our precedent paper,[30] a self-consistent effective medium theory had been used to take into account the influence of hydrodynamic interactions between particles on the propagation of acoustical waves in suspensions of rigid particles. However, spatial correlations were not considered and the theory will now be modified to include these effects.

B. The long wavelength regime

Before delving into this crucial problem, the balance equations will be simplified in the neighborhood of a test particle, lying at position $\mathbf{x}$ to calculate the surface integrals (8) and (9). For that purpose, a mesoscopic scale l such that $a \ll l \ll \lambda$ can be introduced whenever the wavelength λ is much larger than the radius a of the particle, that is to say in the LWR. Within a cell of characteristic length l around the test particle, all terms linked to the compressibility of the continuous phase can be neglected and thus Eqs. (1)–(7) reduce to the following form after Fourier transform:

$$\mathrm{div}(\mathbf{v_c}) = \mathrm{div}(\mathbf{v_d}) = 0, \tag{10}$$

$$-\alpha_{co}\rho_{co}(i\omega)\mathbf{v_c} = -\nabla(\alpha_c p_c) + \mu_c \Delta \mathbf{v} + \mathrm{div}\,\mathbf{S} - \mathbf{F}, \tag{11}$$

$$-\alpha_{do}\rho_{do}(i\omega)\mathbf{v_d} = \mathbf{F}. \tag{12}$$

If we rewrite them in the convective frame of reference related to the velocity of the test particle, we simply obtain

$$\mathrm{div}(\mathbf{V_c}) = \mathrm{div}(\mathbf{V_d}) = 0, \tag{13}$$

$$-\alpha_{co}\rho_{co}(i\omega)\mathbf{V_c} = -\nabla(\alpha_c p_c) + \mu_c \Delta \mathbf{V} + \mathrm{div}\,\mathbf{S} - \mathbf{F} - \alpha_{co}\rho_{co}\nabla\Psi, \tag{14}$$

$$-\alpha_{do}\rho_{do}(i\omega)\mathbf{V_d} = \mathbf{F} - \alpha_{do}\rho_{do}\nabla\Psi, \tag{15}$$

where $\mathbf{V_k} = \mathbf{v_k} - \mathbf{v_d}|_{r=0}$ is the average velocity of phase k in the new frame of reference, $\Psi = -i\omega\mathbf{r}$. $\mathbf{v_d}|_{r=0}$ is a function introduced to take into account the acceleration of the test particle, $\mathbf{r} = \mathbf{x}' - \mathbf{x}$ is the distance from the test particle, and ω the frequency of the propagating wave.

Similar balance equations can also be derived for the conditionally averaged fields but this time, the conditional volume fraction $\alpha_{ko,\mathbf{x}}$ of phase k stands instead of the unconditional one:

$$\mathrm{div}(\alpha_{co,\mathbf{x}}\mathbf{V_{c,x}}) = \mathrm{div}(\alpha_{do,\mathbf{x}}\mathbf{V_{d,x}}) = 0, \tag{16}$$

$$-\alpha_{co,\mathbf{x}}\rho_{co}(i\omega)\mathbf{V_{c,x}} = -\nabla(\alpha_{c,\mathbf{x}} p_{c,\mathbf{x}}) + \mu_c \Delta \mathbf{V_x} + \mathrm{div}\,\mathbf{S_x} - \mathbf{F_x} - \alpha_{co,\mathbf{x}}\rho_{co}\nabla\Psi, \tag{17}$$

$$-\alpha_{do,\mathbf{x}}\rho_{do}(i\omega)\mathbf{V_{d,x}} = \mathbf{F_x} - \alpha_{do,\mathbf{x}}\rho_{do}\nabla\Psi, \tag{18}$$

where $\mathbf{V_{k,x}} = \mathbf{v_{k,x}} - \mathbf{v_d}|_{r=0}$.

On the test sphere $(r=a)$, the conditional velocity of the continuous phase is null and far from it $(r \to \infty)$, the influence of the test particle vanishes. We therefore obtain the following boundary conditions:

$$\mathbf{V_{c,x}} = 0 \quad \text{in } r = a, \tag{19}$$

$$\{\mathbf{V_{c,x}}, \mathbf{V_{d,x}}, p_{c,\mathbf{x}}\} \rightarrow \{\mathbf{V_c}, \mathbf{V_d}, p_c\} \quad \text{when } r \rightarrow \infty. \tag{20}$$

III. SPATIAL CORRELATIONS

A. The conditional volume fraction

For pointlike particle, there would be no difference between the conditional volume fraction $\alpha_{do,\mathbf{x}}$ and the unconditional one α_{do}. However, the nonoverlapping property of hard spheres modifies the distribution of particles in the neighborhood of the test particle. We will now see how the conditional volume fraction can be estimated for hard spheres.

First, the so-called distribution function $p(t,\mathbf{x}|\mathbf{x}')$ (which is nothing but the probability of finding one of the sphere center in $\mathbf{x}'$ when another particle is lying in $\mathbf{x}$) can be calculated with models inherited from statistical physics such as the PY (Refs. 31 and 32 or hypernetted chain[36,37] (HNC) models. Numerical methods (for example, Monte Carlo simulations) could also be used but PY theory provides accurate and easily computable estimate of this function.

Then the conditional volume fraction $\alpha_{do,\mathbf{x}}(\mathbf{x}')$ can be deduced from the distribution function $p(t,\mathbf{x}|\mathbf{x}')$ with the following formula[38] for hard spheres:

$$\alpha_{do,\mathbf{x}}(\mathbf{x}') = \int_{|\mathbf{x}''-\mathbf{x}'|\leqslant a} p(t,\mathbf{x}''|\mathbf{x}) d\mathbf{x}''. \tag{21}$$

For isotropically distributed spheres, the distribution function depends only on the distance $r' = |\mathbf{x}'' - \mathbf{x}|$ and thus this formula reduces to

$$\alpha_{do,\mathbf{x}}(r) = \int_{\max(2a,r-a)}^{r+a} p(t,r') \frac{\pi r'}{r} [2rr' - r'^2 - r^2 + a^2] dr', \tag{22}$$

with $r = |\mathbf{x}' - \mathbf{x}|$. Figure 1 illustrates the evolution of the distribution function and the conditional volume fraction with the distance r from the test particle center, calculated from PY theory for hard spheres. We can note that the nonoverlapping condition does not mean that some parts of the particles cannot lie in the region $a \leqslant r \leqslant 2a$ but just that the centers of the particles are excluded from it. That is why the volume fraction progressively increases in this region contrarily to the distribution function which is null.

B. The self-consistent effective medium closure scheme

We will now apply the self-consistent condition (called method A in papers from Chang *et al.*[25,39] and Yao and Sangani[26,27]) in order to close the infinite hierarchy previously described. We can note that contrarily to the scheme proposed by Buyevich[38,40] (called method B in the papers of Chang *et al.*), closure will be obtained for the conditionaly averaged field and not for the perturbation (as defined by Buyevich in his paper).

To achieve closure, the force $\mathbf{F}$ and the stresslet $\mathbf{S}$ must necessarily be expressed in terms of the average fields and their space and time derivatives of appropriate tensor dimensionality.

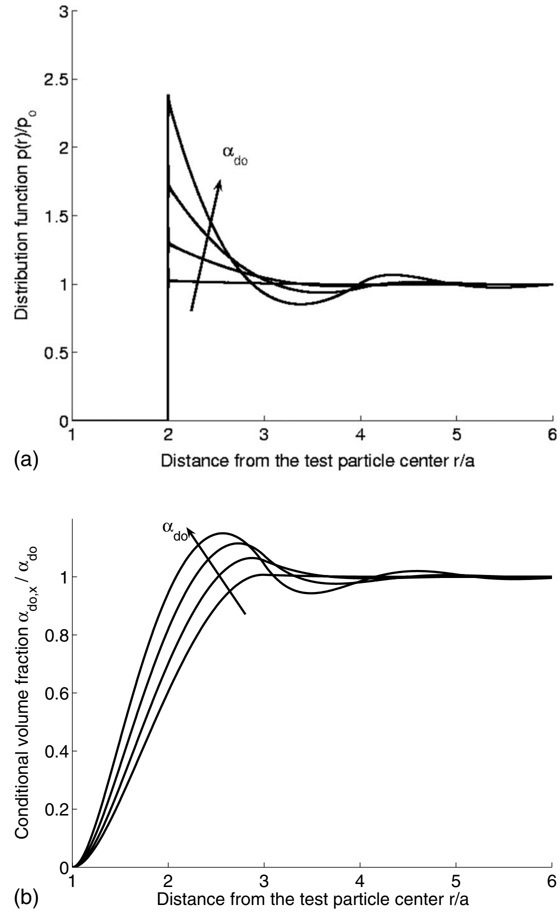

FIG. 1. Evolution of the distribution function and the conditional volume fraction with the distance r/a from the test particle center for volume fraction α_{do} of, respectively, 1%, 10%, 20%, and 30%. These curves are calculated with PY theory.

$$\mathbf{F} = f\langle \mathbf{V_c}, \mathbf{V_d}, \nabla p_c, \nabla\psi, \Delta\mathbf{V_c}, \Delta\mathbf{V_d}, \dots \rangle, \tag{23}$$

$$\text{div}(\mathbf{S}) = s\langle \mathbf{V_c}, \mathbf{V_d}, \nabla p_c, \nabla\psi, \Delta\mathbf{V_c}, \Delta\mathbf{V_d}, \dots \rangle. \tag{24}$$

Since the two-phase equations considered here are linear, f and s must be linear functions of their arguments. Moreover, these functions must depend on the frequency ω to take into account the time derivatives in the Fourier space. For a moving sphere embedded in a pure ambient fluid, the expressions of f and s are well known:[41,42]

$$\mathbf{F} = \alpha_{do}[n_1(\mathbf{V_c} - \mathbf{V_d}) + n_2\Delta\mathbf{V_c} + n_3 \nabla \Psi], \tag{25}$$

$$\text{div}(\mathbf{S}) = \nabla(\alpha_d p_c) + \alpha_{do} n_o \Delta\mathbf{V_c}, \tag{26}$$

where n_o, n_1, n_2, and n_3 are the coefficients that depend on the pure fluid properties $(\mu_c\rho_{co})$ and on the frequency ω. In these expressions, $n_1(\mathbf{V_c} - \mathbf{V_d})$ corresponds to the sum of the Stokes drag, the Basset hereditary, and the total inertial forces, $n_2\Delta\mathbf{V_c}$ is the Faxen correction due to the nonuniformity of the ambient fluid velocity, and $n_3\nabla\Psi$ is due to the acceleration of the test particle.

If we now take into account the influence of the other distributed spheres, the particle is no more embedded in the pure fluid but in an effective medium whose properties are unknown at this stage of the derivation. In this case, the force $\mathbf{F}$ and the stresslet $\mathbf{S}$ will be related to the averaged fields in the same way but with new coefficients $\tilde{n}_k$ which depend on the effective properties of the surrounding fluid $(\mu_{\text{eff}}, \rho_{\text{eff1}}, \rho_{\text{eff2}})$:

$$\mathbf{F} = \alpha_{do}[\tilde{n}_1(\mathbf{V_c} - \mathbf{V_d}) + \tilde{n}_2 \Delta \mathbf{V_c} + \tilde{n}_3 \nabla \Psi], \tag{27}$$

$$\text{div}(\mathbf{S}) = \nabla(\alpha_d p_c) + \alpha_{do}\tilde{n}_0 \Delta \mathbf{V_c}, \tag{28}$$

where ρ_{eff1} and ρ_{eff2} are some effective densities, respectively, linked to the inertial phenomena and the change of frame of reference, and μ_{eff} is the effective viscosity of the solution. We must underline how important is the hypothesis of homogeneity of the suspension at this stage of the derivation because an inhomogeneous distribution of the particles might induce extra forces related to the gradient of the volume fraction.

The self-consistent condition consists of keeping the *same* coefficients $\tilde{n}_k$ to express $\mathbf{F_x}$ and $\mathbf{S_x}$ in terms of the conditionally averaged fields:

$$\mathbf{F_x} = \alpha_{do,\mathbf{x}}[\tilde{n}_1(\mathbf{V_{c,x}} - \mathbf{V_{d,x}}) + \tilde{n}_2 \Delta \mathbf{V_{c,x}} + \tilde{n}_3 \nabla \Psi], \tag{29}$$

$$\text{div}(\mathbf{S_x}) = \nabla(\alpha_{d,\mathbf{x}} p_{c,\mathbf{x}}) + \alpha_{do,\mathbf{x}}\tilde{n}_0 \Delta \mathbf{V_{c,x}}. \tag{30}$$

Of course, the conditional volume fraction replaces the unconditional one because the distribution of the particles is modified by the presence of the test sphere.

Now, the effective properties of the surrounding fluid must be determined in a consistent way. For that purpose, Eqs. (13)–(15) [with the expressions of $\mathbf{F}$ and $\mathbf{S}$ given by Eqs. (27) and (28)] must be properly combined to obtain a final set of equations in the effective medium similar to the equations which would stand in a pure fluid:

$$\text{div}(\mathbf{V_c}) = 0, \tag{31}$$

$$-\rho_{\text{eff1}}(i\omega)\mathbf{V_c} = -\nabla p_c + \mu_{\text{eff}} \Delta \mathbf{V_c} - \rho_{\text{eff2}} \nabla \Psi. \tag{32}$$

As mentioned earlier by the authors,[30] Eqs. (31), (32), and (13)–(15) form a closed system and thus the effective properties can be expressed in terms of the properties of the continuous and dispersed phases, and the coefficients $\tilde{n}_k$ (see Ref. 41 for more details about this calculation):

$$\rho_{\text{eff1}} = \alpha_{co}\rho_{co} + \frac{\alpha_{do}\rho_{do}\tilde{n}_1}{\tilde{n}_1 - i\omega\rho_{do}}, \tag{33}$$

$$\mu_{\text{eff}} = \alpha_{co}\mu_c + \alpha_{do}\tilde{n}_0 + \frac{\alpha_{do}\rho_{do} i\omega \tilde{n}_2 + \alpha_{do}\mu_c(\tilde{n}_1 - i\omega\tilde{n}_2\rho_{\text{eff1}}/\mu_{\text{eff}})}{\tilde{n}_1 - i\omega\rho_{do}}, \tag{34}$$

$$\rho_{\text{eff2}} = \alpha_{co}\rho_{co} + \alpha_{do}\rho_{do}\frac{\tilde{n}_1 - \tilde{n}_3 i\omega}{\tilde{n}_1 - i\omega\rho_{do}}. \tag{35}$$

Our expression of μ_{eff} slightly differs from the expression obtained by Buyevich because of a different choice in the definition of $\tilde{n}_0$, which is more appropriate for our study.

The final step consists of calculating integrals (8) and (9) to express the coefficients $\tilde{n}_k$ in terms of the effective properties of the surrounding fluid. For that purpose, Eqs. (16)–(18) with $\mathbf{F_x}$ and $\mathbf{S_x}$ given by Eqs. (29) and (30) have to be solved.

The solution of Eqs. (31) and (32) for the averaged fields (equivalent to the so-called Brinkman equations) is well known: it was independently solved by Howells[43] for porous media and Buyevich and Markov[35] for the calculation of the force applied on a moving sphere embedded in an unsteady nonuniform velocity field. However, the resolution of the equations for the conditionally averaged fields would be a challenging task because the variation of the conditional volume fraction with the distance r introduces new terms in the equation.

C. Approximation of the conditional volume fraction by a step function

To simplify this calculation, the evolution of the volume fraction obtained with PY theory in Sec. III A will be approximated by a step function:

$$\alpha_{co,\mathbf{x}} = \chi_p + \alpha_{co}\chi_e, \tag{36}$$

$$\alpha_{do,\mathbf{x}} = \alpha_{do}\chi_e, \tag{37}$$

where

$$\chi_p = \begin{cases} 1 & \text{if } a < r < R_c \\ 0 & \text{if } r > R_c \end{cases}, \quad \chi_e = \begin{cases} 0 & \text{if } a < r < R_c \\ 1 & \text{if } r > R_c, \end{cases}$$

and R_c is given by the following formula:

$$\int_{r=a}^{2a} \alpha_{do,\mathbf{x}}(r)d\mathbf{r} + \int_{r=2a}^{\infty} (\alpha_{do,\mathbf{x}}(r) - \alpha_{do})d\mathbf{r} = \int_{R_c<r<2a} \alpha_{do} d\mathbf{r}. \tag{38}$$

With this definition of R_c, the volume occupied by the particles is conserved and also the asymptotic behavior when $r \to \infty$. In this way, a "core-shell" model is obtained (see Fig. 2). The particle is surrounded by a layer of pure fluid which is itself embedded in a homogeneous effective medium. The condition (38) introduced here to calculate the evolution of the "core radius" R_c with the volume fraction (as illustrated by Fig. 3) is equivalent to the one introduced by Spelt *et al.*[29] The only difference is that these authors expressed R_c in terms of the number density, but it does not affect its estimation.

We can now split the conditionally averaged fields into their value in the pure fluid layer and their value in the homogeneous effective medium:

$$\alpha_{co,\mathbf{x}}\mathbf{V_{c,x}} = \chi_p \mathbf{V^p_{c,x}} + \chi_e \alpha_{co}\mathbf{V^e_{c,x}},$$

$$\alpha_{do,\mathbf{x}}\mathbf{V_{d,x}} = \chi_e \alpha_{do}\mathbf{V^e_{d,x}},$$

$$p^e_{c,x} = \chi_p p^p_{c,x} + \chi_e p^e_{c,x},$$

and thus deduce the balance equations in both parts. In the pure fluid layer $(a < r < R)$,

$$\text{div}(\mathbf{V^p_{c,x}}) = 0, \tag{39}$$

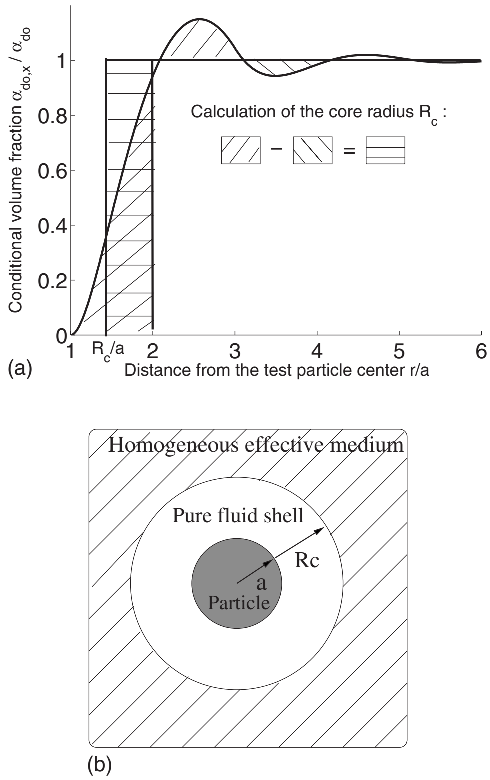

FIG. 2. Approximation of the conditional volume fraction by a step function: core shell model.

$$-\rho_{co}(i\omega)\mathbf{V}^{\mathbf{p}}_{\mathbf{c,x}} = -\nabla p^{p}_{c,x} + \mu_c \Delta \mathbf{V}^{\mathbf{p}}_{\mathbf{c,x}} - \rho_{co} \nabla \Psi. \tag{40}$$

In the homogeneous effective medium ($r > R$),

$$\text{div}(\mathbf{V}^{\mathbf{e}}_{\mathbf{c,x}}) = 0, \tag{41}$$

$$-\rho_{\text{eff1}}(i\omega)\mathbf{V}^{\mathbf{e}}_{\mathbf{c,x}} = -\nabla p^{e}_{c,x} + \mu_{\text{eff}} \Delta \mathbf{V}^{\mathbf{e}}_{\mathbf{c,x}} - \rho_{\text{eff2}} \nabla \Psi. \tag{42}$$

The problem is therefore reduced to the study of a particle embedded in a pure fluid shell (with a viscosity μ_c and a density ρ_{co}), which is itself surrounded by a homogeneous effective medium (with effective properties μ_{eff}, ρ_{eff1}, and ρ_{eff2}).

Now, the boundary conditions must be expressed for the different fields. On the particle surface ($r=a$), the conditionally averaged velocity of the continuous phase is equal to zero that is to say

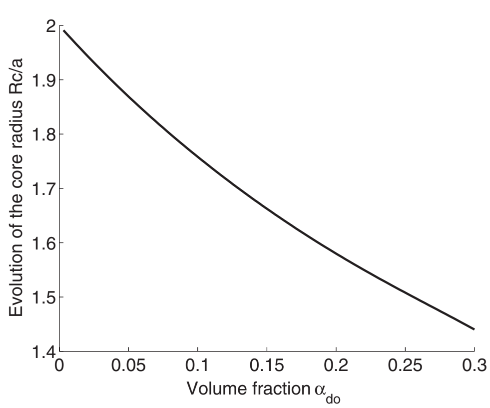

FIG. 3. Evolution of the core radius with the volume fraction.

$$\alpha_{co,\mathbf{x}} \mathbf{V}_{\mathbf{c,x}} = \mathbf{V}^{\mathbf{p}}_{\mathbf{c,x}} = 0.$$

Then, the mass conservation equation for the mean velocity $[\text{div}(\mathbf{V}_{\mathbf{x}}) = 0]$ imposes the following condition at the core-shell surface ($r = R_c$):

$$\mathbf{V}^{\mathbf{p}}_{\mathbf{c,x}} = \alpha_{co} \mathbf{V}^{\mathbf{e}}_{\mathbf{c,x}} + \alpha_{do} \mathbf{V}^{\mathbf{e}}_{\mathbf{d,x}}, \tag{43}$$

where $\mathbf{V}^{\mathbf{e}}_{\mathbf{d,x}}$ can be expressed in terms of $\mathbf{V}^{\mathbf{e}}_{\mathbf{c,x}}$ and $\nabla\Psi$ through Eq. (18).

The momentum conservation gives the following condition in $r = R_c$:

$$\mathbf{\Pi}^{\mathbf{p}}_{\mathbf{x}} \,.\, \mathbf{n} = \mathbf{\Pi}^{\mathbf{e}}_{\mathbf{x}} \,.\, \mathbf{n}, \tag{44}$$

where $\mathbf{n}$ is the normal vector to the surface of the core and $\mathbf{\Pi}^{\mathbf{p}}_{\mathbf{x}}$ and $\mathbf{\Pi}^{\mathbf{e}}_{\mathbf{x}}$ are, respectively, the strain tensors in the pure layer and in the homogeneous effective medium:

$$\mathbf{\Pi}^{\mathbf{p}}_{\mathbf{x}} = -p^{p}_{c,x} + 2\mu_c \mathbf{D}^{\mathbf{p}}_{\mathbf{c,x}} \quad \text{with } \mathbf{D}^{\mathbf{p}}_{\mathbf{c,x}} = 1/2(\nabla \mathbf{V}^{\mathbf{p}}_{\mathbf{c,x}} + \nabla^t \mathbf{V}^{\mathbf{p}}_{\mathbf{c,x}}),$$

$$\mathbf{\Pi}^{\mathbf{e}}_{\mathbf{x}} = -p^{e}_{c,x} + 2\mu_{\text{eff}} \mathbf{D}^{\mathbf{e}}_{\mathbf{c,x}} \quad \text{with } \mathbf{D}^{\mathbf{e}}_{\mathbf{c,x}} = 1/2(\nabla \mathbf{V}^{\mathbf{e}}_{\mathbf{c,x}} + \nabla^t \mathbf{V}^{\mathbf{e}}_{\mathbf{c,x}}).$$

Finally, far from the test particle ($r \to \infty$), the perturbation induced by its presence vanishes so that

$$\{\mathbf{V}^{\mathbf{e}}_{\mathbf{c,x}}, p^{e}_{c,x}\} \to \{\mathbf{V}_{\mathbf{c}}, p_c\}. \tag{45}$$

With Eqs. (31), (32), and (39)–(45), we have derived all the equations and boundary conditions necessary to compute integrals (8) and (9), which reduce to

$$\mathbf{F} = \frac{3\alpha_d}{4\pi a^3} \oint \mathbf{\Pi}^{\mathbf{p}}_{\mathbf{x}} \,.\, \mathbf{n} dS, \tag{46}$$

$$\mathbf{S} = \frac{3\alpha_d}{4\pi a^3} \oint^{(s)} \mathbf{a} \otimes (\mathbf{n} \,.\, \mathbf{\Pi}^{\mathbf{p}}_{\mathbf{x}}) dS. \tag{47}$$

D. Calculation of the closure terms

To solve these equations, the velocity and pressure fields must be expressed in terms of spherical functions according to the method developed by Buyevich and Markov.[42] The details of this calculation can be found in Appendixes A and B. In this section, we only give the final expressions of the coefficient $\tilde{n}_k$ which result from the identification of the expression of $\mathbf{F}$ and $\mathbf{S}$ calculated in the Appendix and formulas (27) and (28).

1. Expression of $\tilde{n}_0$

$$\tilde{n}_0 = 3\frac{\mu_c \beta_c}{\beta_{\text{eff}}}\left[\left(\dot{S}_2(\gamma) + \left(\frac{4}{\gamma} + \frac{\gamma}{2}\right)S_2(\gamma)\right)V_{C_1} + \left(\dot{Q}_2(\gamma) + \left(\frac{4}{\gamma} + \frac{\gamma}{2}\right)Q_2(\gamma)\right)V_{C_2} - \left(\frac{10}{\gamma} + \gamma\right)V_{C_3} - \gamma V_{C_4}\right],$$

where

$$S_2(X) = \frac{(X^2 - 3X + 3)e^{X} - (X^2 + 3X + 3)e^{-X}}{2X^4},$$

$$Q_2(X) = \frac{(X^2 + 3X + 3)e^{-X}}{X^4},$$

and

$$\epsilon = \beta_{\text{eff}}a, \quad \gamma = \beta_c a,$$

$$\eta = \beta_{\text{eff}}R_c, \quad \delta = \beta_c R_c,$$

$$\beta_c^2 = -(i\omega)\rho_{co}/\mu_c, \quad \beta_{\text{eff}}^2 = -(i\omega)\rho_{\text{eff }1}/\mu_{\text{eff}},$$

$$\kappa = \beta_c\mu_c/\beta_{\text{eff}}\mu_{\text{eff}}.$$

Finally, $V_C=[V_{C_1}V_{C_2}V_{C_3}V_{C_4}V_{C_5}V_{C_6}]^t$ is a column vector whose expression is given by

$$V_C = M_2^{-1}V_{A2}, \tag{48}$$

where the expressions of M_2 and V_{A2} are given in Appendix C.

2. Expression of $\tilde{n}_1$, $\tilde{n}_2$, and $\tilde{n}_3$

$$\tilde{n}_1 = -\mu_c\beta_c^2[S_1(\gamma)V_{E_1} + Q_1(\gamma)V_{E_2} - V_{E_3} - V_{E_4}], \tag{49}$$

$$\tilde{n}_2 = \frac{\mu_c\beta_c^2}{\beta_{\text{eff}}^2}[S_1(\gamma)(3V_{D_1} + V_{E_1}) + Q_1(\gamma)(3V_{D_2} + V_{E_2}) - (3V_{D_3} + V_{E_3}) - (3V_{D_4} + V_{E_4})], \tag{50}$$

$$\tilde{n}_3 = \rho_{co} + \mu_c\beta_c^2[S_1(\gamma)V_{F_1} + Q_1(\gamma)V_{F_2} - V_{F_3} - V_{F_4}], \tag{51}$$

where

$$S_1(X) = \frac{(X-1)e^X + (X+1)e^{-X}}{2X^3},$$

$$Q_1(X) = \frac{(X+1)e^{-X}}{X^3},$$

and V_D, V_E, and V_F are some column vectors whose expressions are given by

$$V_D = M_1^{-1}V_{A1}, \tag{52}$$

$$V_E = M_1^{-1}V_{M1}, \tag{53}$$

$$V_F = M_1^{-1}V_{P1}, \tag{54}$$

where the expression of M_1, V_{A1}, V_{M1}, and V_{P1} are given in Appendix C.

With these expressions, the coefficients $\tilde{n}_k$ are related to the effective parameters μ_{eff}, ρ_{eff1}, and ρ_{eff2}, which are themselves related to the coefficients $\tilde{n}_k$ through Eqs. (33)–(35). Thus, the system is closed and the coefficients $\tilde{n}_k$ can be calculated either with a simple iterative procedure or with more elaborate numerical schemes such as the "globally convergent Newton's method."

IV. RESULTS AND COMPARISON WITH EXPERIMENTS

A. Final dispersion equation

Then, the expression of the force (27) and the stresslet (28) can be substituted in the linearized system (1)–(7), and the dispersion equation can be derived for a plane wave by considering fields of the form: $G=G_o+\bar{G}e^{i(k_* x-\omega t)}$, where $\bar{G}$ is the amplitude of the wave, G_o the equilibrium state, and k_* the complex effective wave number. As calculated in a previous paper,[30] the effective wave number k_* is the solution of the following quadratic equation:

$$Ak_*^4 + Bk_*^2 + C = 0,$$

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

$$B = -d_r\left[h_c + \frac{N_0^* + (\lambda_c + 2\mu_c)(\alpha_{co} + \alpha_{do}h_v)/\alpha_{do}\rho_{do}}{i\omega}\right] - \frac{c_{co}^2}{\alpha_{co}\omega^2}\frac{[1 + d_r r h_v]}{[1 + \alpha_{do}\xi_d/\alpha_{co}\xi_c]}, \tag{55}$$

$$C = 1 + d_r h_v,$$

where

$$h_v = \frac{N_1^*}{N_1^* + i\omega(N_3^* - 1)}, \quad h_c = \frac{N_2^*}{N_1^* + i\omega(N_3^* - 1)}$$

and

$$N_k^* = \frac{n_k^*}{\rho_{do}}, \quad d_r = \frac{\alpha_{do}\rho_{do}}{\alpha_{co}\rho_{co}}, \quad r = \frac{\rho_{co}}{\rho_{do}}.$$

We can note that the coefficient $\alpha_{do}\rho_{do}$ was missing in the expression of B in our previous paper (just in the manuscript, the good expression had been considered for the computation). We can also note that a new coefficient $[1+\alpha_{do}\xi_d/\alpha_{co}\xi_c]$ (with ξ_k the compressibility of phase k) appears, as we have taken into account the compressibility of the dispersed phase in Eq. (2).

B. Comparison of the different theories with experiments

Now, the results of this corrected effective medium theory (that take into account spatial correlations) can be compared to previous results[30] obtained with the same theory but without spatial correlations (that is to say when a homogeneous effective medium is considered around the test particle) and also with the classical coupled phase theory (when the test particle is supposed to be surrounded by the pure continuous phase). For that purpose, we will consider the experiments performed by Hipp *et al.*[33] who measured the attenuation of acoustical waves in solutions of silica particle in water for different concentrations, frequencies, and particle sizes. Before analyzing these curves, let us recall some elements which will be useful to understand the influence of spatial correlations. When an acoustical wave propagates through a solution of rigid particles, the particles do not

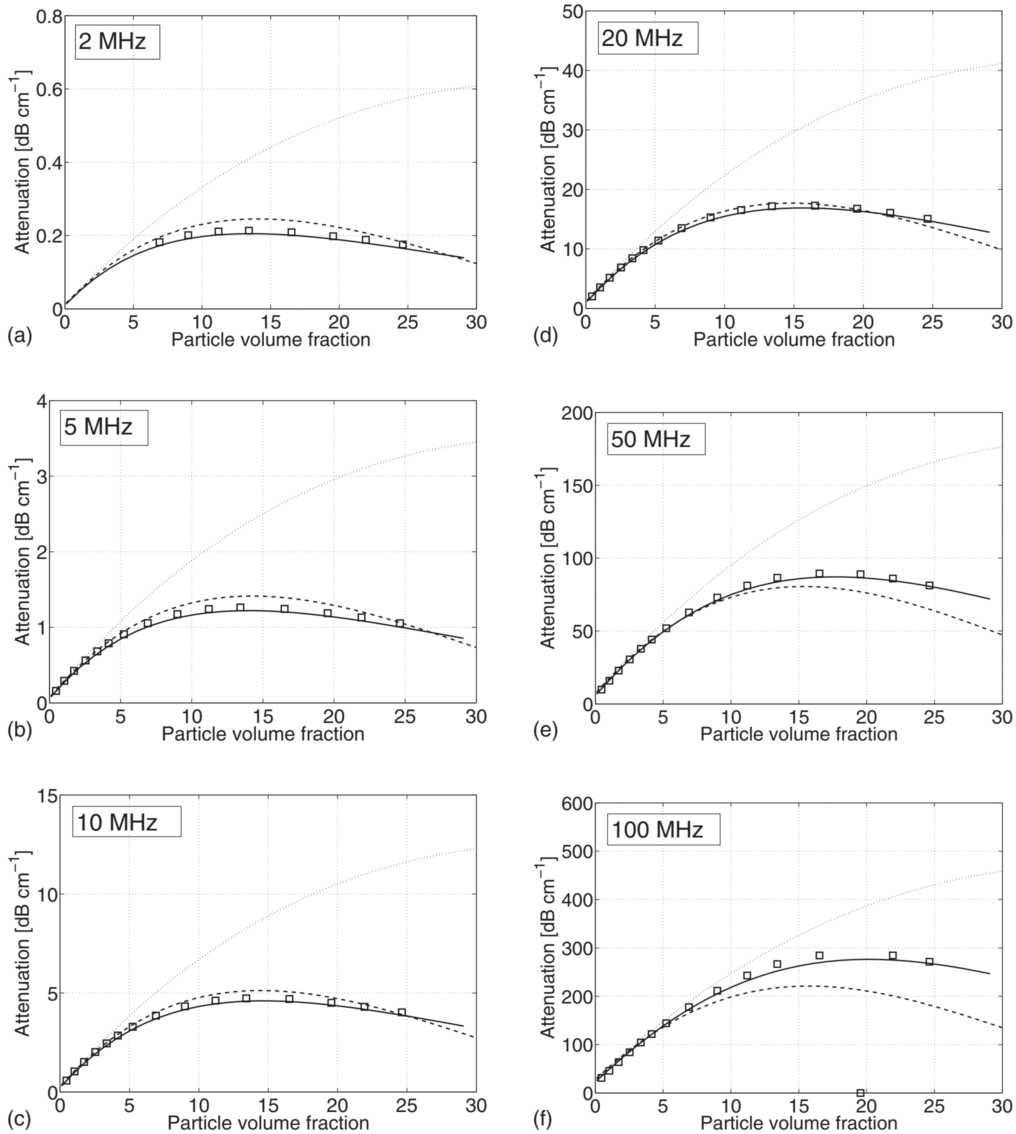

FIG. 4. Attenuation as a function of the volume fraction at various frequencies for silica particles of 56 nm radius in water. The solid lines (-) correspond to the new effective theory, the broken lines (–) to the effective theory without spatial correlationss, the dotted line (..) to the classical coupled phase theory, and the symbols to the experimental data.

move with the same velocity as the surrounding fluid because of the difference of density between them and the surrounding fluid. As a consequence, a dipolar wave is scattered and a part of the energy of the impinging wave is therefore redirected, inducing a loss of spatial coherence. Some energy is also dissipated because of the viscosity of the surrounding fluid which slows down the movement of the particle and therefore converts energy from the compressional propagation mode to a viscous lossy mode. The combination between these two scattering mechanisms is referred to as the "viscoinertial" phenomena. The characteristic length for the decrease of the viscous lossy wave is the size of the boundary layer δ_v which is related to the frequency ω according to the following formula:

$$\delta_v = \sqrt{\frac{2\mu_c}{\omega\rho_c}}.$$

In concentrated suspensions, viscous interactions between neighboring particles may appear according to their concentration and the frequency of the propagating wave. As long as $\delta_v \ll R_c - a$, the properties of the fluid in the boundary layer are very close to the properties of the surrounding pure fluid (because $\alpha_{do,\mathbf{x}} \approx 0$ in this area) and thus the force and the stresselet can be estimated by considering a particle embedded in the pure liquid (as it is done in the classical coupled phase theory). However, when $\delta_v \gg R_c - a$, the variation of the effective properties due to the spatial correlations only concerns a thin part of the boundary layer and thus the approximation of the surrounding fluid by a homogeneous effective medium for the calculation of the closure terms (as it was done in a previous paper[30]) should give good results. As $R_c - a \approx a$, the transition between these two limiting case should happen when $\delta_v \approx a$. For the two suspensions considered here, the corresponding characteristic frequencies are, respectively, of $f_c = 101$ MHz for Fig. 4 and $f_c = 11$ MHz for Fig. 5. Finally, as δ_v is inversely proportional to the frequency, the condition $\delta_v \ll a$ correspond to high frequencies $f \gg f_c$ and the condition $\delta_v \gg a$ to low frequencies $f \ll f_c$.

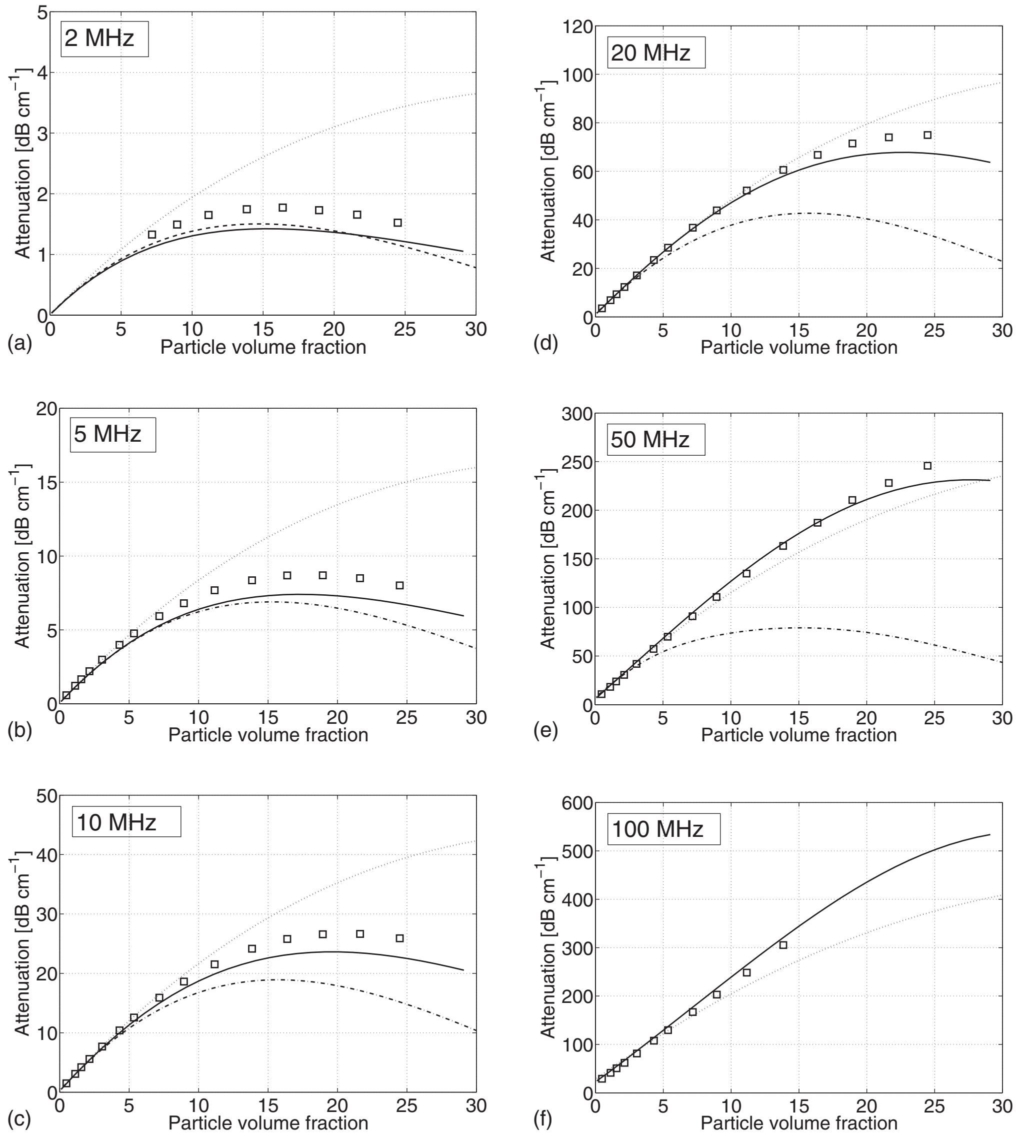

FIG. 5. Attenuation as a function of the volume fraction at various frequencies for silica particles of 164.5 nm radius in water. The solid lines (-) correspond to the new effective theory, the broken lines (–) to the effective theory without spatial correlationss, the dotted line (..) to the classical coupled phase theory, and the symbols to the experimental data.

Effectively, we can see on the different figures that for frequencies $f \ll f_c$, the experimental data are close to the results obtained with the homogeneous effective medium[30] (dash dotted lines), whereas for frequencies $f \gg f_c$, the results are closer to the curves obtained with the classical coupled phase theory (dotted line). As a consequence, none of these theories can properly describe the sound propagation in concentrated suspensions for a wide range of frequencies and particle sizes. For high frequencies and particle concentration, the homogeneous effective theory even gives unphysical effective parameters; that is why we have not plotted the corresponding curves (see Fig. 5 at 100 MHz). With the introduction of spatial correlations, we obtain a model (solid lines) which gives good results for both limiting cases. As expected, the results are less accurate for the transition ($f \approx f_c$) because the progressive evolution of the conditional volume fraction has been approximated by a step function. We can note at this point that equivalent results should be obtained with the model of Spelt *et al.*[29] based on the ECAH[4,5] decomposition. Of course, for low volume fraction ($\alpha_{do} < 5\%$), all these theories give the same results because in this case, the effective properties of the surrounding fluid are close to the properties of the pure fluid. Concerning the remaining discrepancies between our theory and the experiments, different phenomena might explain them such as the polydispersity of the solution or collisions between neighboring particles. Another possible effect might be the increase of the importance of thermal effects in concentrated suspension. It is well known that in dilute suspensions of silica particles in water, viscoinertial effects are more important than thermal ones[44,45] because the first ones are proportional to $(1-r)=O(1)$ in these suspensions and thermal effects to $(\gamma-1) \ll 1$ (where γ is the specific heat ratio). However, viscous interactions make the attenuation induced by viscoinertial effects decrease. In the same way, there will also be some thermal interactions due to the overlapping of viscous boundary layer, which will make the attenuation induced by this scattering mechanism decrease. However, as the viscous boundary layer δ_v is usually larger than the thermal boundary layer δ_t for aqueous solutions (see, for example, Hipp *et al.*[46]), viscoinertial interactions will be more important than thermal interactions and therefore thermal attenuation might become more significant in concentrated suspensions. It would be interesting to investigate this question in a future work.

To conclude this discussion, we can notice that unlike interactions of compressional waves, viscous coupling tends to diminish the attenuation induced by the suspension when the concentration increases, which is quite unusual.

V. CONCLUSION

An effective medium coupled phase theory has been derived to properly describe the sound propagation in concentrated suspensions of rigid particles. An excellent agreement is obtained between this theory and the experimental data of Hipp *et al.* who measured the attenuation induced by the presence of silica particles in water for different particle sizes, concentration up to 30%, and frequencies between 2 and 100 MHz. Moreover, the influence of spatial correlations on the propagation in solutions of rigid particles has been clearly identified. This theory could be improved by considering the exact evolution of the conditional volume fraction with the distance from the test particle center instead of the core-shell aproximation. It could be also extended to polydisperse suspensions but it would require the knowledge of the distribution function in polydisperse suspensions which is not an easy matter.[47] Finally, we can underline that the expressions obtained here for the force and the stresslet could also be used for hydrodynamic studies of concentrated suspensions of particles, as long as the characteristic macroscopic length of the flow is much larger than the size of the particles, and as long as the flow does not modify the distribution of the particles.

ACKNOWLEDGMENTS

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APPENDIX A: RESOLUTION OF THE SYSTEM

First, we can subtract Eqs. (31) and (32) for the averaged fields from Eqs. (41) and (42) for the conditionally averaged fields in the region $r>R_c$ to obtain equations for the perturbation due to the presence of the test sphere:

$$\mathrm{div}(\mathbf{V}_\mathbf{c}^{\mathbf{e}*}) = 0, \tag{A1}$$

$$-\rho_{\mathrm{eff}\,1}(i\omega)\mathbf{V}_\mathbf{c}^{\mathbf{e}*} = -\nabla p_c^{e*} + \mu_{\mathrm{eff}}\Delta \mathbf{V}_\mathbf{c}^{\mathbf{e}*}, \tag{A2}$$

where

$$\mathbf{V}_\mathbf{c}^{\mathbf{e}*} = \mathbf{V}_\mathbf{c,x}^{\mathbf{e}} - \mathbf{V}_\mathbf{c},$$

$$p_c^{e*} = p_{c,x}^{e} - p_c.$$

Then, the boundary conditions can easily be rewritten in terms of the perturbation fields:

$$\mathbf{V}_\mathbf{c,x}^{\mathbf{p}} = 0 \quad \text{in } r=a \tag{A3}$$

$$\mathbf{V}_\mathbf{c,x}^{\mathbf{p}} - \alpha_{co}\mathbf{V}_\mathbf{c}^{\mathbf{e}*} - \alpha_{do}\mathbf{V}_\mathbf{d}^{\mathbf{e}*} = \alpha_{co}\mathbf{V}_\mathbf{c} + \alpha_{do}\mathbf{V}_\mathbf{d} \quad \text{in } r=R_c, \tag{A4}$$

$$\mathbf{\Pi}_\mathbf{x}^{\mathbf{p}}\,.\,\mathbf{n} - \mathbf{\Pi}^{\mathbf{e}*}\,.\,\mathbf{n} = \mathbf{\Pi}_\mathbf{c}\,.\,\mathbf{n} \quad \text{in } r=R_c, \tag{A5}$$

$$\{\mathbf{V}_\mathbf{c}^{\mathbf{e}*}, p_c^{e*}\} \to 0 \quad \text{when } r\to\infty. \tag{A6}$$

Equations (31), (32), (39), (40), (A1), and (A2) can all be written under the form

$$\mathrm{div}(\mathbf{U}) = 0, \tag{A7}$$

$$(\Delta - \beta^2)\mathbf{U} = \nabla R, \tag{A8}$$

where

$$\{U=\mathbf{V}_\mathbf{c}, R = 1/\mu_{\mathrm{eff}}(p_c + \rho_{\mathrm{eff}\,2}\psi), \beta = \beta_{\mathrm{eff}}\} \quad \text{for the first set of equations,}$$

$$\{U=\mathbf{V}_\mathbf{c,x}^{\mathbf{p}}, R = 1/\mu_c(p_{c,x}^p + \rho_{co}\psi), \beta=\beta_c\} \quad \text{for the second,}$$

$$\{U=\mathbf{V}_\mathbf{c}^{\mathbf{e}*}, R = 1/\mu_{\mathrm{eff}}(p_c^{e*}), \beta=\beta_{\mathrm{eff}}\} \text{ for the third one.}$$

Then the velocity and pressure fields can be expressed in terms of spherical functions:

$$U(\mathbf{r}) = \sum_{k=0}^{\infty}\left[F_k(r)s_k(\theta,\phi)\frac{\mathbf{r}}{r} + G_k(r)r\,\nabla\, s_k(\theta,\phi) + H_k(r)\mathbf{r} \times \nabla s_k(\theta,\phi)\right], \tag{A9}$$

$$R(\mathbf{r}) = \sum_{k=0}^{\infty} L_k(r)s_k(\theta,\phi), \tag{A10}$$

where r, θ, and ϕ are the spherical coordinates. $F_k s_k$ denotes the summation

$$F_k s_k = F_k^0(r)P_k(\cos(\theta)) + \sum_{k'=1}^{k}[F_{k+}^{k'}(r)P_k^{k'}(\cos(\theta))\cos(k'\phi) \tag{A11}$$

$$+ F_{k-}^{k'}(r)P_k^{k'}(\cos(\theta))\sin(k'\phi)], \tag{A12}$$

and P_k and $P_k^{k'}$ are, respectively, the principal and associated Legendre functions. Of course, $G_k s_k$ and $H_k s_k$ denote similar summations.

As the inertial terms due to the rotation of the particle are neglected $H_k=0$ and because of the symmetry of the problem (which is invariant by any rotation of ϕ angle), only the principal Legendre functions are required:

$$F_k s_k = F_k^0(r) P_k(\cos(\theta)), \tag{A13}$$

$$G_k s_k = G_k^0(r) P_k(\cos(\theta)). \tag{A14}$$

Then, by replacing the preceding expressions of **U** and R in the conservation Eqs. (A7) and (A8), Buyevich and Markov[42] obtain the following expressions for F_k^0 and G_k^0:

$$F_k^0 = A_k S_k(\xi) + B_k Q_k(\xi) - k M_k \left(\frac{\xi}{\beta}\right)^{k-1} + (k+1) N_k \left(\frac{\xi}{\beta}\right)^{-k-2}, \tag{A15}$$

$$G_k^0 = \frac{2}{k(k+1)} A_k \left(S_k(\xi) + \frac{\xi}{2}\dot{S}_k(\xi)\right) + \frac{2}{k(k+1)} B_k \left(Q_k(\xi) + \frac{\xi}{2}\dot{Q}_k(\xi)\right) - M_k \left(\frac{\xi}{\beta}\right)^{k-1} - N_k \left(\frac{\xi}{\beta}\right)^{-k-2}, \tag{A16}$$

where the coefficients A_k, B_k, M_k, and N_k are some constants which must be determined from the boundary conditions for each order k; $\dot{S}_k$ and $\dot{Q}_k$ are the derivatives of S_k and Q_k with respect to ξ; and the expressions of S_k and Q_k are given by the following expressions:

$$S_k = 2^k \xi^{k-1} \frac{d^k}{d(\xi^2)^k} \frac{\sinh \xi}{\xi}, \tag{A17}$$

$$Q_k = (-2)^k \xi^{k-1} \frac{d^k}{d(\xi^2)^k} \frac{\exp(-\xi)}{\xi}, \tag{A18}$$

with $\xi \equiv \beta r$.

We can now introduce the following notations: the constants $\{A_k, B_k, M_k, N_k\}$ are, respectively, equal to the following:

- $\{a_k, b_k, m_k, n_k\}$ for the first set of equations (for the average fields in the original frame of reference),
- $\{a_k^p, b_k^p, m_k^p, n_k^p\}$ for the second set of equations (for the conditionally averaged field in the pure fluid shell), and
- $\{a_k^e, b_k^e, m_k^e, n_k^e\}$ for the third set of equations (for the perturbation in the effective homogeneous medium).

We can easily deduce from the boundary conditions that: (a) b_k and n_k are null because the averaged fields are bounded when $r \to 0$, and (b) a_k^e and m_k^e are null because the perturbation vanishes when $r \to \infty$.

Now, there only remains to express the relations between the remaining coefficients, deduced from the boundary conditions (A3)–(A6). At first order ($k=1$), we obtain

$$M_1 V_1 = a_1 V_{A1} + m_1 V_{M1} - (i\omega)\, v_d|_{r=0} V_{P1} \tag{A19}$$

and at second order ($k=2$)

$$M_2 V_2 = a_2 V_{A2} + m_2 V_{M2}, \tag{A20}$$

where the expression of the column vectors V_{A1}, V_{M1}, V_{A2}, and V_{M2} and the matrices M_1 and M_2 are given in Appendix C and the expressions of V_1 and V_2 are given by

$$V_1 = \begin{bmatrix} a_1^p \\ b_1^p \\ m_1^p \\ n_1^p/a^3 \\ b_1^e \\ n_1^e/R_c^3 \end{bmatrix}, \quad V_2 = \begin{bmatrix} a_2^p \\ b_2^p \\ am_2^p \\ n_2^p/a^4 \\ b_2^e \\ n_2^e/R_c^4 \end{bmatrix}.$$

APPENDIX B: CALCULATION OF THE FORCE AND THE STRESSLET

The next step consists of expressing the force **F** and the stresslet **S** in terms of the coefficients a_k^p, b_k^p, m_k^p, and n_k^p. It is important to note that only the coefficients of first order ($k=1$) are required for the calculation of the force and the coefficients of second order ($k=2$) for the calculation of the stresslet because the contribution of the other terms vanishes when the integration over the surface of the sphere is performed. The following expressions are obtained:

$$\mathbf{F} = \alpha_{do}\mu_c\beta_c^2 \left[S_1(\gamma) a_1^p + Q_1(\gamma) b_1^p - m_1^p - \frac{n_1^p}{a^3}\right] \mathbf{e_z} + \alpha_{do}\rho_{co} \nabla \Psi,$$

$$\mathbf{S} = \frac{\alpha_{do}\mu_c\beta_c}{5}(3\mathbf{e_z}\otimes\mathbf{e_z} - \mathbf{I}) \times \left[\left(\dot{S}_2(\gamma) + \left(\frac{4}{\gamma} + \frac{\gamma}{2}\right) S_2(\gamma)\right) a_2^p + \left(\dot{Q}_2(\gamma) + \left(\frac{4}{\gamma} + \frac{\gamma}{2}\right) Q_2(\gamma)\right) b_2^p - \left(\frac{10}{\gamma} + \gamma\right) a m_2^p - \gamma\frac{n_2^p}{a^4}\right].$$

The final step consists of expressing the coefficients a_1, m_1, a_2, and m_2 in terms of the averaged fields in $r=0$:

$$a_1 \mathbf{e_z} = 3/\beta_{\text{eff}}^2 \, \Delta \mathbf{V_c}|_{r=0},$$

$$m_1 \mathbf{e_z} = 1/\beta_{\text{eff}}^2 \, \Delta \mathbf{V_c}|_{r=0} - \mathbf{V_c}|_{r=0},$$

$$a_2(3\mathbf{e_z}\otimes\mathbf{e_z} - \mathbf{I}) = 30/\beta_{\text{eff}}^2 \, \nabla^s \Delta \mathbf{V_c}|_{r=0},$$

$$m_2(3\mathbf{e_z}\otimes\mathbf{e_z} - \mathbf{I}) = -\nabla^s \mathbf{V_c}|_{r=0} + 1/\beta_{\text{eff}}^2 \, \nabla^s \Delta \mathbf{V_c}|_{r=0}.$$

Finally, with the relation $\Delta^2 \mathbf{V_c} = \beta_{\text{eff}}^2 \Delta \mathbf{V_c}$ [which can be easily deduced from Eqs. (31) and (32)] and by comparing the above expressions with Eqs. (27) and (28), we obtain the expression of the coefficients $\tilde{n}_k$.

APPENDIX C: EXPRESSION OF THE MATRICES M_1, M_2 AND THE VECTORS V_{A1}, V_{M1}, V_{P1}, V_{A2},V_{M2}

$$M_1 = \begin{bmatrix} S_1(\gamma) & Q_1(\gamma) & -1 & 2 & 0 & 0 \\ S_1(\gamma) + \gamma/2\dot{S}_1(\gamma) & Q_1(\gamma) + \gamma/2\dot{Q}_1(\gamma) & -1 & -1 & 0 & 0 \\ S_1(\delta) & Q_1(\delta) & -1 & 2(a/R_c)^3 & -\alpha_A Q_1(\eta) & -2\alpha_B \\ S_1(\delta) + (\delta/2)\dot{S}_1(\delta) & Q_1(\delta) + (\delta/2)\dot{Q}_1(\delta) & -1 & -(a/R_c)^3 & -\alpha_A(Q_1(\eta) + \eta/2\dot{Q}_1(\eta)) & \alpha_B \\ 2\kappa\dot{S}_1(\delta) & 2\kappa\dot{Q}_1(\delta) & -\kappa\delta & -\kappa(a/R_c)^3(\delta + 12/\delta) & -2\dot{Q}_1(\eta) & \eta + 12/\eta \\ \kappa(\delta/2\ddot{S}_1(\delta) + \dot{S}_1(\delta)) & \kappa(\delta/2\ddot{Q}_1(\delta) + \dot{Q}_1(\delta)) & 0 & 6\kappa a^3/\delta R_c^3 & -(\eta/2\ddot{Q}_1(\eta) + \dot{Q}_1(\eta)) & -6/\eta \end{bmatrix},$$

$$M_2 = \begin{bmatrix} S_2(\gamma) & Q_2(\gamma) & -2 & 3 & 0 & 0 \\ 1/3(S_2(\gamma) + \gamma/2\dot{S}_2(\gamma)) & 1/3(Q_2(\gamma) + (\gamma/2)\dot{Q}_2(\gamma)) & -1 & -1 & 0 & 0 \\ S_2(\delta) & Q_2(\delta) & -2R_c/a & 3(a/R_c)^4 & -\alpha_A Q_2(\eta) & -3\alpha_B \\ 1/3(S_2(\delta) + (\delta/2)\dot{S}_2(\delta)) & 1/3(Q_2(\delta) + (\delta/2)\dot{Q}_2(\delta)) & -R_c/a & -(a/R_c)^4 & -\alpha_A/3(Q_2(\eta) + (\eta/2)\dot{Q}_2(\eta)) & \alpha_B \\ 2\kappa\dot{S}_2(\delta) & 2\kappa\dot{Q}_2(\delta) & -\kappa R_c/a(\delta + 4/\delta) & -\kappa a^4/R_c^4(\delta + 24/\delta) & -2\dot{Q}_2(\eta) & (\eta + 24/\eta) \\ \kappa((4/\delta + \delta/2)S_2(\delta) - \dot{S}_2(\delta)) & \kappa((4/\delta + \delta/2)Q_2(\delta) - \dot{Q}_2(\delta)) & -6\kappa R_c/a\delta & 24\kappa a^4/\delta R_c^4 & \dot{Q}_2(\eta) - (4/\eta + \eta/2)Q_2(\eta) & -24/\eta \end{bmatrix},$$

$$V_{A1} = \begin{bmatrix} 0 \\ 0 \\ \alpha_A S_1(\eta) \\ \alpha_A(S_1(\eta) + \eta/2\dot{S}_1(\eta)) \\ 2\dot{S}_1(\eta) \\ \dot{S}_1(\eta) + \eta/2\ddot{S}_1(\eta) \end{bmatrix}, \quad V_{M1} = \begin{bmatrix} 0 \\ 0 \\ -\alpha_B \\ -\alpha_B \\ -\eta \\ 0 \end{bmatrix}, \quad V_{P1} = \begin{bmatrix} 0 \\ 0 \\ \alpha_C \\ \alpha_C \\ (\rho_{\mathrm{eff2}} - \rho_{co})R_c/\mu_{\mathrm{eff}}\beta_{\mathrm{eff}} \\ 0 \end{bmatrix},$$

$$V_{A2} = \begin{bmatrix} 0 \\ 0 \\ \alpha_A S_2(\eta) \\ \alpha_A/3(S_2(\eta) + \eta/2\dot{S}_2(\eta)) \\ 2\dot{S}_2(\eta) \\ (4/\eta + \eta/2)S_2(\eta) - \dot{S}_2(\eta) \end{bmatrix}, \quad V_{M2} = \begin{bmatrix} 0 \\ 0 \\ -2\alpha_B R_c \\ -2\alpha_B R_c \\ -(4/\eta + \eta/2)R_c \\ -\dfrac{6}{\eta}R_c \end{bmatrix},$$

with

$$\alpha_A = \alpha_{co} + \alpha_{do}(c_1 + c_2), \quad \alpha_B = \alpha_{co} + \alpha_{do}c_1, \quad \alpha_C = \alpha_{do}\frac{\tilde{n}_3 - \rho_{do}}{\tilde{n}_1 - i\omega\rho_{do}},$$

$$c_1 = \frac{\tilde{n}_1}{\tilde{n}_1 - i\omega\rho_{do}}, \quad c_2 = \frac{\tilde{n}_2\beta_{\mathrm{eff}}^2}{\tilde{n}_1 - i\omega\rho_{do}}.$$

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