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SUPPLEMENTARY INFORMATION
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"Soft 3D Acoustic Metamaterial with Negative Index"

Details of the calculation method:

The complex-valued effective wavenumber k (or the effective refractive index $n = k/k_0$ with k_0 the wavenumber for the reference medium) for a medium consisting of a random distribution of polydisperse scatterers can be calculated within the framework of various multiple-scattering theories based on the scatterer volume fraction. For an overview of these models, one can refer to the book by Tsang *et al.* for more information[1]. In all these perturbative approaches, the effective wavenumber k is written as follows:

$$k^2 = \left(\frac{\omega}{v} + \mathrm{j}\alpha\right)^2 = k_0^2 + \int_a \left(\delta_1 \eta(a) + \delta_2 \eta^2(a) + \ldots\right) \mathrm{d}a \qquad \text{(S1)}$$

where $\eta(a)$ is the number of spheres of radius a per unit volume, $\phi = \int_a \eta(a) \times \left(4\pi a^3/3\right) \mathrm{d}a$ is the total volume fraction of spheres. Although the value for δ_1 is the same regardless of the model[1], there is some controversy over the proper value for δ_2. Although many authors have provided more sophisticated expressions for the effective wavenumber k (see, for example, the Lloyd-Berry formula[2], revisited by Linton and Martin[3]), in our work, we applied the formula proposed by Waterman-Truell[4], which is widely used in physics for somewhat concentrated suspensions:

$$k^2 = k_0^2 + \int_a \left(4\pi\eta(a) f_a(0) + \frac{4\pi^2\eta^2(a)}{k_0^2}\left\{\left[f_a(0)\right]^2 - \left[f_a(\pi)\right]^2\right\}\right) \mathrm{d}a \qquad \text{(S2)}$$

where $f_a(0)$ and $f_a(\pi)$ are the forward and backward scattering functions for a single sphere of radius a, respectively.

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In the framework of Waterman and Truell, Aristégui and Angel[5] have derived the complex-valued mechanical constitutive parameters (effective mass density ρ & bulk modulus B) for a random distribution of polydisperse scatterers immersed in a non-viscous fluid:

$$\rho = \rho_0 \left\{ 1 + \int_a \left(\frac{2\pi\eta(a)}{k_0^2} \left[f_a(0) - f_a(\pi) \right] \right) \mathrm{d}a \right\} \quad \text{(S3a)}$$

$$B = \rho_0 c_0^2 \left\{ 1 + \int_a \left(\frac{2\pi\eta(a)}{k_0^2} \left[f_a(0) + f_a(\pi) \right] \right) \mathrm{d}a \right\}^{-1} \quad \text{(S3b)}$$

From these relations, one can also easily obtain the effective acoustic impedance Z:

$$Z = \rho \frac{\omega}{k} = \frac{kB}{\omega} \quad \text{(S4)}$$

Measurements of the material parameters for metafluid constituents:

The calculations of the complex-valued effective acoustic properties (k and Z) and the mechanical constitutive parameters (ρ and B) of our metafluid require knowledge of the material parameters of the matrix and inclusions. Our host matrix acoustically behaved like water: its mass density and phase velocity are $\rho_0 = 1000$ kg/m^3 and $v_0 = 1500$ m/s, respectively. On the other hand, our microbeads made of a macroporous soft silicone rubber, which required proper acoustic characterization. Using this material, we were able to produce large monoliths in the shape of thin disks with a 30-mm diameter and thicknesses varying from 2 to 4 mm (Fig. S1a). Thus, we directly measured the following material parameters: mass density $\rho_1 = 600$ kg/m^3; phase velocity $v_L = 80$ m/s and attenuation $\alpha_L = 60$ Np/mm/MHz$^{1.5}$, for the longitudinal waves; and $v_T = 40$ m/s and $\alpha_T = 200$ Np/mm/MHz$^{1.5}$, for the shear waves. As mentioned in the main text, we then produced microbeads of this macroporous material (Fig. S1b) using a simple microfluidic device.

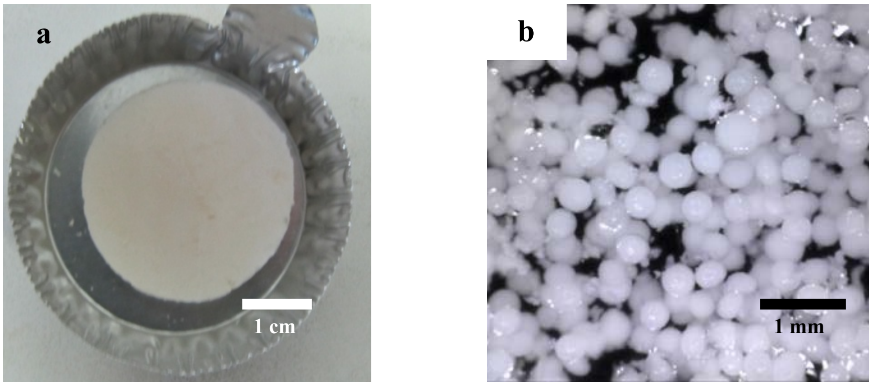

Figure S1 (**a**) Photograph of a slab of our macroporous soft silicon rubber with a porosity of approximately 40%. The smoothness of the disk surface allowed for the direct measurement of its acoustic properties. (**b**) Optical microscopy image of macroporous soft silicone rubber microbeads embedded in a water-based gel matrix. The mean radius <*a*> was 160 µm, and the size dispersion was 25%. The volume fraction Φ_0 was 20%.

Calculations of the effective acoustic properties:

By substituting the measured material parameters into Eqs. S2 and S4, we calculated the real and imaginary parts of both the effective acoustic wavenumber k and effective impedance Z for our metafluid sample displayed in Fig. S2. The model calculations indicated that $\alpha \geq 3$ mm^{-1} (with $\alpha = \text{Im}(k)$ from Eq. S1) in the investigated ultrasonic frequency range of 50 kHz - 500 kHz. Such a large value of the attenuation coefficient α demonstrates that in our experiments, the multi-reflected echoes S_2, S_3, … between the transmitter and the receiver were drastically attenuated in comparison with the directly transmitted pulse S_1. When a typical millimeter distance z between the transducers (z = 1 mm) is considered, the multi-reflected echoes S_2, S_3, … can be neglected because their amplitudes are much lower than that of the directly transmitted pulse S_1: $\left|S_2 / S_1\right| = \exp(-\alpha \times 2z) \leq 2.5 \times 10^{-3}$, $\left|S_3 / S_1\right| = \exp(-\alpha \times 4z) \leq 6 \times 10^{-6}$, ...

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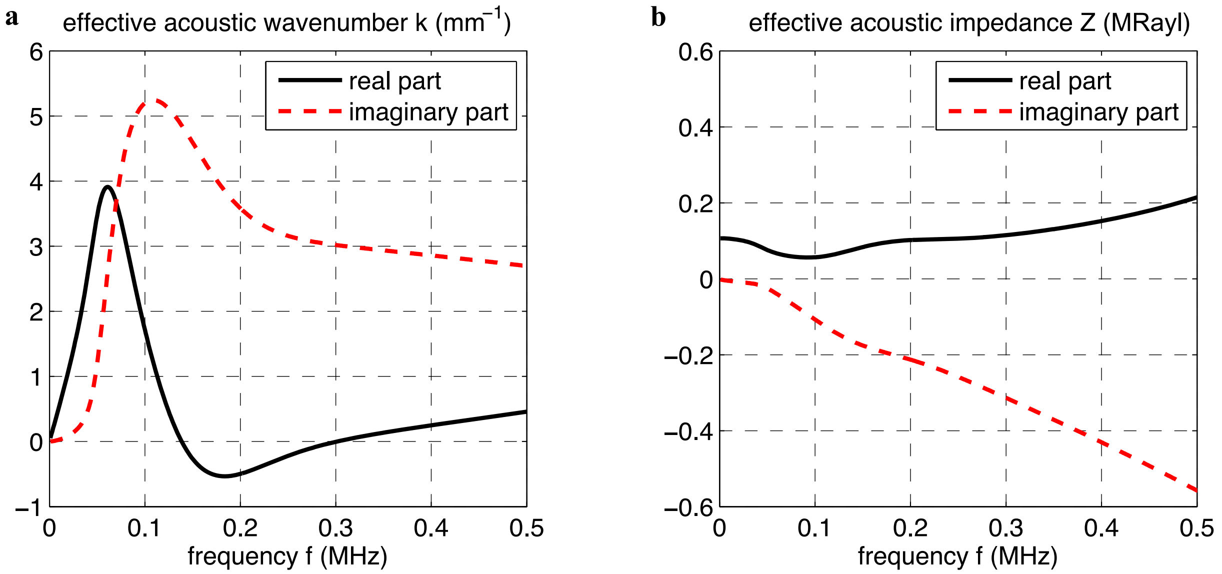

Figure S2 Predictions of the real and imaginary parts of the effective acoustic properties of our metafluid sample. (**a**) The complex-valued effective acoustic wavenumber *k* and (**b**) the effective acoustic impedance *Z* were determined using Eqs. S2-S4, and the material parameters were obtained through direct-contact measurements performed on large soft silicon rubber monoliths and on the pure water-based gel matrix. The mean radius <*a*> was 160 µm, and the size dispersion was 25%. The volume fraction Φ_0 was 20%.

Calculations of the effective mechanical constitutive parameters:

From Eqs. S3 and the measured material parameters, we also determined the real and imaginary parts of both the effective mass density ρ and the effective bulk modulus *B* for our metafluid sample, as shown in Figs. S3. Although the real part of the effective wavenumber *k* (or the effective acoustic index $n = k/k_0$) is negative near 200 kHz (Fig. S2a), the model predicts that the real part of the mass density ρ is positive (Fig. S3a), whereas that of the bulk modulus *B* is negative (Fig. S3b). Therefore, the term “double-negative metamaterials” may be inappropriate to refer to such dissipative metamaterials, as explained by Dubois *et al.*[6]

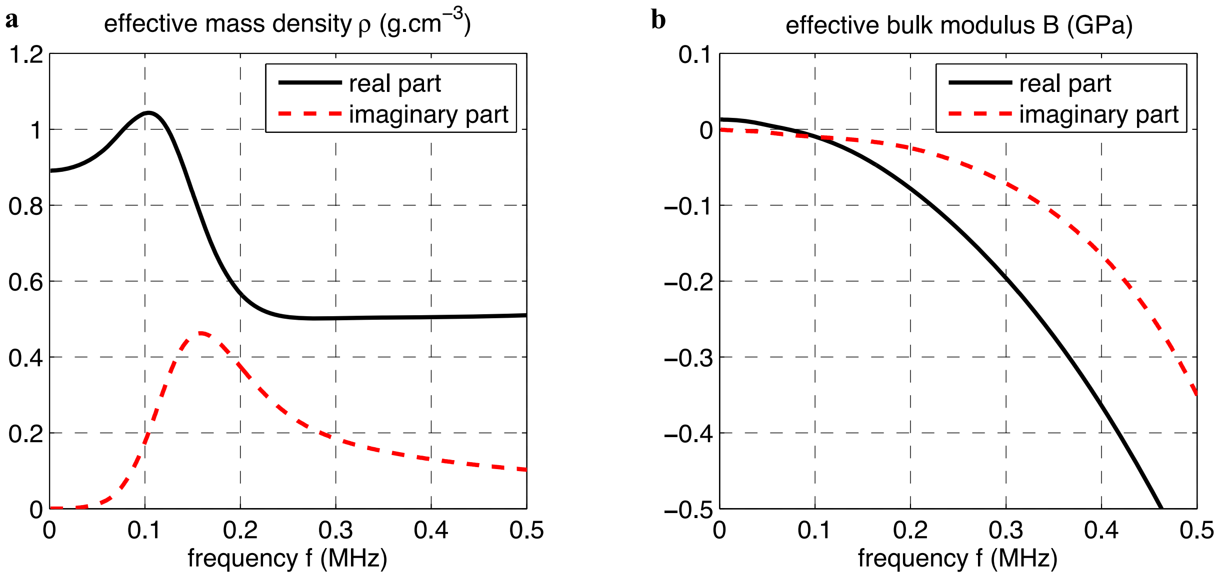

Figure S3 Predictions of the real and imaginary parts of the effective mechanical constitutive parameters of our metafluid sample. (**a**) The complex-valued effective mass density ρ and (**b**) the complex-valued effective bulk modulus B were determined using Eq. 3, and the material parameters were obtained through direct contact measurements performed on soft silicon rubber monoliths and the pure water-based gel matrix. The mean radius $\langle a \rangle$ was 160 µm, and the size dispersion was 25%. The volume fraction Φ_0 was 20%.

References:

1. L. Tsang, J. A. Kong, K.-H. Ding, and C. O. Ao, *Scattering of Electromagnetic Waves: Numerical Simulations*. Wiley, New York (2001).

2. Lloyd, P. & Berry, M. V. Wave propagation through an assembly of spheres IV. Relations between different multiple scattering theories. *Proc. Phys. Soc.* **91**, 678-688 (1967).

3. Linton, C. M. & Martin, P. Multiple scattering by multiple spheres: A new proof of the Lloyd-Berry formula for the effective wavenumber. *Siam J. Appl. Math.* **66**, 1649-1668 (2010).

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4. Waterman, P. C. & Truell, R. Multiple scattering of waves. *J. Math. Phys.* **2**, 512-537 (1961).

5. Aristégui, C. & Angel, Y. C. Effective mass density and stiffness derived from P-wave multiple scattering. *Wave Motion* **44**, 153-164 (2007).

6. Dubois, J., Aristégui, C. & Poncelet, O. Spaces of electromagnetic and mechanical constitutive parameters for dissipative media with either positive or negative index. *J. Appl. Phys.* **115**, 024902 (2014).