Double-negative acoustic metamaterial

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We show here the existence of acoustic metamaterial, in which both the effective density and bulk modulus are simultaneously negative, in the true and strict sense of an effective medium. Our double-negative acoustic system is an acoustic analogue of Veselago's medium in electromagnetism, and shares many unique consequences, such as negative refractive index. The double negativity in acoustics is derived from low-frequency resonances, as in the case of electromagnetism, but the negative density and modulus are derived from a single resonance structure as distinct from electromagnetism in which the negative permeability and negative permittivity originates from different resonance mechanisms.

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The idea of negative refraction in an electromagnetic wave was proposed by Veselago [1] and the concept was recently demonstrated through the notion of metamaterial [2]. Veselago's medium is an isotropic homogeneous medium having a negative refractive index. The electromagnetic (EM) metamaterials that are realized in practice are composites with built-in resonance structures that exhibit effective negative permittivity and negative permeability for some frequency ranges. These "double negative" media have generated tremendous interest due to unique phenomena, such as negative refraction and subwavelength imaging [3]. We note that the phenomena of negative refraction is closely related but not identical to "double negativity." Negative refraction can be achieved through a band-folding effect due to Bragg scattering [4]. In an acoustic wave, we can also engineer the dispersion surface of solids to achieve the same effect as in Notomi's medium, and it is traditionally called phonon focusing [5,6]. With this comparison, it becomes natural and interesting to ask whether we can have an analog of Veselago's medium in an acoustic wave, namely a composite with built-in resonances such that the effective response functions are "double negative" in the effective medium limit.

In EM metamaterials, a negative refractive index in Veselago's medium is derived from the double negativity of permittivity and permeability. In an acoustic wave, the continuity and Newton's second law (with harmonic field dependence $e^{-i\omega t}$) can be expressed, respectively, as

$$\nabla \cdot \vec{v} - \frac{i\omega}{\kappa} p = 0,$$

$$\nabla p - i\omega\rho \vec{v} = 0,$$
 (1)

where $p(\vec{v})$ is the pressure (velocity) field. The density ρ and bulk modulus κ are position dependent in general. By considering a plane-wave solution with wave vector \vec{k} inside a homogeneous medium of constant density and bulk modulus, the refractive index *n* should be defined by

$$k = |n|\omega/c$$
 with $n^2 = \rho/\kappa$. (2)

Therefore, in order to have a propagating plane wave inside the medium, we should have either both positive ρ and κ or both negative ρ and κ . Moreover, the Poynting vector for a propagating plane wave should be defined by

$$\vec{S} = \frac{i}{2\omega\rho} p \nabla p^* = \frac{|p|^2 \vec{k}}{2\omega\rho}.$$
(3)

Now, if we can have Veselago's medium in an acoustic wave, \vec{S} and \vec{k} should point in the opposite directions. This requires negativity in bulk modulus and density. Physically, it means that the medium displays an anomalous response at some frequencies such that it expands upon compression (negative bulk modulus) and moves to the left when being pushed to the right (negative density) at the same time.

This sounds impossible, but the point of this paper is to show that it is mathematically possible and can be realized by dispersing soft rubber in water. Unlike the case in EM metamaterials, that permittivity and permeability can be negative intrinsically, natural materials neither have a negative density nor a negative bulk modulus. Even for composite materials, the effective bulk modulus and density should be normally bounded by the values of the constituents or by the more sophisticated Hashin and Shtrikman bounds [7]. Therefore, we still expect positive bulk modulus and positive density. For instance, for spherical solid particles dispersed in a fluid, the effective bulk modulus κ_{eff} and effective density ρ_{eff} in the long wavelength and in the limit of small volume filling ratio f [8] are governed by

$$\frac{1}{\kappa_{eff}} = \frac{f}{\kappa_s} + \frac{1-f}{\kappa_0},$$
$$\frac{\rho_{eff} - \rho_0}{2\rho_{eff} + \rho_0} = f\frac{\rho_s - \rho_0}{2\rho_s + \rho_0},$$
(4)

where the subscripts "s" and "0" denote, respectively, the properties for the sphere and the background fluid. Then, it can be easily proven from the formulas that κ_{eff} and ρ_{eff} are positive definite for natural materials. However, the above effective medium formulas and the traditional bounds on the effective parameters do not apply if there are low-frequency resonances. Standard homogenizations assume that the wavelengths in each local region (sphere and background)

are all much larger than the average distance between particles. At resonance, the wavelength within the sphere is now comparable to the size of it although the wavelength in the background still remains much larger than the average distance between particles in order to have a valid effective medium description. Under such a condition, Eq. (4) can be generalized to be

$$-1 + \frac{\kappa_0}{\kappa_{eff}} = \frac{3f}{i(k_0 R)^3} D_0,$$

$$\frac{\rho_{eff} - \rho_0}{2\rho_{eff} + \rho_0} = \frac{3f}{i(k_0 R)^3} D_1,$$
 (5)

where D_l is the scattering coefficient of angular momentum l of a single particle with radius R and k_0 is the wave number in the background fluid. These new formulas are derived using the coherent potential approximation method [9,10] by seeking the self-consistent solution to ensure that the inhomogeneous system embedded within an effective medium generates no scattering in the lowest order of frequency. The new formulas reduce to the original effective medium formulas if we substitute the scattering coefficients for a spherical particle in its long wavelength (within sphere) limit. We will see that it becomes possible to achieve negative effective bulk modulus and negative effective density through resonance behavior in D_0 and D_1 , which are functions in frequency.

From now on, we will establish the existence of acoustic double negativity using a composite of soft rubber spheres suspended in water as an example. We choose to use soft rubber such that sound travels much slower in it than in water. Then, the Mie resonances (monopolar and dipolar) can be brought to very low frequency due to the high contrast of sound speed between rubber and water. Let us start with a system of rubber spherical particles suspended in water of a low volume filling ratio 0.1, where Eq. (5) is reasonably accurate. We have ignored, for simplicity, the shear wave within the rubber spheres due to the high velocity contrast [11] between the rubber and water, and we emphasize that the main features stay the same if we also include the shear wave within the particles [12]. The spheres are assumed to be made of a kind of silicone rubber [13]. The effective medium result using the generalized effective medium formulas is shown in Fig. 1.

From Fig. 1, the effective bulk modulus and density near the static limit are positive as predicted by Eq. (4). The monopolar resonance creates a negative bulk modulus above the normalized frequency at about 0.035 while the dipolar resonance creates a negative density above the normalized frequency at about 0.04. Here, a is the lattice constant if the spheres are arranged in a fcc lattice. Hence, there is a narrow frequency range where we have both negative bulk modulus and negative density. The imaginary part of the effective parameters is due to the diffusive scattering loss.

The Mie resonances at low frequency in acoustics are the analog of the resonances created by split rings and wires in electromagnetic left-handed medium. In EM left-handed medium, the wires and split rings create a negative electric di-



FIG. 1. Effective density and bulk modulus [using Eq. (5)] for rubber (ρ =1300 kg m⁻³, κ =6.27×10⁵ Pa) spheres of filling ratio 0.1 within water (ρ =1000 kg m⁻³, κ =2.15×10⁹ Pa).

polar and magnetic dipolar response by two different mechanisms, while in our case a single structure gives rise to two kinds of resonances to achieve double negativity. The monopolar resonance creates a negative response such that the volume dilation of a single particle is out of phase with a hydrostatic pressure field. The dipolar resonance creates a negative response such that the motion of the center of mass of the particle is out of phase with an incident directional pressure field. If these negative responses are large enough to compensate the background fluid, we can have both negative effective bulk modulus and negative effective density.

We note from Fig. 1 that in the regime of double negativity, both the density and the reciprocal of the bulk modulus are decreasing in magnitude fast enough so that the group velocity becomes negative according to Eq. (2). It gives rise to negative refraction.

Under the condition that the background wavelength is much larger than the average interparticle distance and for slow spatially varying volume averaged wave field, homogenization of the composite to give effective bulk modulus and density is meaningful and it is valid to replace the whole composite by an effective medium in considering its acoustic properties. However, the CPA formulas [Eq. (4)] are quantitatively accurate only at a low filling ratio although it gives the physical origin of the double negativity. We need other procedures to extract the effective parameters. In the following, we demonstrate the extraction and the usefulness of assigning negative bulk modulus and density in considering acoustic properties of composites of a higher filling ratio. For this purpose, we consider the transmittance at different incidence angles through eight layers of a fcc colloidal crystal of silicone rubber spheres within water. For simplicity, we take the density of the rubber spheres (1000 kg m^{-3}) to match with that of water and the sound speed within the rubber to be 46.4 ms⁻¹. The radius of the rubber spheres is fixed at 1 cm. We look at two different filling ratios of 40% and 74%, respectively. We first calculate the dispersion at zero transverse wave vector $K_z(\omega; \vec{k_t} = \vec{0})$ using the layer-multiplescattering formalism [14], where the fcc (111) planes of the crystal are aligned perpendicular to the z axis. The square of the effective refractive index $n_{eff}^2(\omega)$ is then extracted from it by $K_z^2 = (\omega/c)^2 n_{eff}^2$, where c is the speed of sound in the background medium. Note that the eigenstate can be spanned by plane-wave components consisting of both normal and dif-



FIG. 2. Double acoustic negativity and negative refractive index of rubber spheres in water, fcc arrangement with a filling ratio of 40%. (a) Dispersion; (b) and (c) The transmittance through eight layers of the (111) planes at normal and 60° incidence. The open circles are calculated by the multiple-scattering method and the solid line is the approximation using homogeneous media with effective bulk modulus and density [as shown in panel (d)] extracted from the dispersion.

fracted plane waves. Conventionally, we can only extract the refractive index from the dispersion curve. In fact, by recognizing that the information of half-space reflection amplitude is already embedded in the details of the eigenmode, we can get an additional parameter. In an effective medium, it can be proved that the half-space reflection amplitude $(r_{h.s.})$ is the ratio between the amplitudes of the backward and forward propagating normal plane-wave components at the middle between two (111) planes of spheres. Therefore, we can extract the effective surface impedance $Z_{eff}(\omega)$ from it using $r_{h.s.} = (Z_{eff} - c\rho_0)/(Z_{eff} + c\rho_0)$. Since we have assumed that the crystal can be effectively replaced by a homogeneous medium, the effective density and effective bulk modulus can be found from

$$Z_{eff} = \frac{\omega \rho_{eff}}{K_z},$$

$$n_{eff}^2 = \frac{\rho_{eff}}{\rho_0} \frac{\kappa_0}{\kappa_{eff}}.$$
(6)

Figure 2(a) shows the dispersion of the rubber/water composite calculated using the layer-multiple-scattering formalism for a filling ratio of 0.4, and the extracted effective bulk modulus and effective density are shown in Fig. 2(d). Below 2.65 kHz, both the effective bulk modulus and density are negative. In the double-negative regime, there exists a singly degenerate band of effective medium with negative group velocity in the dispersion. It is called the double negative (DNG) band. Above 2.65 kHz, the effective density becomes positive and the negative modulus give rise to a gap at the dispersion.

In Figs. 2(b) and 2(c), the transmittance at normal incidence and 60° off normal incidence through the eight layers



FIG. 3. Double acoustic negativity and negative refractive index of rubber spheres in water, fcc arrangement with filling ratio of 74%. (a) Dispersion; (b) and (c) The transmittance through eight layers of the (111) planes at normal and 60° incidence. The open circles are calculated by the multiple-scattering method and the solid line is the approximation using homogeneous media with effective bulk modulus and density [as shown in panel (d)] extracted from the dispersion.

of particles calculated by layer-multiple-scattering formalism, is shown as open circles, compared with the transmittance calculated by assuming the same thickness of the effective medium using parameters just extracted, in solid lines. The excellent agreement reaffirms that it is both qualitatively meaningful and quantitatively accurate to use negative effective bulk modulus and density extracted to describe the medium, and that we have a double-negative medium in exactly the same spirit as Veselago's EM medium. We emphasize here that the result of the double-negative band originates from resonances, and is not a band-folding effect from Bragg scattering. The composite system can be treated as a homogeneous medium in the same spirit as the Veselago's medium in the EM wave.

In Fig. 2(a), there is an extra deaf band of double degeneracy just above the double-negative band and they meet at the zone center. The physical origin of this band can be understood also from the effective medium. When we put a plane-wave solution into Eq. (1), in addition to the expected longitudinal mode $(\vec{v}//\vec{k}, \text{ with dispersion } k^2 = \omega^2 \rho/\kappa)$, we get two extra transverse modes $(\vec{k} \cdot \vec{v} = 0)$ when the effective density is zero. It is equivalent to the longitudinal plasmon mode in EM at a frequency of zero permittivity. The deaf band has a Λ_3 symmetry and cannot be excited by a normal-incidence wave [14], which has a Λ_1 symmetry, and it couples weakly even to the incident wave of an oblique incidence angle. Since this deaf band does not couple with the incident wave and it is not the expected longitudinal mode, we do not use the deaf band in calculating the effective parameters.

When the filling ratio is further increased with the particle fixed in size, Fig. 3 shows the dispersion, effective density/ bulk modulus, and the transmittance through 0° or 60° across eight layers of the composite. As the concentration of particles becomes higher, the resonance becomes stronger. It

results in an even larger frequency width of the doublenegative band of negative density and negative bulk modulus. We see that the effective medium also represents the composite very well in calculating the transmittance of various incidence angles. Again, this is the evidence that the composite can be really treated as an effective medium.

The high contrast between the sound speed in rubber and water makes the double-negative band stay at a very low frequency so that we can use negative density and negative bulk modulus to represent the band. If the contrast in sound speed becomes smaller, we expect the effective medium description degrades in the intermediate frequency regime. For example, the sound speed in the rubber is now set to a higher value of 150 ms⁻¹. Figure 4 shows the corresponding dispersion and transmittance at both normal and 60° off normal incidence. At this case, the double-negative band is shifted to higher frequencies. If we insist on extracting effective bulk modulus and density, we find that they still provide a quantitative description for normal incidence but the agreement between effective medium and exact multiple-scattering results is not good at oblique incidence. In this case, the effective medium description is qualitatively meaningful but not quantitatively correct.

In short, we have demonstrated theoretically the concept of a double-negative acoustic medium (Poynting vector in opposite direction with wave vector) which has a simultaneously negative effective bulk modulus and density. It is the acoustic analog of Veselago's medium having simultaneously negative values of ε and μ . While the negative ε and negative μ in EM metamaterials are typically derived from two types of resonances, the negative modulus and density originate from the monopolar and dipolar resonances of the same structure, one example being soft rubber in water. We emphasize that the double negativity is a consequence of resonance and the resulting negative refraction properties are not





FIG. 4. Double acoustic negativity and negative refractive index of rubber spheres in water, fcc arrangement with filling ratio of 40% and sound speed in rubber being 150 ms⁻¹. (a) Dispersion; (b) and (c) The transmittance through eight layers of the (111) planes at normal and 60° incidence. The open circles are calculated by the multiple-scattering method and the solid line is the approximation using homogeneous media with effective bulk modulus and density [as shown in panel (d)] extracted from the dispersion.

a consequence of a band-folding effect due to Bragg scattering. We note in passing that our results remain true even if we include shear wave components within the particles. We also note that some unique properties of a double-negative medium, such as negative refractive index and subwavelength focusing, are natural consequences.

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