Effective dynamic constitutive parameters of acoustic metamaterials with random microstructure

Mihai Caleap¹, Bruce W Drinkwater and Paul D Wilcox
Department of Mechanical Engineering, University of Bristol, Queen’s Building, University Walk, Bristol BS8 1TR, UK
E-mail: Mihai.Caleap@bristol.ac.uk

New Journal of Physics 14 (2012) 033014 (29pp)
Received 23 November 2011
Published 12 March 2012
Online at http://www.njp.org/
doi:10.1088/1367-2630/14/3/033014

Abstract. A multiple scattering analysis in a non-viscous fluid is developed in order to predict the effective constitutive parameters of certain suspensions of disordered particles or bubbles. The analysis is based on an effective field approach, and uses suitable pair-correlation functions to account for the essential features of densely distributed particles. The effective medium that is equivalent to the original suspension of particles is a medium with space and time dispersion, and hence, its parameters are functions of the frequency of the incident acoustic wave. Under the quasi-crystalline approximation, novel expressions are presented for effective constitutive parameters, which are valid at any frequency and wavelength. The emerging possibility of designing fluid–particle mixtures to form acoustic metamaterials is discussed. Our theory provides a convenient tool for testing ideas in silico in the search for new metamaterials with specific desired properties. An important conclusion of the proposed approach is that negative constitutive parameters can also be achieved by using suspensions of particles with random microstructures with properties similar to those shown in periodic arrays of microstructures.

¹ Author to whom any correspondence should be addressed.
1. Introduction

The recent interest in effective material properties, especially for the effective mass density \( \rho_{\text{eff}} \), comes from the study of metamaterials [1]. Metamaterials are artificially fabricated structures (often periodic) which are designed so that they display unusual macroscopic properties. For example, various engineered materials with negative effective density and/or negative bulk modulus have been demonstrated [2–4]. Applications of these metamaterials include improved acoustic imaging [5, 6] and sound wave manipulation for cloaking [7, 8]. Most of these approaches rely on resonant inclusions and the resulting acoustic metamaterial parameters vary strongly with frequency. The prospect of modelling effective material properties of composites with microstructure brings about the need for fast and accurate computational schemes to test ideas in silico.

Here, our main concern is the dynamic acoustic properties of a mixture of spherical particles suspended in a non-viscous fluid. In order to estimate the effective mass density of fluid–particle mixtures, various different approaches are possible. The simplest starting point is the Archimedes principle in which the effective mass density of a two-phase composite (fluid matrix of density \( \rho_0 \) and inclusions of density \( \rho \)) is given by the volume average as

\[
\rho_{\text{eff}} = (1 - \phi) \rho_0 + \phi \rho,
\]  

(1)
where $\phi$ is the volume fraction occupied by the inclusions. A similar formula can be used to calculate the effective bulk modulus $K_{\text{eff}}$. The inherent assumption in definition (1) is that $\rho_{\text{eff}}$ is insensitive to the relative motion of the phases of the composite.

In some circumstances however, a different estimate for $\rho_{\text{eff}}$ is obtained. The reason is that regions of different phases do not necessarily move in unison, even in the long-wavelength limit, and therefore the local average momentum is not simply the product of the local averaged mass and sound speed. Ament [10] presented another expression, known as the ‘Ament estimate’, for the effective mass density that differs significantly from the intuitive mixture law. In the absence of viscosity, his estimate becomes

$$\frac{\rho_{\text{eff}}}{\rho_0} = \frac{1 + \phi \mathcal{D}}{1 - 2\phi \mathcal{D}}, \quad \mathcal{D} = \frac{\rho - \rho_0}{\rho_0 + 2\rho}. \tag{2}$$

Fikioris and Waterman [11] have provided an independent analytical verification of equation (2) in the long-wavelength limit using a multiple-scattering formulation. The same result was also obtained later by Kuster and Tok Lossz [12] and Guanaurd and Wertman [13] using different methodologies. The effective mass density would satisfy the mixture law (1) if the matrix were an elastic medium. The expression (2) results from the inertia effects [14] which occur when the matrix is a fluid: the fluid can slide past the particle and induce an effect of added mass. Sound speed estimates based on equation (2) have also been shown to agree with experiments [15]. For small $\phi \mathcal{D}$, formula (2) is approximated as

$$\frac{\rho_{\text{eff}}}{\rho_0} \simeq 1 + 3\phi \mathcal{D} + 6\phi^2 \mathcal{D}^2 + O(\phi^3 \mathcal{D}^3). \tag{3}$$

Berryman [17] considered a ‘self-consistent’ version of equation (2) resulting in a quadratic equation for $\rho_{\text{eff}}$,

$$\frac{\rho_{\text{eff}} - \rho_0}{3\rho_{\text{eff}}} = \frac{\phi}{\rho_{\text{eff}} + 2\rho}, \tag{4}$$

which can be compared with equation (2). For small $\phi \mathcal{D}$, equation (4) is approximated as

$$\frac{\rho_{\text{eff}}}{\rho_0} \simeq 1 + 3\phi \mathcal{D} + 6\phi^2 \mathcal{D}^2 + \frac{3\rho}{\rho_0 + 2\rho} + O(\phi^3 \mathcal{D}^3), \tag{5}$$

which differs from equation (3) in the quadratic order.

It is of interest to extend the Ament estimate (2) to finite frequencies and wavelength. In this paper, a comprehensive description of acoustic scattering by multiple spheres using a multiple-scattering formulation is presented. It is based on the approach developed by Fikioris and Waterman [11]. This formulation describes a mixture of spherical particles suspended in a non-viscous fluid as a linearly viscoelastic material, whose elastic parameters with respect to the coherent acoustic wave motion are spatially constant but frequency dispersive and complex valued. Based on this approach (see section 5), the effective mass density depends on the volume fraction $\phi$ and on the scattering dispersion parameter $\tilde{\omega} = ka$, where $k$ is the wavenumber in the fluid matrix and $a$ is the particle radius. This dependence is later shown in section 5 to be as follows:

$$\frac{\rho_{\text{eff}}(\tilde{\omega})}{\rho_0} = \frac{1 + \phi \mathcal{D}}{1 - 2\phi \mathcal{D}} + i\frac{\phi \mathcal{D}^2 \tilde{\omega}^3}{(1 - 2\phi \mathcal{D})^3} + r(\tilde{\omega}), \tag{6}$$

where $i$ is the imaginary unit, and $r(\tilde{\omega})$ is a certain complex-valued function, which tends to zero along with its first derivatives at $\tilde{\omega} = 0$. The real part of equation (6) is recognized as the Ament estimate (2).
Estimating the effective material properties of inhomogeneous composites has been a subject of scientific and engineering interest with a long history. So far, many approaches and predictive schemes have been proposed. Early works were limited to either static or very-long-wavelength approximations, e.g. [10, 12, 17]. More recently, frequency-dependent descriptions have emerged, e.g. [22–26, 28]. Some schemes include the self-consistent and effective medium methods for which a substantial body of literature may be found. Although variants exist, these methods often consider the scattering problem of a coated particle in an effective medium. The coated particle consists of a layered particle of inner radius $a$ and a concentric layer of background material with an outer radius $b$. The actual boundary condition of the problem is imposed on $r = a$; within the coating, the wavenumber is $k$; outside the coating ($r > b$), the wavenumber is $\kappa$ (which is unknown). Transmission conditions are imposed across $r = b$, and the exciting field is a plane wave travelling with the wavenumber $\kappa$. This leads to what Twersky [18] called the ‘schizoid’ or ‘two-space’ problem (see also [19]). Given $b$ and $\kappa$ the coated-particle problem can be solved exactly. Common choices for the radius $b$ are $b = a$ (no coating) or such that $(a/b)^3 = \phi$, the volume fraction occupied by the particles. The effective medium parameters are determined by applying the condition that total scattering vanishes in the limit when $\kappa b \ll 1$. This approach allows determination of the effective mass density; however, the result is formally restricted to the low-frequency and long-wave ranges (see, e.g., [20] and references therein). Using only the leading-order terms, this estimate for the effective mass density is reduced to equation (6), as is shown in the remainder of the paper. It is necessary to note that a deficiency of all current versions of the effective medium methods is their failure to describe the influence of the spatial distribution of particles on the effective constitutive parameters. Such a description is possible in the framework of a self-consistent scheme called the effective field method (see, e.g., [37]) and our work is within the framework of this scheme.

The effective material properties of periodic acoustic structures can be easily calculated in the long-wavelength limit (see, e.g., [21, 69]). The periodicity means that the equation of motion can be solved exactly. For random microstructures however, this equation can be solved exactly only for a small number of particles. In order to simplify the problem of solving the equation of motion in the presence of multiple particles, an approximation which neglects the finite size of the particle is customary. Several authors, for example, have treated a random suspension as a mixture of particles distributed in a fluid and the effective wavenumber $\kappa$ has been calculated explicitly [9, 22, 26, 27]. This approach yields a simple formula for the effective mass density $\rho_{\text{eff}}$, which is linear in volume fraction [28]. At higher concentrations, however, this formula is unjustified and leads to incorrect results. This point is further discussed in the present paper.

Acoustic metamaterials can be made by setting up a periodic distribution of resonant particles in a fluid [29]. When the particles are arranged in structures having a square or hexagonal symmetry, the resulting system effectively behaves like a homogeneous and isotropic fluid. This behaviour is obtained when the wavelength of the propagating acoustic wave is larger than about a few times the lattice separation between individual scatterers. Materials behaving with an effective anisotropic mass density can also be engineered by arranging particles on non-symmetric lattices [30]. Acoustic parameters depending on the local coordinates can be achieved by simply changing the dimensions of the particles [31]. We show in the present paper that acoustic metamaterials can also be engineered by using suspensions of particles with random microstructures.

The paper proceeds as follows. The problem is formulated in section 2 where the multiple-scattering theory is outlined. In section 3, an implicit form of the wavenumber equation is
obtained using matrix notation. Although closed-form asymptotics of the effective wavenumber can be derived with the desired accuracy [33], the remainder of section 3 corroborates results for the effective wavenumber accurate to second order in volume fraction $\phi$ for finite-size or point-like particles. The main results of the paper are then derived in sections 4 and 5. Based on the above-mentioned matrix formulation and the asymptotic approximations of section 3, the reflection coefficient for the random array of spheres is obtained in section 4. Using the ansatz that the sought effective medium is described acoustically by the effective wavenumber and the reflection coefficient for the associated semi-infinite suspension of particles, the corresponding effective material properties are derived in section 5. A significant outcome of the proposed approach is that explicit expressions are obtained for the effective material parameters. Although the statement of the problem was completed by employing a specific pair-correlation function, the entire analysis is generalized to arbitrary pair-correlation functions in section 6. The possibility of designing random fluid–particle mixtures to form acoustic metamaterials is discussed in section 7. Low-frequency, long-wavelength expansions of the effective parameters are also derived in the appendices.

2. Background

Spherical polar coordinates $(r, \theta, \varphi)$ are defined from the origin $O$, so that a typical point $(x, y, z)$ has position vector $\mathbf{r} = r\hat{r} = r(\sin \theta \cos \varphi, \sin \theta \sin \varphi, \cos \theta)$, where $r = |\mathbf{r}|$. We then define local spherical polar coordinates $(\rho_j, \theta_j, \varphi_j)$ at $O_j$, so that $\mathbf{r} = \rho_j + \mathbf{r}_j$ and $\rho_j = \rho_j \hat{\rho}_j = \rho_j(\sin \theta_j \cos \varphi_j, \sin \theta_j \sin \varphi_j, \cos \theta_j)$, $\rho_j = |\rho_j|$. We assume that $\theta_j = 0$ is in the $z$-direction.

We consider a semi-infinite suspension of particles in a non-viscous fluid of density $\rho_0$, bulk modulus $\kappa_0$ and sound speed $c_0$. The spheres are of identical geometry and elastic properties and uniformly and randomly distributed in the half-space $z > 0$, where their number density is $n_0$.

A plane wave is incident on the half-space and propagates in the $z$-direction, as shown in figure 1. Since viscosity is neglected, only pressure waves can propagate in the fluid. These waves cause the fluid particles to move in the $z$-direction. The dynamics of the current multiple-scattering problem can be expressed in terms of appropriate scalar potentials. If the time
dependence is of the form \( \exp(-i\omega t) \), the displacement potential \( \phi^{in} \) is given by

\[
\phi^{in}(\mathbf{r}) = \phi_0 \exp(ikz), \quad k = \omega/c_0, \tag{7}
\]

where \( \phi_0 \) is an amplitude factor, \( \omega \) is the angular frequency, and the time dependence has been omitted for brevity.

We assume that the scattering properties of an isolated sphere are fully described by a set of coefficients \( T_n \), which are known. Therefore, the transition operator \( T \) for every sphere is assumed to have translational invariance with

\[
T(\mathbf{r})\phi^{in}(\mathbf{r}) = \phi_0 \sum_{n=0}^{\infty} i^n(2n + 1)T_n \psi_n(\mathbf{r}), \tag{8}
\]

where \( \psi_n(\mathbf{r}) = h_n(kr)P_n(\cos \theta) \) is an outgoing spherical wavefunction, \( h_n \equiv h^{(1)}_n \) is a spherical Hankel function, and \( P_n \) is a Legendre polynomial. Observe in equation (8) that there is no dependence on the azimuthal angle \( \phi \), owing to the axial symmetry of the sphere.

The far-field scattering amplitude of an isolated sphere is then given by

\[
T(\mathbf{r})\phi^{in}(\mathbf{r}) \simeq \frac{1}{kr} \exp(ikr) f(\theta) \quad (r \to \infty). \tag{9}
\]

The angular shape function \( f(\theta) \) is an infinite series with coefficients equal to the scattering amplitudes \( T_n \), i.e.

\[
f(\theta) = \frac{1}{ik} \sum_{n=0}^{\infty} (2n + 1)T_n P_n(\cos \theta). \tag{10}
\]

Note that \( f(0) \) and \( f(\pi) \) are measures, in the forward and backward directions, of the displacement potential due to scattering by a single sphere.

The incident wave (7) is subjected to multiple scattering between the spheres. For any given configuration (ensemble) of a finite number of spheres, the scattered motion may be computed exactly by solving the appropriate system of equations. However, for the case in hand the distribution of spheres is known in a probabilistic sense only. Thus, it is common to regard the volume containing the spheres as a random medium with certain average (homogenized) properties. We perform ensemble averaging [19, 22, 34, 35] in order to calculate the average (coherent) motion. There exist a number of approaches to solving such problems; a survey can be found, for example, in [36]. The method of effective fields [37] is used in our work. This allows us to take into account the spatial distributions of particles via specific pair-correlation functions \( g(\rho_{ji}) \). For simplicity, the spatial distribution of particles is assumed homogeneous and isotropic, and the ‘hole correction’ (that prevents the overlapping of the particles in the averaging process) [11] is adopted in the following, for which,

\[
g(\rho_{ji}) = H(\rho_{ji} - b), \tag{11}
\]

where \( H \) is the Heaviside unit function: \( H(x) = 1 \) for \( x > 0 \) and \( H(x) = 0 \) for \( x < 0 \). The hole radius \( b \) satisfies \( b \geq 2a \) so that spheres are not allowed to overlap. More complex forms of pair-correlation functions are considered in [33]. Appendix A outlines the relevant multiple-scattering theory.

Subject to the special case of the hole-correction (11), the subsequent analysis provides a comprehensive description of the coherent wave propagation in fluid–particle mixtures in terms of two distinct scattering processes: single and multiple scattering. A generalization of the main results to arbitrary pair-correlation functions is also provided in section 6.
3. The wavenumber matrix and the associated eigenvector

The quasi-crystalline approximation [32] allows us to develop the dispersion equation for the effective wavenumber $\kappa$ of the coherent acoustic field in the fluid–particle mixture. This equation follows from equation (A.7a) and can be written in matrix form [33]

$$\begin{bmatrix} I - \varepsilon R Q - \varepsilon e e' / \xi Q \end{bmatrix} x = 0,$$

(12)

where $e = (1, 1, \ldots)'$ is a constant vector, $I$ is the identity matrix and the scalars $\varepsilon$ and $\xi$ are

$$\xi = \kappa^2 - k^2, \quad \varepsilon = 4\pi n_0.$$  

(13)

The infinite square matrices $R$ and $Q$ have elements

$$R_{n\nu} = \sum_p G(0, v|0, n|p) R_p(\kappa b),$$

(14)

$$Q_{n\nu} = \frac{1}{ik} \tilde{T}_v \delta_{n\nu},$$

(15)

where

$$R_p(\kappa b) = \frac{1}{\xi} [\mathcal{M}_p(\kappa b) - 1]$$

(16)

and

$$\tilde{T}_v = (2v + 1) T_v.$$  

(17)

In equation (14), $G$ is a Gaunt coefficient [38] and the summation over $p$ is finite, covering the range $|n - \nu|$ to $(n + \nu)$ in steps of 2, so that $(p + n + \nu)$ is even.

A solution $x \neq 0$ can only exist for the particular value of $\xi$ that makes the matrix premultiplying $x$ in equation (12) singular. We then obtain an implicit equation for the effective wavenumber $\kappa$,

$$\kappa^2 = k^2 + \varepsilon \mathcal{F}(\kappa),$$  

(18)

where the effective scattering amplitude $\mathcal{F}$ is given by [33]

$$\mathcal{F}(\kappa) = e' \left( Q^{-1} - \varepsilon R \right)^{-1} e.$$  

(19)

Equation (18) is an exact expression for the effective wavenumber. (The only assumption made to arrive at this result is the quasi-crystalline approximation). Note that the formula (19) separates the implicit form of the effective wavenumber into two distinct parts. One part is defined by the scattering matrix $Q$, which describes the response of a single particle to a plane incident harmonic wave. The other part, $R$, is defined by the spatial arrangements of the particles and accounts for multiple scattering. Note that for a regular array of particles, the quasi-crystalline approximation is exact, in which case the multiple-scattering matrix $R$ can be reduced to a known lattice sum [39].

The infinite eigenvector $x$ of amplitudes $X_n$, associated with the wavenumber equation (18), follows from equation (A.7b) of appendix A and equation (12) as

$$x = \frac{2k}{\kappa^2 + k} (I - \varepsilon R Q)^{-1} e.$$  

(20)
The eigenvector \( \mathbf{x} \) will be useful later in the analysis for calculating the reflection coefficient for the coherent wave reflected at the interface \( z = 0 \).

Equations (18) and (20) are the most convenient starting point for the calculation of the effective parameters of the fluid–particle mixture. We will now study some important specific cases to render the wavenumber equation (18) more tractable.

3.1. Asymptotic expansion

At low concentration \( (\varepsilon a^3 \ll 1) \), we assume an asymptotic expansion in powers of \( \varepsilon \) as follows:

\[
K^2 = k^2 + \varepsilon \xi_1 + \varepsilon^2 \xi_2 + \cdots .
\]

Following the procedure detailed by Caleap et al [33], the coefficients in the expansion (21) are obtained as

\[
\xi_1 = e' Qe, \tag{22a}
\]

\[
\xi_2 = e' Q R_0 Q e, \tag{22b}
\]

where \( R_0 = R(0) \). The elements of the matrix \( R_0 \) are then found by expanding the function \( R_p(kb) \) in equation (16) for small \( (kb - kb) \). One finds that

\[
R_{0n}(kb) = \sum_p \mathcal{G}(0, v|0, n|p) R_p^{(0)}(kb), \tag{23}
\]

where

\[
R_p^{(0)}(x) = \frac{i b^2}{2x} \left\{ p(p + 1) - x^2 \right\} h_p(x) j_p(x) - x \left[ x \frac{d}{dx} h_p(x) + h_p(x) \right] \frac{d}{dx} j_p(x) \right\}. \tag{24}
\]

In terms of the Fourier series, the coefficients in equations (22) may be written as

\[
\xi_1 = \frac{1}{ik} \sum_v \tilde{T}_v, \tag{25a}
\]

\[
\xi_2 = \frac{1}{(ik)^2} \sum_{v,n} \tilde{T}_v \tilde{T}_n \tilde{R}_{0v}(kb). \tag{25b}
\]

The results in classical multiple scattering theories are usually defined in terms of the angular shape function \( f(\theta) \) for the scattering of a plane wave by a single particle. By using the definition of the Gaunt coefficients (A.10) and the angular shape function (10), equations (22) (or (25)) may be expressed as

\[
\xi_1 = f(0), \tag{26a}
\]

\[
\xi_2 = \frac{1}{2} \int_0^\pi d\theta \sin \theta \mathcal{H}_0(\theta) S(\theta), \tag{26b}
\]

where

\[
S(\theta) = \sum_p (2p + 1) P_p(\cos \theta) R_p^{(0)}(kb) \tag{27}
\]
and
\[ H_0(\theta) = [f(\theta)]^2. \] (28)

It is useful to describe yet another form of the coefficients (26). This is done in the next section, which considers point-particle approximation: in addition to \( \varepsilon a^3 \ll 1 \), we also require \( kb \ll 1 \).

3.2. Point particles: the small \( kb \) limit

The leading order term in \( kb \) of equation (A.8) is \( M_p(kb) \approx (\kappa/k)^p \). Then, it follows from equation (16) that
\[ R^{(0)}_P \simeq \frac{P}{2k^2}. \] (29)

By means of these approximations, equation (27) simplifies. Then, by using the generating function for the Legendre polynomials,
\[ S(\theta) = \frac{1}{2k^2} \left( 2 \frac{\partial^2}{\partial z^2} + 3 \frac{\partial}{\partial z} \right) \left( 1 - 2z \cos \theta + z^2 \right)^{-1/2} \bigg|_{z=1}, \] (30)
we obtain
\[ \kappa^2 = k^2 + 4\pi n_0 f(0) + \left( \frac{2\pi n_0}{k} \right)^2 [H_0(\pi) - H_0(0) - I_0] + O(n_0^3) \quad (kb \ll 1), \] (31)
where
\[ I_0 = \int_0^\pi d\theta \frac{1}{\sin(\theta/2)} \frac{d}{d\theta} H_0(\theta). \] (32)

The result in equation (31) is accurate to second order in terms of the concentration expansion term. Note that the first two terms of equation (31) give the thin slab result of Fermi [42] who considered the propagation of neutrons through a thin sheet of scatterers. Lloyd and Berry [43] have applied the multiple scattering treatment of Lloyd [44, 45] (developed for the treatment of scattering in electronic structures of liquids and disordered alloys) and obtained precisely the result in equation (31). Equation (31) defines what is called the ‘Lloyd–Berry estimate’ in the remainder.

4. The reflection coefficient

The interaction of the incident wave (7) with the scatterers causes it to be reflected. The reflection coefficient represents the average back-scattered amplitude in the domain \( \{ z < 0 \} \). As shown in [11], equation (A.1) (with \( n(r_j) = n_0 \)) yields for the reflected field
\[ \langle \phi(r) \rangle = \mathcal{R} \exp(-ik z) \quad (z < 0), \] (33)
where the reflection coefficient \( \mathcal{R} \) is defined as
\[ \mathcal{R} = -\frac{\varepsilon}{2k(\kappa + k)} \mathcal{F}_\pi, \quad \mathcal{F}_\pi = \frac{1}{ik} \sum_{n=0}^\infty (-1)^n (2n + 1) T_n X_n. \] (34)
As expected, \( \mathcal{R} \) depends on the infinite eigenvector of amplitudes \( X_n \).
Using the notation from section 3, equation (A.7b) may be written as
\[ \varepsilon \frac{K + k}{2K} e^{iQx} = 1. \]  
(35)

Once the eigenvector \( x \) is determined, the effective forward- and back-scattered amplitudes, \( F_0 \) and \( F_\pi \), follow from equations (A.7b) and (34), respectively, which can be recast in matrix form as
\[ F_0 = e^{iQx} \quad \text{and} \quad F_\pi = e^{iQx}, \]  
(36)

where the additional infinite matrix \( J \) has elements \( J_{\nu\nu} = \cos \nu \pi \). Then, the reflection coefficient is obtained from
\[ R = -\frac{\varepsilon F_\pi}{4k^2 + \varepsilon F_0}. \]  
(37)

Equation (37) is an exact expression for the reflection coefficient.

As in section 3.1, we consider next the low-concentration asymptotic approximation. We seek an asymptotic expansion of \( R \) in powers of the parameter \( \varepsilon \):
\[ R = R_0 + \varepsilon R_1 + \cdots. \]  
(38)

Using the form (20) in equations (36) and (37), the individual terms in (38) may be obtained as
\[ R_0 = -\frac{\varepsilon}{4k^2} e^{iJQe}, \]  
(39a)
\[ R_1 = -\frac{\varepsilon}{8k^4} \left[ 2k^2 e^{iJQR_0} - (e^{iQe})(e^{iQe}) \right]. \]  
(39b)

Subsequent terms become more complicated but the procedure for finding them is straightforward.

An alternative expression to equation (38) can be obtained in terms of Fourier series, i.e.
\[ R = \frac{\varepsilon}{4k^3} \sum_v (-1)^v \tilde{T}_v + \frac{\varepsilon^2}{8k^6} \sum_{v,\pi} (-1)^v \tilde{T}_v \tilde{T}_\pi \left[ \tilde{R}_{00}(kb) - 1 \right] + O(\varepsilon^3). \]  
(40)

By using the definition of the Gaunt coefficients (A.10) and the angular shape function (10), equation (40) may also be expressed
\[ R = -\frac{\varepsilon}{4k^2} f(\pi) + \frac{\varepsilon^2}{8k^4} \left[ \mathcal{H}_\pi(0) - k^2 \int_0^\pi d\theta \sin \theta \mathcal{H}_\pi(\theta) S(\theta) \right] + O(\varepsilon^3), \]  
(41)
where the function \( S \) is defined in equation (27), and
\[ \mathcal{H}_\pi(\theta) = \mathcal{H}_\pi(\pi - \theta) = f(\theta) f(\pi - \theta). \]  
(42)

To conclude this section, we give the point-particle approximation of the low-concentration expansion (41). In addition to \( \varepsilon a^3 \ll 1 \), we also require \( kb \ll 1 \). We use the same procedure as that described in section 3.2. In terms of \( n_0 \), we obtain
\[ R = -\frac{\pi n_0}{k^2} f(\pi) + \left( \frac{\pi n_0}{k^2} \right)^2 [2\mathcal{H}_\pi(0) + \mathcal{I}_\pi] + O(n_0^3) \quad (kb \ll 1), \]  
(43)
where
\[
I_\pi = \int_0^\pi d\theta \frac{1}{\sin(\theta/2)} \frac{d}{d\theta} \mathcal{H}_\pi(\theta) .
\] (44)

Based on the knowledge of the effective wavenumber and the reflection coefficient of the associated semi-infinite suspension of particles in a fluid, we calculate next the effective dynamic properties of the fluid–particle mixture.

5. Effective dynamic constitutive parameters

In this section, we extend the effective mass density expressions (2) and (3) to finite frequencies and wavelength. Specifically, we show that a frequency-dependent effective mass density can be obtained from the reflection coefficient of the previous section, which reduces to equations (2) and (3) in the long-wavelength limit and which can yield complex values. The bulk modulus \( \kappa_{\text{eff}} \) of the fluid composite is also found to be complex-valued and frequency-dependent. Note that energy dissipation is induced only by scattering between multiple scatterers. Therefore, the imaginary part of the effective parameters is only due to the diffusive scattering loss.

5.1. Effective interface conditions

The effective medium corresponding to a suspension of particles in a fluid can be described as a linearly viscoelastic fluid from the standpoint of coherent wave propagation (see, e.g., [28, 46]). The viscoelastic fluid has mass density \( \rho_{\text{eff}} \) and coherent acoustic waves propagate with the effective wavenumber \( K \) of equation (18). The reflection coefficient at the interface between an inviscid fluid and a viscoelastic fluid may then be written as
\[
R = \frac{\rho_{\text{eff}} k - \rho_0 K}{\rho_{\text{eff}} k + \rho_0 K} .
\] (45)

Observe that \( \rho_{\text{eff}} k / \rho_0 K \) is the ratio of the acoustic impedance of the viscoelastic fluid to that of the lossless surrounding fluid. Equation (45) is obtained by imposing continuity of pressure and normal velocity at the interface. Fikioris and Waterman [11] showed that these continuity conditions are justified, at least in the low-frequency limit, and effective parameters can be determined. (Note that pressure is given by the product of a scalar potential with density). A similar approach has been used in [28, 46, 47] in the framework of Waterman and Truell [26]. Effective interface conditions at the boundary of the half-space can also be derived in the analytical framework of Fikioris–Waterman (see [48] for two-dimensional (2D) problems).

5.2. Effective mass density and bulk modulus

Equating the reflection coefficients of equations (37) and (45), and using equation (A.7b), one finds that
\[
\frac{\rho_{\text{eff}}}{\rho_0} = \frac{1}{2k^2} \frac{4k^2 + \varepsilon (F_0 - F_\pi)}{4k^2 + \varepsilon (F_0 + F_\pi)} (2k^2 + \varepsilon F_0) .
\] (46)

Equation (46) is the generalization of the Ament estimate for the effective mass density. It reduces exactly to equation (2) in the long-wavelength limit (see equation (B.13) in appendix B).
If one defines the effective bulk modulus $\kappa_{\text{eff}}$ by

$$\frac{\kappa_{\text{eff}}}{\kappa_0} = \frac{\rho_{\text{eff}} k^2}{\rho_0 K^2},$$

(47)

where the effective wavenumber $K$ is given by equation (18) and the effective mass density $\rho_{\text{eff}}$ by equation (46), then $\kappa_{\text{eff}}$ is given by

$$\frac{\kappa_{\text{eff}}}{\kappa_0} = \frac{4k^2 + \varepsilon (F_0 - F_\pi)}{4k^2 + \varepsilon (F_0 + F_\pi)} \frac{2k^2}{2k^2 + \varepsilon F_0}.$$  

(48)

Once the effective scattering amplitudes $F_0$ and $F_\pi$ are determined from equations (36), the effective mass density and bulk modulus follow from equations (46) and (48).

Note that the low-frequency approximations of the effective mass density and bulk modulus are derived in appendix B, equations (B.5) and (B.6), respectively. Li and Chan [54] have derived the same formulae for $\rho_{\text{eff}}$ and $\kappa_{\text{eff}}$ by using an effective medium method (i.e. the coherent potential approximation). They showed that it becomes possible to achieve negative effective bulk modulus and negative effective mass density through resonance behaviour of the scattering coefficients $T_0(\omega)$ and $T_1(\omega)$. Structures are designed in which the resonances occur at very low frequency using silicone rubber spherical particles in water. Our general results for $\rho_{\text{eff}}$ and $\kappa_{\text{eff}}$, equations (46) and (48), extend this previous analysis to include as many diffraction orders (monopole, dipole, quadrupole and so on) as needed for convergence, and are not restricted to the low-frequency range.

Note that equations (46) and (48) are the basis of two different closed-form formulae for effective parameters to be derived in the remainder of this section, depending on the level of approximation (i.e. low concentration and/or point particles).

5.3. Explicit formulae for $\rho_{\text{eff}}(\omega)$ and $\kappa_{\text{eff}}(\omega)$

Here we combine the results of the previous two sections to give explicit formulae for the effective parameters $\rho_{\text{eff}}$ and $\kappa_{\text{eff}}$ valid in two limits: low concentration and point particles, respectively. First the low-concentration asymptotic approximation is considered. From equation (46), one obtains for the effective mass density

$$\frac{\rho_{\text{eff}}}{\rho_0} \simeq 1 + \tilde{\rho}_1 \frac{\varepsilon}{2k^2} + \tilde{\rho}_2 \frac{\varepsilon^2}{2k^2} + \mathcal{O}(\varepsilon^3),$$

(49)

where the coefficients $\tilde{\rho}_1$ and $\tilde{\rho}_2$ are given in matrix notation by

$$\tilde{\rho}_1 = e' Qe - e' JQe,$$

(50a)

$$\tilde{\rho}_2 = e' Q R_0 Qe - e' J Q R_0 Qe - \frac{1}{4k^2} \left[ (e' Qe)^2 - (e' JQe)^2 \right].$$

(50b)

In terms of the angular shape function $f(\theta)$ the coefficients (50) may be expressed as

$$\tilde{\rho}_1 = f(0) - f(\pi),$$

(51a)

$$\tilde{\rho}_2 = \frac{1}{2} \int_0^\pi d\theta \sin \theta \left[ H_0(\theta) - H_\pi(\theta) \right] S(\theta) - \frac{1}{4k^2} \left[ H_0(0) - H_\pi(0) \right].$$

(51b)
By combining the expansions in $\varepsilon$ of equations (21) and (49) with equation (47), the effective bulk modulus becomes

$$
\frac{\kappa_{\text{eff}}}{\kappa_0} = 1 + 2\frac{k^2 + \rho_1 \varepsilon + \rho_2 \varepsilon^2}{k^2 + \xi_1 \varepsilon + \xi_2 \varepsilon^2},
$$

$$
\simeq 1 + \frac{\varepsilon}{2k^2} (\rho_1 - 2\xi_1) + \frac{\varepsilon^2}{2k^2} \left[ (\rho_2 - 2\xi_2) - \frac{1}{k^2} (\rho_1 - 2\xi_1) \xi_1 \right] + \mathcal{O}(\varepsilon^3),
$$

(52a)

(52b)

where $\xi_1$ and $\xi_2$ are given by equations (22) (or (26)).

We now consider the point-particle limit. In addition to $\varepsilon a^3 \ll 1$, we also require $kb \ll 1$. We use the same procedure as that described in the previous sections. Equations (49) and (52) become, in terms of $n_0$,

$$
\frac{\rho_{\text{eff}}}{\rho_0} \simeq 1 + \frac{2\pi n_0}{k^2} \left[ f(0) - f(\pi) \right] + \frac{(2\pi n_0)^2}{k^4} \left[ \mathcal{H}_0(\pi) - \mathcal{H}_0(0) - \frac{1}{2} (I_0 - I_\pi) \right]
$$

$$
+ \mathcal{O}(n_0^3) \quad (kb \ll 1),
$$

(53)

$$
\frac{\kappa_{\text{eff}}}{\kappa_0} = \frac{\left\{ k^2 + 2\pi n_0 \left[ f(0) - f(\pi) \right] \right\} k^2 + (2\pi n_0)^2 \left[ \mathcal{H}_0(\pi) - \mathcal{H}_0(0) - \frac{1}{2} (I_0 - I_\pi) \right]}{\left[ k^2 + 4\pi n_0 f(0) \right] k^2 + (2\pi n_0)^2 \left[ \mathcal{H}_0(\pi) - \mathcal{H}_0(0) - I_0 \right]} \quad (kb \ll 1),
$$

(54a)

$$
\simeq 1 - \frac{2\pi n_0}{k^2} \left[ f(0) + f(\pi) \right] + \frac{(2\pi n_0)^2}{k^4} \left[ 2\mathcal{H}_0(0) + 2\mathcal{H}_\pi(0) + \frac{1}{2} (I_0 + I_\pi) \right] + \mathcal{O}(n_0^3).
$$

(54b)

The closed-form formulae (53) and (54) are direct consequences of the Lloyd–Berry estimate (31) for the effective wavenumber and the reflection coefficient (43) for the associated semi-infinite suspension of particles, which are both correct to quadratic terms in the concentration. Note that equation (49) (or (53)) agrees precisely with the small-$\phi$ estimate of equation (3) in the long-wavelength limit (see equation (B.21) in appendix B). This agreement provides a further check on the calculations. A comparison with previous work is also made in appendix C.

Apart from their dependence on $k$ and $n_0$, the effective dynamic parameters given by (31), (43), (53) and (54) are all completely determined when the angular shape function $f(\theta)$ for an isolated particle is known. If this scattering amplitude can be determined analytically, numerically or experimentally, then the effective medium equivalent to the fluid–particle mixture is fully described.

It is worth noting that if one wants to study the behaviour of effective parameters at high concentrations (where such expansions are no longer appropriate), the general implicit equations (46) and (48) have to be used and/or more accurate pair-correlation functions have to be considered. In the following section, the entire analysis is generalized to account for arbitrary pair-correlation functions.

6. Generalization to an arbitrary pair-correlation function

The advantage of the quasi-crystalline approximation is that statistics, describing the spatial distribution of scatterers, can be incorporated into the effective field method. All the above
developments start with the general equations (18) and (20). They are both given explicitly in terms of the multiple-scattering matrix $\mathbf{R}$. Statement of the problem was completed by using the hole correction (11), for which the matrix $\mathbf{R}$ has elements given by equation (14). We shall now render our analysis more general; that is, an arbitrary pair-correlation function $g(r)$ (that satisfies the conditions (A.4)) is used to derive expressions for $\mathbf{K}$, $\mathbf{R}$, $\rho_{\text{eff}}$ and $\kappa_{\text{eff}}$.

Note first that the form of $\mathbf{R}^p$ in equation (16) is the result of hole correction. More generally, this function has values

$$
\mathbf{R}^p(\mathbf{K}b) = \frac{M_p(\mathbf{K}b) - 1}{\mathbf{K}^2 - \mathbf{K}^2} + \mathbf{i}k \int_b^\infty \mathbf{d}r \left[ g(r) - 1 \right] r^2 h_p(kr) j_p(\mathbf{K}r).
$$

(55)

The integral in equation (55) describes the statistics of particle locations in terms of an arbitrary pair-correlation function $g(r)$.

Next, in the limit of $\kappa \rightarrow k$, equation (55) gives

$$
\mathbf{R}^p_{(0)}(x) = \frac{i\mathbf{b}}{2k} \left\{ p(p+1) - x^2 \right\} h_p(x) j_p(x) - x \left[ x \mathbf{d}h_p(x) + h_p(x) \right] \mathbf{d}x j_p(x)
+ \mathbf{i}k \int_b^\infty dr \left[ g(r) - 1 \right] r^2 h_p(kr) j_p(kr), \quad x = kb.
$$

(56)

Finally, in the point-particle limit, equation (56) reduces to

$$
\mathbf{R}^p_{(0)}(kb) \simeq \frac{p}{2k^2} + \int_b^\infty dr \left[ g(r) - 1 \right] \left[ (2p+1)^{-1} + \delta_{p0}ikr \right] r \quad (kb \ll 1).
$$

(57)

The above equations enable us to calculate the effective dynamic parameters accounting for pair-correlated particles. If the simple hole correction (11) is used in equations (55)–(57), then equations (16), (24) and (29) are recovered.

To generalize equations (31), (43), (53) and (54) to an arbitrary pair-correlation function $g(r)$, one only needs to calculate $\mathbf{e}' \mathbf{Q} R_0 \mathbf{Q} \mathbf{e}$ and $\mathbf{e}' \mathbf{Q} J R_0 \mathbf{Q} \mathbf{e}$ in terms of the angular shape function $f(\theta)$. Combining equations (27) and (57) and using the generating function for Legendre polynomials, equation (30) is replaced with

$$
\mathcal{S}(\theta) = \frac{1}{2k^2} \left( 2 \frac{\partial^2}{\partial z^2} + 3 \frac{\partial}{\partial z} \right) \left[ 1 - 2z \cos \theta + z^2 \right]^{1/2} + \mathcal{N}^{(1)}(2 - 2 \cos \theta)^{-1/2} + ikn^{(2)},
$$

(58)

where

$$
\mathcal{N}^{(p)} = \int_b^\infty dr \left[ g(r) - 1 \right] r^p \quad (p = 1, 2).
$$

(59)

One obtains

$$
\mathbf{e}' \mathbf{Q} R_0 \mathbf{Q} \mathbf{e} \simeq \frac{1}{4k^2} \left[ \mathcal{H}_0(\pi) - \mathcal{H}_0(0) - \mathcal{I}_0 \right] + \mathcal{N}^{(1)} \left[ \mathcal{H}_0(\pi) - \mathcal{J}_0 \right] + \frac{1}{2}i kn^{(2)} \mathcal{E}_0,
$$

(60)

$$
\mathbf{e}' \mathbf{Q} J R_0 \mathbf{Q} \mathbf{e} \simeq -\frac{1}{4k^2} \mathcal{I}_\pi + \mathcal{N}^{(1)} \left[ \mathcal{H}_\pi(0) - \mathcal{J}_\pi \right] + \frac{1}{2}i kn^{(2)} \mathcal{E}_\pi,
$$

(61)

where

$$
\mathcal{J}_\gamma = \int_0^\pi \mathbf{d} \theta \sin(\theta/2) \frac{\mathbf{d}}{\mathbf{d} \theta} \mathcal{H}_\gamma(\theta), \quad \mathcal{E}_\gamma = \int_0^\pi \mathbf{d} \theta \sin \theta \mathcal{H}_\gamma(\theta) \quad (\gamma = 0, \pi).
$$

(62)
The effective wavenumber and the reflection coefficient are then obtained as
\[ k^2 \simeq k_0^2 + 4\pi n_0 f(0) + (2\pi n_0)^2 \left\{ \frac{1}{k^2} [H_0(\pi) - H_0(0) - J_0] + 4N_1 [H_0(\pi) - J_0] + 2ikN_2 \varepsilon_0 \right\} \],
(63)
\[ \Re \simeq -\frac{\pi n_0}{k^2} f(\pi) + \left( \frac{\pi n_0}{k} \right)^2 \left\{ \frac{1}{k^2} [2H_1(0) + I_\pi] - 4N_1 [H_1(0) - J_\pi] - 2ikN_2 \varepsilon_\pi \right\} . \]
(64)

Equations (63) and (64) are accurate to second order in concentration for point-like particles with an arbitrary two-point correlation function \( g(r) \). Similar expressions can be derived for the effective mass density and bulk modulus by using equations (60) and (61).

7. Numerical illustrations and discussion

In order to illustrate the general behaviour of different approximations for effective dynamic parameters, some numerical examples are presented. Before starting, note that in a previous paper [33] we have rigorously compared the three solutions for the effective wavenumber (one implicit: equation (18), and two explicit: equations (21) and (31)). We have also highlighted differences in the effective wavenumber due to the choice of the pair-correlation function. It was noted that the hole correction yielded some unphysical results in some cases, whereas the Virial expansion [55] and the Percus–Yevick results [56] were, in the cases considered, physically acceptable. In the following, the pair-correlation function \( g(r) \) satisfying the conditions (A.4) is chosen to be given by the Percus–Yevick relation (see [33]). Also, whenever the exclusion distance \( b \) (i.e. the distance of the closest approach between the centres of adjacent spheres) needs to be specified in the calculations, the value \( b = 2a \) is taken, which is thought to be physically reasonable.

We consider spherical particles of radius \( a \), mass density \( \rho \) and longitudinal and shear sound speeds, \( c_L \) and \( c_T \), respectively. Two different fluid–particle mixtures are considered: steel and polystyrene particles, both suspended in water\(^2\).

7.1. Reflection (transmission) from an effective layer of particles

Results are presented for the reflection coefficient \( \Re \) for a half-space of steel particles suspended in water. The reflection coefficient is calculated by using the general expression (37) and its two expansions in volume fraction \( \phi \): finite-size and point-like particles, equations (41) and (43), respectively. The comparison of these formulations is shown in figure 2, for two volume fractions \( \phi = 0.15 \) and \( \phi = 0.35 \). On the horizontal axis, the scattering dispersion parameter \( \tilde{\omega} \) is defined as \( \tilde{\omega} = ka = 2\pi a/\Lambda \), where \( \Lambda \) is the wavelength of the plane incident wave in water. Thus, for a value of \( \tilde{\omega} \simeq 1 \), the incident wavelength is approximately three times the diameter of the particle. It is therefore a fairly long wavelength. We observe from figure 2 that the values for the reflection coefficient predicted via the general expression (37) and the expansion (41) for finite-size particles are almost identical to each other, but differ slightly from the results based on the point-particle approximation (43), and this difference increases as the distribution becomes denser (\( \phi = 0.35 \)). The expansion (41) for finite-size particles is shown to

\(^2\) Parameter values for these materials are \( \rho_0 = 1 \text{ kg m}^{-3}, c_0 = 1.48 \text{ m ms}^{-1} \) for water; \( \rho = 7.932 \text{ kg m}^{-3}, c_L = 5.96 \text{ m ms}^{-1}, c_T = 3.26 \text{ m ms}^{-1} \) for steel; \( \rho = 1.06 \text{ kg m}^{-3}, c_L = 2.38 \text{ m ms}^{-1}, c_T = 1.18 \text{ m ms}^{-1} \) for polystyrene.
Figure 2. Modulus and phase of the reflection coefficient $\Re$ for a half-space of steel particles suspended in water versus the dimensionless frequency $\tilde{\omega}$, for two values of volume fraction $\phi = 0.15$ and 0.35. Solid lines represent results obtained with the general expression (37), dashed lines—with the expansion (41) for actual-size particles and dash-dotted lines—with the point-particle approximation (43). The Percus–Yevick result is used for the pair-correlation function.

be an excellent approximation of the true expression (37) even for quite dense distributions. As such, the expansion formulae of effective parameters for finite-size particles are exploited in the following.

The explicit dependence of the reflection coefficient (45) on the effective mass density $\rho_{\text{eff}}$ means that, in principle, measurement of $\Re$ via experiment can provide useful knowledge for its determination. Knowing the effective wavenumber $K$ and mass density $\rho_{\text{eff}}$, the effective acoustic impedance $Z$ can easily be calculated, such as

$$Z = \rho_{\text{eff}} \frac{\omega}{K} = \frac{z_0}{4k^2 + \varepsilon (F_0 - F_\pi)} \frac{4k^2 + \varepsilon (F_0 + F_\pi)}{4k^2 + \varepsilon (F_0 - F_\pi)}, \quad (65)$$

where $z_0 = \rho_0 \omega / k$ is the acoustic impedance of the inviscid fluid. With $Z$, one can describe acoustic wave propagation inside and outside a finite layer of particles suspended in a fluid by keeping track of the successive internal reflections from the boundaries of the equivalent homogeneous layer. There is an infinite number of reflections, which yields the reflection and transmission coefficients in the form of two infinite series. It is easy to interpret physically each term in the series. The two series converge, respectively, to the expressions [73]

$$R_h = \frac{\Re \left[ 1 - \exp(2iK\kappa) \right]}{1 - \Re^2 \exp(2iK\kappa)}, \quad T_h = \frac{(1 - \Re^2) \exp[i(K - k)\kappa]}{1 - \Re^2 \exp(2iK\kappa)}, \quad (66)$$
Figure 3. Moduli of the reflection and transmission coefficients corresponding to a fluid layer containing steel particles immersed in water versus the dimensionless effective wavelength \( \lambda_{\text{eff}} / h \), for two values of layer thickness \( h = a \) and \( h = 10a \). Comparison of four predictions, which correspond, respectively, to the four mass densities given by equations (1), (3), (5) and (49)—dotted, dashed, dash-dotted and solid lines. The volume fraction of particles is \( \phi = 0.35 \).

where \( h \) is the thickness of the layer and

\[
\rho = \lim_{h \to \infty} \rho_h = \frac{Z - z_0}{Z + z_0}
\]

is the reflection coefficient of the associated semi-infinite layer.

It is important to note that the solution of the multiple-scattering problem derived in this paper does not require a priori knowledge of an effective mass density or of an effective bulk modulus for the mixture of particles inside the fluid. Furthermore, no assumptions concerning these quantities are needed. There exist works, however, where such assumptions are made in order to evaluate reflection and transmission on either side of a finite layer [57–59]. The resulting effective mass densities are real valued and frequency independent. Figure 3 illustrates the importance of using a complex-valued frequency-dependent mass density as in equation (49). This figure represents the moduli of the reflection and transmission coefficients corresponding to a fluid layer containing steel particles immersed in water (see footnote 2). On the horizontal axis, the effective wavelength \( \lambda_{\text{eff}} \) of the plane coherent wave is defined as \( \lambda_{\text{eff}} = 2\pi / \text{Re} \kappa \). The volume fraction of particles is \( \phi = 0.35 \); two layer thicknesses are considered: \( h = a \) and \( h = 10a \). Four results are compared, which correspond, respectively, to the four mass densities given by equations (1), (3), (5) and (49)—the mixture law, Ament estimate, Berryman estimate and effective dynamic mass density. There are large differences for both the reflection and
transmission coefficients. This suggests that serious errors can result from using inappropriate expressions to model the effective mass density in experimental approaches. Only in the low-frequency limit does the mass density formula reduce to that corresponding to the Ament estimate.

7.2. Acoustic metamaterials with random microstructure

Acoustic metamaterials, in which either the mass density or the bulk modulus is negative, can be used, for example, to design acoustic lenses to overcome the diffraction limit [60, 61] or for the design of acoustic panels for sound insulation [62]. ‘Double negative’ acoustic metamaterials, in which both the mass density and the bulk modulus are negative, can cause negative refraction [63] and, as is known from electromagnetic wave theory, they can also be used to increase the resolution of conventional lenses [64, 65].

The existence of frequency ranges where the effective medium presents negative constitutive parameters is related to resonances of the individual scatterers that constitute the metamaterial, i.e. either soft-particle resonances (e.g. [54]) or Helmholtz-like resonances (e.g. [66]). It is known that the monopolar resonances in the individual particles are responsible for negative bulk modulus and that the dipolar resonances are responsible for negative mass density (see, e.g., [54, 69]). However, the full effect of the ensemble of scatterers that constitute the effective medium has only been explained partially, as higher diffraction orders (monopole, dipole, quadrupole and so on) are not included in the models; therefore such arguments are restricted only to the low-frequency range. In the low-frequency range, the effective medium is independent of whether the microstructure is random or periodic [21]. Not surprisingly, previous
Figure 5. Real and imaginary parts of the effective bulk modulus $\kappa_{\text{eff}}/\kappa_0$ ($= \tilde{\kappa}_\text{eff}' - i \tilde{\kappa}_\text{eff}''$) for polystyrene particles in water.

studies on acoustic metamaterials treated the microstructure as a regular lattice of scatterers, since for a periodic system, as opposed to a disordered one, the normal modes of the system may be easily obtained in the low-frequency limit (e.g. [67–69]).

In the following, we show that negative constitutive parameters can also be achieved by using suspensions of particles with random microstructures and are not restricted to the low-frequency, long-wave limit. Here we report only one example of particles to realize acoustic metamaterials, i.e. homogeneous spherical particles. Obviously, more complex particles (e.g. homogeneous particles but with spherical anisotropy, oblate/prolate ellipsoidal particles, etc) could be used to enhance the frequency response, but a full analysis of this type is beyond the scope of this work.

Figures 4 and 5 illustrate the effective mass density $\rho_{\text{eff}}/\rho_0$ ($= \tilde{\rho}_\text{eff}' + i \tilde{\rho}_\text{eff}''$) and bulk modulus $\kappa_{\text{eff}}/\kappa_0$ ($= \tilde{\kappa}_\text{eff}' - i \tilde{\kappa}_\text{eff}''$) corresponding to a suspension of polystyrene particles in water (see footnote 2). As can be seen, in particular frequency regions, both the effective mass density and bulk modulus yield negative values in the vicinity of local resonances of individual particles. Figure 6 displays a contour plot of these negative regions. Also shown in figure 6 are the resonant frequencies for an isolated polystyrene sphere in water. Each resonance may be labelled by two symbols ($n, \ell$) in the Regge trajectories, where the first index defines the ordinal number of the resonance and the second indicates the family of surface waves. The $\ell = 1$ family is usually referred to as the Rayleigh waves and the others (i.e. $\ell = 2, 3, \ldots$) are the Whispering Gallery waves. It is interesting to note that the negative regions for both $\tilde{\rho}_\text{eff}'$ and $\tilde{\kappa}_\text{eff}'$ are mainly due to the Rayleigh wave family of modes ($\ell = 1$). The first negative region for $\tilde{\kappa}_\text{eff}'$ is related to the $(n, \ell) = (2, 1)$ resonance, while for $\tilde{\rho}_\text{eff}'$ the first negative region is associated with the
Figure 6. Contour plot of negative regions of the effective mass density $\tilde{\rho}_{\text{eff}}'$ (blue curves) and bulk modulus $\tilde{\kappa}_{\text{eff}}'$ (red curves) shown in figures 4 and 5. Also shown in this figure are the resonant frequencies for an isolated polystyrene sphere in water.

Figure 7. Real and imaginary parts of the effective mass density $\rho_{\text{eff}}/\rho_0$ (blue curves) and bulk modulus $\kappa_{\text{eff}}/\kappa_0$ (red curves) versus the dimensionless frequency $\tilde{\omega}$, for values of volume fraction $\phi \in (0.345, 0.355)$. The effect of every diffraction order is clearly seen in this figure. In particular, note that in the frequency region where $\tilde{\omega} \in (2.16, 2.22)$ (which corresponds to the $(3, 1)$ resonance), both $\tilde{\rho}_{\text{eff}}'$ and $\tilde{\kappa}_{\text{eff}}'$ exhibit negative values simultaneously.

$(n, \ell) = (3, 1)$ resonance. Note from figure 6 that the monopolar $(n = 0)$ and dipolar $(n = 1)$ resonances occur in a higher-frequency range.

Figure 7 shows the behaviour of $\rho_{\text{eff}}/\rho_0$ and $\kappa_{\text{eff}}/\kappa_0$ versus the dimensionless frequency $\tilde{\omega}$, for values of volume fraction $\phi \in (0.345, 0.355)$. The effect of every diffraction order is clearly seen in this figure. In particular, note that in the frequency region where $\tilde{\omega} \in (2.16, 2.22)$ (which corresponds to the $(3, 1)$ resonance), both $\tilde{\rho}_{\text{eff}}'$ and $\tilde{\kappa}_{\text{eff}}'$ exhibit negative values simultaneously.
Figure 8. Real and imaginary parts of the effective mass density $\rho_{\text{eff}}/\rho_0$ (blue curves) and bulk modulus $\kappa_{\text{eff}}/\kappa_0$ (red curves) versus the volume fraction $\phi$, for values of the dimensionless frequency $\tilde{\omega} \in (2.16, 2.22)$.

In figure 8, the behaviour of $\rho_{\text{eff}}/\rho_0$ and $\kappa_{\text{eff}}/\kappa_0$ versus the volume fraction $\phi$ is presented for values of the dimensionless frequency $\tilde{\omega} \in (2.16, 2.22)$.

In summary, figures 4 and 5 display a few narrow regions in which the effective mass density and/or bulk modulus are negative. These regions depend on both the frequency and the concentration of the suspended particles. To enlarge the negative regions of both $\rho_{\text{eff}}$ and $\kappa_{\text{eff}}$, one could design the particles to broaden their resonant behaviour. For instance, considering heavier core particles, e.g. lead-cored polystyrene particles, will enhance the field oscillation in the polystyrene shell, which, in turn, will widen the resonant regions of $\rho_{\text{eff}}$. On the other hand, if one engineers the particles to make them easier to deform and compress [70], e.g. air-filled polystyrene particles, this will widen the resonant regions of $\kappa_{\text{eff}}$. The radius of the inner core of the particles is chosen such that the negative regions of both $\rho_{\text{eff}}$ and $\kappa_{\text{eff}}$ overlap. This and other applications will be exploited elsewhere.

8. Conclusion and perspectives

Using the multiple-scattering approach proposed by Fikioris and Waterman [11], we have determined the coherent wave motion in a non-viscous fluid containing disordered heterogeneities, such as particles, bubbles or contrast agents. Scatterers can be homogeneous spheres, layered, shell-like with encapsulated liquids or gas, non-absorbing, or absorbing. They are randomly distributed in a uniform half-space. Based on the quasi-crystalline approximation,
the fluid–particle mixture is then shown to behave as an effective dissipative medium from
the standpoint of the coherent wave motion. The effective medium is fully described once the
effective wavenumber and the reflection coefficient for the associated semi-infinite suspension
of particles are known. The effective mass density $\rho_{\text{eff}}$ and bulk modulus $\kappa_{\text{eff}}$ of the effective
medium are determined by solving the general equations (46) and (48). As expected, $\rho_{\text{eff}}$ and $\kappa_{\text{eff}}$
are complex valued and frequency dependent. The imaginary part of the effective parameters is
only due to the diffusive scattering loss. Equations (46) and (48) are the basis of two closed-form
formulae for $\rho_{\text{eff}}$ and $\kappa_{\text{eff}}$ valid in two limits: low concentration and point particles. The low-
concentration expansions (49) and (52) are accurate to second order in the number of scatterers
per unit volume, $n_0$, for finite-size particles. The acoustic limiting case of small spheres is
then considered, in the process taking $kb \ll 1$. This limit corresponds to point particles with
a spherical excluded volume (i.e. a hole of radius $b$). The resulting new formulae for $\rho_{\text{eff}}$ and
$\kappa_{\text{eff}}$, equations (53) and (54), depend only on the angular shape function $f(\theta)$ for an isolated
particle, apart from their dependence on $k$ and $n_0$.

We have shown that the effective mass density and bulk modulus can be made negative
near resonances by choosing appropriate resonant particles. The theory provides a technique for
searching for acoustic metamaterials with specific desired properties. To obtain negative mass
density $\rho_{\text{eff}}$ and bulk modulus $\kappa_{\text{eff}}$, one must engineer the particles to coincide and enlarge the
negative regions of both $\rho_{\text{eff}}$ and $\kappa_{\text{eff}}$. One way to achieve this might be to simply change the
dimensions of the particles or to consider core–shell particles. Another way is to consider a two-
phase suspension comprising a fluid matrix and a distribution of clustered particles (e.g. [72]).
The application of this work to more complex acoustic metamaterials is the subject of ongoing
research.

Acknowledgments

The authors acknowledge the UK Research Centre in Non-destructive Evaluation for financial
support. This work was carried out using the computational facilities of the Advanced
Computing Research Centre, University of Bristol (http://www.bris.ac.uk/acrc/).

Appendix A

A.1. The Foldy–Lax integral equation

The coherent motion in the fluid is, according to the Foldy–Twersky approach, the sum of
the incident motion and the wave motions scattered by all the scatterers. In terms of the
displacement potential, one has [35]

$$
\langle \phi(\mathbf{r}) \rangle = \phi^{\text{in}}(\mathbf{r}) + \int \mathbf{n}(\mathbf{r}) f(\mathbf{r}) \langle \phi^{\text{ex}}(\mathbf{r}|\mathbf{r}_j) \rangle d\mathbf{r}_j,
$$

(A.1)

where $\mathbf{n}(\mathbf{r}_j)$ is the conditional number density of the $j$th scatterer. The second term on the
right-hand side of (A.1) represents the global contribution of all scatterers located at all
possible positions $\mathbf{r}_j$, under the assumption that the distribution is random and uniform. The
term $\mathbf{f}(\mathbf{r}_j) \langle \phi^{\text{ex}}(\mathbf{r}|\mathbf{r}_j) \rangle$ is an average displacement potential corresponding to the wave motion
scattered by the scatterer at $\mathbf{r}_j$, while $\langle \phi^{\text{ex}}(\mathbf{r}|\mathbf{r}_j) \rangle$ corresponds to the coherent wave motion acting
on the scatterer at \( r_j \). The average on disorder is taken over all configurations keeping the scatterer at \( r_j \) fixed. Thus, it is a partial average. The integral of this scattered potential over all positions in the volume accessible to scatterers represents the total contribution of all scattered motions. The point at which the wave motion is evaluated is \( r \).

When all the scatterers occupy deterministic positions, a scatterer at \( r_i \) ‘sees’ an exciting wave motion that is the sum of the incident motion and the motions scattered by all the other scatterers. Under the ‘quasi-crystalline approximation’ [32], the integral equation for the exciting displacement potential is given by

\[
\langle \phi^{ex}(r|r_i) \rangle = \phi^{in}(r) + \int n(r_j|r_i)T(r_j) \langle \phi^{ex}(r|r_j) \rangle \, dv_j,
\]

(A.2)

where \( n(r_j|r_i) \) is the conditional number density of the scatterer at \( r_j \) if a scatterer is fixed at \( r_i \).

For a uniform and random array of spheres of radius \( a \), the conditional number density must be specified in terms of a pair-correlation function, such that

\[
n(r_j|r_i) = n_0 g(\rho_{ji})
\]

(A.3)

where the pair-correlation function \( g \) satisfies the conditions

\[
g(\rho_{ji}) = 0 \quad \text{for} \quad \rho_{ji} < 2a, \quad \text{and} \quad \lim_{\rho_{ji} \to \infty} g(\rho_{ji}) = 1.
\]

(A.4)

The former condition holds for non-overlapping sets of spheres; the later condition is correct if the correlation of particle locations disappears when the distance between their centres tends to infinity.

A.2. The Fikioris–Waterman dispersion relation

Assuming that the exciting displacement potential (A.2) is a ‘bounded’ solution of the Helmholtz equation in the fluid and is a regular function at the point \( r_i \), one finds that the most general expression of \( \langle \phi^{ex} \rangle \) is

\[
\langle \phi^{ex}(r|r_i) \rangle = \sum_{n=0}^{\infty} E_n(r_i) \hat{\psi}_n(\rho_i),
\]

(A.5)

where \( \hat{\psi}_n(\rho_i) = j_n(k\rho_i)P_n(\cos \theta_i) \) is a regular spherical wavefunction and \( j_n \) is a spherical Bessel function. The unknown modal coefficients \( E_n(r_i) \) characterize the average displacement potential of the exciting motion on the scatterer at \( r_i \).

In order to solve the ensemble average equation (A.2), the exciting coefficients \( E_n(r) \) are decomposed into refracted plane waves in the forward direction (\( z > 0 \)) with a propagation constant \( \kappa \). The effective wavenumber \( \kappa \) defines the coherent motion according to

\[
E_n(r) = X_n \exp(i\kappa z).
\]

(A.6)

Both \( \kappa \) and \( X_n \) are unknown. Substituting (A.5), with (A.6), into (A.2) and using the addition theorem for spherical wavefunctions [33] yields an infinite set of linear algebraic equations for the determination of \( \kappa \) and the coefficients \( X_n \). The details of the calculation are not shown here since they merely reproduce results obtained in [11] and [33].

New Journal of Physics 14 (2012) 033014 (http://www.njp.org/)
The Fikioris–Waterman dispersion relation and the extinction theorem are then recovered, and are given, respectively, by

\[
X_n + \frac{4i\pi n_0}{k(k^2 - k^2)} \sum_{\nu=0}^{\infty} (2\nu + 1) T_{\nu} X_{\nu} \sum_p G(0, v|0, n|p) \left[ M_p(kb) + (k^2 - k^2) N_p(kb) \right] = 0,
\]

(A.7a)

\[
\kappa = k + \frac{2\pi n_0}{k} \mathcal{F}_0, \quad \mathcal{F}_0 = \frac{1}{ik} \sum_{\nu=0}^{\infty} (2\nu + 1) T_{\nu} X_{\nu}.
\]

(A.7b)

where

\[
M_p(z) = izh_p(x) \frac{d}{dz} j_p(z) - xj_p(z) \frac{d}{dx} h_p(x), \quad x = kb,
\]

(A.8)

\[
N_p(z) = ik \int_{b}^{\infty} dr [g(r) - 1]r^2 h_p(kr) j_p(zr/b).
\]

Equation (A.7a), known as the Lorentz–Lorenz law [40], defines the effective wavenumber and the associated infinite eigenvector, whereas equation (A.7b) defines the amplitude of the eigenvector. The Gaunt coefficients \( G \) appear in the linearization expansion [41]

\[
P_n(x) P_\nu(x) = \sum_p G(0, n|0, \nu|p) P_p(x),
\]

(A.9)

and are defined by

\[
G(0, n|0, \nu|p) = \frac{2p + 1}{2} \int_0^{\pi} d\theta \sin \theta P_n(\cos \theta) P_\nu(\cos \theta) P_p(\cos \theta).
\]

(A.10)

**Appendix B**

**B.1. Asymptotic approximations**

In the Rayleigh limit, the size of the particles \( a \) is assumed to be small compared to the incident wavelength \( \Lambda \), i.e. \( \Lambda \gg a \). It can be shown that only the monopole \( (n = 0) \) and dipole \( (n = 1) \) terms are significant in this limit. The single-scattering operator (matrix) \( Q \) is therefore compact and has only two eigenvalues of finite size. Furthermore, the infinite matrix \( R \) is reduced to a rank 2 matrix. One obtains for spherical particles

\[
Q = \frac{1}{ik} \begin{pmatrix} T_0 & 0 \\ 0 & 3T_1 \end{pmatrix}, \quad R = \frac{1}{k} \begin{pmatrix} 0 & \frac{1}{k^2 + \epsilon} \\ \frac{1}{k^2 + \epsilon} & \frac{1}{k^2} \end{pmatrix}.
\]

(B.1)

Substituting equations (B.1) into equation (12), a closed form solution for the effective wavenumber \( \kappa \) is obtained by letting the determinant of the matrix premultiplying \( x \) be zero, i.e.

\[
\frac{\kappa^2}{k^2} = \frac{(k^3 - i\epsilon T_0)(k^3 - i\epsilon T_1)}{k^3(k^3 + 2i\epsilon T_1)}.
\]

(B.2)

Once the effective wavenumber is determined from equation (B.2), the eigenvector of amplitudes \( X_0 \) and \( X_1 \) then follows from equation (A.7b). They are given in explicit form...
in [11] and need not be repeated here. Next, the reflection coefficient is obtained by noting that
\( \mathcal{R} = \frac{k^2 \mathcal{K} - k^3 + i \varepsilon \mathcal{T}_0}{k^2 \mathcal{K} + k^3 - i \varepsilon \mathcal{T}_0} \) (B.3)

The calculation of the effective mass density is accomplished by noting that equation (46) is equivalent to
\[ \frac{\rho_{\text{eff}}}{\rho_0} = \frac{(1 + \mathcal{R})}{(1 - \mathcal{R})} \frac{\mathcal{K}}{\tilde{k}}. \]
(B.4)

Therefore, by using equations (B.2) and (B.3) we infer that
\[ \frac{\rho_{\text{eff}}}{\rho_0} = \frac{k^3 - i \varepsilon \mathcal{T}_1}{k^3 + 2i \varepsilon \mathcal{T}_1}. \]
(B.5)

The effective bulk modulus follows as
\[ \frac{\kappa_{\text{eff}}}{\kappa_0} = \frac{k^3}{k^3 - i \varepsilon \mathcal{T}_0}. \]
(B.6)

We can see that the effective bulk modulus and mass density are related to monopolar \((n = 0)\) and dipolar \((n = 1)\) scattering coefficients, respectively. The effective parameters thus obtained are real valued if the materials of the system are lossless.

In general, the scattering coefficients are complex and are given in the form \( \mathcal{T}_n = \mathcal{T}_n' + i \mathcal{T}_n'' \), with \( \mathcal{T}_n', \mathcal{T}_n'' \in \mathbb{R} \). Keeping the leading terms in the real and the imaginary parts and noting that \( \mathcal{T}_n' = -\mathcal{T}_n'' \), \((n = 0, 1)\), the solution of \( \mathcal{K} \) is such that \( \text{Re}(\mathcal{K}) = \mathcal{O}(1) \) and \( \text{Im}(\mathcal{K}) = \mathcal{O}(\kappa a)^3 \), i.e.
\[ \frac{k^2}{k^2} = \frac{(k^3 + i \varepsilon \mathcal{T}_1'') (k^3 + i \varepsilon \mathcal{T}_1'')}{k^3 (k^3 - 2i \varepsilon \mathcal{T}_1'')} + i \varepsilon \frac{3 \mathcal{T}_1''^2 (k^3 + i \varepsilon \mathcal{T}_1'')}{(k^3 - 2i \varepsilon \mathcal{T}_1'')^2} + \frac{T_0''^2 (k^3 + i \varepsilon \mathcal{T}_1'')}{k^3 (k^3 - 2i \varepsilon \mathcal{T}_1'')}. \]
(B.7)

The effective mass density and bulk modulus may then be expressed as
\[ \frac{\rho_{\text{eff}}}{\rho_0} = \frac{k^3 + i \varepsilon \mathcal{T}_1''}{k^3 - 2i \varepsilon \mathcal{T}_1''} + \frac{3k^3 \mathcal{T}_1''^2}{(k^3 - 2i \varepsilon \mathcal{T}_1'')^2}, \]
(B.8)

\[ \frac{\kappa_{\text{eff}}}{\kappa_0} = \frac{k^3 + i \varepsilon \mathcal{T}_0''}{k^3 + i \varepsilon \mathcal{T}_0''} - \frac{k^3 \mathcal{T}_0''^2}{(k^3 + i \varepsilon \mathcal{T}_0'')^2}. \]
(B.9)

Let \( \tilde{\omega} = ka \); one has to the leading order in \( \tilde{\omega} \)
\[ \mathcal{T}_0'' = \tilde{\omega}^3 \left[ \frac{\mathcal{M}}{3} + \mathcal{O}(\tilde{\omega}^2) \right], \quad \mathcal{T}_1'' = \tilde{\omega}^3 \left[ \frac{\mathcal{D}}{3} + \mathcal{O}(\tilde{\omega}^2) \right], \]
(B.10)

where
\[ \mathcal{D} = \frac{\rho - \rho_0}{\rho_0 + 2 \rho} \quad \text{and} \quad \mathcal{M} = \frac{\kappa_0}{\kappa} - 1. \]
(B.11)

Then, in terms of the fractional volume \( \phi = \frac{1}{3} \varepsilon a^3 \) \((= \frac{4}{3} \pi n_0 a^3)\) occupied by the particles, equations (B.7)–(B.9) yield
\[ \frac{k^2}{k^2} \simeq \frac{(1 + \phi \mathcal{M}) (1 + \phi \mathcal{D})}{1 - 2 \phi \mathcal{D}} + i \phi \left[ \frac{\mathcal{M}^2 (1 + \phi \mathcal{D})}{3 (1 - 2 \phi \mathcal{D})} \frac{\mathcal{D}^2 (1 + \phi \mathcal{M})}{(1 - 2 \phi \mathcal{D})^2} \right] \tilde{\omega}^3, \]
(B.12)
\[ \rho_{\text{eff}} \approx \frac{1 + \phi \mathcal{D}}{1 - 2\phi \mathcal{D}} + i \frac{\phi \mathcal{D}^2 \bar{\omega}^3}{(1 - 2\phi \mathcal{D})^2}, \quad (\text{B.13}) \]
\[ \kappa_{\text{eff}} \approx \frac{1}{1 + \phi \mathcal{M}} - i \frac{\phi \mathcal{M}^2 \bar{\omega}^3}{3 (1 + \phi \mathcal{M})^2}. \quad (\text{B.14}) \]

The real parts of effective parameters in (B.12)–(B.14) are identified as the corresponding results due to Fikioris and Waterman [11]. The real part of equation (B.13) is identical to the Ament estimate (2), whereas the real part of equation (B.14) is recognized as the Reuss average. Note that the signs \( \text{Im} \rho_{\text{eff}} > 0 \) and \( \text{Im} \kappa_{\text{eff}} < 0 \) are in agreement with the sign of dissipation.

### B.2. Second-order expansions

It is straightforward to expand the low-frequency formulae obtained in the previous section. Instead, the point-particle expressions (31), (43), (53) and (54) are approximated in the low-frequency limit \( (ka \ll 1) \). This provides an additional check of the correctness of the results obtained in this paper.

The leading order contribution to the angular shape function \( f(\theta) \) clearly only involves \( T_0 \) and \( T_1 \), i.e.
\[ f(\theta) \approx \frac{1}{i k} (T_0 + 3T_1) \cos \theta. \quad (\text{B.15}) \]

In terms of equation (B.15), the integrals of equations (32) and (44) yield
\[ I_0 \approx \frac{24}{k^2} T_1 (T_0 + T_1), \quad (\text{B.16a}) \]
\[ I_\pi \approx -\frac{24}{k^2} T_1^2. \quad (\text{B.16b}) \]

Finally, using the results (B.16), the effective acoustic \((\kappa \text{ and } \Re)\) and material \((\rho_{\text{eff}} \text{ and } \kappa_{\text{eff}})\) parameters are obtained as
\[ k^2 \approx k^2 - \epsilon \frac{i}{k} (T_0 + 3T_1) - \epsilon^2 \frac{3}{k^4} T_1 (T_0 + 2T_1), \quad (\text{B.17}) \]
\[ \Re \approx \epsilon \frac{i}{4k} (T_0 - 3T_1) - \epsilon^2 \frac{1}{8k^6} (T_0^2 + 3T_1^2), \quad (\text{B.18}) \]
\[ \frac{\rho_{\text{eff}}}{\rho_0} \approx 1 - \epsilon \frac{3i}{k^3} T_1 - \epsilon^2 \frac{6}{k^6} T_1^2, \quad (\text{B.19}) \]
\[ \frac{\kappa_{\text{eff}}}{\kappa_0} \approx \frac{k^6 - 3i k^3 T_1 - 6 \epsilon^2 T_1^2}{k^6 - i k^3 (T_0 + 3T_1) - 3 \epsilon^2 T_1 (T_0 + 2T_1)}, \quad (\text{B.20a}) \]
\[ \approx 1 + \epsilon \frac{i}{k^3} T_0 - \epsilon^2 \frac{1}{k^6} T_0^2 + \mathcal{O}(\epsilon^3). \quad (\text{B.20b}) \]

As expected, equations (B.17)–(B.20) agree to second order in the concentration with equations (B.2)–(B.6). Similar expressions in terms of \( \mathcal{D} \) and \( \mathcal{M} \) (of equation (B.11) may also be obtained. In particular, for the effective mass density we obtain in terms of the fractional volume \( \phi \)
\[ \frac{\rho_{\text{eff}}}{\rho_0} \approx 1 + 3\phi \mathcal{D} + 6\phi^2 \mathcal{D}^2 + i\phi \mathcal{D}^2 \bar{\omega}^3 (1 + 4\phi \mathcal{D}) + \mathcal{O}(\phi^3). \quad (\text{B.21}) \]
Appendix C

C.1. Comparison with previous work

It is relevant to recall some results based on the widely used formula for the effective wavenumber

\[ K^2 = k^2 + 4\pi n_0 f(0) + \left( \frac{2\pi n_0}{k} \right)^2 \left[ H_0(0) - H_0(\pi) \right], \]  

(C.1)

obtained originally by Urick and Ament [9] for a slab of spheres. This formula was rederived later by Waterman and Truell [26], and more recently by Angel and Aristegui [27], by using different arguments. Further restriction to monopole and dipole scatterers gives the form derived by Reiche [49], Foldy [34] and Lax [50]. For the case of small scatterers all the above-mentioned forms reduce to the one given essentially by Rayleigh [51] (by expanding the square root of \( K^2 \) to the first order in \( n_0 \)). Twersky [19] extended equation (C.1) to arbitrary angles of incidence and arbitrary scatterers by generalizing Reiche’s approximation procedure for a wave normally incident on a slab of dipoles [49]. However, we discount this since the effective wavenumber itself should not depend on the incidence wave angle. Equation (C.1) defines what is called the ‘Waterman–Truell estimate’ in the following.

It is known that the Waterman–Truell estimate is not correct as far as the term in \( n_0^2 \) is concerned [43]; the correct expression is given by the Lloyd–Berry estimate (31). Nevertheless, this result has been used to estimate effective properties. Indeed, Aristegui and Angel [28] have used the Waterman–Truell estimate (C.1) to obtain the effective mass density and bulk modulus of a mixture of particles suspended in a fluid. Their estimate for the effective mass density is given in our notation by

\[ \frac{\rho_{\text{eff}}}{\rho_0} = 1 + \tilde{\rho}_1 \frac{\varepsilon}{2k^2} \left( f(0) - f(\pi 40) \right), \]

(C.2)

which may be compared with equation (49). It is noted that equation (C.2) is linear in \( \varepsilon \), whereas equation (49) gives a more refined estimate for \( \rho_{\text{eff}} \). Observe that equation (C.2) does not depend on whether the particles are point-like or have finite sizes. Indeed, the Waterman–Truell analysis is based on the point-particle approximation, whereas our estimate (49) is valid for finite-size particles. Therefore, to the first order in \( \varepsilon \) the effective mass density is independent of the finite excluding volume of the particles. The effective bulk modulus based on the Waterman–Truell estimate (C.1) is given by [28]

\[ \frac{\kappa_{\text{eff}}}{\kappa_0} = \left[ 1 - \frac{\varepsilon}{2k^2} (\tilde{\rho}_1 - 2\xi_1) \right]^{-1} = \left[ 1 + \frac{\varepsilon}{2k^2} [f(0) + f(\pi)] \right]^{-1}, \]

(C.3)

\[ \simeq 1 + \frac{\varepsilon}{2k^2} (\tilde{\rho}_1 - 2\xi_1) + \frac{\varepsilon^2}{4k^2} (\tilde{\rho}_1 - 2\xi_1)^2 + O(\varepsilon^3). \]

(C.4)

As expected, equation (C.4) differs from equation (52b) in the \( \varepsilon^2 \) term. Note that the formulae (C.2) and (C.3) have been previously derived by Twersky [22] (in terms of permittivity and permeability within the framework of electromagnetic scattering) by neglecting the ‘hole corrections’. Moreover, the long-wavelength limit of equations (C.2) and (C.3) has been obtained by Poujol–Pfefer [53], based on different arguments, well before being established by Aristegui and Angel. Note also that a more sophisticated form of the effective mass density has been obtained by Twersky in another paper [19], by applying the ‘two-space’ scatterer.
formalism [18] to equations (C.2) and (C.3). It is worth noting that the self-consistent model set out by Yang and Mal [71] is also based on the Waterman–Truell estimate.

More recently, Martin et al [16] and Parnell and Abrahams [52] derived the effective properties for 2D composites, in the quasi-static limit, based on arguments similar to those of Poujol–Pfefer [53]. In particular, the former work leads to a formula for the effective mass density in two-dimensions that is reminiscent of the Ament estimate (2) (more precisely, a small-$\phi$ approximation of the Ament estimate).

References

[1] Smith D R and Pendry J B 2006 J. Opt. Soc. Am. 23 391
[2] Yang Z, Mei J, Yang M, Chan N H and Sheng P 2008 Phys. Rev. Lett. 101 204301
[3] Lee S H, Park C M, Seo Y M, Wang Z G and Kim C K 2009 Phys. Lett. A 373 4464
[4] Liu X N, Hy G K, Huang G L and Sun C T 2011 Appl. Phys. Lett. 98 251907
[5] Li J, Liu Z and Qiu C 2006 Phys. Rev. B 73 054302
[6] Zhu J, Christensen J, Jung J, Martin-Moreno L, Yin X, Fok L, Zhang X and Garcia-Vidal F J 2010 Nat. Phys. 7 52
[7] Zhang S, Yin L and Fang N 2009 Phys. Rev. Lett. 102 194301
[8] Li J, Fok L, Yin X, Bartal G and Zhang X 2009 Nature Mater. 8 931
[9] Urick R J and Ament W S 1949 J. Acoust. Soc. Am. 21 115
[10] Ament W S 1953 J. Acoust. Soc. Am. 25 638
[11] Fikioris J G and Waterman P C 1964 J. Math. Phys. 5 1413
[12] Kuster G T and Toksöz M N 1974 Geophysics 39 587
[13] Gaunaurd G C and Wertman W 1989 J. Acoust. Soc. Am. 85 541
[14] Landau L D and Lifshitz E M 1959 Fluid Mechanics (Reading, MA: Addison Wesley)
[15] Harker A H and Temple J A G 1988 J. Phys. D: Appl. Phys. 21 1576
[16] Martin P A, Maurel A and Parnell W J 2010 J. Acoust. Soc. Am. 128 571
[17] Berryman J G 1980 J. Acoust. Soc. Am. 68 1809
[18] Twersky V 1962 J. Math. Phys. 3 716
[19] Twersky V 1962 J. Math. Phys. 3 724
[20] Wu Y, Li J, Zhang Z-Q and Chan C T 2006 Phys. Rev. B 74 085111
[21] Sheng P, Mei J, Liu Z and Wen W 2007 Phys. B: Condens. Matter 394 256
[22] Twersky V 1962 J. Math. Phys. 3 700
[23] Twersky V 1977 J. Math. Phys. 18 2468
[24] Twersky V 1978 J. Acoust. Soc. Am. 64 1710
[25] Twersky V 1978 J. Math. Phys. 18 215
[26] Waterman P C and Truell R 1961 J. Math. Phys. 2 512
[27] Angel Y C and Aristégui C 2005 J. Acoust. Soc. Am. 118 72
[28] Aristégui C and Angel Y C 2007 Wave Motion 44 153
[29] Zhang X and Liu Z 2004 Appl. Phys. Lett. 85 341
[30] Torrent D and Sánchez-Dehesa J 2008 New J. Phys. 10 023004
[31] Torrent D and Sánchez-Dehesa J 2008 New J. Phys. 10 063015
[32] Lax M 1952 Phys. Rev. 85 621
[33] Caleap M, Drinkwater B W and Wilcox P D 2012 J. Acoust. Soc. Am. 131 2036
[34] Foldy L L 1945 Phys. Rev. 67 107
[35] Ishimaru A 1997 Wave Propagation and Scattering in Random Media (Piscataway, NJ: IEEE)
[36] Markov K Z 1994 Advances in Mathematical Modelling of Composite Materials (Singapore: World Scientific Publishing)
[37] Kanaun S K and Levin V M 1993 Effective Field Method in Mechanics of Composite Materials (Petrozavodsk: Petrozavodsk State University)

[38] Gaunt J A 1929 Phil. Trans. R. Soc. A 228 151

[39] Linton C M and Thompson I 2009 J. Comput. Phys. 228 1815

[40] Born M and Wolf E 1975 Principles of Optics (New York: Oxford Pergamon)

[41] Xu Y-L 1996 Math. Comput. 65 1601

[42] Fermi E 1950 Nucl. Phys. (Chicago, IL: University of Chicago Press)

[43] Lloyd P and Berry M V 1967 Proc. Phys. Soc. 91 678

[44] Lloyd P 1967 Proc. Phys. Soc. 90 207

[45] Lloyd P 1967 Proc. Phys. Soc. 90 217

[46] Aristégui C and Angel Y C 2010 Wave Motion 47 199

[47] Conoir J M, Robert S, El Mouhtadi A and Luppé F 2009 Wave Motion 46 522

[48] Martin P A 2011 J. Acoust. Soc. Am. 129 1685

[49] Reiche F 1916 Ann. Phys., Lpz. 355 1

[50] Lax M 1951 Rev. Mod. Phys. 23 287

[51] Rayleigh L 1899 Phil. Mag. 47 375

[52] Parnell W J and Abrahams D 2010 Waves Random Complex Media 20 678

[53] Poujol-Pfefer M-F 1994 PhD Thesis Université d’Aix-Marseille II

[54] Li J and Chan C T 2004 Phys. Rev. E 70 055602

[55] Green H S 1952 The Molecular Theory of Fluids (New York: Interscience)

[56] Percus J K and Yevick G J 1958 Phys. Rev. 110 1

[57] Carstensen E L and Foldy L L 1947 J. Acoust. Soc. Am. 19 481

[58] Commander K W and Prosperetti A 1989 J. Acoust. Soc. Am. 85 732

[59] Poujol-Pfefer M-F 1995 J. Sound Vib. 184 665

[60] Ambati M, Fang N, Sun C and Zhang X 2007 Phys. Rev. B 75 195447

[61] Deng K, Ding Y, He Z, Zhao H, Shi J and Liu Z 2009 J. Appl. Phys. 105 124909

[62] Yang Z, Dai H M, Chan N H, Ma G C and Sheng P 2010 Appl. Phys. Lett. 96 041906

[63] Pendry J B 2000 Phys. Rev. Lett. 85 3966

[64] Zhang X and Liu Z 2008 Nature Mater. 7 435

[65] Li J, Fok L, Yin X, Bartal G and Zhang X 2009 Nature Mater. 8 931

[66] Hu X, Ho K M, Chan C T and Zi J 2008 Phys. Rev. B 77 172301

[67] Mei J, Liu Z Y, Wen W J and Sheng P 2009 Phys. Rev. Lett. 96 024301

[68] Huang H H and Sun C T 2009 New J. Phys. 11 013003

[69] Torrent D and Sánchez-Dehesa J 2011 New J. Phys. 13 093018

[70] Wu Y, Lai Y and Zhang Z-Q 2007 Phys. Rev. B 76 205313

[71] Yang R-B and Mal A K 1994 Int. J. Solids Struct. 42 1945

[72] Caleap M, Drinkwater B W and Wilkox P D 2011 NDT&E Int. 44 456

[73] Angel Y C, Aristegui C and Caleap M 2009 arXiv:0903.5191v1