Effective Mass Density of Fluid-Solid Composites

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We show through rigorous derivation and experimental support that the dynamic effective mass density of an inhomogeneous mixture, used in the prediction of wave velocities in the long wavelength limit, can differ from the static version—the volume average of the component mass densities. The physical reason for this difference is explained. The dynamic mass density expression, first derived by Berryman more than two decades ago, is shown to give a closer correspondence between the acoustic and electromagnetic metamaterials by allowing for negative mass densities at frequencies around resonances.

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The effective mass density of a composite is one of the most basic quantities in the study of materials. It is common sense that the effective mass density of a mixture of materials should be their volume average, \( D_{\text{eff}} = D_v = D_1(1-f) + D_2 f \), where \( D_1 \) and \( D_2 \) are the mass densities for the matrix and the inclusions, respectively, and \( f \) is the filling ratio of the solid inclusions. This expression is denoted below as the volume-averaged mass density (VAMD). An important application of the composite effective mass density is in the prediction of wave velocities in the low frequency limit, where the relevant wavelength is much larger than the typical feature sizes in the composite. More than two decades ago, Berryman [1] derived a different effective mass density expression for the prediction of (fluid matrix–solid) composite wave properties in the long wavelength limit, based on the average \( T \)-matrix approach:

\[
\frac{D_{\text{eff}} - D_1}{(d-1)D_{\text{eff}} + D_1} = f \frac{D_2 - D_1}{(d-1)D_2 + D_1},
\]

where \( d \) denotes the spatial dimensionality of the problem.

The Berryman effective mass density expression is noted to differ significantly from the intuitive VAMD, and for all the intervening years after the initial derivation it has remained a curiosity rather than extensively used, mainly owing to the lack of experimental support as well as to the strong sense that the intuitive VAMD must be correct, since otherwise it would be equivalent to stating the rather radical principle that the static mass density for a composite should be different from its dynamic mass density, even in the long wavelength limit. An additional objection to the Berryman expression is that the derivation treats the multiple scatterings inherent in the inhomogeneous system only in an averaged sense, and therefore not rigorously.

In this Letter, we show through rigorous derivation that the recent experimental evidence [2] has indeed provided strong support to the radical principle that the Berryman effective mass density expression should be the correct one to use in predicting the dynamic wave properties of fluid (matrix)–solid composites in the long wavelength limit, and that the VAMD is only a special case, obtainable from the Berryman expression when the components’ mass density contrasts are small. The implications of this finding, especially relevant to the acoustic correspondence with the doubly negative response functions (electric and magnetic) of electromagnetic metamaterials, are presented.

The starting point of our considerations is the recent experiment [2] of Cervera et al., in which the sound velocity was measured in a two-dimensional phononic crystal [2–17] composed of regular arrays of rigid cylinders in air, confirming that in the low frequency (long wavelength) regime the sound propagates at subsonic velocity. In Fig. 1, we show the comparison of experimental results with the prediction of the usual effective medium theory (EMT) [18], in which the effective bulk modulus \( B_{\text{eff}} \) is given by

\[
\frac{1}{B_{\text{eff}}} = \frac{1-f}{B_1} + \frac{f}{B_2},
\]

where \( B_1 = \lambda_1 \) and \( B_2 = \lambda_2 + \frac{2}{3} \mu_2 \) are the bulk moduli for the liquid matrix and the solid inclusions, respectively, with \( \lambda \) and \( \mu \) being the Lamé constants. The effective mass density used in the effective medium theory prediction shown in Fig. 1 is given by VAMD. A very large discrepancy is seen. As the experiment is done with a regular array of solid cylinders, the system is amenable to accurate numerical predictions using the multiple scattering theory (MST) [17]. The MST-calculated results are shown as the solid triangles. Excellent agreement is seen.

Since the experiment was clearly in the long wavelength regime (with the wavelength at least 9 times the lattice constant), it becomes an intriguing question as to the form of the long wavelength (or the low frequency) limit of the exact theory, the prediction of which is shown to give good agreement with the experiment. This is because at the low frequency limit the dispersion relation for phononic crys-
function and Hankel function of the first kind, respectively. Since the incident wave on scatterer $i$ comes from the scattered waves by all the scatterers except scatterer $i$, we have

$$\vec{u}_{i}^{\text{sc}}(\vec{\rho}) = \sum_{j \neq i} \sum_{n} b_{n}^{j} \vec{H}_{n}^{j}(\vec{\rho}).$$

(5)

With the help of the addition theorem, we can prove that

$$\vec{H}_{n}^{j}(\vec{\rho}) = \sum_{n} G_{n}^{i,j} \vec{J}_{n}(\vec{\rho}).$$

(6)

where $G_{n}^{i,j} = G_{nn}(\vec{R}_{j} - \vec{R}_{i})$ denotes the translation (from scatterer $i$ to scatterer $j$) coefficients, with $\vec{R}_{i,j}$ denoting the position of scatterer $i(j)$. We refer to Ref. [17] for the precise definition of $G_{nn}(\vec{R})$. The expansion coefficients $A = \{a_{n}\}$ for the incident field and $B = \{b_{n}\}$ for the scattered field with reference to a given scatterer are related through the elastic Mie scattering matrix $T = \{T_{nn}\}$ for the scatterer by

$$b_{n} = \sum_{n} T_{nn} a_{n}.$$  

(7)

Substituting Eqs. (3), (6), and (7) into Eq. (5), we arrive at

$$\sum_{j} \left( \delta_{ij} \delta_{nn} - \sum_{n} c_{ij}^{n} T_{i,n}^{a} \right) a_{i}^{n} = 0.$$  

(8)

For a periodic system, the normal modes of the system may be obtained by solving the following secular equation:

$$\det\left[ T_{nn}^{-1} - G_{nn}(k) \right] = 0,$$  

(9)

where $G_{nn}(k) = \sum_{\vec{R}} G_{nn}(\vec{R}) \exp(i \vec{k} \cdot \vec{R})$. By taking the low frequency limit of $\alpha_{1} \rightarrow 0$ and by retaining the dominant terms, Eq. (9) is simplified to a $3 \times 3$ matrix equation [19]:

$$\begin{vmatrix}
D_{1} x^{2} + D_{2} - D_{3} \frac{x^{2}}{1-x^{2}} & \frac{ix f}{1-x^{2}} & - \frac{f}{1-x^{2}} \\
\frac{-ix f}{1-x^{2}} & B_{2} - B_{1} + \frac{x^{2}}{1-x^{2}} & \frac{ix f}{1-x^{2}} \\
\frac{f}{1-x^{2}} & \frac{-ix f}{1-x^{2}} & D_{1} + D_{2} + \frac{x^{2}}{1-x^{2}} 
\end{vmatrix} = 0,$$  

(10)

in which $x = V_{\text{eff}}/V_{1}$ is the constant to be evaluated, while $f$, $B_{1}$, and $B_{2}$ are the filling ratio and the elastic moduli, respectively [20]. It can easily be verified that Eq. (10) is a quadratic equation of $x^{2}$, and by omitting the trivial root $x^{2} = 1$, we obtain the root

$$x^{2} = \frac{(D_{2} + D_{1}) - (D_{2} - D_{1})f}{(D_{2} + D_{1}) + (D_{2} - D_{1})f} B_{2}.$$  

(11)

By using the expression $V_{\text{eff}} = \frac{\rho_{\text{eff}}}{\rho_{\text{air}}}$ and the effective medium expression for $B_{\text{eff}}$, Eq. (2), we arrive at precisely the Berryman effective mass density in 2D. Equation (11)
is noted to be valid for both the square and the hexagonal lattices. Hence, it is plausible that the Berryman effective mass density expression is generally valid for isotropic composites. This is the case especially since our derivation verifies, at the same time, the effective bulk modulus formula, Eq. (2), which is valid in general for isotropic composites consisting of solid inclusions in fluid [18].

As the static version of the effective mass density must be the VAMD (verifiable through simple weighing of the composite and its constituents, and measuring the volumes), the reason for the different (long wavelength) dynamic version can be found in the fact that, for wave properties, VAMD contains the implicit assumption of wave field homogeneity in the long wavelength limit. This assumption can be violated (even in the long wavelength limit) when there is a very large impedance mismatch between the two components, such as in the experiment of Cervera et al. In Fig. 2(a) we show the calculated wave field intensities, in color, for the relevant experiment. It is noted that the wave field is nearly zero inside the cylinders. Hence, it is almost impossible to have the condition for the validity of VAMD. The observed decrease in wave velocity can be ascribed to the wave paths' increased tortuosity. However, when the impedance mismatch is relatively moderate, e.g., when the mass density contrast is small, then the Berryman expression yields the VAMD. This is shown in Fig. 3, where the two expressions yield the almost identical effective mass density for the PMMA cylinders in water. For comparison with Fig. 2(a), we have also plotted the displacement field intensities for the PMMA-water system in Fig. 2(b), in which the wave field homogeneity is evident. As our derivation is obtained by taking the long wavelength limit in which the wave field homogeneity is evident. It is noted that the wave vector is along the y direction, and a is the lattice constant. It is seen that the wave amplitude is nearly zero inside the Al cylinders. Decreasing the frequency further does not alter this fact. (b) The same for PMMA cylinders in water. Wave field is seen to be much more homogeneous than that in (a).

It should be noted that the effective bulk modulus expression, Eq. (2), derived via either the average T matrix approximation [1] or the coherent potential approximation [18] (and verified through the low frequency limit of the MST), represents the low frequency limit of the n = 0 angular scattering channel, whereas the D_B expression is the low frequency limit of the n = 1 channel. In the case examined above, the confinement of the wave amplitude in the interstitial space also implies that the effective bulk modulus is dominated by that of the fluid, just as predicted by Eq. (2), i.e., the two are consistent in this case. But even under general considerations D_B and Eq. (2) should both be valid, since they represent separate, yet at the same time parallel, wave scattering channels.

An important implication of our conclusion is in regard to the correspondence between acoustic [21,22] and electromagnetic [23,24] metamaterials. As VAMD can never be negative, it follows that if it is valid, then there can never be a one to one correspondence between the two classes of metamaterials in the effective medium limit. From Eq. (1), it is easily perceived that the expression allows a simple extension to the case when there is a n = 1 resonance [19]:

\[
\frac{D_{\text{eff}} - D_1}{D_{\text{eff}} + D_1} = \frac{4f}{i\pi(\alpha_1 R)^2} S_1, \tag{12}
\]

where

\[
S_1 = T_{11} = -\frac{F_1[J_1(\alpha_1 R) - \alpha_1 R J_2(\alpha_1 R)] + F_2[4J_2(\alpha_2 R) - \alpha_1 R J_3(\alpha_1 R)]}{F_1[H_1(\alpha_1 R) - \alpha_1 R H_2(\alpha_1 R)] + F_2[4H_2(\alpha_2 R) - \alpha_1 R H_3(\alpha_1 R)]}
\]
can be negative for Eq. (12) it is easily seen that the effective mass density numbers in the solid scatterers, respectively. From background fluid, and longitudinal and transverse wave properties, regardless of the impedance mismatch density to be the correct one for predicting dynamic compression was lacking in that case.

This fact was noted in Ref. [22], but the justification for using the Berryman mass density expression was lacking in that case.

In conclusion, we find the Berryman effective mass density to be the correct one for predicting dynamic wave properties, regardless of the impedance mismatch between the components. Our conclusion is based on taking the long wavelength limit of the rigorous multiple scattering theory, and on excellent agreement with the experimental result of Cervera et al.

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[1] James G. Berryman, J. Acoust. Soc. Am. 68, 1809 (1980); 68, 1820 (1980). Please note that the $(d-1)$ factor in Eq. (1b) is situated differently from the well-known Maxwell-Garnett formula. This is because the dielectric constant $\varepsilon$ corresponds with $1/\rho$.


[19] The derivation is straightforward but tedious. It will be published elsewhere.

[20] The shear wave excitations are fully accounted for in the MST, but at low frequency expansions they appear only in orders higher than $n = 0, 1$ and are negligible.


