

Sound propagation in dilute suspensions of spheres: Analytical comparison between coupled phase model and multiple scattering theory

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(Received 16 March 2015; revised 10 September 2015; accepted 21 September 2015; published online 30 October 2015)

Sound propagation in dilute suspensions of small spheres is studied using two models: a hydrodynamic model based on the coupled phase equations and an acoustic model based on the ECAH (ECAH: Epstein-Carhart-Allegra-Hawley) multiple scattering theory. The aim is to compare both models through the study of three fundamental kinds of particles: rigid particles, elastic spheres, and viscous droplets. The hydrodynamic model is based on a Rayleigh-Plesset-like equation generalized to elastic spheres and viscous droplets. The hydrodynamic forces for elastic spheres are introduced by analogy with those of droplets. The ECAH theory is also modified in order to take into account the velocity of rigid particles. Analytical calculations performed for long wavelength, low dilution, and weak absorption in the ambient fluid show that both models are strictly equivalent for the three kinds of particles studied. The analytical calculations show that dilatational and translational mechanisms are modeled in the same way by both models. The effective parameters of dilute suspensions are also calculated. © 2015 Acoustical Society of America.

[<http://dx.doi.org/10.1121/1.4932171>]

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NOMENCLATURE

R_0	Static particles radius	k_m and k_m^p	Wavenumbers in the ambient fluid and in particles ($m = L, T$)
R	Dynamic particles radius	$K_m = k_m R_0$ and $K_m^p = k_m^p R_0$	Dimensionless wavenumbers ($m = L, T$)
ρ_0	Density of the ambient fluid	F_p	Hydrodynamic forces
c_0	Adiabatic sound speed in the ambient fluid	v_p	Velocity of particles
λ and μ	Lamé constants of the ambient fluid	k_H and k_A	Effective wavenumbers of the hydrodynamic and acoustic models
η_v and η_s	Bulk and shear viscosities of the ambient fluid	φ_0	Amplitude of the incident wave in the acoustic model
$\tau_v = \frac{\eta_v + \frac{4}{3}\eta_s}{\rho_0 c_0^2}$	Characteristic viscous time	c_φ	Phase velocity of the coherent wave
ρ_p	Density of particles	α	Attenuation of the coherent wave
λ_p and μ_p	Lamé constants of particles	F_p	Hydrodynamic forces
c_p	Adiabatic sound speed in droplets	C_p and d_p	Coefficients representing dilatational effects
$K_p = \lambda_p + \frac{2}{3}\mu_p$	Bulk modulus of particles	t_p	Coefficient representing translational effects
$V_{p0} = \frac{4}{3}\pi R_0^3$	Volume of particles	A	Coefficient due to the Archimedes force
$V_p = \frac{4}{3}\pi R^3$	Dynamic volume of particles	B_p	Coefficient representing the Stokes viscous drag, the Basset-Boussinesq history force, and the added mass effect
n_0	Number of particles per unit volume	$\tilde{\rho} = \frac{\rho_p}{\rho}$	Density ratio
$\Phi_0 = n_0 V_{p0}$	Volume fraction of particles	$\tilde{\mu} = \frac{\mu_p}{\mu}$	Ratio of shear moduli
$m_p = \rho_p V_{p0}$	Mass of particles		
c_L and c_T	Longitudinal and shear bulk wave velocities in the ambient fluid		
c_L^p and c_T^p	Longitudinal and shear bulk wave velocities in particles		

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I. INTRODUCTION

The propagation of sound waves through dilute suspensions has been the subject of a large number of studies since the pioneering articles of Sewell¹ who considered immovable rigid particles suspended in a viscous atmosphere and Lamb² who took into account particle motion. Two different methods have been particularly developed to determine the effective wavenumber of the coherent wave propagating in suspensions: the coupled phase theory based on the two-phase hydrodynamic equations^{3,4} and the scattering theory, also called ECAH theory, historically based on the works of Epstein and Carhart⁵ and Allegra and Hawley.⁶ The primary advantage of the ECAH theory is to be valid over the whole frequency range whatever the nature of the spherical particle, whereas the coupled phase theory is limited to the long wavelength regime. However, the coupled phase theory gives a good framework to incorporate phenomena that would be difficult to include in the scattering theory such as nonlinear effects, mass transfers, or chemical reactions. After linearization, it also leads to an explicit dispersion equation that is generally simpler to calculate and which can be useful when dealing with the inverse problem. The comparison of both modeling approaches has already been widely discussed in the literature and particularly in the review of Challis *et al.*⁷ However, most of the comparisons are based on numerical results.^{4,8} In this context, the aim of this article is to compare numerically but also analytically both models through the study of fundamental kinds of particles: rigid particles, elastic spheres, and viscous droplets. To this end, three important hypotheses are used. First, because the hydrodynamic model is valid only in the long wavelength regime, the frequency range investigated is limited to frequencies for which the wavelength is much greater than the sphere radii. Second, the ambient fluid is assumed to be Newtonian and viscous with weak bulk absorption and without thermal dissipation. Finally, spherical particles are supposed to be identical (no polydispersion effect) and their concentration is assumed to be low.

In this study, the hydrodynamic model takes into account two mechanisms of interaction: the first one due to the dilatational effects of particles (volume change) and the second one to the visco-inertial forces associated to the translational motion of particles relative to the ambient fluid. The dilatational effects which depend on the compressibility of the particle are modeled through Rayleigh-Plesset-like equations for gas bubbles⁹ or incompressible shells.¹⁰ A key point here is to modify these equations to take into account the elasticity of elastic spheres and droplets. To this end, we have generalized a Rayleigh-Plesset-like equation under the hypothesis of long wavelength. The result is quite similar to that of Guédra *et al.*¹¹ dealing with compressible shells but is obtained on the basis of simpler assumptions. The translational effects of the particles are described by the visco-inertial forces exerted by a fluid on a single particle: the Stokes viscous drag, the so-called Basset-Boussinesq history force, the Archimedes force, and the added mass force. If the expressions of these hydrodynamic forces for incompressible liquids are well known in the case of rigid particles,¹² bubbles,¹³ and droplets,¹⁴ there is no expression in the literature of the hydrodynamic forces for elastic spheres. Moreover, these forces are rarely taken into

account in the coupled phase theory except for rigid particles¹⁵ and shells.¹⁶ This is why there is a fundamental interest to introduce such forces for deformable particles. Contrary to the coupled phase models used in Refs. 4, 7, and 8, the compressibility of particles is taken into account by coupling the hydrodynamic equations with a Rayleigh-Plesset-like equation describing the dilatational effect of particles. This is the reason why the agreement of our hydrodynamic model with the multiple scattering theory is generally better than observed previously in Refs. 4, 7, and 8.

The ECAH theory models the scattering of a plane longitudinal wave by a spherical particle in a viscous fluid. The scattered waves are decomposed into compressional and shear modes which are expressed in terms of spherical harmonics, each of them with amplitudes calculated from the boundary conditions at the surface of the particle. Then, the wavenumber describing the sound propagation in suspensions is straightforwardly obtained from the multiple scattering theory based on Foldy's approximation.¹⁷ As the concentration of particles is assumed to be small, it is not necessary to use more general theories which are based either on the quasicrystalline approximation^{18,19} or the self-consistent approaches.²⁰ In the long wavelength regime, it is well established that the propagation is mainly governed by the two first modes of vibration of the particles.⁷ The first mode incorporates the compressibility effects of the spheres and of the surrounding viscous fluid. It corresponds to a scattered radiation in a monopole form. The second mode represents viscous losses owing to the to-and-fro motion of the particle with respect to the surrounding fluid as well as scattered radiation in dipole form.⁷ A key point here is to get the analytical expressions of the scattering coefficients corresponding to the two first modes of vibration at low frequency. This allows to compare the analytical calculations obtained by both models in order to make the physical interpretations easier.

The outline of the article is as follows. The elastic parameters used in both models for the ambient fluid and the particles are introduced in Sec. II. Section III deals with the "hydrodynamic model," the Rayleigh-Plesset-like equation, and the hydrodynamic forces. A general presentation of the "acoustic model" based on the ECAH theory and its modifications is performed in Sec. IV. Section V is devoted to numerical simulations. In Sec. VI, analytical calculations of the amplitudes of modes are carried out and a comparison of the effective wavenumbers of both models is then performed. Section VII deals with the comparison of the modeling of dilatation mechanisms in both models. Finally, the link between the modes $n=1$ of scattered waves and the hydrodynamic forces is explicitly shown in Sec. VIII.

II. DEFINITIONS OF THE ELASTIC PARAMETERS

The ambient fluid is characterized by its mass density ρ_0 , sound speed c_0 , shear viscosity η_s , and bulk viscosity η_v . Analogy between solid and fluid acoustic equations allows to define formally the elastic parameters for the fluid²¹

$$\begin{cases} \lambda = \rho_0 c_0^2 - i\omega \left(\eta_v - \frac{2}{3} \eta_s \right), \\ \mu = -i\omega \eta_s. \end{cases} \quad (1)$$

The compressional and shear wave velocities in the ambient viscous fluid are given by

$$\begin{cases} c_L = \sqrt{(\lambda + 2\mu)/\rho_0} = c_0 \sqrt{1 - i\omega\tau_v}, \\ c_T = \sqrt{\mu/\rho_0} = \frac{1-i}{2} \sqrt{\frac{2\omega\eta_s}{\rho_0}}, \end{cases} \quad (2)$$

with

$$\tau_v = \frac{4}{\rho_0 c_0^2} \left(\eta_v + \frac{2}{3} \eta_s \right) \quad (3)$$

the characteristic time of viscosity.

The particles which can be either solid or fluid are characterized by their mass density ρ_p and the “elastic” parameters λ_p and μ_p . In the case of solid spheres, λ_p and μ_p are the Lamé constants. These constants tend obviously to infinity for rigid particles. In the case of viscous droplets, given the above analogy, the elastic parameters are expressed as

$$\begin{cases} \lambda_p = \rho_p c_p^2 - i\omega \left(\eta_v^p - \frac{2}{3} \eta_s^p \right), \\ \mu_p = -i\omega \eta_s^p, \end{cases} \quad (4)$$

with sound speed c_p , shear viscosity η_s^p , and bulk viscosity η_v^p . It is also useful to introduce the bulk modulus of the particle defined by

$$K_p = \lambda_p + \frac{2}{3} \mu_p. \quad (5)$$

The compression and shear bulk wave velocities in the ambient fluid are defined by Eq. (2) and the compression and shear waves velocities in the particle are given in full generality by

$$c_L^p = \sqrt{(\lambda_p + 2\mu_p)/\rho_p} \quad \text{and} \quad c_T^p = \sqrt{\mu_p/\rho_p}. \quad (6)$$

The wavenumbers associated to compressional and shear waves are expressed by

$$k_m = \omega/c_m \quad \text{and} \quad k_m^p = \omega/c_m^p \quad (m = L, T), \quad (7)$$

and the non-dimensional wavenumbers are given by

$$K_m = k_m R_0 \quad \text{and} \quad K_m^p = k_m^p R_0 \quad (m = L, T), \quad (8)$$

where R_0 is the static particles radius.

III. HYDRODYNAMIC MODEL

The hydrodynamic model presented in this section is based on that of Coulouvrat *et al.*¹⁶ The main objective is to adapt this theory to elastic spheres and droplets. First, a Rayleigh-Plesset-like equation is derived in order to describe the dynamics of elastic spheres and droplets. Second, the hydrodynamic forces are expressed for each kind of particle. Finally, the effective wavenumber k_H describing the sound

propagation in dilute suspensions is calculated, and some approximations are introduced in order to compare the hydrodynamic and acoustic models analytically.

A. Coupled phase theory

Spherical particles of static radius R_0 are assumed to be identical and randomly distributed within a compressible and viscous fluid. The particle volume denoted $V_p = \frac{4}{3} \pi R^3$ depends on the dynamic radius R of spherical particles. The volume fraction of particles within the total suspension volume is denoted as Φ_0 with $\Phi_0 = n_0 V_{p0}$ where n_0 is the number of particles per unit volume and $V_{p0} = \frac{4}{3} \pi R_0^3$ is the particle volume at the equilibrium state. The mass m_p of the particle is given by $m_p = \rho_p V_{p0}$. In the following, p and v are the mean pressure and velocity fields of the ambient fluid, and v_p is the mean velocity field of the particles.

Considering a linear acoustical regime and assuming the static field is constant and homogeneous, the average fields are governed by the following set of equations including the mass and momentum balance equations and Newton’s second law applied to the spheres:¹⁶

$$\begin{cases} (1 - \Phi_0) \left(\frac{\partial p}{\partial t} + \rho_0 \frac{\partial v}{\partial z} \right) = \rho_0 n_0 \frac{\partial V_p}{\partial t} - \rho_0 \Phi_0 \frac{\partial v_p}{\partial z}, \\ (1 - \Phi_0) \rho_0 \frac{\partial v}{\partial t} + \frac{\partial p}{\partial z} - \left(\eta_v + \frac{4}{3} \eta_s \right) \frac{\partial^2 v}{\partial z^2} = -n_0 F_p, \\ m_p \frac{\partial v_p}{\partial t} = F_p, \end{cases} \quad (9)$$

where F_p represents the hydrodynamic forces which depend on the nature of the particles. As for bubbles,²² the compressibility is taken into account by the first term on the right-hand side of the mass conservation. In the case of rigid particles, this term vanishes because there is no variation of the volume V_p . Contrary to bubbly liquids, the density of the disperse phase is not neglected here, explaining the second term on the right-hand side of the mass conservation.

Compressional waves are considered, for which the velocity field is parallel to the direction z of wave propagation. The suspension is assumed to be dilute but sufficiently dense to be treated as a volume-averaged continuum. The study is therefore carried out in the low frequency regime implying that the particles spacing is smaller than the wavelength. Thus any component ψ of the different fields is written $\psi = \psi_0 + \hat{\psi} \exp[i(k_H z - \omega t)]$ as the sum of a static field ψ_0 and of an acoustic perturbation $\hat{\psi}$ with $|\hat{\psi}| \ll \psi_0$, where k_H denotes the effective wavenumber. In the static case, the fluid is not moving and $v_0 = v_{p0} = 0$. In the linear regime, the pressure and density fluctuations of the ambient fluid are proportional to one another, $p = c_0^2 \rho$, so that previous equations become

$$\begin{cases} \frac{\omega}{\rho_0 c_0^2} \hat{p} - k_H \hat{v} = \frac{\Phi_0}{1 - \Phi_0} \left(3\omega \frac{\hat{R}}{R_0} + k_H \hat{v}_p \right), \\ [i\omega(1 - \Phi_0) - c_0^2 k_H^2 \tau_v] \rho_0 \hat{v} - ik_H \hat{p} = n_0 \hat{F}_p, \\ -i\omega m_p \hat{v}_p = \hat{F}_p. \end{cases} \quad (10)$$

In order to solve this system of equations, it is necessary to express the amplitude of the radius perturbation \hat{R} as a function of the amplitude of the acoustic pressure \hat{p} and the hydrodynamic forces \hat{F}_p as functions of the velocities \hat{v} and \hat{v}_p . Such relations depend on the nature of the particles and they will be given separately for rigid particles, elastic spheres, and droplets. It is worth noting that the last equation in Eq. (10) is questionable because the velocity \hat{v}_p is not clearly defined for particles which are not rigid. We will come back to this point later.

B. Dynamics of elastic spheres and viscous droplets

The particle dynamics is taken into account in the hydrodynamic model by the term \dot{R}/R_0 appearing in the first equation of the system (10). In order to close this system, the goal is to express this term as a function of the acoustic pressure. For deformable particles, the starting point is the Keller-Kolodner equation²³

$$R\ddot{R}\left(1 - \frac{\dot{R}}{c}\right) + \frac{3}{2}\dot{R}^2\left(1 - \frac{\dot{R}}{3c}\right) = \left(1 + \frac{\dot{R}}{c}\right)H + \frac{R}{c}\dot{H}, \quad (11)$$

where the function H is given by

$$H(t) = \frac{1}{\rho_0} \left[p(R) - \tau_{rr}(R) - 4\eta_s \frac{\dot{R}}{R} - p(t) \right]. \quad (12)$$

In Eq. (12), $p(R)$ and $\tau_{rr}(R)$ are the pressure and normal viscous stress at the surface of the sphere and $p(t)$ is the pressure in the fluid.

The particle here is an elastic sphere or a viscous droplet of natural radius R_{ref} ($R_0 \approx R_{\text{ref}}$) characterized by the parameters ρ_p , λ_p , and μ_p . The size of the particle being much smaller than the acoustic wavelength, the starting point is to neglect the inertial terms of the momentum conservation equation. This leads to assume a quasi-static motion, namely,

$$\frac{\partial \sigma_{rr}^p}{\partial r} + \frac{1}{r} \left(2\sigma_{rr}^p - \sigma_{\theta\theta}^p - \sigma_{\varphi\varphi}^p \right) = 0, \quad (13)$$

where the non-zero components of the stress tensor σ_{rr}^p , $\sigma_{\theta\theta}^p$, and $\sigma_{\varphi\varphi}^p$ are related to the normal displacement u_r^p as follows:

$$\begin{cases} \sigma_{rr}^p = (\lambda_p + 2\mu_p) \frac{\partial u_r^p}{\partial r} + 2\lambda_p \frac{u_r^p}{r}, \\ \sigma_{\theta\theta}^p = \sigma_{\varphi\varphi}^p = \lambda_p \frac{\partial u_r^p}{\partial r} + 2(\lambda_p + \mu_p) \frac{u_r^p}{r}. \end{cases} \quad (14)$$

Substituting Eq. (14) into Eq. (13) yields

$$(\lambda_p + 2\mu_p) \left(\frac{\partial^2 u_r^p}{\partial r^2} + \frac{2}{r} \frac{\partial u_r^p}{\partial r} - \frac{2}{r^2} u_r^p \right) = 0, \quad (15)$$

an equation whose regular solution at $r=0$ is in the form

$$u_r^p(r, t) = F(t)r. \quad (16)$$

The continuity of the normal velocity at the interface $r=R(t)$,

$$\dot{u}_r^p(R, t) = \dot{F}(t)R(t) = \dot{R}(t), \quad (17)$$

allows to express the function $\dot{F}(t)$ which can then be integrated with respect to time

$$F(t) = \int_{-\infty}^t \frac{\dot{R}}{R} dt = \ln\left(\frac{R}{R_{\text{ref}}}\right). \quad (18)$$

Hence, the normal stress at the interface $r=R(t)$ takes the following form:

$$\sigma_{rr}^p(R) = 3K_p \ln\left(\frac{R}{R_{\text{ref}}}\right). \quad (19)$$

Neglecting surface tensions, the continuity of normal stresses at the interface yields

$$\sigma_{rr}^p(R) = -p(R) + \tau_{rr}(R). \quad (20)$$

Finally, inserting Eqs. (19) and (20) into Eq. (12) leads to the following expression:

$$H(t) = -\frac{1}{\rho_0} \left[3K_p \ln\left(\frac{R}{R_{\text{ref}}}\right) + 4\eta_s \frac{\dot{R}}{R} + p(t) \right]. \quad (21)$$

The Keller-Kolodner equation (11) with $H(t)$ defined by Eq. (21) gives a generalized Rayleigh-Plesset-like equation for elastic spheres and viscous droplets immersed in a viscous fluid. This is an important result that allows us to apply the hydrodynamic model to particles other than bubbles or rigid particles.

The pressure p and the dynamic radius R are assumed to be written in the following forms:

$$\begin{cases} p(t) = p_0 + \hat{p} e^{i(k_H z - \omega t)}, \\ R(t) = R_0 + \hat{R} e^{i(k_H z - \omega t)}, \end{cases} \quad (22)$$

with \hat{p} the amplitude of the acoustic pressure ($|\hat{p}| \ll p_0$) and \hat{R} the amplitude of the radius perturbation ($|\hat{R}| \ll R_0$). Substituting Eq. (22) into Eqs. (11) and (21) and taking into account the continuity of normal stresses at equilibrium

$$3K_p \ln\left(\frac{R_0}{R_{\text{ref}}}\right) = -p_0 \quad (23)$$

gives

$$\hat{p} = C_p \frac{\hat{R}}{R_0}, \quad (24)$$

with

$$C_p = \frac{\rho_0 (\omega R_0)^2}{1 - iK_L} + 4i\omega\eta_s - 3K_p. \quad (25)$$

The bulk modulus K_p being usually very superior to the static pressure p_0 , Eq. (23) shows that the radius at equilibrium R_0 is approximately equal to the static radius R_{ref} . Moreover, if the bulk modulus tends to infinity, the coefficient C_p tends also to infinity as in the case of rigid particles.

C. Hydrodynamic forces

The hydrodynamic forces in the Fourier space can be put in the form¹⁶

$$\hat{F}_p = B_p(\hat{v} - \hat{v}_p) + A\hat{v}, \quad (26)$$

where the coefficient A representing the Archimedes force¹³ is given by

$$A = -\frac{4}{3}\pi R_0 \eta_s K_T^2 = -i\frac{4}{3}\pi R_0^3 \rho_0 \omega = -i\frac{\omega m_p}{\tilde{\rho}}, \quad (27)$$

with

$$\tilde{\rho} = \rho_p / \rho_0 \quad (28)$$

the ratio of the particle density to the ambient fluid density. The coefficient B_p standing for the Stokes viscous drag, the Basset-Boussinesq history force, and the added mass effect depends on the nature of the particle. For viscous droplets, it is given by¹⁴

$$B_{\text{drop}} = 6\pi R_0 \eta_s \left[1 - iK_T - \frac{K_T^2}{9} - \frac{(1 - iK_T)^2}{3 - iK_T + \tilde{\mu} \frac{g_1}{g_2}} \right], \quad (29)$$

with

$$\tilde{\mu} = \eta_s^p / \eta_s. \quad (30)$$

The functions g_1 and g_2 depending on the dimensionless wavenumbers associated to shear waves

$$K_T = \sqrt{i\omega \frac{\rho_0}{\eta_s} R_0} \quad \text{and} \quad K_T^p = \sqrt{i\omega \frac{\rho_p}{\eta_s^p} R_0} \quad (31)$$

are defined by

$$\begin{cases} g_1 = (K_T^p)^3 - 6K_T^p + 3[2 - (K_T^p)^2] \tan(K_T^p), \\ g_2 = (K_T^p)^2 \tan(K_T^p) + 3K_T^p - 3 \tan(K_T^p). \end{cases} \quad (32)$$

The coefficient B_{rig} for rigid particles is defined by the relation¹²

$$B_{\text{rig}} = 6\pi R_0 \eta_s \left(1 - iK_T - \frac{1}{9} K_T^2 \right). \quad (33)$$

The hydrodynamic forces for elastic spheres are not known, but the analogy between elastic parameters of elastic spheres and viscous droplets (cf. Sec. II) can be used here. Thus the coefficient B_{elas} can be defined using Eq. (29) if the coefficient $\tilde{\mu}$ defined by Eq. (30) is rewritten as follows:

$$\tilde{\mu} = \frac{-i\omega \eta_s^p}{-i\omega \eta_s} = \frac{\mu_p}{\mu}. \quad (34)$$

In this form, the coefficient $\tilde{\mu}$ is defined not only for droplets but also for elastic spheres. In both cases, $K_T^p = \omega R_0 / c_T^p$ is the non-dimensional wavenumber associated to shear waves with velocity c_T^p given by Eq. (6).

D. Effective wavenumber k_H

Combining Eq. (26) and the last equation of Eq. (10), the velocity of the sphere and the hydrodynamic forces can be expressed according to the velocity \hat{v} as follows:

$$\begin{cases} \hat{v}_p = \frac{A + B_p}{\tilde{\rho}A + B_p} \hat{v}, \\ \hat{F}_p = \frac{\tilde{\rho}A(A + B_p)}{\tilde{\rho}A + B_p} \hat{v}. \end{cases} \quad (35)$$

Hence, taking into account Eqs. (35) and (24), the first equation of the system (10) can be written

$$(1 - \Phi_0 d_p) \hat{p} = [1 - \Phi_0(1 - t_p)] \frac{\rho_0 c_0^2 k_H}{\omega} \hat{v}, \quad (36)$$

where the coefficients d_p and t_p are given by

$$\begin{cases} d_p = 1 + 3 \frac{\rho_0 c_0^2}{C_p}, & \text{with } p = \text{rig, elas, drop}, \\ t_p = \frac{A + B_p}{\tilde{\rho}A + B_p}, & \text{with } p = \text{rig, elas, drop}. \end{cases} \quad (37)$$

As C_{rig} tends to infinity, the coefficient d_{rig} is equal to unity; d_p represents the dilatational effects due to the variations of volume described by the Rayleigh-Plesset-like equation and t_p represents the translational particle effects due to the visco-inertial forces exerted on the particles (hydrodynamic forces).

Substituting Eq. (37) into Eq. (35) yields

$$\begin{cases} \hat{v}_p = t_p \hat{v}, & \text{with } p = \text{rig, elas, drop}, \\ \hat{F}_p = \tilde{\rho} A t_p \hat{v}, & \text{with } p = \text{rig, elas, drop}. \end{cases} \quad (38)$$

Noting that

$$n_0 = \Phi_0 / \left(\frac{4}{3} \pi R_0^3 \right) = -i \frac{\rho_0 \omega}{A} \Phi_0, \quad (39)$$

and taking into account Eqs. (35), (36), and (10), the effective wavenumber k_H is finally given by

$$\left(\frac{k_H}{k_L} \right)^2 = \frac{(1 - i\omega\tau_v) [1 - \Phi_0(1 + d_p - \tilde{\rho}t_p) + \Phi_0^2 d_p (1 - \tilde{\rho}t_p)]}{1 - i\omega\tau_v - \Phi_0(1 - t_p) + i\omega\tau_v \Phi_0 d_p}, \quad (40)$$

with $k_L = \omega / c_L$ the wavenumber of compressional waves propagating in the ambient fluid. This expression is the most general formula of the article describing the propagation of the coherent wave in a suspension of particles whatever the nature of these particles.

E. Analytical approximations

In order to compare analytically the hydrodynamic and acoustic models, asymptotic expansions must be carried out with respect to Φ_0 . For dilute suspensions ($\Phi_0 \ll 1$), and in the practical case of liquids with very weak absorption ($\omega\tau_v \ll 1$), as water for example, Eq. (40) simplifies to

$$\left(\frac{k_H}{k_L} \right)^2 \approx 1 - \Phi_0 [d_p + (1 - \tilde{\rho})t_p]. \quad (41)$$

It is worthwhile to note here that, if the ambient fluid and particles densities are equal, hydrodynamic forces have no effect on the coherent waves. Moreover, because the hydrodynamic model is based on the hypothesis of long wavelengths, the following approximation

$$|K_L| = |\omega R_0/c_L| \approx \omega R_0/c_0 \ll 1 \quad (42)$$

can be used. Substituting Eq. (25) into Eq. (37) and using Eq. (42) yields

$$d_p \approx \frac{3K_p - 3\rho_0 c_0^2}{3K_p - 4i\omega\eta_s}. \quad (43)$$

In general for elastic spheres or liquid droplets, the bulk modulus K_p is typically greater than, or of the same order of magnitude as, $\rho_0 c_0^2$. However in some cases like porous spheres²⁴ it can be significantly smaller and then the viscous term at the denominator is not negligible.

For rigid particles, the bulk modulus K_p tends to infinity. It follows that $d_p = 1$ and Eq. (41) simplifies to

$$\left(\frac{k_H}{k_L}\right)^2 \approx 1 - \Phi_0 + \Phi_0(\tilde{\rho} - 1) \frac{A + B_{\text{rig}}}{\tilde{\rho}A + B_{\text{rig}}}. \quad (44)$$

If the rigid particle is assumed to be infinitely heavy ($\tilde{\rho} = \rho_p/\rho_0 \rightarrow \infty$), it is therefore fixed ($v_p = 0$) and Eq. (44) reduces to

$$\left(\frac{k_H}{k_L}\right)^2 \approx 1 + \Phi_0 \frac{B_{\text{rig}}}{A}. \quad (45)$$

F. Effective (constitutive) parameters of dilute suspensions

A dilute suspension can be considered as an effective medium with space and time dispersion, and therefore, its elastic parameters are functions of the frequency. In general, as in the case of fluids, the effective density $\rho_H(\omega)$ and bulk modulus $K_H(\omega)$ are often calculated from the reflexion coefficient of a plane wave normally incident on the interface between the fluid and the dilute suspension.^{25,26} In the following, we briefly show that the effective parameters can be directly calculated from Eq. (36), without having to use the reflection coefficient. Indeed, the effective impedance $Z_H(\omega)$ of the dilute suspension is by definition given by

$$Z_H(\omega) = \frac{\hat{p}}{\hat{v}} = \frac{[1 - \Phi_0(1 - t_p)] \rho_0 c_0^2 k_H}{\omega(1 - \Phi_0 d_p)}. \quad (46)$$

As a direct result, we get

$$\begin{cases} \frac{\rho_H(\omega)}{\rho_0} = \frac{1 - \Phi_0(1 - t_p)}{1 - \Phi_0 d_p} \left(\frac{k_H}{k_0}\right)^2, \\ \frac{K_H(\omega)}{\rho_0 c_0^2} = \frac{1 - \Phi_0(1 - t_p)}{1 - \Phi_0 d_p}. \end{cases} \quad (47)$$

As discussed in Refs. 25 and 26, the above relations provide a convenient tool in the search for new metamaterials with specific desired properties such as negative effective properties.

IV. ACOUSTIC MODEL

As discussed in Challis *et al.*,⁷ the ECAH theory is historically based on Foldy's approximation for dilute suspensions. It can also be derived from the quasi-crystalline approximation if the concentration Φ_0 of particles tends to increase. The quasi-crystalline approximation leads to more or less complicated equations depending on the level of approximation.^{18,19,27} For this study, due to the low concentration approximation, it is sufficient to use Foldy's theory for which the effective wavenumber k_A describing the sound propagation in suspensions is given by¹⁷

$$\left(\frac{k_A}{k_L}\right)^2 = 1 - i \frac{3\Phi_0}{K_L^3} \sum_{n=0}^{\infty} (2n+1) A_n^{(s,L)}, \quad (48)$$

where $A_n^{(s,L)}$ is the amplitude of the n th mode of the longitudinal wave scattered in the ambient fluid.

The scattering amplitudes $A_n^{(s,L)}$ depend on the boundary conditions at the interface between the particle and the ambient viscous fluid. Obviously they have to be calculated separately for each kind of particle. Assumptions and notations are not thoroughly the same in all publications that deal with the acoustic scattering by different kind of particles;^{21,28} this is one of the reasons for which the calculation of $A_n^{(s,L)}$ is briefly summarized in this section.

A. General equations

Assuming the Helmholtz decomposition of the displacement in the form

$$\mathbf{u} = \mathbf{grad} \varphi_L + \mathbf{rot}(r\varphi_T \mathbf{e}_r), \quad (49)$$

the potentials φ_L and φ_T associated to the longitudinal (compressional) and transverse (shear) components of the waves can be expanded into spherical harmonics ($m = L, T$)

$$\varphi_m(r, \theta) = \varphi_0 \sum_{n=0}^{\infty} A_n^m z_n(k_m r) \tilde{Y}_n(\theta), \quad (50)$$

with

$$\tilde{Y}_n(\theta) = i^n (2n+1) P_n(\cos \theta), \quad (51)$$

where P_n are Legendre polynomials, z_n are spherical Bessel functions of order n , and A_n^m ($m = L, T$) are the unknown scattering coefficients calculated from the appropriate boundary conditions. The normal u_r and tangential u_θ displacements, and the normal σ_{rr} and tangential $\sigma_{r\theta}$ stresses at the interface $r = R_0$ can therefore be expressed as

$$\left\{ \begin{aligned} u_r(R_0, \theta) &= \frac{\varphi_0}{R_0} \sum_{n=0}^{\infty} (A_n^L U_n^L + A_n^T U_n^T) \tilde{Y}_n(\theta), \\ u_\theta(R_0, \theta) &= \frac{\varphi_0}{R_0} \sum_{n=1}^{\infty} (A_n^L V_n^L + A_n^T V_n^T) \frac{d\tilde{Y}_n(\theta)}{d\theta}, \\ \sigma_{rr}(R_0, \theta) &= \frac{\varphi_0}{R_0^2} \sum_{n=0}^{\infty} (A_n^L \Sigma_n^L + A_n^T \Sigma_n^T) \tilde{Y}_n(\theta), \\ \sigma_{r\theta}(R_0, \theta) &= \frac{\varphi_0}{R_0^2} \sum_{n=1}^{\infty} (A_n^L T_n^L + A_n^T T_n^T) \frac{d\tilde{Y}_n(\theta)}{d\theta}, \end{aligned} \right. \quad (52)$$

where

$$\left\{ \begin{aligned} U_n^L &= n z_n(K_L) - K_L z_{n+1}(K_L), \\ U_n^T &= n(n+1) z_n(K_T), \\ V_n^L &= z_n(K_L), \\ V_n^T &= (1+n) z_n(K_T) - K_T z_{n+1}(K_T), \\ \Sigma_n^L &= [2n(n-1)\mu - (\lambda+2\mu)K_L^2] z_n(K_L) + 4\mu K_L z_{n+1}(K_L), \\ \Sigma_n^T &= 2n(n^2-1)\mu z_n(K_T) - 2n(n+1)\mu K_T z_{n+1}(K_T), \\ T_n^L &= 2\mu[(n-1)z_n(K_L) - K_L z_{n+1}(K_L)], \\ T_n^T &= \mu(2(n^2-1) - K_T^2) z_n(K_T) + 2\mu K_T z_{n+1}(K_T), \end{aligned} \right. \quad (53)$$

with $K_m = k_m R_0$ the dimensionless wavenumbers. For the tangential displacement u_θ and the tangential stress $\sigma_{r\theta}$, the sum starts from $n=1$ because the term $n=0$ vanishes due to the derivative of Y_n with respect of θ . It implies that the boundary conditions must be applied separately for the mode $n=0$ and for the other modes.

B. Definition of the potentials

The scalar potential of the incident wave φ_L^i , those of scattered waves in the ambient fluid φ_m^s ($m=L, T$) and those corresponding to the fields in the elastic sphere φ_m^p ($m=L, T$) are given by

$$\left\{ \begin{aligned} \varphi_L^i(r, \theta) &= \varphi_0 e^{ik_L z} = \varphi_0 \sum_{n=0}^{\infty} j_n(k_L r) \tilde{Y}_n(\theta), \\ \varphi_m^s(r, \theta) &= \varphi_0 \sum_{n=0}^{\infty} A_n^{(s,m)} h_n(k_m r) \tilde{Y}_n(\theta), \\ \varphi_m^p(r, \theta) &= \varphi_0 \sum_{n=0}^{\infty} A_n^{(p,m)} j_n(k_m^p r) \tilde{Y}_n(\theta), \end{aligned} \right. \quad (54)$$

where j_n is the spherical Bessel function of order n and h_n the spherical Hankel function of the first kind and order n which is an outgoing spherical wave when considering the $\exp(-i\omega t)$ time harmonic dependence. The functions $U_n^{(i,L)}$, $V_n^{(i,L)}$, $\Sigma_n^{(i,L)}$, and $T_n^{(i,L)}$ corresponding to the incident wave propagating in the ambient viscous fluid and those $U_n^{(s,m)}$, $V_n^{(s,m)}$, $\Sigma_n^{(s,m)}$, and $T_n^{(s,m)}$ and $U_n^{(p,m)}$, $V_n^{(p,m)}$, $\Sigma_n^{(p,m)}$, and $T_n^{(p,m)}$ corresponding to the scattered waves in the ambient fluid and in the sphere, respectively, are given by Eq. (53) with $K_m = k_m R_0$ and $K_m^p = k_m^p R_0$, respectively [cf. Eq. (8)].

C. Calculations of the scattering coefficients

1. Fixed rigid particles

If the sphere is rigid ($\lambda_p \rightarrow \infty$, $\mu_p \rightarrow \infty$) and infinitely heavy ($\rho_p \rightarrow \infty$) the particle is assumed to be fixed and the boundary conditions correspond to the cancellation of displacements u_r and u_θ of the fluid at the interface $r=R_0$. For the mode $n=0$, the tangential displacement u_θ is always zero and the normal displacement associated to shear wave is also zero ($U_0^T = 0$). The amplitude of the mode $n=0$ of the shear wave is therefore zero and that of the longitudinal wave is given by the expression

$$A_0^{(s,L)} = -\frac{U_0^{(i,L)}}{U_0^{(s,L)}}. \quad (55)$$

The amplitudes of the modes $n \neq 0$ scattered in the ambient fluid satisfy the matrix equation

$$\begin{pmatrix} U_n^{(s,L)} & U_n^{(s,T)} \\ V_n^{(s,L)} & V_n^{(s,T)} \end{pmatrix} \cdot \begin{pmatrix} A_n^{(s,L)} \\ A_n^{(s,T)} \end{pmatrix} = -\begin{pmatrix} U_n^{(i,L)} \\ V_n^{(i,L)} \end{pmatrix}. \quad (56)$$

2. Moving rigid particles

For moving rigid particles, λ_p and μ_p both tend to infinity but ρ_p keeps a finite value. Thus contrary to the fixed rigid particle, the moving rigid particle can move and the boundary conditions do not correspond to the cancellation of displacements u_r and u_θ of the fluid at the interface $r=R_0$. The velocity and therefore the mass of the particle have to be taken into account in the modeling, which requires modifying the ECAH theory. For that purpose the analytical calculations follow broadly those of Temkin and Leung.²⁸

The velocity of the particle \mathbf{v}_p is related to the linearized hydrodynamic forces \mathbf{F}_p acting on the sphere by the relation

$$m_p \frac{\partial \mathbf{v}_p}{\partial t} = \mathbf{F}_p, \quad (57)$$

where \mathbf{F}_p is given by

$$\mathbf{F}_p = \int_0^{2\pi} \int_0^\pi \bar{\bar{\sigma}}(R_0, \theta) \cdot \mathbf{e}_r R_0^2 \sin \theta d\theta d\phi e^{-i\omega t}, \quad (58)$$

with $\bar{\bar{\sigma}}$ the stress tensor in the ambient fluid. We have

$$\bar{\bar{\sigma}} \cdot \mathbf{e}_r = \sigma_{rr} \mathbf{e}_r + \sigma_{r\theta} \mathbf{e}_\theta, \quad (59)$$

with

$$\begin{cases} \mathbf{e}_r = \sin \theta \cos \phi \mathbf{e}_x + \sin \theta \sin \phi \mathbf{e}_y + \cos \theta \mathbf{e}_z, \\ \mathbf{e}_\theta = \cos \theta \cos \phi \mathbf{e}_x + \cos \theta \sin \phi \mathbf{e}_y - \sin \theta \mathbf{e}_z, \end{cases} \quad (60)$$

where the non-zero components of the stress tensor $\sigma_{rr}(R_0, \theta)$ and $\sigma_{r\theta}(R_0, \theta)$ are defined in Eq. (52). It follows that

$$\mathbf{F}_p = \hat{F}_p e^{-i\omega t} \mathbf{e}_z, \quad (61)$$

with

$$\hat{F}_p = 2\pi R_0^2 \int_0^\pi (\sigma_{rr} \cos \theta - \sigma_{r\theta} \sin \theta) \sin \theta d\theta. \quad (62)$$

As the incident wave is a compressional wave that propagates in a direction parallel to the z -axis, the motion of the fluid will be symmetric about this axis. This explains why the hydrodynamic forces are directed towards the z -axis. Substituting Eq. (52) into Eq. (62) gives

$$\hat{F}_p = 4i\pi\phi_0[A_1^{(s,L)}(\Sigma_1^{(s,L)} + 2T_1^{(s,L)}) + A_1^{(s,T)}(\Sigma_1^{(s,T)} + 2T_1^{(s,T)}) + \Sigma_1^{(i,L)} + 2T_1^{(i,L)}], \quad (63)$$

and using Eq. (57) the velocity of the particle can be put in the following form:

$$\mathbf{v}_p = \hat{v}_p e^{-i\omega t} \mathbf{e}_z = j \frac{F_p}{\omega m_p} e^{-i\omega t} \mathbf{e}_z. \quad (64)$$

The amplitudes of scattered waves are then determined from the boundary conditions which are the continuity of normal and tangential velocities at the interface $r = R_0$,

$$\begin{cases} -i\omega(u_r^s + u_r^i) = \hat{v}_p \cos \theta, \\ -i\omega(u_\theta^s + u_\theta^i) = -\hat{v}_p \sin \theta. \end{cases} \quad (65)$$

Substituting Eq. (52) into the above boundary conditions and using the orthogonality of Legendre polynomials, the amplitudes of the modes $n \neq 1$ are solutions of Eqs. (55) and (56), and those of the mode $n = 1$ are solutions of the matrix equation

$$\begin{pmatrix} U_1^{(s,L)} + X_{(s,L)} & U_1^{(s,T)} + X_{(s,T)} \\ V_1^{(s,L)} + X_{(s,L)} & V_1^{(s,T)} + X_{(s,T)} \end{pmatrix} \begin{pmatrix} A_1^{(s,L)} \\ A_1^{(s,T)} \end{pmatrix} = - \begin{pmatrix} U_1^{(i,L)} + X_{(i,L)} \\ V_1^{(i,L)} + X_{(i,L)} \end{pmatrix}, \quad (66)$$

with

$$X_m = \frac{\Sigma_1^m + 2T_1^m}{\rho_p(\omega R_0)^2}, \quad m = (s, L), (s, T), (i, L). \quad (67)$$

As expected, the mass and velocity of the sphere have an influence only on the mode $n = 1$.

3. Elastic spheres and droplets

For elastic spheres and droplets immersed in a viscous fluid, the boundary conditions are the continuity of normal and tangential displacements, and of normal and tangential stresses at the interface $r = R_0$. For the mode $n = 0$, the tangential displacement u_θ and stress $\sigma_{r\theta}$ are always zero. Taking into account Eq. (53), the amplitudes of the shear waves $A_0^{(s,T)}$ and $A_0^{(p,T)}$ are therefore zero. Substituting Eqs. (52) and (53) into the boundary conditions leads to the matrix equation

$$\begin{pmatrix} U_0^{(s,L)} & -U_0^{(p,L)} \\ \Sigma_0^{(s,L)} & -\Sigma_0^{(p,L)} \end{pmatrix} \cdot \begin{pmatrix} A_0^{(s,L)} \\ A_0^{(p,L)} \end{pmatrix} = - \begin{pmatrix} U_0^{(i,L)} \\ \Sigma_0^{(i,L)} \end{pmatrix}, \quad (68)$$

for the mode $n = 0$ and

$$\begin{pmatrix} U_n^{(s,L)} & U_n^{(s,T)} & -U_n^{(p,L)} & -U_n^{(p,T)} \\ V_n^{(s,L)} & V_n^{(s,T)} & -V_n^{(p,L)} & -V_n^{(p,T)} \\ \Sigma_n^{(s,L)} & \Sigma_n^{(s,T)} & -\Sigma_n^{(p,L)} & -\Sigma_n^{(p,T)} \\ T_n^{(s,L)} & T_n^{(s,T)} & -T_n^{(p,L)} & -T_n^{(p,T)} \end{pmatrix} \cdot \begin{pmatrix} A_n^{(s,L)} \\ A_n^{(s,T)} \\ A_n^{(p,L)} \\ A_n^{(p,T)} \end{pmatrix} = - \begin{pmatrix} U_n^{(i,L)} \\ V_n^{(i,L)} \\ \Sigma_n^{(i,L)} \\ T_n^{(i,L)} \end{pmatrix}, \quad (69)$$

for the modes $n \neq 0$.

V. NUMERICAL COMPARISON OF THE TWO MODELS

The goal of this section is to perform numerical calculations in order to compare both models for three different kinds of particles, e.g., rigid particles, elastic spheres, and viscous droplets, and to highlight the influence of the particles density and elastic parameters on the coherent wave. Numerical comparisons in the literature^{4,7,8} show good agreement for the evolution of the phase velocity as a function of the concentration at a given frequency between the ECAH theory and the coupled phase model. However, the translational effects of particles cannot be neglected at long wavelengths. As the rigid particles are supposed to be fixed when using the usual ECAH theory⁷ it was necessary to make changes in this case. More precisely, scattering coefficients of rigid particles are modified according to analytical calculations that follow broadly those of Temkin and Leung.²⁸ In this article, for each kind of particle, numerical simulations are carried out for a concentration $\Phi_0 = 1\%$ of particles of static radius $R_0 = 10 \mu\text{m}$. Because the hydrodynamic model is valid only in the long wavelength regime, the study is made in the frequency range so that the non-dimensional wavenumber is in the range 10^{-4} to 1. In all cases, particles are immersed in water of mass density $\rho_0 = 1000 \text{ kg m}^{-3}$, adiabatic sound speed $c_0 = 1500 \text{ m s}^{-1}$, and bulk $\eta_v = 2.4 \text{ mPa s}$ and shear $\eta_s = 1 \text{ mPa s}$ viscosities.

In the following the effective wavenumbers k_H and k_A are compared through the normalized phase velocity c_φ/c_0 and the attenuation α defined by the following expressions:

$$\begin{cases} \frac{c_\varphi}{c_0} = \frac{k_0}{\text{Re}\{k_{A,H}\}}, \\ \alpha = \text{Im}\{k_{A,H}\}. \end{cases} \quad (70)$$

Solid lines represent the results obtained by the hydrodynamic model [Eq. (40)] and dashed lines those obtained by the acoustic model [Eq. (48)].

A. Rigid particles

In this section, numerical results are compared for rigid particles in order to understand the influence of the particles mass on the coherent wave. Four different values of the coefficient $\tilde{\rho} = \rho_p/\rho_0$ are considered. Normalized phase velocity and attenuation are plotted in Fig. 1 as functions of the non-dimensional wavenumber $k_0 R_0$ for four cases: $\tilde{\rho} \rightarrow \infty$

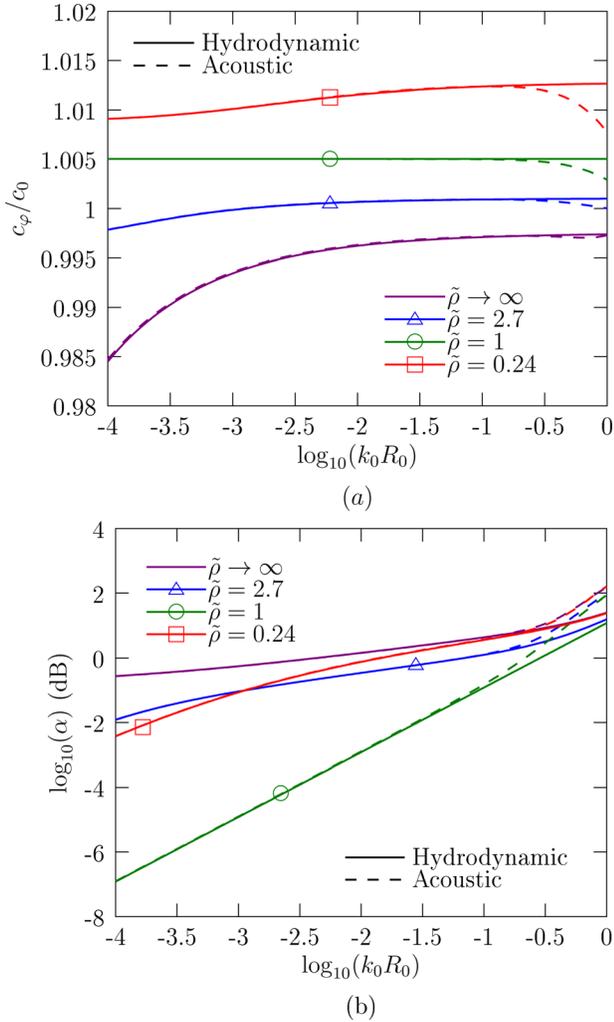


FIG. 1. (Color online) Evolutions of (a) the normalized phase velocity c_ϕ/c_0 and (b) the attenuation α as functions of the non-dimensional wavenumber $k_0 R_0$ in the case of rigid particles. The solid/dashed lines represent the results of the hydrodynamic/acoustic model [Eq. (40)/Eq. (48)].

(curves without symbol), $\tilde{\rho} = 2.7$ (curves with triangle), $\tilde{\rho} = 1$ (curves with circle), and $\tilde{\rho} = 0.24$ (curves with square).

It appears that both models give strictly equivalent results for values of the non-dimensional wavenumber $k_0 R_0$ inferior to 10^{-1} . Curves for the case $\tilde{\rho} = 1$ are very different from other ones, because the velocity is not dispersive and the attenuation is very weak, close to the one due to the ambient fluid viscosity. Observing the effective wavenumber in the case of moving rigid particles [cf. Eq. (44)], it appears clearly that the translational motion disappears if $\tilde{\rho} = 1$. The more $\tilde{\rho}$ deviates from $\tilde{\rho} = 1$, the more phase velocity is dispersive and attenuation is large. This shows that inertia effects included in the hydrodynamic forces play an important role on the dispersion of coherent waves.

B. Solid elastic spheres

In this section, numerical results are compared for four kinds of solid elastic spheres: poly(methyl methacrylate) (PMMA), Aluminum, Rubber, and Cork. The values of the mechanical parameters are given in Table I.

TABLE I. Values of mechanical parameters used for numerical results of Fig. 2.

Solid	ρ_p (g cm ⁻³)	λ_p (GPa)	μ_p (GPa)	K_p (GPa)	$\tilde{\rho}$
Aluminum	2.7	55	25	72	2.7
PMMA	1.2	3.5	0.85	4	1.2
Rubber	0.9	2	0.005	2	0.9
Cork	0.24	0.2	10	7	0.24

The normalized phase velocity and attenuation are plotted in Fig. 2 for the four materials of this study: PMMA (curves without symbol), Aluminum (curves with triangle), Rubber (curves with circle), and Cork (curves with square).

Once again it appears that both models yield strictly equivalent results for the values of the non-dimensional wavenumber $k_0 R_0$ lower than 10^{-1} . There is only a small difference when $k_0 R_0$ is close to 10^{-1} in the case of rubber spheres. This difference corresponds to the resonance of the mode $n = 2$ which is not taken into account by the hydrodynamic model.

In all considered cases, elastic curves are close to those of the moving rigid particles of the same density. This is not really surprising since the coefficient $\tilde{\mu}$ is large in the frequency range of interest $k_0 R_0 \leq 0.1$. As expected, the results

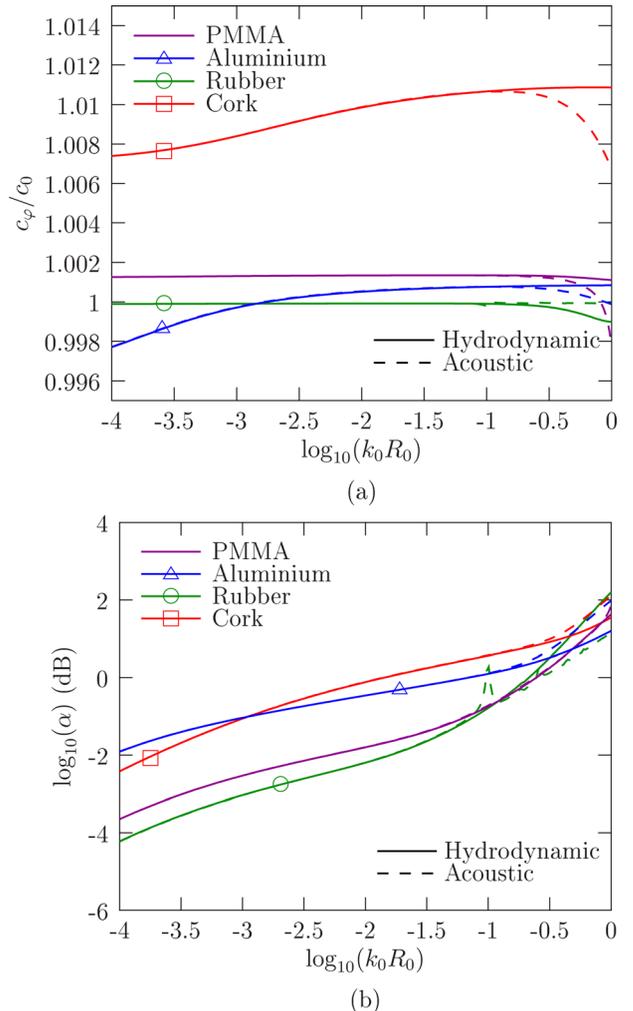


FIG. 2. (Color online) Same as Fig. 1 for elastic spheres.

TABLE II. Values of mechanical parameters used for numerical results of Fig. 3. Values for PFOB and Fluorinated oil are issued from Refs. 16 and 29, respectively.

Liquid	ρ_p (g m ⁻³)	c_p (m s ⁻¹)	η_v^p (mPa s)	η_t^p (mPa s)	$\tilde{\rho}$
PFOB	1.918	624	5	1.8	1.918
Fluorinated oil	1.9	670	2850	950	1.9
Olive oil	0.85	1430	252	84	0.85
Motor oil	0.9	1750	1620	540	0.9

for PMMA and rubber are very similar because their densities are of the same order of magnitude. The compressibility has no effect on coherent waves, contrary to the hydrodynamic forces that depend on the ratio of particle to ambient fluid densities.

C. Droplets

In this section, numerical results are compared for droplets containing PFOB (Perfluorooctylbromide), Fluorinated oil, olive oil, or motor (SAE50) oil. The values of the mechanical parameters are given in Table II.

The normalized phase velocity and the attenuation are plotted in Fig. 3 for the four kinds of droplets: PFOB (curves without symbol), Fluorinated oil (curves with triangle), olive oil (curves with circle), and motor oil (curves with square).

As for solid spheres, both models are strictly equivalent in the long wavelength regime. In the cases of PFOB, fluorinated oil, and olive oil, the bulk modulus of the inner liquid is inferior to the ambient fluid one, leading to a phase velocity inferior to c_0 contrary to the motor oil case.

In a general way, the conclusions for the droplets and elastic particles are similar. Compressibility has a weak effect on coherent waves, and hydrodynamic forces are well approximated by rigid ones as soon as coefficient $\tilde{\mu}$ is large enough. Even if the coefficient $\tilde{\mu}$ of PFOB, which is of order unity, is much smaller than the coefficient $\tilde{\mu}$ of the other droplets, the behavior of coherent waves is the same. It means that coefficient $\tilde{\mu}$ must be quite small to observe a behavior different from the one of moving rigid spheres. This is an important point to consider but out of the scope of the present paper.

VI. ANALYTICAL COMPARISON OF THE TWO MODELS

As previously shown, both models give similar numerical results for any kind of particles. The goal of this section is now to support these observations by comparing the two models analytically under the long wavelength assumption. For that purpose, asymptotic expansions of Bessel and Hankel spherical functions with small arguments have to be used. The bulk absorption in the ambient viscous fluid being supposed to be small, the velocity of shear waves in the ambient fluid is much smaller than that of longitudinal waves. So the non-dimensional wavenumber $|K_T|$ associated to shear waves is much greater than the non-dimensional wavenumber $|K_L|$ associated to compressional waves, and is not necessarily small. As a consequence, asymptotic expansions are not used for the spherical functions of variable K_T ,

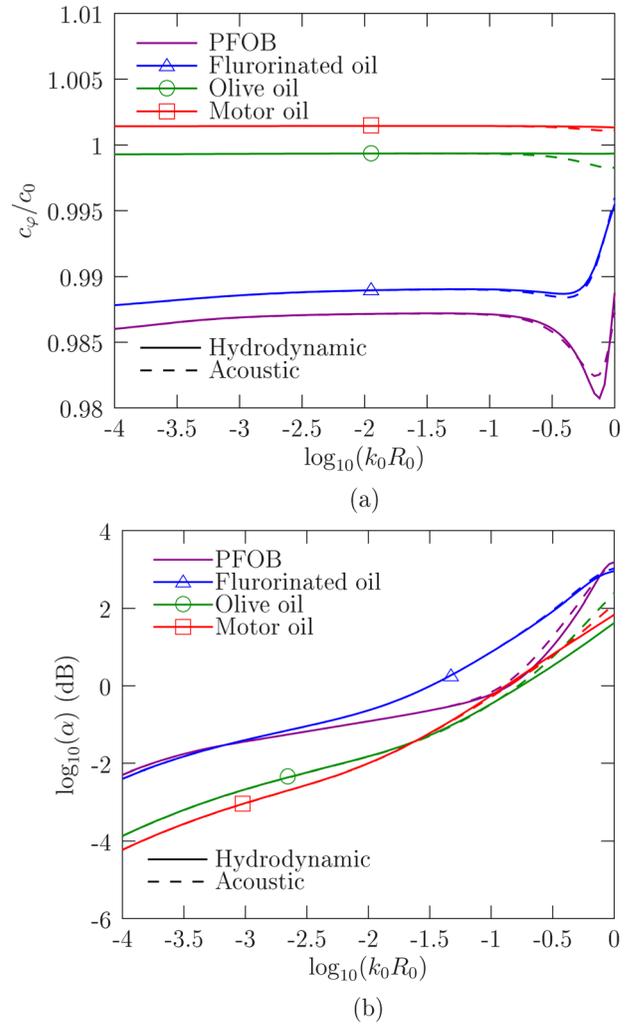


FIG. 3. (Color online) Same as Fig. 1 for viscous droplets.

neither for the spherical functions of variable K_T^p associated to shear waves in viscous droplets.

Moreover, it is well established in the long wavelength regime that the propagation is governed by the two first modes of vibration of the particles⁷ allowing to simplify the expression (48) as follows:

$$\left(\frac{k_A}{k_L}\right)^2 = 1 - i \frac{3\Phi_0}{K_L^3} \left[A_0^{(s,L)} + 3A_1^{(s,L)} \right]. \quad (71)$$

A. Asymptotic approximations of the scattering coefficients

For each kind of particle, the amplitudes of modes are obtained using the Cramer's method. In the following the amplitudes $A_0^{(s,L)}$, $A_1^{(s,L)}$, and $A_1^{(s,T)}$ are calculated for the cases of rigid particles (fixed or moving), elastic spheres, and viscous droplets.

1. Fixed rigid particles

In the case of fixed rigid particles, the amplitudes of modes are obtained by solving Eq. (56) and are given by

$$\begin{cases} A_0^{(s,L)} = -i\frac{K_L^3}{3}, \\ A_1^{(s,L)} = -\frac{K_L^3}{6K_T^2}(3i + 3K_T - iK_T^2), \\ A_1^{(s,T)} = -\frac{iK_L}{2}e^{-iK_T}. \end{cases} \quad (72)$$

Taking into account Eqs. (27) and (33), the amplitude $A_1^{(s,L)}$ can still be expressed by

$$A_1^{(s,L)} = \frac{iK_L^3}{9} \left(1 + \frac{B_{\text{rig}}}{A}\right). \quad (73)$$

2. Moving rigid particles

As previously mentioned, the amplitude of the mode $n=0$ of the longitudinal scattered waves in the ambient fluid is the same for fixed and moving rigid particles, namely, the first equation of the system (72). Solving the matrix Eq. (66), the amplitudes of the modes $n=1$ of the longitudinal and transverse scattered waves in the ambient fluid can be written

$$\begin{cases} A_1^{(s,L)} = \frac{iK_L^3(\tilde{\rho} - 1)(3i + 3K_T - iK_T^2)}{3 \cdot 9(i + K_T) - (2\tilde{\rho} + 1)iK_T^2}, \\ A_1^{(s,T)} = -\frac{(\tilde{\rho} - 1)K_L K_T^2 e^{-iK_T}}{9(i + K_T) - (2\tilde{\rho} + 1)iK_T^2}, \end{cases} \quad (74)$$

or using Eqs. (27) and (33),

$$\begin{cases} A_1^{(s,L)} = -\frac{iK_L^3}{9}(1 - \tilde{\rho})\frac{A + B_{\text{rig}}}{\tilde{\rho}A + B_{\text{rig}}}, \\ A_1^{(s,T)} = \frac{K_L K_T^2 e^{-iK_T}}{18(i + K_T)}(1 - \tilde{\rho})\frac{2B_{\text{rig}} - A}{\tilde{\rho}A + B_{\text{rig}}}. \end{cases} \quad (75)$$

As expected, contrary to fixed rigid particles, the mass of the particle is now present in the amplitudes of the modes $n=1$. Moreover if the particle and the ambient fluid have the same

mass density, namely, if $\tilde{\rho} = 1$, the amplitudes of the mode $n=1$ vanish.

3. Elastic spheres and droplets

Solving Eq. (68) we get

$$A_0^{(s,L)} = i\frac{K_L^3}{3} \frac{\lambda + \frac{2}{3}\mu - \lambda_p - \frac{2}{3}\mu_p}{\lambda_p + \frac{2}{3}\mu_p + \frac{4}{3}\mu}, \quad (76)$$

so that amplitude $A_0^{(s,L)}$ can be expressed by

$$A_0^{(s,L)} = -i\frac{K_L^3}{3}d_p, \quad \text{with} \quad d_p = \frac{3K_p - 3\rho_0 c_0^2}{3K_p - 4i\omega\eta_s}. \quad (77)$$

We recover here the expression of the hydrodynamic model given by Eq. (43). The important point to note is that the amplitude takes into account the viscosity of the ambient liquid contrary to that given by Settnes and Bruus.³⁰ If the bulk modulus K_p increases, the sphere tends to become rigid, and the amplitude of the mode $n=0$ behaves similarly to the rigid case [first part of Eq. (72)] either fixed or moving. It has to be noted here that the mass of the sphere has no influence on the amplitude of the mode $n=0$, that is to say on the dilatational effect. It is also worth noting that dilatational effects disappear if the bulk moduli of both media are equal ($K_p = \rho_0 c_0^2$).

Calculations leading to the expressions for the mode $n=1$ amplitudes for either elastic spheres or viscous droplets are much more complicated than in the previous cases, because determinants of 4×4 matrices are difficult to handle analytically. For that purpose, determinants used for the Cramer's method are computed with the help of Maxima 5.24.0. Numerous factorizations of expressions given by Maxima have had to be performed "manually." After these tedious but not really complicated calculations, the amplitudes $A_1^{(s,L)}$ and $A_1^{(s,T)}$ can be written

$$\begin{cases} A_1^{(s,L)} = -\frac{i(1 - \tilde{\rho})K_L^3[\tilde{\mu}g_1(iK_T^2 - 3K_T - 3i) + g_2(K_T^3 + 3iK_T^2 - 6K_T - 6i)]}{3\tilde{\mu}g_1[iK_T^2(2\tilde{\rho} + 1) - 9(i + K_T)] + 3g_2[(2\tilde{\rho} + 1)(K_T^3 + 3iK_T^2) - 18(i + K_T)]}, \\ A_1^{(s,T)} = -\frac{K_L K_T^2 e^{-iK_T}(1 - \tilde{\rho})(\tilde{\mu}g_1 + 2g_2)}{\tilde{\mu}g_1[iK_T^2(2\tilde{\rho} + 1) - 9(i + K_T)] + g_2[(2\tilde{\rho} + 1)(K_T^3 + 3iK_T^2) - 18(i + K_T)]}, \end{cases} \quad (78)$$

where functions g_1 and g_2 are given by Eq. (32). Then taking into account Eqs. (27) and (29), the amplitudes of the modes $n=1$ can finally be put in the forms

$$\begin{cases} A_1^{(s,L)} = -i\frac{(1 - \tilde{\rho})K_L^3}{9}\frac{A + B_p}{\tilde{\rho}A + B_p}, \\ A_1^{(s,T)} = (1 - \tilde{\rho})\frac{K_L K_T^2 e^{-iK_T}}{18(i + K_T)}\frac{2B_p - A}{\tilde{\rho}A + B_p}. \end{cases} \quad (79)$$

It is worth pointing out that the bulk modulus of elastic spheres and droplets does not appear in the above expressions, contrary to the shear modulus μ_p that appears in the coefficients B_{elas} or B_{drop} through the parameter $\tilde{\mu}$ [cf. Eq. (34)]. The amplitude $A_1^{(s,L)}$ is different and more general than the one derived by Settnes and Bruus³⁰ that does not take into account the elasticity of the sphere.

B. Effective wavenumbers

Whatever the kind of particle, the asymptotic analytical expressions of the scattering coefficients are formally the same. Indeed the amplitude of the mode $n=0$ of the longitudinal wave scattered in the ambient fluid always takes the form

$$A_0^{(s,L)} = -\frac{iK_L^3}{3}d_p. \quad (80)$$

Similarly, taking into account Eq. (37) the amplitudes A_1^L and A_1^T are always given by

$$A_1^{(s,L)} = \frac{iK_L^3}{9}(\tilde{\rho} - 1)t_p \quad (81)$$

and

$$A_1^{(s,T)} = \frac{K_L K_T^2 e^{-iK_T r}}{18(i + K_T)} [3 - (2\tilde{\rho} + 1)t_p]. \quad (82)$$

Equation (81) is proportional to the coefficient $(\tilde{\rho} - 1)t_p$ which can be expressed by

$$(\tilde{\rho} - 1)t_p = \frac{\rho_p \hat{v}_p - \rho \hat{v}}{\rho \hat{v}}. \quad (83)$$

The right-hand side of Eq. (83) involves the difference between the momentum of the particle ($\rho_p \hat{v}_p$) and the momentum of the fluid that would occupy the particles position and follow its movement ($\rho \hat{v}$): this difference of momentum explains the acoustic scattering associated to the mode of vibration $n=1$ at long wavelengths.

In order to demonstrate analytically that both models are equivalent, the expressions of the effective wavenumbers given by the acoustic model have to be expressed and compared to those obtained by the hydrodynamic model. Reporting Eqs. (80) and (81) in Eq. (71) leads straightforwardly to the relation

$$\left(\frac{k_A}{k_L}\right)^2 = 1 - \Phi_0 [d_p + (1 - \tilde{\rho})t_p], \quad (84)$$

which is the same as Eq. (41).

VII. DILATATIONAL MECHANISMS

The aim of this section is to show that dilatational mechanisms are modeled in the same way by both models. This amounts to show that Eq. (24) is also satisfied by the acoustic model. It is not obvious because \hat{p} and \hat{R} are averaged quantities which are not explicitly introduced in the acoustic model. For that purpose, the amplitude of the acoustic pressure and the radial displacement on the particles surface appearing in Eq. (24) has to be identified.

Due to the hypothesis $|K_L| \ll 1$, the coefficients C_p given by the hydrodynamic model are very well approximated by

$$C_p \approx 4i\omega\eta_s - 3K_p, \quad \text{with } p = \text{elas, drop}. \quad (85)$$

In the following, we consider that the field incident on the test particle is the mean field of an effective medium. This is

the independent scattering approximation. Now restricting to incident plane wave, this yields

$$p(t) = p_0 + \hat{p}e^{i(k_H z - \omega t)} \approx p_0 + \hat{p}_{\text{inc}}e^{i(k_H z - \omega t)}. \quad (86)$$

To compute the scattering, we consider a small volume of ambient fluid surrounding the test particle so that \hat{p}_{inc} is the amplitude of the incident wave given by

$$p_{\text{inc}}(t) = \hat{p}_{\text{inc}}e^{i(k_L z - \omega t)}. \quad (87)$$

This approximation is based on the assumption that scattering due to particles modifies mostly the signal phase, that is to say wave velocity and attenuation, but not too much the amplitude. Clearly, this is possible only if the particles concentration is low enough. It follows directly from Eqs. (49) and (54) that

$$p_{\text{inc}} = -\rho_0 c_0^2 \text{div } \mathbf{u}_{\text{inc}} = \rho_0 c_0^2 k_L^2 \varphi_0 e^{i(k_L z - \omega t)}. \quad (88)$$

As a result, the approximation performed in Eq. (86) leads to

$$\hat{p} = \rho_0 c_0^2 k_L^2 \varphi_0. \quad (89)$$

The amplitude \hat{R} of the perturbation of the dynamic radius appearing in Eq. (24) is identified to the radial displacement of the fluid averaged on the surface of the particle. The averaged radial displacement $\langle u_r \rangle_S$ is expressed by

$$\langle u_r \rangle_S = \frac{\varphi_0}{R_0} \left(\frac{iA_0^{(s,L)}}{K_L} - \frac{K_L^2}{3} \right), \quad (90)$$

and using Eq. (80) we get

$$\langle u_r \rangle_S = -\frac{K_L^2 \varphi_0}{3R_0} (1 - d_p). \quad (91)$$

The amplitude of the acoustic pressure \hat{p} can finally be expressed as follows:

$$\hat{p} = C_p \frac{\langle u_r \rangle_S}{R_0} = C_p \frac{\hat{R}}{R_0}, \quad (92)$$

with

$$C_p = 4i\omega\eta_s - 3K_p, \quad \text{with } p = \text{elas, drop}. \quad (93)$$

These expressions are strictly equal to those of the hydrodynamic model [cf. Eq. (85) for elastic spheres and droplets]. Therefore, relation (24) is well satisfied under the hypothesis of low enough concentrations. Retrospectively, this result shows that the hypothesis of quasi-static motion used to derive the new Rayleigh-Plesset-like equation for elastic spheres and droplets is validated by the acoustic model.

VIII. TRANSLATIONAL MECHANISMS AND NEWTON'S LAW

Because the amplitude of the mode $n=1$ is directly related to the coefficient t_p , it appears of fundamental interest

to understand how the hydrodynamic forces are implicitly taken into account in the acoustic model. This amounts to show that Eq. (38) is also satisfied by the acoustic model. For that purpose it is first necessary to determine the velocities of the fluid and of the particles appearing in the hydrodynamic model (\mathbf{v} and \mathbf{v}_p , respectively).

A. Velocity of the fluid

As in the previous section, we assume that the concentration is small enough to introduce the following approximation:

$$\mathbf{v}(z, t) = \hat{v} e^{i(k_H z - \omega t)} \mathbf{e}_z \approx v_{\text{inc}}(z, t), \quad (94)$$

where

$$v_{\text{inc}}(z, t) = -i\omega \frac{\partial \varphi_L^i}{\partial z} e^{-i\omega t} = \frac{\omega^2}{c_L} \varphi_0 e^{i(k_L z - \omega t)}. \quad (95)$$

It follows that

$$\hat{v} = \frac{\omega^2}{c_L} \varphi_0, \quad (96)$$

whatever the kind of particle.

B. Velocity of deformable particles

For rigid particles, the velocity is clearly defined and is uniform all over the particle. For deformable spheres, we define this velocity by averaging the acoustic velocity over the particles volume

$$\langle \mathbf{v}_p \rangle_V = -\frac{i\omega}{\frac{4}{3}\pi R_0^3} \int \int \int_V (u_r^p \mathbf{e}_r + u_\theta^p \mathbf{e}_\theta) e^{-i\omega t} dV. \quad (97)$$

Taking into account Eq. (60), the velocity is shown to be polarized in the direction \mathbf{e}_z , namely $\langle \mathbf{v}_p \rangle_V = \hat{v}_p e^{-i\omega t} \mathbf{e}_z$. Using expressions of displacements of the sphere, we get

$$\hat{v}_p = \frac{3\omega\varphi_0}{R_0} \left[A_1^{(p,L)} j_1(k_L^p) + 2A_1^{(p,T)} j_1(k_T^p) \right], \quad (98)$$

and the long wavelength approximation yields

$$\hat{v}_p \approx \frac{3\omega\varphi_0}{R_0} \left[A_1^{(p,L)} \frac{K_L^p}{3} + 2A_1^{(p,T)} j_1(K_T^p) \right]. \quad (99)$$

Using the continuity of normal displacements, the particles velocity takes the form

$$\hat{v}_p = \frac{3\omega\varphi_0}{R_0} \left[\frac{2i}{K_L^2} A_1^{(s,L)} - 2A_1^{(s,T)} (i + K_T) \frac{e^{iK_T}}{K_T^2} + \frac{K_L}{3} \right]. \quad (100)$$

Reporting Eqs. (81) and (82) in Eq. (100) leads to

$$\hat{v}_p = t_p \hat{v}. \quad (101)$$

Obviously this velocity is zero for fixed rigid particles because t_{rig} tends to 0 when $\tilde{\rho}$ tends to infinity [cf. Eq. (37)].

C. Hydrodynamic forces of the acoustic model

In the long wavelength regime, the hydrodynamic forces given by Eq. (63) can be put directly in the form

$$\hat{F}_p = \frac{4}{3} i\pi\varphi_0 \left[(\lambda + 2\mu)(3iA_1^{(s,L)} - K_L^3) + 6\mu(i + K_T)e^{iK_T} A_1^{(s,T)} \right]. \quad (102)$$

Reporting Eqs. (81) and (82) into this last relation leads to

$$\hat{F}_p = -i\frac{4}{3}\pi R_0^3 \rho_0 \omega^2 k_L \varphi_0 \tilde{\rho} t_p, \quad (103)$$

and using Eqs. (27) and (96) we obtain

$$\hat{F}_p = \tilde{\rho} A t_p \hat{v}, \quad \text{with } p = \text{rig, elas, drop}. \quad (104)$$

As a result, the hydrodynamic forces appearing in the acoustic model are the same as those used in the hydrodynamic model in the cases of rigid particles, solid elastic spheres, and viscous droplets. In addition, the hydrodynamic forces for elastic spheres have the same expression as for droplets, with the coefficient $\tilde{\mu}$ defined by Eq. (34). This shows that the analogy introduced in Sec. III C in order to define the hydrodynamic forces for elastic particles is perfectly valid.

D. Second Newton's law applied to the particles

Using the continuity of normal and tangential stresses at the surface of elastic particles, the hydrodynamic forces given by Eq. (63) can be expressed as

$$\hat{F}_p = 4i\pi\varphi_0 \left[A_1^{(p,L)} (\Sigma_1^{(p,L)} + 2T_1^{(p,L)}) + A_1^{(p,T)} (\Sigma_1^{(p,T)} + 2T_1^{(p,T)}) \right]. \quad (105)$$

Although the expression of \hat{F}_p involves stress amplitudes, \hat{F}_p is a force because the unit of φ_0 is the square meter. Reporting Eq. (53) into this last expression leads to

$$\hat{F}_p = -m_p \omega^2 \frac{3i\varphi_0}{R_0} \left[j_1(K_L^p) A_1^{(p,L)} + 2j_1(K_T^p) A_1^{(p,T)} \right]. \quad (106)$$

Taking into account the exact expression (98) of the velocity of the particle, the hydrodynamic forces satisfy the equation

$$\hat{F}_p = -i\omega m_p \hat{v}_p, \quad (107)$$

which is nothing else than the second Newton's law applied to a particle. The second Newton's law is here obtained without any assumption or approximation. This justifies retrospectively its use in the hydrodynamic model even for deformable particles.

IX. CONCLUSION

The comparison between the hydrodynamic and acoustic models describing the sound propagation in dilute suspensions of spheres has been performed under the

assumption of long wavelengths and small bulk absorption in the ambient viscous fluid. The hydrodynamic model of Coulouvrat *et al.*¹⁶ previously developed for incompressible shells has been generalized to compressible particles, particularly to elastic spheres and droplets. The acoustic model based on the ECAH theory is also generalized in order to take into account the translational effects of rigid moving spheres. Numerical and analytical comparisons show that both models are strictly equivalent for long wavelengths and small enough concentrations of particles. This is actually the most important result of the article. Numerical and analytical calculations highlighted the following significant results.

The coherent wavenumber of the hydrodynamic model fundamentally depends on the dilatational and translational coefficients d_p and t_p . The first one is directly calculated from the Rayleigh-Plesset-like equation and the second one from the hydrodynamic forces. Both play a great role in the physical insight of the propagation of waves. Analytical calculations show that they are closely identified to the scattering coefficients associated to the modes of vibration $n=0$ and $n=1$ calculated from the ECAH theory.

We have derived a new Rayleigh-Plesset-like equation for describing the dilatational effects of elastic spheres and droplets at long wavelengths. The starting point is the Keller-Kolodner equation, which takes into account the compressibility of the ambient fluid. As the particles size is much smaller than the acoustic wavelength, the inertial term in the momentum equation can be neglected. This makes it possible to calculate the normal stress at the fluid-solid interface.

The hydrodynamic forces for elastic spheres are introduced by redefining the coefficient $\tilde{\mu}$ as being the ratio of the shear moduli rather than the ratio of the shear viscosities. Thus, the coefficient $\tilde{\mu}$ is well adapted both for the elastic spheres and droplets. The hydrodynamic forces have exactly the same analytical form because of the close analogy between the solid and fluid acoustic equations (cf. Sec. II). The $\tilde{\mu}$ coefficient plays a very important role for analyzing the behavior of hydrodynamic forces. If $\tilde{\mu}$ is large enough, elastic hydrodynamic forces are well approximated by the usual rigid hydrodynamic forces. This is what happens in most of the cases encountered in the nature.

Analytical expressions of the scattering amplitudes for modes $n=0$ and $n=1$ have been obtained in the long wavelength approximation for elastic spheres immersed in a viscous fluid. First, contrary to the result of Ref. 30, it appears that shear viscosity of the ambient fluid has an influence on the amplitude of mode $n=0$ [cf. Eq. (77)]. Second, the amplitude of mode $n=1$ of the longitudinal scattered wave depends on the shear modulus of the particle [cf. Eq. (79)]. In the case of soft spheres for which the shear modulus is small, this behavior becomes very important.

The second Newton's law is recovered whatever the kind of particles, even for elastic particles and droplets, if the particles velocity is defined as the averaging of the acoustic velocity field over the particle volume. Note that the low concentration hypothesis here is important in order to identify the amplitudes of the acoustic fields appearing in both models.

Finally, it is worth noting that numerical simulations have been performed for different concentrations and the two models give similar results until $\Phi_0 = 5\%$, which is approximately the limit of validity of Foldy's approximation.

ACKNOWLEDGMENTS

The authors acknowledge funding from the French National Agency for Research (ANR-11BS09-007-03) project DiAMAN.

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