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Thomas Brunet, Aurore Merlin, Benoit Mascaro, Kevin Zimny ...+4 more authors

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Published on: 01 Apr 2015 - Nature Materials (Nature Research)

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To cite this version:

Thomas Brunet, Aurore Merlin, Benoit Mascaro, Kevin Zimny, Jacques Leng, et al.. Soft 3D acoustic metamaterial with negative index. Nature Materials, Nature Publishing Group, 2014, 14, pp. 384-388. 10.1038/nmat4164 . hal-01146463

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Soft 3D acoustic metamaterial with negative index

Thomas Brunet[1]*, Aurore Merlin[2], Benoit Mascaro[1], Kevin Zimny[2], Jacques Leng[3],

Olivier Poncelet[1], Christophe Aristégui[1], Olivier Mondain-Monval[2]

[1]University of Bordeaux, CNRS, I2M-APy, UMR 5295, 33405 Talence, France.

[2]University of Bordeaux, CNRS, CRPP, UPR 8641, 33600 Pessac, France.

[3]University of Bordeaux, CNRS, Solvay, LOF, UMR 5258, 33608 Pessac, France.

*Correspondence to: thomas.brunet@u-bordeaux.fr

Introductory paragraph:

Many efforts have been devoted to the design and the achievement of negative-refractive-index metamaterials since the 2000s[1-8]. One of the current challenges in that field is to extend beyond electromagnetism by realizing three-dimensional (3D) media with negative acoustic indices[9]. We report a new class of locally resonant ultrasonic metafluids consisting of a concentrated suspension of macroporous microbeads engineered using soft-matter techniques. The propagation of Gaussian pulses within these random distributions of “ultra-slow” Mie resonators is investigated through *in situ* ultrasonic experiments. The acoustic index is shown to be negative over broad frequency bandwidths, depending on the volume fraction of the microbeads as predicted by multiple-scattering calculations. These soft 3D acoustic metamaterials open the way for key applications such as sub-wavelength imaging and transformation acoustics, which require the production of acoustic devices with negative or zero-valued indices.

Main text:

Unlike periodic structures, *i.e.,* photonic crystals for light[2] or phononic crystals for sound[3], in which negative refraction is a consequence of band-folding effects, the negative index of a metamaterial originates from low-frequency resonances of sub-wavelength particles. A negative refractive index in electromagnetism was first experimentally verified at microwave frequencies[4]; since then, other experimental demonstrations of negative-index metamaterials have been reported in the near-infrared domain[5,6] and at optical wavelengths[7]. However, the metamaterial concept applies not only to electromagnetism[8] but also to different fields such as acoustics[9]. Since the first experimental realization of "locally resonant sonic materials[10]", negative-acoustic-index metamaterials have been achieved for only the audible domain through the use of two-dimensional solid structures consisting of periodic arrays of interspaced membranes and side holes[11] or arrays of Helmholtz and rod-spring resonators[12]. Coiling up space has also proven to be an effective method of providing acoustic metamaterials with extreme constitutive parameters[13,14]. Possible future applications such as perfect acoustic lenses, by analogy with John Pendry's perfect electromagnetic lens[15], call for three-dimensional negative-acoustic-index metamaterials, which have not yet been achieved[16].

Unlike the mechanical engineering techniques that are usually used to machine 1D[17] or 2D[18] solid structures, soft-matter techniques are very promising for the fabrication of metamaterials because they should yield 3D locally-resonant metafluids, or so-called soft acoustic metamaterials[19], which present the following advantages: i) macroscopic isotropy, unlike the most general class of acoustic metafluids with anisotropic inertia[20,21]; ii) broad versatility, as microfluidics[22] allows the up-scaled production of micro-resonators with controlled sizes, shapes and compositions; iii) potential tunability, as soft inclusions can be either deformed or oriented under external stimuli[23]; and iv) facile shaping and molding, as the host matrix is fluid.

A unique method of achieving 3D negative-acoustic-index metamaterials is to benefit from strong low-frequency Mie resonances (monopolar and dipolar) of "ultra-slow" inclusions[24]. However, no dense homogeneous material possesses a sufficiently low sound speed to exhibit such a feature, not even the so-called "soft silicone rubbers[25]". By contrast, the use of *porous* soft silicone rubbers should be more promising because the sound speed in porous media is very low[26], with the latter depending on the ratio between the elastic moduli and mass density. Indeed, the large volume of air cavities present in porous materials causes them to be very soft (or highly compressible), thereby allowing these materials to exhibit very low elastic moduli while maintaining relatively high mass densities because of their solid skeletons. Particles composed of such "ultra-slow" materials ($v_1 \approx 100$ m/s) randomly dispersed in water ($v_0 \approx 1500$ m/s) should exhibit an extremely large monopolar resonance, like air bubbles[27], thereby causing the suspension to exhibit a negative effective bulk modulus B. These fairly dense particles should also exhibit a strong dipolar resonance, producing a negative effective mass density ρ in an overlapping frequency region. If they are considered as perfect (non-dissipative) media, such "double-negative acoustic metamaterials" are expected to have negative acoustic indices[24].

Such a straightforward analysis in terms of negative constitutive parameters may be inappropriate for a dissipative medium with a negative refractive index[28]. In both electromagnetism and acoustics, the refractive index n is complex-valued because it is defined as the ratio of the complex-valued wavenumber k ($= \omega/v + j\alpha$) in the metamaterial to that in a reference medium k_0 ($= \omega/v_0 + j\alpha_0$), where v denotes the phase velocity, α represents the attenuation coefficient, and ω is the angular frequency. For a non-dissipative reference medium

(α_0 = 0), the refractive index $n = v_0/v + j(\alpha v_0/\omega)$ is often referred to as its real part (= v_0/v). By convention, its imaginary part is taken to be positive ($\alpha > 0$), thus defining the forward direction along which the wave energy flows. Conventional dissipative media or positive-index materials support only forward wave motions ($v > 0$). Metamaterials, however, can sustain backward waves ($v < 0$) in the spectral vicinity of the particle resonances; such materials are referred to as negative-index materials[1].

In this study, we exploited strong low-frequency Mie resonances of sub-wavelength macroporous silicone rubber microbeads to achieve negative-index metamaterials in the ultrasonic domain. We produced particles with a mean radius $\langle a \rangle$ of 160 μm and a size dispersion of approximately 25% (Fig. 1a and b) by generating high-internal-phase emulsion droplets in a simple microfluidic device[29]. These macroporous microbeads, with a porosity of approximately 40%, were then randomly dispersed in a water-based gel matrix, *i.e.,* a Bingham fluid[30] that behaves acoustically like water ($v_0 \approx 1500$ m/s), thus forming a concentrated suspension of "ultra-slow" particles with a volume fraction Φ_0 of approximately 20%. Because the macroporous microbeads are closed (Fig. 1c), they are stable over time (a few days), unlike air bubbles (a few minutes), which are very sensitive to numerous undesirable effects such as Oswald ripening or dissolution in the host matrix[31].

To determine the acoustic refractive index n of the investigated macroporous-particle suspensions, we measured the angular phase shifts of Gaussian pulses propagating through varying thicknesses in direct-acoustical-contact measurements (Fig. 1e). Ultrasonic pulses, with central frequencies f_0 ranging from 50 kHz to 500 kHz, were emitted and detected using a pair of large broadband ultrasonic transducers (transmitter/receiver) with a diameter of 30 mm. Because the size of our macroporous particles is approximately 300 μm, *i.e.,* 10 times smaller than the smallest incident wavelength used in our experiments (3 mm at 500 kHz for water), our microbeads can be considered to be sub-wavelength resonators, as sought for metamaterials[1]. When the propagation distance z was varied from 1.0 mm to 1.5 mm, the transmitted temporal signals shifted on the time-delay axis, allowing us to directly infer the phase velocity v and the acoustic index n (= v_0/v).

Indeed, such *in situ* acoustical measurements can avoid additional phase delays caused by the interfaces, as reported in electromagnetism[6]. Because the strong resonant scattering induces high attenuation in such a random medium, we detect only the ballistic coherent pulse[32], which propagates from the transmitter to the receiver. Therefore, pulses reflected on the transducer surfaces are not propagated, thus avoiding spurious phase delays caused by interferences between the successive echoes (see the Supplementary Information).

For all operating frequencies f_0, the amplitude of the transmitted pulses decreases when the propagation distance z is increased in such a passive medium, as illustrated at 140 kHz (Fig. 2a). However, the temporal shift of the pulse oscillations, which contains information regarding the phase velocity v, depends on the frequency f_0. Note that in our conditions, *i.e.* by investigating "thin" samples, we are able to infer unambiguously the phase velocity because the measured phase delays (~ 0.1 μs) are much smaller than the time period of the transmitted signals (> 1 μs). Since all the oscillations shift as a whole, we focus our attention on the center of the pulses, denoted by t_{phase} and for which the amplitude is zero, as depicted in Fig. 2b to d.

The forward shift of t_{phase} observed at 110 kHz (Fig. 2b) indicates a positive phase velocity (or positive acoustic index, Fig. 2e). By contrast, the backward shift of t_{phase} at 170 kHz (Fig. 2d) reveals a negative phase velocity (or negative acoustic index, Fig. 2g). For the intermediate frequency f_0 = 140 kHz, the transmitted pulses are not shifted on the time-delay axis (Fig. 2c), indicating "infinite" phase velocities (or zero acoustic index, Fig. 2f).

Additionally, we obtained the phase-velocity and acoustic-index spectra over a broad frequency range [50–500 kHz] from the angular-phase measurements performed on the fast Fourier transforms (FFTs) of the temporal signals recorded for two different propagation distances (z = 1.0 mm and 1.5 mm). The phase velocity v and the acoustic index n (= v_0/v) were observed to be negative between 140 kHz and 275 kHz (see the black curves in Fig. 3a and b).

The initial sample ($\Phi_0 \approx 20\%$) was then diluted to produce another sample with $\Phi_1 \approx 15\%$, which resulted in a clear reduction of the frequency width of the negative bands (red curves in Fig. 3a and b). When the volume fraction was drastically lowered ($\Phi_2 \approx 0.2\%$), the negative features disappeared (green curves in Fig. 3a and b), giving way to classical dispersion ($v > 0$), as previously observed in bubbly media[31].

Finally, we compared our acoustical measurements to theoretical predictions produced through multiple-scattering modeling revealing fairly good qualitative agreement (Fig. 3a to d). To obtain the theoretical acoustic index n, we calculated the effective wavenumber k of our samples using the Waterman-Truell formula[33] (see the Supplementary Information), often used for concentrated random media ($\Phi \approx 10\%$). The values of the parameters for the porous silicone rubber that were used for the calculations (the velocity and attenuation coefficient for both longitudinal and shear waves) were measured independently on large monolithic cylindrical samples (30 mm in diameter and 3 mm in thickness) of the same material as the microbeads. The material parameters used for the calculations are ρ_1 = 600 kg/m^3, v_L = 80 m/s, α_L = 60 Np/mm/MHz$^{1.5}$ (for longitudinal waves), and v_T = 40 m/s, α_T = 200 Np/mm/MHz$^{1.5}$ (for shear waves) for the porous silicone rubber and ρ_0 = 1000 kg/m^3, v_0 = 1500 m/s for the water-based gel matrix.

Through direct-pulse-propagation experiments on soft acoustic metamaterials, we confirmed that the phase velocity could be negative over a broad range of ultrasonic frequency bandwidths, thus demonstrating a three-dimensional negative-acoustic-index metamaterial. Our approach based on the use of soft-matter techniques means that these metafluids can be produced on a large scale, paving the way toward the production of acoustic devices with negative or zero-valued indices, as required for key applications such as sub-wavelength imaging and transformation acoustics.

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Acknowledgments:

This work was supported by the Agence Nationale pour la Recherche (Grant 2011-BS0902101 Metakoustik-Aerospace Valley) and the US Air Force European Office of Aerospace Research and Development (Grant FA8655-12-1-2067). This work was performed under the auspices of the Labex AMADEUS ANR-10-LABX-0042-AMADEUS with the help of French state Initiative d'Excellence IdEx ANR-10-IDEX-003-02. The authors thank G. Pibre from Blue Star Company for providing us with silicone rubber and J. Valencony from Siltech Company for providing the surfactants.

Contributions:

T.B., O.M.M., J.L., O.P. and C.A. designed the project. A.M. synthesized the macroporous soft silicone rubber microbeads with the help of K.Z. Formulation aspects were supervised by O.M.M. A.M. set up the microfluidics on the advice of J.L. T.B. and B.M. conducted the acoustical measurements and performed the calculations under the guidance of C.A. and O.P. for the theoretical aspects. T.B. wrote the paper in collaboration with C.A., O.P., J.L. and O.M.M.

Competing financial interests:

The authors declare that they have no competing financial interests.

Figure 1 **Soft 3D acoustic metamaterials composed of "ultra-slow" Mie resonators.** **a**, Optical microscopy image of macroporous silicone rubber microbeads embedded in a water-based gel matrix. **b**, Corresponding histogram showing the microbead size distribution. The mean radius $<a>$ is 160 µm, and the standard deviation σ of the distribution is 25%, as deduced from a Gaussian fit (solid line). The particle porosity is approximately 40%. **c** and **d**, SEM images of both the surface and core of a microbead. **e**, Photograph of the acoustical experimental setup for the *in situ* phase-velocity measurements. The propagation distance z between the pair of large broadband ultrasonic transducers (emitter/receiver) could be varied precisely because the receiver was mounted on a z-axis motorized linear stage.

Figure 2 **Phase delays of the transmitted signals, as a function of propagation distance.** **a**, Examples of typical measured ballistic coherent pulses (f_0 = 140 kHz) transmitted through a concentrated suspension of macroporous silicone rubber microbeads ($\Phi_0 \approx 20\%$, $<a>$ = 160 µm, σ = 25%) for five propagation distances z (= 1.0, 1.1, 1.2, 1.3, 1.4 and 1.5 mm). The time t_{phase} refers to the center of the pulses for which the amplitude is zero. **b**-**d**, Zoomed-in views of the oscillations around t_{phase} for f_0 = 110, 140 and 170 kHz. **e**-**g**, Values of the time t_{phase} as a function of the propagation distance z, yielding experimental values for the phase velocity v (= $\delta z/\delta t_{phase}$) and acoustic index n (= v_0/v) from linear regressions for f_0 = 110, 140 and 170 kHz.

Figure 3 Comparisons between acoustical experiments and theoretical predictions. **a**, Experimental phase velocities v extracted from fast Fourier transforms performed on ultrasonic Gaussian pulses with central frequencies f_0 ranging from 50 kHz to 500 kHz. **b**, Experimental acoustic indices n (= v_0/v) deduced from the above phase-velocity spectra (**a**) given the knowledge of the phase velocity of the host matrix ($v_0 \approx 1500$ m/s). **c-d**, Predicted phase velocities and acoustic indices calculated in the framework of multiple scattering (see the Waterman-Truell formula given in the Supplementary Information). The volume fractions are approximately $\Phi_0 \approx 20\%$ (black lines), $\Phi_1 \approx 15\%$ (red lines) and $\Phi_2 \approx 0.2\%$ (green lines).