Coherent wave propagation in viscoelastic media with mode conversions and pair-correlated scatterers

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HIGHLIGHTS

• The effective wavenumber is expanded up to order 3 in concentration (c).
• The effect of correlation between scatterers is studied.
• It is of order 3 in concentration but of order 2 in scattering.
• It can modify greatly the wave velocity around a subwavelength resonance.
• It increases the attenuation by a small constant amount at higher frequency.

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ABSTRACT

The influence of correlation between scatterers on coherent waves propagation is studied in the case of a viscoelastic medium hosting a random configuration of either spherical or cylindrical scatterers. A distinction is made between the hole correction and the additional disturbances to the pair correlation function beyond the excluded volume via a radial and concentration dependent Ursell function. The effect of the Ursell function on the effective wavenumber is shown to be of order 3 in concentration and order 2 in scattering, and the corresponding formulas generalize those of Caleap et al. (2012) for an ideal fluid host medium. The whole order 3 in concentration is calculated; its other part is of order 3 in scattering. Both parts of the order 3 in concentration are the sum of two terms, one related to mode conversions, the other not. The numerical study is performed mostly for aluminum spheres in epoxy, which is a rather illustrative situation of the different phenomena that participate to the coherent propagation. The Ursell function effect is enhanced at low frequency, while counteracted partly at higher frequency, by the other term of order 3 in concentration. The most visible effects of both terms are on the attenuation. The Ursell term related to mode conversions is larger than the one with no mode conversions included in the low frequency regime.

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1. Introduction

Multiple scattering of waves in random media is of practical interest in many fields, ranging from ultrasonic monitoring of particulate mixtures [1] to the detection of flaws in industrial [2], biological [3,4], or other natural media [5]. Metamaterials
2. Statistics of the medium

We consider \( N \) identical spheres of radius \( a \) randomly distributed in a given region of volume \( V \) with a concentration \( c \) equal to \( (4/3) \pi a^3 / V \). The probability density of finding one sphere centered at \( r \) is known as the 'Hole Correction'. Usually, it is assumed that the hole correction, which physically reasonable for sparse concentration, becomes a poor approximation with the increase of concentration. More generally, we can use \( \frac{1}{1+U(r,\rho_0)} \) for \( |r-\mathbf{r}_1| > b \) and \( 0 \) otherwise. However, it is known that the hole correction, which physically reasonable for sparse concentration, becomes a poor approximation with the increase of concentration. More generally, we can use \( \frac{1}{1+U(r,\rho_0)} \) for \( |r-\mathbf{r}_1| > b \) and \( 0 \) otherwise.

The simplest choice for the conditional number density is the "Hole Correction". Usually, it is assumed that the hole correction, which physically reasonable for sparse concentration, becomes a poor approximation with the increase of concentration. More generally, we can use \( \frac{1}{1+U(r,\rho_0)} \) for \( |r-\mathbf{r}_1| > b \) and \( 0 \) otherwise.

The pair distribution function, in addition to decaying rapidly to zero as \( r \to \infty \), tends to infinity obeying

\[
\lim_{r \to \infty} \mathcal{C}(r) = \frac{1}{N} n(\mathbf{r}, \rho_0).
\]

The pair distribution function is often built by the periodic arrangement of scatterers in a host matrix [6,8] but it is also possible to get interesting properties from and in such cases the concentration of scatterers is rather large (approximately 20%) and it becomes important to have models well adapted to such concentrations.
3. Dispersion equation

This section is written so as to avoid useless and cumbersome calculations. Starting from Ref. [22] we highlight the most important formulas and show how they can be modified or supplemented by the introduction of the Ursell function of Eqs. (3), (4). The notations followed are mostly the same as in Ref. [22]. The host medium allows the propagation of \( P \) different types of waves (\( P = 1 \) for ideal fluids, \( P = 2 \) for elastic solids, \( P = 3 \) for thermo-viscous fluids or Biot-porous solid,...), and each is associated to a different coherent wave.

All effective theories follow the same procedure. An incident plane wave is supposed to enter the volume hosting the scatterers, giving rise to a multiple scattering process. The multiple scattering equations express the incident fields on a given scatterer as the sum of an incident plane wave plus all the waves scattered by the other scatterers, as well as scattered waves in terms of incident waves on a given scatterer via the use of the transition matrix of that scatterer. The conditional ensemble average, over the random positions of the scatterers, of the incident field on one given scatterer at a given position, leads to a hierarchy of equations that is closed usually with the quasi-crystalline approximation. The average field incident on one given scatterer, the coherent field, is then expanded into spherical (or cylindrical, depending on the geometry of the scatterers) harmonics, in the same way as if it were a plane wave, propagating with a complex wavenumber, the effective wave number. Equating the coefficients of the exponential terms related to the effective wavenumber \( \xi \) to zero provides the so-called Lorentz–Lorenz equation to which the partial amplitudes \( A_{\xi}^{p} \) in the expansion of a coherent wave of type \( p \) must obey. It is given in Eq. (18) in Ref. [22] as:

\[
A_n^{(p)} = \frac{-i\pi b}{2\xi} \sum_{q=0}^{P} \sum_{\ell=\max(0,|n-v|)}^{n+v} (1)^{n+\ell} (2n+1) T_\ell^{(qp)} A_\ell^{(q)} \left( \xi b \right) G(0, v|0, n|\ell) = 0,
\]

with

\[
y_p = \xi^2 - k_p^2 \quad \text{and} \quad \epsilon = -4\pi n_0.
\]

In these equations, \( k_p \) is the wave number of a \( p \)-type wave in the absence of scatterers, \( T^{(qp)} \) is the diagonal scattering matrix of a wave of type \( q \) into one of type \( p \), and the Gaunt coefficients \( G(0, v|0, n|\ell) \) are defined by

\[
P_{\ell}^{m}(\cos \theta) P_{\ell}^{\mu}(\cos \theta) = \sum G(m, n|\mu, \nu|\ell) P_{\ell}^{m+\mu}(\cos \theta).
\]

The \( N^{(p)}_\ell(\xi) \) function, defined as

\[
N^{(p)}_\ell(\xi) = \xi b^{\ell} (\xi b) h^{(1)}(k_p b) - k_p b^{\ell} (\xi b) h^{(1)}(k_p b),
\]

is a direct consequence of imposing the hole correction Eq. (2). The generalization to an arbitrary pair distribution function as in Eq. (3) is done in Ref. [15]; it leads to the introduction of an additional term to Eq. (5), which becomes

\[
A_n^{(p)} = \frac{-i\pi b}{2\xi} \sum_{q=0}^{P} \sum_{\ell=\max(0,|n-v|)}^{n+v} (1)^{n+\ell} (2n+1) T_\ell^{(qp)} A_\ell^{(q)} \left[ N^{(p)}_\ell(\xi) + \frac{1}{k_p b^2} \right] G(0, v|0, n|\ell) = 0,
\]

with

\[
L^{(p)}_\ell(\xi) = \int_0^{+\infty} j_m (\xi r) h^{(1)}_m (k_p r) U(r, n_0) k_p^2 r^2 dr.
\]

The procedure to recover the effective wavenumbers \( \xi \) from Eq. (10) is exactly the same as in Ref. [22], which followed Ref. [21]. It consists in writing the system of equations obtained for all values of \( p \) and \( n \) in Eq. (9) in a matrix form with block matrices whose coefficients are matrices related to wave types \( (p, q) \), while the coefficients of these latter matrices are related to orders \( (n, v) \) of the afore-mentioned expansions in spherical harmonics. The trick is then to obtain the dispersion equation by setting to zero the determinant of a \((P, P)\) block matrix,

\[
\text{Det} \left[ Y(\xi) - \epsilon M(\xi) \right] = 0,
\]

where \( Y(\xi) \) and \( M(\xi) \) are matrices of rank \( P \), \( Y(\xi) \) is diagonal with \( Y_{pp}(\xi) = y_p \). The \( M_{qp}(\xi) \) elements of \( M(\xi) \) are given by (misprints in Eqs. (27, 28) of Ref. [22])

\[
M_{qp}(\xi) = M_{qp}^{(p)}\left[ |T_{pq}| T_{pq} \right] e^n, \quad (12a)
\]

\[
M_{qp}(\xi) = \frac{1}{e_q} \sum_{n=1}^{+\infty} M_{qp}^{(n)} e^n, \quad (12b)
\]
\(|e_p|\) are block vectors whose components other than the \(p\)th, which is equal to \(\sqrt{\pi / k_p}\), are zero vectors, \(T\) is a block matrix of coefficients \((T)_{pq} = T^{(pq)}\), and the \(n\)th component of \(|e|\) is \((-1)^n\). The \(\overline{Q}\) block matrix is diagonal, with \((\overline{Q})_{pp} = Q^{(pp)}\), and, as we start here with Eq. (9), in place of Eq. (5), Eq. (19) in Ref. [22] is to be replaced by the following:

\[
\overline{Q}_{nn}^{(p)}(\xi) = \frac{\pi}{k_p y_p} \left\{ -1 + i k_p b \sum_{\ell = n}^{n + m} \left[ N_{\ell}^{(p)}(\xi) + \frac{1}{k_p b k_p^2} \right] \right\} G(0, v|0, 0|\ell) \left(-1\right)^{v+n}. \tag{13}
\]

As expected, we recover the characteristic equation of Ref. [15] when \(P = 1\). We look now for the expansion of the solutions of the dispersion equation into powers of the concentration of scatterers.

### 4. Wavenumbers of the coherent waves

For a given type of wave \(p\), we assume the corresponding effective wavenumber to obey the following formal asymptotic expansion \([20, 21]\) up to order \(N_0\) in concentration,

\[
\xi_{p}^2 = k_p^2 + \sum_{n=1}^{N_0} \varepsilon^n y_p^{(n)}, \tag{14}
\]

and we look for the \(y^{(p)}_p\)s by inserting Eq. (14) into the dispersion equation, Eq. (11). In order to do so, we need to expand as well the Ursell and \(Q_{nn}^{(p)}\) functions that depend on the concentration, but, because of Eqs. (4), (12)–(13), only up to order \(N_0 - 2\):

\[
U(r, n_0) = \sum_{j=1}^{N_0-2} \left[ \frac{\partial^j}{\partial n_0^j} U(r, n_0) \right]_{n_0 = 0} + \cdots = n_0 u(r) + \cdots, \tag{15}
\]

\[
\overline{Q}_{nn}^{(q)}(\xi_p) = \overline{Q}_{nn}^{(q)}(k_p) + \sum_{j=1}^{N_0-2} \varepsilon^j \left[ \frac{d^j \overline{Q}_{nn}^{(q)}(\xi_p(\varepsilon))}{d\varepsilon^j} \right]_{\varepsilon = 0}, \tag{16}
\]

\(\overline{Q}_{nn}^{(q)}(k_p)\) is the zero order term in \(\varepsilon = -4i n_0\) of \(\overline{Q}_{nn}^{(q)}(\xi)\), and, as such, it is found from replacing \(n_0\) with 0 in Eq. (13), so that its expression remains the same as in Ref. [22],

\[
\overline{Q}_{nn}^{(q)}(k_p) = -\frac{\pi}{k_q (k_p^2 - k_q^2)} (-1)^{n+m} \left[ 1 - ik_q b \sum_{v = |n-m|}^{n+m} N_v^{(q)}(k_p) G(0, m|0, n|v) \right], \tag{17a}
\]

\[
\overline{Q}_{nn}^{(p)}(k_p) = \frac{\pi}{2k_p^2} (-1)^{n+m} \sum_{v = |n-m|}^{n+m} D_v^{(p,0)}(k_p) G(0, m|0, n|v), \tag{17b}
\]

with

\[
D_v^{(p,0)}(k_p) = ik_q b \begin{Bmatrix} v (v + 1) - (k_b b)^2 j_v(k_b b) h_v^{(1)}(k_q b) \\ -k_b b j_v(k_b b) h_v^{(1)}(k_q b) - k_q b j_v(k_b b) h_v^{(1)}(k_q b) \end{Bmatrix}. \tag{18}
\]

In the following, we extend the results of Ref. [22] up to order \(N_0 = 3\). We thus need the \(\overline{Q}_{nn}^{(q,j)}(k_p)\) terms for \(j = 1\):

\[
\overline{Q}_{nn}^{(q,1)}(k_p) = \frac{\pi}{2} (-1)^{n+m} \sum_{v = |n-m|}^{n+m} G(0, m|0, n|v) \]

\[
\times \begin{Bmatrix} M_{pp}^{(q,0)} \frac{1}{k_q^2 (k_p^2 - k_q^2)} \left[ \frac{1}{k_p^2 k_p^2} D_v^{(q,0)}(k_p) + 2 \left[ 1 - ik_q b N_v^{(q)}(k_p) \right] \right] \\ - \frac{1}{2k_p^2} A_v^{(q)}(k_p) \end{Bmatrix} (q \neq p), \tag{19}
\]

\[
\overline{Q}_{nn}^{(p,1)}(k_p) = -\frac{\pi}{4} (-1)^{n+m} \sum_{v = |n-m|}^{n+m} G(0, m|0, n|v) \left[ M_{pp}^{(q,0)} \frac{1}{2k_p^2} \left[ 2D_v^{(p,0)}(k_p) + d_v(k_p) \right] + \frac{1}{k_p^2} A_v^{(p)}(k_p) \right], \tag{20}
\]
with

\[ d_v (k_p) = i k_p b \left[ (k_p b)^2 + v (v + 1) \right] j_v (k_p b) \, h_v^{(1)} (k_p b) + (k_p b)^2 \]

\[- v (v + 1) - i (k_p b)^2 f_v (k_p b) \, h_v^{(1) \prime} (k_p b) - i (k_p b)^2 f_v (k_p b) \, h_v^{(1)} (k_p b), \]

\[ M_{pp}^{(0)} = \frac{\pi}{k_p} \sum_{n=0}^{+\infty} (2n + 1) \, T_n^{(pp)}, \]

and

\[ A_v^{(q)} (\xi) = \int_b^{+\infty} j_v (\xi r) \, h_v^{(1)} (k_q r) \, u (r) \, k_r^2 \, dr. \]

It is worth noticing that the \( q_{nnv}^{(q)} (k_p) \) terms, which are related to order 3 in concentration, contain not only the effect of the Ursell function through the \( A_v^{(q)} (k_p) \) integrals, as usual [14–18], but that also of the hole correction through the functions evaluated at \( k_q b \) in Eqs. (19), (20). This point had never been clearly discussed in the literature as far as we know.

Collecting together Eqs. (11), (12), (14)–(23) provides the same orders 1 and 2 of the asymptotic expansion in Eq. (14) as in the case of the hole correction, i.e.

\[ y_p^{(1)} = M_{pq}^{(0)}, \]

\[ y_p^{(2)} = M_{pp}^{(1)} + \sum_{q=1}^{p} \frac{M_{pq}^{(0)} M_{qp}^{(0)}}{k_p^2 - k_q^2}, \]

with

\[ M_{qp}^{(0)} = \frac{\pi}{k_p^{1/2} k_q^{1/2}} \sum_{n=0}^{+\infty} (2n + 1) \, T_n^{(pq)}, \]

\[ M_{ap}^{(1)} = \frac{\pi}{k_q^{1/2} k_p^{1/2}} \sum_{r=1}^{p} \sum_{n} \sum_{m} (-1)^{n+m} (2n + 1) (2m + 1) T_n^{(ar)} T_m^{(aq)} \overline{Q}_{nm}^{(l)} (k_p). \]

The third order term is found as

\[ y^{(3)} = M_{pp}^{(2)} + \sum_{q=1}^{p} \frac{M_{pq}^{(0)} M_{qp}^{(0)} - M_{pq}^{(0)} M_{qp}^{(0)}}{k_p^2 - k_q^2} \frac{2}{2} \]

\[ + \sum_{q=1}^{p} \sum_{r=1}^{p} \sum_{n} \sum_{l} \frac{M_{pq}^{(0)} M_{qp}^{(0)} M_{pp}^{(0)}}{k_p^2 - k_q^2} \frac{2}{2} \]

\[ + \sum_{q=1}^{p} \sum_{r=1}^{p} \sum_{n} \sum_{l} \frac{M_{pq}^{(0)} M_{qp}^{(0)}}{k_p^2 - k_q^2} \frac{2}{2} \]

\[ \] with

\[ M_{pp}^{(2)} = \frac{\pi}{k_p} \sum_{q=1}^{p} \sum_{n} \sum_{m} (-1)^{n+m} (2n + 1) (2m + 1) \]

\[ \times \left[ T_n^{(qp)} T_m^{(pp)} \overline{Q}_{nm}^{(l)} (k_p) + \sum_{r=1}^{p} \sum_{l} (2l + 1) T_r^{(qp)} T_m^{(pp)} \overline{Q}_{rl}^{(l)} (k_p) \overline{Q}_{em}^{(l)} (k_p) \right]. \]

All second and third order terms may be collected together and Eq. (14) can be written in the more comprehensive and practical form

\[ \frac{\xi_p^2}{k_p^2} = 1 + \frac{E}{k_p^3} \delta_v^{(pp)} + \frac{E^2}{k_p^6} \left( \delta_v^{(pp)} + \sum_{q=1}^{p} \delta_v^{(pq)} \right) + \frac{E^3}{k_p^9} \left( \delta_v^{(pp)} + \sum_{q=1}^{p} \delta_v^{(pq)} + \sum_{q=1}^{p} \delta_v^{(pq)} \right) + \cdots , \]

with the \( \delta \) terms given in Appendix B, and the order 3 terms being the main result of this paper.
5. On the correlation

Many authors consider that the correlation as a whole can be regarded as second order in concentration \([14,15,23,24]\). The asymptotic expansion, Eq. (28), shows that while the hole correction effect is indeed, the additional Ursell function term in the pair distribution function is third order only in concentration.

In order to understand better the difference of order between those two correlation-related terms, let us consider the correlation term given for spherical scatterers in a fluid \((P = 1, k_p = k)\) in Refs. [14,24] through the following integral,

\[
l_v(\xi) = e^2 \int_b^{+\infty} \left[ g(r) - 1 \right] r^2 h^{(1)}_v(kr) j_v(\xi r) \, dr,
\]

with the pair correlation (or distribution) function \(g\) defined, with our notations, as (cf. Eq. (15))

\[
g(r) = 1 + U(r, n_0) = 1 + n_0 u(r) + \cdots.
\]

The key point here is that the pair distribution function \(g\) does not depend on \(r\) only, but on the concentration as well. Moreover, it tends to unity when the concentration vanishes, as discussed in Section 2 of this paper as well as in the first paper of Ref. [11]. Considering for example the Percus–Yevick’s pair correlation function, its Virial expansion, up to third order in concentration, is given in Ref. [15] as

\[
g(r) = \begin{cases} 1 & \text{for } r \geq 2b \\ 1 + n_0 \frac{4\pi}{3} b^3 \left(1 - \frac{3r}{4b} + \frac{1}{16b^3}\right) & \text{for } b \leq r \leq 2b,
\end{cases}
\]

which is clearly the same form as the right hand side of Eq. (30), and, used in Eq. (29), provides a third order term in concentration:

\[
l_v^{(3)}(\xi) = e^2 n_0 \frac{4\pi}{3} b^3 \int_b^{2b} \left(1 - \frac{3r}{4b} + \frac{1}{16b^3}\right) r^2 h^{(1)}_v(k_q r) j_v(\xi r) \, dr.
\]

It follows also that, while correctly taken into account in Ref. [15], the effect of the Ursell function should not be \textit{a priori} separated from the (other part of) order 3 in concentration that the authors had found too long to warrant including in their paper, and we focus now on the third order in concentration. The \(\delta_{3^{(pp)}}\) and \(\delta_{3^{(pq)}}\) coefficients include the hole correction, and each of them is a sum of terms that involve the product of three scattering coefficients. The last coefficient, \(\delta_{3^{(pq)}}\), which vanishes with the Ursell function, involves the product of two scattering coefficients only. In other words, while all other terms of a given order in concentration are of the same order in scattering, those due to the Ursell function are of one order less in scattering than in concentration. It might thus be that the \(\delta_{3^{(pp)}}\) and \(\delta_{3^{(pq)}}\) terms are negligible, compared to \(\delta_{3^{(qq)}}\). The relative importance of all correlation terms, in both the second and third orders in concentration, is studied numerically in the next section.

6. Numerical study

We focus here on the coherent wave whose wavenumber is closest to that of a longitudinal wave in the absence of scatterers \((p = 1)\). The aim of this section is to investigate numerically the influence of the different terms of the expansion of its wavenumber on both its velocity and its attenuation.

Silica nano-spheres in (viscous) water have been studied in Ref. [25] in the sub-wavelength domain for the compression wave and concentrations in the range 5%–20%; it has shown that agreement between theory and experiment could not be achieved if one neglected the compression/viscous–shear mode conversions, and that, for the highest values of the concentration, third order terms in concentration were needed as well. Ref. [26] was about the sub-wavelength multiple scattering by Tungsten–Carbide spheres in an epoxy resin, with concentrations ranging from 2% to 10%; again, the influence of the mode conversions was clearly demonstrated from the comparison with experiment, along with the need of taking into account the hole correction (as opposed to the low frequency \(k\rightarrow 0\) assumption). In both references, however, discrepancy between theory and experiment appeared as the concentration increased: at concentrations larger than 25% in Ref. [25], in which third order terms in concentration were included but the Ursell function was neglected, and at concentrations larger than 5% in Ref. [26], in which there were no third order terms. The Tungsten spheres in Ref. [26] exhibited a strong sub-wavelength resonant behavior that was not present in the case of Ref. [25], and we believe it might be the reason why the model failed at relatively low concentrations in Ref. [26]. In the following, we avoid this very peculiar case where the mass density ratio between the scatterers and the matrix is very large, which may lead to negative attenuation, as discussed later in this section. We consider here at first different concentrations of aluminum spheres in an epoxy matrix. The aluminum beads are then replaced by much softer spheres, made of Polydimethylsiloxane (PDMS), whose mass density, contrary to that of aluminum, is smaller than that of epoxy. Both Epoxy and PDMS are viscoelastic isotropic media, modeled
with frequency dependent complex Lamé coefficients. There are thus $P = 2$ different types of waves in the epoxy host medium: longitudinal ($p = 1$) and shear ($p = 2$) waves. The Zener model used for epoxy [27],

$$C = C_0 + \sum_{n=1}^{4} C_n \frac{(\omega \tau_n)^2 - i \omega \tau_n}{1 + (\omega \tau_n)^2},$$

with $C$ standing for either $\lambda$ or $\mu$, is derived from the experiment and described in Ref. [26], while the Kelvin–Voigt model is used for the PDMS,

$$\lambda = \lambda_0 - i \omega \left( \eta_B - \frac{2}{3} \eta_S \right), \quad \mu = \mu_0 - i \omega \eta_S,$$

with $\eta_B = 1.3$ Pa s and $\eta_S = 0.6$ Pa s.

The elastic Lamé coefficients $\lambda_0$ and $\mu_0$, along with the mass density, are given in Table 1 for (elastic) aluminum, epoxy and PDMS, while the relaxation times in Eq. (33) are given in Table 2. These values lead to a practically real wavenumber $k_1$ of the longitudinal wave in epoxy on the frequency range we studied.

Most figures in the two following sections will show the dispersion curves (velocity and attenuation) versus frequency obtained from different approximations of the effective wavenumber. The (green) curves with no marks correspond to $k_1 b \rightarrow 0$, i.e. no hole correction is taken into account, contrary to the (black) curves with triangles for which the exclusion radius is equal to the diameter of the scatterers. Both sets of curves correspond to the asymptotic expansion of the wavenumber limited to order two in concentration and scattering. The additional correlation term due to the Ursell function is added to provide the (blue) curves with stars. The pair distribution function considered is that of Percus–Yevick; its Virial expansion, Eq. (31), is used with the exclusion radius $b$ equal to twice the radius $a$ of the cylinders. Finally, the (magenta) curves with squares have been drawn with the whole order three in concentration taken into account.

### 6.1. Aluminum spheres in epoxy

We consider here an epoxy matrix with aluminum spheres within. As can be seen from Table 1, aluminum is ten to twenty times stiffer than epoxy: its resonant behavior, thus is governed mainly by the ratio of its density to that of epoxy. The spheres have a 0.5 mm radius $a$. Figs. 1, 2 show respectively the speed and the attenuation of the longitudinal coherent wave for $c = 15\%$ on the reduced frequency range $0.01 \leq k_1 a \leq 10$, and Figs. 3, 4 the same at low frequency, $0.001 \leq k_1 a \leq 1$. The influence of the concentration is observed through the comparison of the latest with Figs. 5, 6, which are their equivalent at higher concentration, $c = 25\%$.

Same way as in Ref. [26], Figs. 1–4 show clearly a peculiar behavior of the dispersion curves obtained at order 2 when the hole correction is not taken into account (green curves, no marks), when compared to the other curves. Numerically, this is due to the fact that $k_2 b$ is five times greater than $k_1 a$, so that the Hankel functions of argument $k_2 b$ cannot be replaced by their asymptotic expression at low argument as was done in the low frequency section of Ref. [22]: Eq. (36) of that reference, in fact, is rather a quasi-static approximation than a low frequency one. Physically, this underlines the importance of taking into account the size of the scatterers, even at low frequency. However, it may happen that forgetting it provides nonetheless a reasonable agreement with the experiment at larger than unity $k_1 a$ values and non overlapping scatterers [23].

The Lloyd–Berry effective wavenumber $\left[10,11\right]$ corresponds to the dashed green curve with no marks in Figs. 3, 4. It is recovered from the order 2 with no hole correction by neglecting the mode conversions. The difference between those two curves with no marks clearly shows the importance of mode conversions at low frequency.

We focus now on the other curves of Figs. 1–4, obtained with the hole correction accounted for and $b = 2a$. All three curves with marks in Fig. 1 provide practically the same velocity for high frequencies $f \geq 3$ MHz ($k_1 a$ around 4 and larger). By contrast, Fig. 2 shows that the differences between the attenuation curves are negligible only at rather low frequencies,
Fig. 1. (Color online) Aluminum spheres in epoxy, $a = 500 \mu m$, $c = 15\%$. Effective phase velocity $\omega / |\text{Re}(\xi_1)|$ versus frequency, $0.01 \leq k_1 a \leq 10$, $0.025 \leq k_1 a \leq 25$. Green line, no marks: from the expansion of $\xi_1$ up to order 2 in concentration, no hole correction ($b \to 0$). Black line + triangles: from the expansion of $\xi_1$ up to order 2, hole correction included ($b = 2a$). Blue line + stars: third order Ursell term added. Magenta line and squares: from the expansion of $\xi_1$ up to whole order 3 in concentration.

Fig. 2. (Color online) Aluminum spheres in epoxy, $a = 500 \mu m$, $c = 15\%$. Effective attenuation $\text{Im}(\xi_1)$ versus frequency, $0.01 \leq k_1 a \leq 10$, $0.025 \leq k_1 a \leq 25$. Green line, no marks: from the expansion of $\xi_1$ up to order 2 in concentration, no hole correction ($b \to 0$). Black line + triangles: from the expansion of $\xi_1$ up to order 2, hole correction included ($b = 2a$). Blue line + stars: third order Ursell term added. Magenta line and squares: from the expansion of $\xi_1$ up to whole order 3 in concentration.

$f \leq 2 \text{ MHz} (k_1 a \text{ around 2.8 and smaller})$. Other calculations, not shown here, for titanium beads have led us to think that such a difference between velocity and attenuation behavior is quite a general result for scatterers that are heavier than the matrix. In addition, one can check from the comparison of Figs. 5, 6, drawn for $c = 25\%$, to Figs. 3, 4, drawn for $c = 15\%$, that the frequency range in which there are noticeable differences between the different approximations increases with concentration.

Let us concentrate now on the influence of the Ursell function, which can be asserted from the comparison between the (black) curves with triangles and the (blue) ones with stars. The Ursell function has no influence on the velocity at low and high frequency, $f \leq 0.05 \text{ MHz}$ and $f \geq 3 \text{ MHz} (k_1 a \text{ around 4 and larger})$, while the situation is quite different for the attenuation, which is sensitive to this additional correlation term on the whole frequency range $2 \leq f \leq 7 \text{ MHz} (k_1 a \text{ roughly between 2.8 and 10})$. Again, it seems to be a relatively general result that the influence of the Ursell function on the attenuation takes place on a larger frequency band than it does on the velocity: we observed it for the other bead materials tested, as well as the authors of Ref. [23] for steel rods in water and of Ref. [24] for bubbles in PDMS.

The sub-wavelength resonance in Ref. [26] was shown to be due to the dipole mode $n = 1$. It is also the case here for the resonance at 340 kHz (from Eq. (3) in Ref. [26]) that is more clearly seen in Fig. 5, because of the higher concentration, than in Figs. 1–4. In order to investigate its effect, we have plotted in Fig. 7 the velocity obtained from the wavenumber expansion up to order 2 in concentration, with no hole correction taken into account ($b \to 0$), for different values of the concentration. This figure clearly shows a curvature inversion around the resonance frequency (marked by the vertical dashed line) that is
Fig. 3. (Color online) Aluminum spheres in epoxy, \( a = 500 \mu m, c = 15\% \). Effective phase velocity \( \omega / |\text{Re}(\xi_1)| \) versus frequency. Zoom of Fig. 1 at low frequency, \( 0.001 \leq k_1a \leq 1 \). Solid green line, no marks: from the expansion of \( \xi_1 \) up to order 2 in concentration, no hole correction \( (b \to 0) \). Dashed green line, no marks: order 2, no hole correction \( (b \to 0) \) and no mode conversions. Black line + triangles: from the expansion of \( \xi_1 \) up to order 2, hole correction included \( (b = 2a) \). Blue line + stars: third order Ursell term added. Magenta line and squares: from the expansion of \( \xi_1 \) up to whole order 3 in concentration.

Fig. 4. (Color online) Aluminum spheres in epoxy, \( a = 500 \mu m, c = 15\% \). Effective attenuation \( \Im(\xi_1) \) versus frequency. Zoom of Fig. 2 at low frequency, \( 0.001 \leq k_1a \leq 1 \). Solid green line, no marks: from the expansion of \( \xi_1 \) up to order 2 in concentration, no hole correction \( (b \to 0) \). Dashed green line, no marks: order 2, no hole correction \( (b \to 0) \) and no mode conversions. Black line + triangles: from the expansion of \( \xi_1 \) up to order 2, hole correction included \( (b = 2a) \). Blue line + stars: third order Ursell term added. Magenta line and squares: from the expansion of \( \xi_1 \) up to whole order 3 in concentration.

enhanced by the increase of the concentration. Turning then back again to Fig. 5, corresponding to \( c = 25\% \), one can see that the main effect of the Ursell function is a shift towards high frequency of the whole velocity curve: for example, the [0.2 MHz, 0.4 MHz] frequency interval over which the phase velocity decreases is shifted to [0.3 MHz, 0.5 MHz] by the introduction of the Ursell term. A similar shift, due in that case to the hole correction only, can be observed from the comparison of Figs. 5–7. The hole correction is usually seen only as a mean to proscribe inter penetration of scatterers. In the light of the similarity of its effect with that of the Ursell function, it can also be seen as the low concentration part of the correlation term, as described indeed by Eq. (31).

The influence of mode conversions on the terms related to the Ursell function is depicted by Fig. 8, which clearly shows that the amplitude of the Ursell term related to mode conversions can be larger than that with no mode conversion. The same holds for both the real and imaginary parts of those terms: mode conversions must be taken into account at low frequency \((k_1a < 1)\).

As for the remaining part of order three in concentration, it seems to enhance the Ursell function effect at low frequency (the blue curves with stars are in between the black ones with triangles and the magenta ones with squares in Figs. 3, 4) and to counteract it partly at higher frequency (the black curves with triangles are between the blue one with stars and the magenta one with squares in Figs. 1, 2). This concurs with the need of its introduction in the low frequency study of Ref. [25].
Fig. 5. (Color online) Aluminum spheres in epoxy, \( a = 500 \, \mu m, c = 25\% \). Effective phase velocity \( \omega / [\Re (\xi)] \) versus frequency, \( 0.001 \leq k_1 a \leq 1, \ 0.0025 \leq k_2 a \leq 2.5 \). Black line + triangles: from the expansion of \( \xi_1 \) up to order 2 in concentration, hole correction included (\( b = 2a \)). Blue line + stars: third order Ursell term added. Magenta line and squares: from the expansion of \( \xi_1 \) up to whole order 3 in concentration.

Fig. 6. (Color online) Aluminum spheres in epoxy, \( a = 500 \, \mu m, c = 25\% \). Effective attenuation \( \Im (\xi) \) versus frequency, \( 0.001 \leq k_1 a \leq 1, \ 0.0025 \leq k_2 a \leq 2.5 \). Black line + triangles: from the expansion of \( \xi_1 \) up to order 2 in concentration, hole correction included (\( b = 2a \)). Blue line + stars: third order Ursell term added. Magenta line and squares: from the expansion of \( \xi_1 \) up to whole order 3 in concentration.

A last remark should be made before concluding this section: while hardly noticeable in Fig. 6, the attenuation at order 2 in concentration (black curve with triangles) is negative around 0.28 MHz, while it is always positive at the lower concentration of Fig. 4. Such a phenomenon had already been noticed by others [15,28,29]. Introduction of the correlation gives back a positive attenuation in Fig. 6, as it was shown to do in Ref. [15]. However, other studies we have conducted (not shown here) have shown that the correlation term merely increases the critical concentration at which a negative attenuation may occur. This critical concentration, in fact, appears mostly in the vicinity of a sub-wavelength resonance, and depends on the scatterers mass density: the higher the latter, the smaller the critical concentration [26]. Now, the question is: is a negative attenuation merely a manifestation of the limit of the multiple scattering model itself, or a truly physical metamaterial like property, as is, for example, a negative index [9]? Answering this question is beyond our ability and aim at the present time.

6.2. PDMS spheres in epoxy

PDMS is both lighter and much softer than epoxy (see Table 1), and, in contrast with aluminum, its resonant behavior involves more than one parameter (the mass density ratio with the matrix). Its high compressibility gives rise to a large number of Mie resonances [30] on the whole frequency range that cause the modulations observed in Figs. 9, 10. The sub-wavelength resonance is not as strong as that of the aluminum spheres, hence the phase velocity variations (\( f < 0.5 \, MHz, \ k_1 a \) smaller than 1.4) in Fig. 9 are smaller than those in Fig. 5 for the same \( k_1 a \) reduced frequency range. A much stronger dispersion of the phase velocity is observed in Fig. 9 than in Fig. 7 for the aluminum spheres and the same concentration.
Fig. 7. (Color online) Aluminum spheres in epoxy, $a = 500 \, \mu\text{m}$. Effective phase velocity $\omega/\text{Re}(\xi_1)$ versus frequency, $0.001 \leq k_1 a \leq 1$, $0.0025 \leq k_2 a \leq 2.5$, for different values of the concentration. From the expansion of $\xi_1$ up to order 2 in concentration, no hole correction $(b \to 0)$. The vertical dashed line is situated at the resonance frequency of the spheres, $f = 340$ kHz.

Fig. 8. (Color online) Aluminum spheres in epoxy, $a = 500 \, \mu\text{m}$. Comparison of the magnitude of the Ursell related parts of the third order term in concentration in the expansion of $\xi_1^2/k_1^2$ versus frequency $0.01 \leq k_1 a \leq 10$, $0.025 \leq k_2 a \leq 25$. Solid red line: $\left(\frac{k}{k_1}\right)^3 \delta^{(11)}_{\text{Ursell}}$, blue dashed line: $\left(\frac{k}{k_1}\right)^3 \delta^{(12)}_{\text{Ursell}}$, from Eqs. (B.1), (B.3.6). This figure does not depend on the concentration, due to the division parameter.

We have, however tried to identify neither the vibration mode associated to that behavior, nor the resonances observed at higher frequency or the origin of the small variations superposed to those resonances in the attenuation curves of Fig. 10, as it is out of the scope of this paper.

The main thing to be retained, however, is that the previous conclusions made for heavy and “rigid” particles are still valid here: the attenuation is more sensitive to correlation than the velocity, with the attenuation decrease due to the Ursell function counteracted by the remaining terms of order 3 in concentration, so that the attenuation, when all terms at order 3 in concentration are taken into account, is larger than the expansion limited to order 2 in concentration provides.

### 7. Conclusion

We have studied the properties of coherent waves in elastic media through the expansion of the effective wavenumber up to the third power of the concentration. Order three contains two different contributions. The first one is a consequence of correlation between scatterers that we modeled by the introduction of a pair distribution function. While of order three
in concentration, it is of order two only in scattering. The second one is, as all other terms of the expansion, of the same order in scattering as in concentration.

When compared to the influence of the correlation on the properties of the coherent wave, that of the other part of the third order is no different on a qualitative point of view. It needs, however, to be taken into account for quantitative purposes.

The effect of correlation depends strongly on frequency, through the ratio of the longitudinal wavelength to the radius of the scatterers. At low frequency, it may modify the phase velocity rather greatly, as the frequency range over which the sub-wavelength dipolar resonance induces a high velocity dispersion is shifted towards higher frequencies. As a general rule, while the correlation has a rather small effect on the phase velocity, its influence on the attenuation is not negligible, except at very low frequency \( k_1 a < 1 \). Adding only the term related to the Ursell function to the expansion up to order two in concentration decreases the attenuation, but adding the whole order three in concentration increases it by a small and practically constant amount.

The authors wish to thank one of the reviewers who has helped them clarifying the original manuscript by pointing out that the general term of correlation included both the hole correction and the Ursell function.
Appendix A. The 2D case

The expressions needed for the 2D case are given here

$$M_{qp}^{(0)} = \sum_{n=-\infty}^{+\infty} T_n^{(pq)}$$

$$M_{qp}^{(1)} = \sum_{n=-\infty}^{+\infty} \sum_{m=-\infty}^{+\infty} \sum_{r=1}^{+\infty} T_n^{(qr)} T_m^{(pr)} \overline{Q_{nm}^{(r)}} (k_p)$$

$$M_{pp}^{(2)} = \sum_{n=-\infty}^{+\infty} \sum_{m=-\infty}^{+\infty} \sum_{q=1}^{+\infty} T_n^{(qp)} T_m^{(p)} \overline{Q_{nm}^{(p,1)}} + \sum_{n=-\infty}^{+\infty} \sum_{m=-\infty}^{+\infty} \sum_{q=1}^{+\infty} \sum_{r=1}^{+\infty} T_n^{(qr)} T_m^{(pr)} \overline{Q_{n\ell}^{(r)}} (k_p) \overline{Q_{\ell m}^{(q)}} (k_p)$$

$$Q_{nm}^{(q)} (k_p) = \frac{i \pi}{2} N_{nm}^{(q)} (k_p) - 1 \quad (q \neq p)$$

$$Q_{nm}^{(p)} (k_p) = \frac{1}{k_p^2} D_{nm}^{(p)} (k_p)$$

$$N_{nm}^{(q)} (\xi) = \xi b_m^{(q)} (\xi b) H_0^{(1)} (k_q b) - k_q b_m^{(q)} (\xi b) H_0^{(1)} (k_q b)$$

$$D_m^{(q)} (k_p) = \frac{i \pi}{4} \left[ m^2 - (k_p b)^2 \right] J_m (k_p b) H_0^{(1)} (k_q b) - k_p b_m^{(q)} (k_p b) H_0^{(1)} (k_q b)$$

$$\overline{q}_{nm}^{(q,1)} (k_p) = \frac{M_{pp}^{(0)}}{k_p^2} \frac{1}{(k_p^2 - k_q^2)^2} \left\{ (k_p^2 - k_q^2) D_{n-m}^{(q,0)} (k_p) - k_p^2 \left[ \frac{i \pi}{2} N_{n-m}^{(q)} (k_p) - 1 \right] \right\} - \frac{\pi}{8 k_p^2} \Lambda_{n-m}^{(q)} (k_p) \quad (q \neq p)$$

$$\overline{q}_{nm}^{(p,1)} (k_p) = \frac{M_{pp}^{(0)}}{k_p^4} \left[ \frac{1}{2} D_{n-m}^{(p,0)} (k_p) + d_{n-m} (k_p) \right] - \frac{\pi}{8 k_p^2} \Lambda_{n-m}^{(p)} (k_p).$$

$$\Lambda_m^{(q)} (\xi) = \int_b^{+\infty} J_m (\xi r) H_0^{(1)} (k_q r) u (r) k_q^2 r dr,$n

$$d_m (k_p) = \frac{1}{8} \left[ m^2 - (k_p b)^2 - i \pi (k_p b)^2 J_m (k_p b) H_0^{(1)} (k_q b) \right].$$

Appendix B. Final expressions

The most practical form of the wavenumber expansion, up to order 3 in concentration, is:

$$\frac{\xi_p^2}{k_p^2} = 1 + \frac{\epsilon_p^2}{k_p^2} \delta_1^{(pp)} + \frac{\epsilon_p^2}{k_p^2} \left( \delta_2^{(pp)} + \sum_{q=1}^{p} \delta_2^{(pq)} \right) + \frac{\epsilon_p^3}{k_p^3} \left( \delta_3^{(pp)} + \sum_{q=1}^{p} \delta_3^{(pq)} + \sum_{q=1}^{p} \delta_3^{(ppq30real)} \right),$$

with $d = 2$ in the 2D case and $d = 3$ in the 3D one, and the different delta terms given hereafter for both cases.

The 2D case

$$\delta_1^{(pp)} = \sum_{n=-\infty}^{+\infty} T_n^{(pp)}$$
\[
\delta_2^{(pp)} = \sum_{n, m = -\infty}^{+\infty} T_n^{(pp)} T_m^{(pp)} D_{n-m}^{(p,0)} (k_p) \tag{B.2.2}
\]
\[
\delta_2^{(pq)} = \frac{i\pi}{2} \frac{k_p^2}{k_p^2 - k_q^2} \sum_{n, m = -\infty}^{+\infty} T_n^{(pq)} T_m^{(pq)} D_{n-m}^{(p,0)} (k_p) \tag{B.2.3}
\]
\[
\delta_3^{(pq)} = -\frac{1}{2} \sum_{n, m = -\infty}^{+\infty} T_n^{(pp)} T_m^{(pp)} T_{n-m}^{(0,0)} (k_p) + \sum_{n, m, \ell = -\infty}^{+\infty} T_n^{(pp)} T_{m-\ell}^{(pp)} \sum_{m = -\infty}^{+\infty} T_{n-m}^{(pq)} D_{n-m}^{(p,0)} (k_p) \tag{B.2.4}
\]
\[
\delta_3^{(pq)} = i\pi \frac{k_p^2}{k_p^2 - k_q^2} \sum_{n, m = -\infty}^{+\infty} T_n^{(pq)} T_m^{(pq)} D_{n-m}^{(q,0)} (k_p) \tag{B.2.5}
\]
\[
\delta_3^{(pq)}_{\text{3dshell}} = -\frac{\pi^2}{8} (k_p a)^2 \sum_{n, m = -\infty}^{+\infty} T_n^{(pq)} T_m^{(pq)} \int_{b}^{+\infty} J_{n-m} (k_p r) H_{n-m}^{(1)} (k_q r) u_0 (r) k_p^2 r dr, \tag{B.2.6}
\]
with \( u_0 (r) \) the non dimensional function defined from function \( u \),
\[
u_0 (r) = \frac{\partial U}{\partial C} \bigg|_{n_0 = 0} = \frac{1}{\pi a^2} u \left( r \right). \tag{B.2.7}
\]

The 3D case
\[
\delta_1^{(pp)} = \pi \sum_{n = 0}^{+\infty} (2n + 1) T_n^{(pp)} \tag{B.3.1}
\]
\[
\delta_2^{(pp)} = \frac{\pi^2}{2} \sum_{n = 0}^{+\infty} (2n + 1) (2m + 1) T_n^{(pp)} T_m^{(pp)} \sum_{m = 0}^{n+m} D_{m}^{(p,0)} (k_p) G (0, m | 0, n | v) \tag{B.3.2}
\]
\[
\delta_2^{(pq)} = i\pi^2 (k_p b) \frac{k_p^2}{(k_p^2 - k_q^2)} \sum_{n = 0}^{+\infty} (2n + 1) (2m + 1) T_n^{(pq)} T_m^{(pq)} \sum_{m = 0}^{n+m} N_{m}^{(q)} (k_p) G (0, m | 0, n | v) \tag{B.3.3}
\]
\[
\delta_3^{(pp)} = \frac{\pi^3}{8} (k_p b)^2 \frac{k_p^2}{(k_p^2 - k_q^2)} \sum_{n, m = 0}^{+\infty} (2n + 1) (2m + 1) (2\ell + 1) T_n^{(pp)} T_m^{(pp)} T_{\ell}^{(pp)} \sum_{m = 0}^{n+m} D_{m}^{(p,0)} (k_p) d_{\ell} (k_p) G (0, m | 0, n | v) \tag{B.3.4}
\]
\[
\delta_3^{(pq)} = \frac{\pi^3}{8} \sum_{n, m = 0}^{+\infty} (2n + 1) (2m + 1) (2\ell + 1) T_n^{(pq)} T_m^{(pq)} T_{\ell}^{(pp)} \sum_{m = 0}^{n+m} D_{m}^{(p,0)} (k_p) D_{\ell}^{(p,0)} (k_p) G (0, \ell | 0, n | v) G (0, m | 0, \ell | \mu) \tag{B.3.5}
\]
\[
\frac{k_p^3}{2k_q (k_p^2 - k_q^2)} \sum_{n=0}^{+\infty} \sum_{m=0}^{+\infty} \sum_{\ell=0}^{+\infty} (2n + 1) (2m + 1) (2\ell + 1) \left( T_{n}^{(qp)} T_{m}^{(pq)} T_{\ell}^{(pp)} \right)
\]

\[
\times \sum_{v=[n-m]} \sum_{[n]} \sum_{[m]} D_v^{(p,0)} (k_p) G (0, m|0, n|v)
\]

\[
- i (k_p b) \frac{k_p^2}{(k_p^2 - k_q^2)} \sum_{n=0}^{+\infty} \sum_{m=0}^{+\infty} \sum_{\ell=0}^{+\infty} (2n + 1) (2m + 1) (2\ell + 1) \left( T_{n}^{(qp)} T_{m}^{(pq)} T_{\ell}^{(pp)} \right)
\]

\[
\times \sum_{v=[n-m]} N_v^{(q)} (k_p) G (0, m|0, n|v)
\]

\[
\delta_3^{(pq)} = \pi^3
\]

\[
+ i (k_p b) \frac{k_p^2}{2 (k_p^2 - k_q^2)} \sum_{n=0}^{+\infty} \sum_{m=0}^{+\infty} \sum_{\ell=0}^{+\infty} (2n + 1) (2m + 1) (2\ell + 1) \left( T_{n}^{(qp)} T_{m}^{(pq)} T_{\ell}^{(pp)} \right)
\]

\[
\times \sum_{v=[n-m]} \sum_{[n]} \sum_{[m]} D_v^{(p,0)} (k_p) N_v^{(q)} (k_p) G (0, m|0, \ell|v) G (0, \ell|0, n|\mu)
\]

\[
- (k_p b)^2 \frac{k_p^2}{(k_p^2 - k_q^2)} \sum_{r=1}^{p} \frac{1}{(k_p^2 - k_q^2)} \sum_{n=0}^{+\infty} \sum_{m=0}^{+\infty} \sum_{\ell=0}^{+\infty} (2n + 1) (2m + 1) (2\ell + 1) \left( T_{n}^{(qp)} T_{m}^{(pq)} T_{\ell}^{(pp)} \right)
\]

\[
\times \sum_{v=[n-m]} \sum_{[n]} \sum_{[m]} N_v^{(r)} (k_p) N_v^{(f)} (k_p) G (0, \ell|0, n|v) G (0, m|0, \ell|\mu)
\]

\[
\delta_{3\text{Durr}}^{(pq)} = - \frac{\pi^3}{3} (k_p a)^3 \sum_{n=0}^{+\infty} \sum_{m=0}^{+\infty} \sum_{\ell=0}^{+\infty} (2n + 1) (2m + 1) T_{n}^{(qp)} T_{m}^{(pq)}
\]

\[
\times \sum_{v=[n-m]} \int_0^\infty j_v (k_p r) h_v^{(1)} (k_q r) u_0 (r) k_p^2 r^2 dr G (0, m|0, n|v)
\]

with \( u_0 \) the non dimensional function defined from function \( u \),

\[
u_0 (r) = \left( \frac{\partial U}{\partial c} \right)_{n_0=0} = \frac{3}{4} \frac{1}{\pi a^2} u (r).
\]

References


