

# Homogenization of acoustic metamaterials of Helmholtz resonators in fluid

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By using a two-step homogenization approach, we derive analytical formulas of effective mass density  $\rho_e$  and effective bulk modulus  $B_e$  for two- and three-dimensional acoustic metamaterials of Helmholtz resonators (HRs) in fluid. A negative  $B_e$  is found at certain frequencies due to the monopolar resonance, leading to a low-frequency acoustic band gap. A unified picture is presented for metamaterials of HRs and three-component metamaterials of negative  $\rho_e$ . Our work supports recent observations in a one-dimensional array of HRs [N. Fang *et al.*, *Nat. Mater.* **5**, 452 (2006)] and presents important high-dimensional extensions for exploring more fascinating phenomena.

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Recently, acoustic and/or elastic metamaterials (artificial structured materials<sup>1,2</sup>) have received considerable interest due to their exotic acoustic and/or elastic properties.<sup>3-17</sup> In particular, their effective mass density and modulus can be singly or simultaneously negative,<sup>3-13</sup> allowing intriguing phenomena for sound waves such as low-frequency band gaps,<sup>3-10</sup> negative refraction, and superlensing.<sup>1,12-14</sup>

Negative effective mass density or modulus can occur at certain frequencies if an appropriate resonance is included into the structures.<sup>3-13</sup> A famous example is the three-component phononic crystal with locally resonant structures,<sup>3</sup> which exhibits a negative effective mass density  $\rho_e$  due to a dipolar resonance.<sup>4,5</sup> Very recently, a low-frequency band gap was demonstrated in a one-dimensional (1D) array of Helmholtz resonators (HRs).<sup>6</sup> By using further retrieval analysis of wave-scattering coefficients,<sup>7-9</sup> a negative effective bulk modulus  $B_e$  was confirmed in the 1D metamaterials of HRs.<sup>9</sup> Unlike other designs for negative  $B_e$  using air bubbles or soft-rubber spheres,<sup>12,13</sup> this metamaterial consists of a rigid body but still possesses a sizable working frequency range. However, high-dimensional metamaterials of HRs, which are essential for more fascinating phenomena,<sup>6,14,15</sup> remain unexplored.

In this Brief Report, we study two-dimensional (2D) and three-dimensional (3D) acoustic metamaterials consisting of cylindrical and spherical HRs in fluid (such as water or air), respectively. By using a two-step homogenization approach, we can derive analytical formulas for the  $\rho_e$  and  $B_e$  within coherent-potential approximation (CPA).<sup>18</sup> The accuracy of these formulas is confirmed by accurate retrieval results using multiple-scattering techniques. We show that our metamaterials of HRs can exhibit a negative  $B_e$  at certain frequencies due to a monopolar resonance. Systematic analyses are done for the frequency range of negative  $B_e$  and the low-frequency-limit behavior. Based on our derivations, a unified picture is presented for metamaterials of HRs and three-component metamaterials of negative  $\rho_e$ .

Our HRs are cylindrical and/or spherical rigid shells with uniformly distributed slits and/or holes, as shown in Fig. 1(c). The inner and outer radii of the shells are  $r_2$  and  $r_1$ , respectively. The background fluid has a mass density  $\rho_0$  and bulk modulus  $B_0$ . Our HR becomes a conventional HR if only a single slit and/or hole exists in the shell. It has been shown that for acoustic waves of long wavelength ( $\lambda_0$ ), a rigid-body slab with small slits and/or holes (with a size  $\Delta < d/4$  and period  $d < \lambda_0/4$ ) is effectively equivalent to a uniform slab of a thickness  $h/n$ , density  $\rho_0 n^2$ , and bulk modulus  $B_0$ , where  $h$  is the thickness of the rigid-body slab,  $n = 1/f$ , and  $f$  is the filling ratio of slits and/or holes ( $f = d/\Delta$  for slits).<sup>17</sup> In the following, we will use this conclusion and

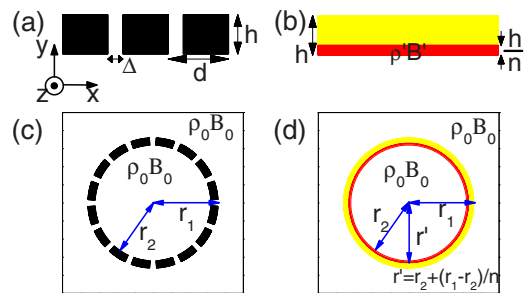


FIG. 1. (Color online) (a) A rigid-body slab with periodic slits (holes) and (b) its equivalence of a two-layer slab. The slabs have a thickness  $h$  and are extending in the  $X$ - $Z$  plane. The slits and/or holes have a size  $\Delta$ , period  $d$ , and filling ratio  $f$  ( $f = d/\Delta$  for slits). The top layer has a thickness  $h - h/n$  but the same fields at their two surfaces, and the bottom layer has a thickness  $h/n$ , density  $\rho' = \rho_0 n^2$  with  $n = 1/f$ , and modulus  $B' = B_0$ . (c) A 2D (3D) HR of rigid cylindrical (spherical) shell with uniformly distributed slits (holes) and (d) its equivalence of a two-layer shell made of the slabs in (a) and (b), respectively.

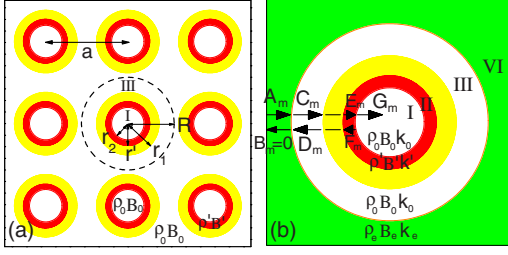


FIG. 2. (Color online) (a) A 2D (3D) acoustic metamaterial consisting of a square (simple cubic) lattice of the cylindrical (spherical) shells in Fig. 1(d) and (b) a coated shell in an effective medium of \$(\rho\_e, B\_e)\$.

replace the wall of the HR by its equivalence of a two-layer shell,<sup>19</sup> where the inner layer has a thickness \$(r\_1 - r\_2)/n\$, density \$\rho' = \rho\_0 n^2\$, and modulus \$B' = B\_0\$, and the outer layer has a thickness \$(r\_1 - r\_2)(1 - 1/n)\$ but the same fields at its two surfaces. Here, we introduce the outer layer to make the total wall thickness unchanged.

We first consider a 2D metamaterial consisting of a square array of above cylindrical HRs in fluid, as shown in Fig. 2(a). The filling ratio of HRs is \$f\_s = \pi r\_1^2/a^2\$, where \$a\$ is the lattice constant. Set \$\mathbf{r} = (x, y) = (r, \theta)\$ in the plane perpendicular to the axes of HRs. For harmonic acoustic waves with angular frequency \$\omega\$, the pressure field \$P(\mathbf{r})e^{i\omega t}\$ obeys the two-dimensional Helmholtz equation,

$$(\nabla^2 + k^2)P = 0, \quad (1)$$

which is subjected to continuities of \$P\$ and \$\frac{1}{\rho} \frac{\partial P}{\partial r}\$ at the layer interfaces of each HR (set \$\mathbf{r} = 0\$ at the center of HR). The wave numbers in the background and inner walls of HRs are given by \$k\_0 = \omega \sqrt{\rho\_0/B\_0}\$ and \$k' = \omega \sqrt{\rho'/B'} = nk\_0\$, respectively.

Then, we derive analytical formulas for the \$\rho\_e\$ and \$B\_e\$ of HRs by the CPA method.<sup>18</sup> We consider a “circular unit cell” (coated HR) of radius \$R = r\_1/\sqrt{f\_s}\$ and replace the metamaterial outside by a uniform effective medium of \$\rho\_e, B\_e\$, and \$k\_e = \omega \sqrt{\rho\_e/B\_e}\$ [see Fig. 2(b)]. The pressure field \$P\$ can be decomposed into incident and scattering cylindrical waves [represented by Bessel (\$J\_m\$) and Hankel (\$H\_m\$) functions, respectively]: \$P^I = \sum\_m G\_m J\_m(k\_0 r) e^{im\theta}\$ when \$r < r\_2\$, \$P^{II} = \sum\_m [E\_m J\_m(k' r) + F\_m H\_m(k' r)] e^{im\theta}\$ when \$r\_2 < r < r' \equiv r\_2 + (r\_1 - r\_2)/n\$, \$P^{III} = \sum\_m [C\_m J\_m(k\_0 r) + D\_m H\_m(k\_0 r)] e^{im\theta}\$ when \$r\_1 < r < R\$, and \$P^{VI} = \sum\_m [A\_m J\_m(k\_e r) + B\_m H\_m(k\_e r)] e^{im\theta}\$ when \$r > R\$. For the outer layer of HR, the fields will be the same at its two surfaces so that \$P^{II}(r') = P^{III}(r\_1)\$ and \$\frac{1}{\rho'} \frac{\partial P^{II}(r')}{\partial r} = \frac{1}{\rho\_0} \frac{\partial P^{III}(r\_1)}{\partial r}\$. Combining \$P^I(r\_2) = P^{II}(r\_2)\$, \$\frac{1}{\rho\_0} \frac{\partial P^I(r\_2)}{\partial r} = \frac{1}{\rho'} \frac{\partial P^{II}(r\_2)}{\partial r}\$, \$P^{III}(R) = P^{VI}(R)\$, and \$\frac{1}{\rho\_0} \frac{\partial P^{III}(R)}{\partial r} = \frac{1}{\rho\_e} \frac{\partial P^{VI}(R)}{\partial r}\$, the \$m\$th order scattering coefficient \$B\_m/A\_m\$ of the coated HR can be obtained and \$B\_m/A\_m = 0\$ (which “defines” the effective medium) when

$$\begin{aligned} & \frac{1}{\rho_0} k_0 J'_m(k_0 R) - J_m(k_0 R) \frac{1}{\rho_e} k_e J'_m(k_e R) / J_m(k_e R) \\ & - \frac{1}{\rho_0} k_0 H'_m(k_0 R) - H_m(k_0 R) \frac{1}{\rho_e} k_e J'_m(k_e R) / J_m(k_e R) \\ & = - \frac{1}{\rho_0} k_0 J'_m(k_0 r_1) - J_m(k_0 r_1) U_m \\ & = - \frac{1}{\rho_0} k_0 H'_m(k_0 r_1) - H_m(k_0 r_1) U_m \end{aligned} \quad (2)$$

where

$$U_m = \frac{1}{\rho'} k' \frac{J'_m(k' r') + H'_m(k' r') P_m}{J_m(k' r') + H_m(k' r') P_m}$$

and

$$P_m = - \frac{\frac{1}{\rho'} k' J'_m(k' r_2) - J_m(k' r_2) \frac{1}{\rho_0} k_0 J'_m(k_0 r_2) / J_m(k_0 r_2)}{\frac{1}{\rho'} k' H'_m(k' r_2) - H_m(k' r_2) \frac{1}{\rho_0} k_0 J'_m(k_0 r_2) / J_m(k_0 r_2)}.$$

We note that the first and second terms in Eq. (2) and \$P\_m\$ are just the \$m\$th order scattering coefficients of a cylinder with radius \$R\$, \$\rho\_e\$, and \$B\_e\$ in a matrix of \$\rho\_0, B\_0\$, the HR, and the core cylinder, respectively. If we only consider the scattering of cylindrical waves of the two lowest orders \$m=0\$ and 1, \$\rho\_e\$ and \$B\_e\$ can be numerically calculated after solving \$\frac{1}{\rho\_e} k\_e J'\_0(k\_e R) / J\_0(k\_e R)\$ and \$\frac{1}{\rho\_e} k\_e J'\_1(k\_e R) / J\_1(k\_e R)\$ from Eq. (2).

Under the approximation of long wavelength [\$k\_0 a, k\_e a \ll 1\$, thus \$k'(r' - r\_2) = k\_0(r\_1 - r\_2) \ll 1\$] and small slit size (\$n = 1/f \gg 1\$), Eq. (2) becomes<sup>20</sup>

$$\frac{1}{B_e} = \frac{1}{B_0} \left( 1 - f_s + \frac{f_s r_2 / r_1}{1 - \omega^2 / \omega_R^2} \right) \quad \text{for } m = 0, \quad (3)$$

$$\rho_e = \rho_0 [1 + f_s / (L - 1)] / (1 - f_s) \quad \text{for } m = 1, \quad (4)$$

where \$L=2\$ in the 2D case and the resonant frequency is given by

$$\omega_R = v_0 \sqrt{L / [n r_2 (r_1 - r_2)]}. \quad (5)$$

Here, \$v\_0 = \sqrt{B\_0/\rho\_0}\$ is the sound velocity in the background fluid. If only a single slit exists in the wall of HR (\$n \approx 2\pi r\_2/\Delta\$, where \$\Delta\$ is the slit width), Eq. (5) will become the conventional HR frequency in 2D, namely, \$\omega\_R = v\_0 \sqrt{\Delta / [\pi r\_2^2 (r\_1 - r\_2)]}\$.<sup>21</sup> For the case without slits (\$n \to \infty\$), \$\omega\_R\$ will approach zero and Eq. (3) will reduce to the formula of \$B\_e\$ for rigid rods in fluid [where \$\rho\_e\$ is still given by Eq. (4)],<sup>16</sup> namely,

$$B_e = B_0 / (1 - f_s). \quad (6)$$

From Eq. (3), we can see that the \$B\_e\$ of HRs depends on frequency. At the low-frequency limit (\$\omega \to 0\$), the effective bulk modulus is given by

$$B_e = B_0 / (1 - f_s + f_s r_2 / r_1). \quad (7)$$

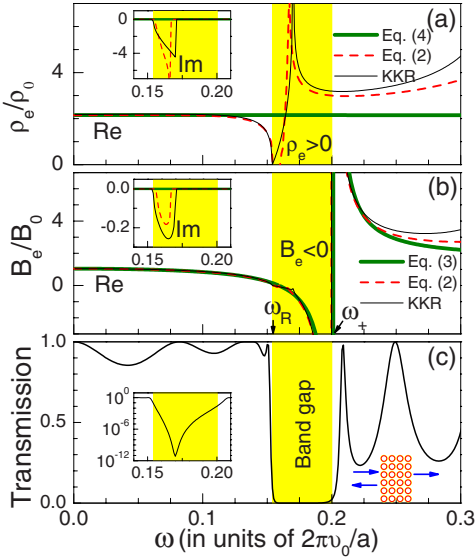


FIG. 3. (Color online) (a) The effective mass density and (b) bulk modulus for the 2D acoustic metamaterial in Fig. 2(a), and (c) the transmission spectrum for normal incidence of a plane wave on a four-layer metamaterial. The parameters are  $r_1=0.36a$ ,  $r_2=0.32a$ , and  $f=6.39 \times 10^{-3}$  (thus  $n=156.5$ ,  $r'=0.320\,256a$ ,  $\rho'=2.45 \times 10^4\rho_0$ , and  $B'=B_0$ ).

Here,  $B_e$  will be zero at the resonant frequency  $\omega_R$ . For a frequency range above  $\omega_R$  ( $\omega_R < \omega < \omega_+$ ),  $B_e$  becomes negative, where the upper frequency edge is given by

$$\omega_+ = \omega_R \sqrt{1 + f_s r_2^2 / [r_1(1 - f_s)]}. \quad (8)$$

Since the effective density  $\rho_e$  will always be positive [by Eq. (4)], an acoustic band gap (with an imaginary  $k_e$ ) will be formed in the frequency range with negative  $B_e$ .

Similar CPA derivations can also be done for 3D metamaterials consisting of HRs of spherical shells with uniformly distributed holes. In 3D,<sup>22</sup> the Bessel ( $J_m$ ) and Hankel ( $H_m$ ) functions in Eq. (2) should be replaced by spherical Bessel ( $j_m$ ) and Hankel ( $h_m$ ) functions, respectively. Under similar approximation, Eqs. (3)–(8) will also be obtained where  $f_s$  becomes the volume ratio of spherical HRs ( $f_s = \frac{4}{3}\pi r_1^3/a^3$  in a simple cubic lattice) and  $L=3$  in the 3D case. For HRs of single hole ( $n \approx 4\pi r_2^2/S_h$ , where  $S_h$  is the area of the hole), Eq. (5) will become the conventional formula of HR frequency, namely,  $\omega_R = v_0 \sqrt{S_h / [\frac{4}{3}\pi r_2^3(r_1 - r_2)]}$ .<sup>21</sup> When no holes exist in the shells ( $n \rightarrow \infty$ ),  $\omega_R$  will approach zero and Eqs. (4) and (6) can be obtained for rigid spheres in fluid.

To check the validity of the above analytic formulas, we apply the multiple-scattering or Koringa-Kohn-Rostoker (KKR) method (including high-order cylindrical waves)<sup>23</sup> to simulate normal incidence of a plane wave on a slab of above 2D acoustic metamaterial [in Fig. 2(a)]. Then, the effective parameters ( $B_e, \rho_e$ ) can be retrieved from the transmission and reflection coefficients.<sup>7–9</sup> The retrieving is easy for a one-layer slab but the results will be the same for more layers. In Fig. 3, we show the accurate KKR results of ( $B_e, \rho_e$ ) and those from Eqs. (2)–(4) for a square lattice of cylindrical HRs in fluid where  $r_1=0.36a$ ,  $r_2=0.32a$ , and

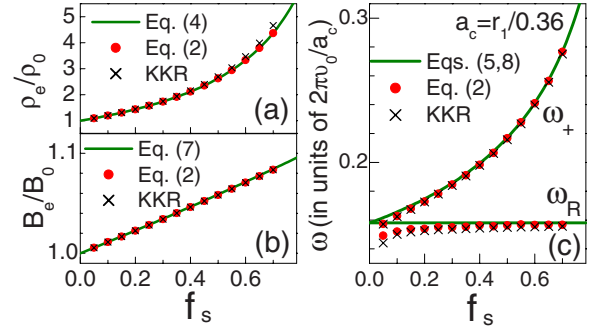


FIG. 4. (Color online) (a) The effective mass density and (b) bulk modulus when  $\omega r_1/v_0 \rightarrow 0$ , and (c) the edges of the frequency range with a negative  $B_e$  as functions of the filling fraction  $f_s \equiv \pi r_1^2/a^2$  for the 2D acoustic metamaterials in Fig. 2(a). The parameters of  $r_1/r_2$  and  $f$  are as in Fig. 3.

$f=6.39 \times 10^{-3}$  (thus,  $n=156.5$ ,  $r'=0.320\,256a$ ,  $\rho'=2.45 \times 10^4\rho_0$ , and  $B'=B_0$ ). A frequency range [ $\tilde{\omega} \equiv \omega_0 a / (2\pi\nu_0) \in (0.154, 0.2)$ ] with a negative  $B_e$  and positive  $\rho_e$  can be found above the resonant frequency. As a result, a low-frequency stop band can be observed in the transmission spectrum for a metamaterial slab [see Fig. 3(c)]. We note that the transmission dip at  $\tilde{\omega}=0.170$  corresponds to a peak of  $\rho_e=9.77-i4.45$  (with  $B_e=-0.194-i0.09$  which will not bring a gain or loss; KKR results). This peak of  $\rho_e$  cannot be captured by Eq. (4) with the long-wavelength approximation and it is usually understood as a result of band folding. However, we find that this feature can still be obtained by numerically solving Eq. (2) for the lowest 2 orders of cylindrical waves. For higher frequencies ( $\tilde{\omega} > 0.25$ ), the scattering of high-order cylindrical waves will be important and more accurate KKR calculations are needed.

Figure 4 shows the  $\rho_e$  and  $B_e$  at the low-frequency limit ( $\omega r_1/v_0 \ll 1$ ) and the frequency edges of negative  $B_e$  with varying the filling fraction  $f_s$  of HRs. The frequency range of negative  $B_e$  is found wider with increasing the filling ratio of HRs. Excellent agreement is found between our analytical formulas and KKR results. Recently, a better impedance match and higher refractive index were found in 2D HRs than rigid rods by finite-difference time-domain simulations.<sup>7</sup> This result was explained by a smaller  $B_e$  of HRs ( $B_e=B_0$ ) than rods. We note that our new Eq. (7) can present a better understanding for this phenomenon.<sup>20</sup>

By now, we have shown that our metamaterials of HRs exhibit a negative effective bulk modulus at certain frequencies due to a monopolar ( $m=0$ ) resonance. For the HR, the fluid inside the heavy shell ( $\rho'=\rho_0 n^2 \gg \rho_0$ ) will retain a cylindrical and/or spherical shape. However, its volume can still periodically vary (since  $B'=B_0$ ) giving rise to the monopolar resonance. Strong monopolar resonances can also exist in air bubbles<sup>11,13</sup> or soft-rubber spheres<sup>12</sup> in water, leading to a negative  $B_e$ .<sup>12,13</sup> Such monopolar resonances can still survive even when a solid matrix is used (e.g., water-coated air bubbles in solid).<sup>13</sup> So if a solid-based structure with a negative  $B_e$  is desired, fluid-coated HRs in a solid matrix can be similarly applied. By combining our HRs with the metamaterials of a negative  $\rho_e$ , a double negative metamaterial can be constructed,<sup>13</sup> enabling negative refrac-

tion and superlensing of acoustic waves. We note that our HRs can also exhibit an arbitrary positive  $B_e$  [ $0 < B_e < B_0/(1-f_s+f_s r_2/r_1)$  or  $B_e > B_0/(1-f_s)$ ] but normal  $\rho_e$  [by Eq. (4)] at certain frequencies ( $\omega < \omega_R$  or  $\omega > \omega_+$ ). When an additional metamaterial with an anisotropic  $\rho_e$  but normal  $B_e$  is provided, a fascinating acoustic cloaking may also be realized.<sup>15</sup>

A coated-cylinder and/or coated-sphere structure exists in our acoustic metamaterials of HRs and the locally resonant sonic materials with soft rubber.<sup>3</sup> However, a negative effective mass density was found in the latter.<sup>4,5</sup> To present a unified picture for these two metamaterials, we mathematically consider metamaterials of soft shells in fluid [by using  $B' = B_0/n^2 \ll B_0$  and  $\rho' = \rho_0$  in Fig. 1(d)]. Under the long-wavelength approximation [ $k_0 a, k_e a \ll 1$ , thus  $k'(r' - r_2) \ll 1$ ], Eq. (2) becomes<sup>20</sup>

$$\frac{1}{B_e} = \frac{1}{B_0} \left[ 1 + f_s (Ln - 1) \left( 1 - \frac{r_2}{r_1} \right) \right], \quad (9)$$

$$\frac{\rho_e - \rho_0}{\rho_e + \rho_0} = -f_s + f_s \frac{Lr_2/(r_1 + r_2)}{1 - \omega^2/\omega_R'^2}, \quad (10)$$

for  $m=0$  and 1, respectively, where the resonant frequency is given by

$$\omega'_R = v_0 \sqrt{(L - 1 + r_1/r_2)/[nr_1(r_1 - r_2)]}. \quad (11)$$

From Eq. (10), a negative effective mass density can be seen due to a dipolar ( $m=1$ ) resonance, which agrees with recent explanations.<sup>4,5</sup> For the soft shell, the fluid inside will retain the shape and volume and will vibrate in a certain direction, giving rise to the dipolar resonance.

In summary, we have proposed a two-step homogenization approach to derive analytical formulas of  $\rho_e$  and  $B_e$  for 2D and 3D acoustic metamaterials of HRs in fluid. A negative  $B_e$  was found at certain frequencies due to the monopolar resonance, leading to a low-frequency acoustic band gap. A unified picture was also presented for metamaterials of HRs and three-component metamaterials of negative  $\rho_e$ . Our work supported recent observations in 1D array of HRs and presented important high-dimensional extensions for exploring more fascinating acoustic phenomena.

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<sup>19</sup>Based on the coordinate transformation method, the two-layer slab can be transformed to a one-layer slab with the same slab thickness. However, anisotropic effective parameters will be found. Here, we use this two-layer slab for convenience of analytical derivation.

<sup>20</sup>See EPAPS Document No. E-PRBMDO-77-015817 for derivation of Eqs. (3), (4), (9), and (10) and comparison of our formulas with FDTD results. For more information on EPAPS, see <http://www.aip.org/pubservs/epaps.html>

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