## Metamaterial with Simultaneously Negative Bulk Modulus and Mass Density

Yiqun Ding, Zhengyou Liu,\* Chunyin Qiu, and Jing Shi

Key Lab of Acoustic and Photonic materials and devices of Ministry of Education and Department of Physics, Wuhan University,

Wuhan 430072, China

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We report a metamaterial which simultaneously possesses a negative bulk modulus and mass density. This metamaterial is a zinc blende structure consisting of one fcc array of bubble-contained-water spheres (BWSs) and another relatively shifted fcc array of rubber-coated-gold spheres (RGSs) in epoxy matrix. The negative bulk modulus and mass density are simultaneously derived from the coexistent monopolar resonances from the embedded BWSs and dipolar resonances from the embedded RGSs. The Poisson ratio of the metamaterial also turns negative near the resonance frequency.

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Conventional materials have a positive permittivity  $\varepsilon$ and permeability  $\mu$ . In 1968, Veselago postulated a material with simultaneously negative  $\varepsilon$  and negative  $\mu$  [1] and found that in this double-negative material (DNM), electromagnetic (EM) waves propagate with the wave vector reversed to the energy flow, and the refractive index nbecomes negative, given by  $n = -\sqrt{\epsilon \mu}$ . A number of peculiar phenomena are predicted for the DNM [1-9], including the reverse Doppler shift, reverse Cherenkov radiation, etc. A design of DNM for EM waves was recently proposed by Pendry et al. [2] and later realized by Smith *et al.* [10]. This DNM is a periodical array of both the split-ring and the metallic rods. In response to incident EM waves, the resonances of the split-ring could provide the effective negative permeability, while the resonances of the metallic rods could provide the effective negative permittivity [3].

With the realization of DNM for EM waves, an interesting question arises: whether or not a "double-negative" material is possible for elastic or acoustic waves. If both the mass density and the elastic constant become negative, negative refraction for elastic waves could be expected similarly, with the refractive index n given by

$$n = -\sqrt{\rho/X},\tag{1}$$

where  $\rho$  is the mass density, X = G for the transverse wave or  $X = E \equiv K + \frac{4}{3}G$  for the longitudinal wave, with *K* and *G* being, respectively, bulk modulus and shear modulus. Parameter *E* governs the velocity of the longitudinal waves by  $v_l = \sqrt{\frac{E}{\rho}}$ . Nevertheless, conventional materials have neither a negative mass density nor a negative elastic constant. Recently, it has been shown that a class of three-component phononic crystal with local resonant structure exhibits negative effective mass density (EMD) at (dipolar) resonant frequency [11,12]. A ultrasonic metamaterial consisting of an array of subwavelength Helmholtz resonators was demonstrated to have a negative effective bulk modulus (EBM) at (monopolar) resonant frequency [13]. A phononic crystal consisting of rubber spheres in water was reported to be a double-negative acoustic medium [14]. However, this structure is not a complete or pure DNM. In Fig. 1, we show the complete band structure for it, where solid circles are the band structure shown in Ref. [14] [Fig. 2(a)]. The lower band was regarded as a double-negative band, and the upper was regarded as a deaf band. However, a deaf band can not be deaf along all directions; hence, the overlap of the doublenegative band with other bands means the failure or incompleteness of the description in terms of isotropic effective material parameters (although it may be correct along [111] direction). The lack of strong monopolar and dipolar resonances in the design of the system (with a single structural unit, i.e., rubber spheres in water, it is difficult to get the monopolar and dipolar resonances both strong) is the reason for the failure in obtaining a pure double-negative band. Thus, to realize a true DNM, it is preferable to use a double unit structure, with one specially



FIG. 1 (color online). Full band structure for the reported fcc structure consisting of rubber spheres in water with filling fraction 40% in Ref. [14], calculated with the MST method. Solid circles are the bands along [111] direction given in Fig. 2a of Ref. [14].

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FIG. 2 (color online). (a) *T*-matrix elements for a BWS in epoxy for monopolar wave component l = 0 and dipolar wave component l = 1. Monopolar resonance (l = 0) can be seen. The inset shows a unit cell of the fcc structure and the insides of a structural unit, *E* standing for epoxy, *B* for air bubble, and *W* for water; (b) Band structure for such a structure with filling fraction 20.4%, calculated with MST method; (c) Effective bulk modulus  $K_e$ , shear modulus  $G_e$  and  $E_e = K_e + \frac{4}{3}G_e$  obtained from CPA method; (d) Band structure calculated with the effective parameters.

designed to have very strong monopolar resonance while the other one to have very strong dipolar resonance. In this Letter, we propose that a true and robust DNM can be realized by combining an array of bubble-contained water spheres (BWS) with an array of rubber-coated gold spheres (RGS) in epoxy matrix. The monopolar resonances of the BWSs give rise to the negative bulk modulus, while the dipolar resonances of RGSs give rise to the negative mass density. This is also as yet a realization of simultaneously negative bulk modulus and mass density in a solid based material. In addition, the Poisson ratio of this solid based DNM also becomes negative, which is distinct from a fluid based DNM.

It is well-known that an air bubble in water exhibits strong monopolar resonance in response to acoustic waves [15], and the monopolar resonances of a periodic array of air bubbles in water give rise to a wide band gap for the structure [16–20]. As the EBM of a fluid based structure may turn to negative by monopolar resonances [14], might the EBM of a solid based structure, if designed to contain structural units with built-in monopolar resonances such as air bubbles in water, turn to negative too? To answer this question, we consider a structure consisting of an array of BWSs in epoxy with a fcc lattice, which is shown in the inset of Fig. 2(a). To realize the structure in the lab, small balloons can be used to replace air bubbles, which can also

provide monopolar resonances. By putting small balloons into voids periodically fabricated in the epoxy matrix and filling the voids with water, the desired structure can be obtained. To demonstrate the existence of monopolar resonance in this system, we calculate the Mie scattering matrix for a single BWS in epoxy (the ratio of the radii of the air bubble to the water sphere being 2/23 for the BWS). The monopolar resonance would correspond to a peak of the modulus of the 0th order matrix element varying with the frequency. The material parameters are chosen as following: for air  $\rho = 1.23 \text{ kg/m}^3$ ,  $K = 1.42 \times$  $10^5$  N/m<sup>2</sup>, for water  $\rho = 1000.0$  kg/m<sup>3</sup>,  $K = 2.22 \times$  $10^9 \text{ N/m}^2$ , and for epoxy  $\rho = 1180.0 \text{ kg/m}^3$ ,  $K = 5.49 \times$  $10^9 \text{ N/m}^2$ ,  $G = 1.59 \times 10^9 \text{ N/m}^2$ . The modulus of the low order (i.e., l = 0, 1) matrix elements varying with the frequency are shown in Fig. 2(a). Symbols L, M, N respectively denote the longitudinal, the first transverse, and the second transverse modes. The matrix element XY(X, Y = L, M, N) stands for the conversion from X mode to Y mode during the scattering procedure. The L mode and the N mode are coupled to each other with LN = NL, while the *M* mode is decoupled [21]. We observe a sharp peak for element *LL* of the monopolar component (l = 0), which is obvious evidence for the monopolar resonance. We then perform a band structure calculation for a fcc array of the BWSs in epoxy with a filling fraction of 20.4% by using a Multiple Scattering Theory (MST) method [22]. As observed in Fig. 2(b), the band structure shows a unique feature: corresponding to the monopolar resonance, an absolute gap opens at the resonant frequency 0.35 (in unit of  $2\pi v_t/a$ , where  $v_t$  is the transverse wave speed in the matrix and a is the lattice constant) for the longitudinal modes, while the transverse modes are not influenced by the monopolar resonance of the BWSs. The monopolar component of elastic waves in nature is purely longitudinal (so, there is only one matrix element, LL for l = 0), which means that monopolar resonance can only be induced by longitudinal waves.

We now use the CPA method [23] to estimate the effective parameters for the present system at low frequency, including the EBM, the effective shear modulus, and the effective mass density. Within the framework of CPA, the main task is to seek the self-consistent solution for these effective parameters for an effective medium to ensure that a building block embedded within the effective medium generates no scattering. Figure 2(c) shows the EBM  $K_e$ , the effective shear modulus  $G_e$ , and effective elastic parameter  $E_e = K_e + \frac{4}{3}G_e$  versus the frequency. It is noticed that at the frequencies around the resonance,  $K_e$  and  $E_e$  turn negative; however,  $G_e$  remains positive all over the range. It is also noticed that the negative region of  $E_e$  coincides with the band gap of the longitudinal modes. In Fig. 2(d), we show the dispersion curves calculated by using the effective parameters; a satisfactory agreement with the rigorous band structure calculation is observed.

So far we have gotten a material with negative EMB. As was already known, negative EMD can be realized in a three-component system with local dipolar resonances [12]. To tune the negative EMD into the same frequency region as the negative EMB obtained above, here, we use a structure consisting of a fcc array of rubber-coated gold spheres (RGSs) in epoxy, with a filling fraction of 9.77%. The rubber coating has an inner-to-outer radius ratio of 15/18. The inset in Fig. 3(a) schematically shows this structure. Figure 3(a) shows the Mie scattering matrix for a single RGS in epoxy; resonant peaks can be clearly seen in the matrix elements of dipolar components (l = 1), which are the evidence of dipolar resonances. The dipolar resonance opens an absolute band gap extending from 0.35 to 0.5 (in unit of  $2\pi v_t/a$ ), as shown in Fig. 3(b). The material parameters used in these calculations are for gold,  $\rho = 19500.0 \text{ kg/m}^3$ ,  $K = 1.80 \times 10^{11} \text{ N/m}^2$ , G = $2.99 \times 10^{10} \text{ N/m}^2$ ; and for rubber,  $\rho = 1300.0 \text{ kg/m}^3$ ,  $K = 2.20 \times 10^9 \text{ N/m}^2$ ,  $G = 9.98 \times 10^6 \text{ N/m}^2$ . Since both the longitudinal and the transverse waves can induce dipolar resonances [12], a common band gap opens for both the longitudinal and the transverse waves.

A simple model had shown that the dipolar resonance can result in a negative EMD [12]. Here, we use the CPA method to calculate the EMD for the structure, as shown in Fig. 3(c). As expected, the EMD turns to negative in the gap region, which makes both the longitudinal and the



FIG. 3 (color online). (a) *T*-matrix elements for a RGS in epoxy for l = 0 and l = 1, dipolar resonance (l = 1) is clearly seen. The inset shows a unit cell of the fcc structure, and the insides of a structural unit, *E* standing for epoxy, *G* for gold and *R* for rubber; (b) Band structure for such a structure with filling fraction 9.77%, calculated with MST method; (c) EMD derived from CPA method; (d) Band structure calculated with the effective parameters.

transverse waves attenuated. Now, we calculate the dispersion relation for the structure based on the effective parameters. As shown in Fig. 3(d), the dispersion relation resembles the rigorous band structure in Fig. 3(b) very much, indicating that the effective parameters obtained from CPA method characterize this structure very well. It is noted that there is an overlap in frequency domain for the negative EMD and the negative EMB discussed above, which is critical to obtain a DNM as shown below.

To obtain simultaneously negative EMB and EMD, we combine the two fcc lattices, say, the fcc lattice by BWSs and the fcc lattice by RGSs, with a shift of [1/4, 1/4, 1/4]in epoxy, to form a zinc blende structure as shown in the inset of Fig. 4(a). This new structure contains both the monopolar resonances and the dipolar resonances. Figure 4(a) shows the band structure calculated with the MST method. It is interesting to note that in the overlap region of the negative EBM and EMD mentioned above, a new pass band, with normalized frequency extending from 0.373 to 0.414, emerges, which is a typical negative refraction band with a shape of negative group velocity [24]. Therefore, this is a band with simultaneously negative EBM and EMD. To demonstrate that the pass band is due to the double negativity of the material parameters, we calculate the dispersion relation for this structure with the effective parameters obtained with CPA method, which is shown in Fig. 4(b). We note that the dispersion relation satisfactorily replicates the rigorous band structure at low frequency. The pass band is exactly the region with the EMD and the elastic parameter  $E_e$  both negative. Comparing Fig. 4(a) with Fig. 2(b), it is found that the transverse waves in the gap region of Fig. 2(b) disappear now because of the negative EMD due to the dipolar resonances in the new structure. It is interesting to note that with the EMB becoming negative, the Poisson ratio becomes singular at the resonant frequency too. As shown in Fig. 5, we observe a frequency region in which the Poisson ratio also turns negative. This feature is distinct from a structure with a fluid base. Although a zinc blende



FIG. 4 (color online). (a) Band structure for a zinc blende structure consisting of both BWSs and RGSs in epoxy, calculated with the MST method. The inset shows a unit cell of the zinc blende structure. (b) Band structure for this structure calculated with the effective parameters.



FIG. 5. Poisson ratio for the zinc blende structure, calculated with the effective parameters from CPA method.

structure is employed to demonstrate the realization of a DNM for the convenience of calculation, any structure composed of these BWSs and RGSs, either order or disorder, also possesses the same double negativity. Since local resonances (either monopolar or dipolar) are the main mechanism for gap opening, order is not an important issue for the structure design. Therefore, the DNM obtained in this way is robust.

In summary, we demonstrate that a metamaterial, possessing simultaneously negative bulk modulus and mass density, can be achieved by combining two types of structural units with built-in monopolar and dipolar resonances. While the monopolar resonances give rise to the negative bulk modulus, the dipolar resonances give rise to the negative mass density. This brings up a new class of material for elastic or acoustic waves with fascinating properties, such as negative refraction, inverse Doppler Effect, and so on.

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<sup>\*</sup>To whom all correspondence should be addressed: zyliu@whu.edu.cn