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Locally Resonant Sonic Materials

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We have fabricated sonic crystals, based on the idea of localized resonant structures, that exhibit spectral gaps with a lattice constant two orders of magnitude smaller than the relevant wavelength. Disordered composites made from such localized resonant structures behave as a material with effective negative elastic constants and a total wave reflector within certain tunable sonic frequency ranges. A 2-centimeter slab of this composite material is shown to break the conventional mass-density law of sound transmission by one or more orders of magnitude at 400 hertz.

Complete sound attenuation for a certain frequency range can be achieved through the concept of a "classical wave spectral gap," originally introduced in relation to the electromagnetic wave, denoted the "photonic band gap" (*1*). Subsequently extended to elastic waves (*2*–*5*), the idea states that a strong periodic modulation in density and/or sound velocity can create spectral gaps that forbid wave propagation. However, the spatial modulation must be of the same order as the wavelength in the gap. It is thus not practical for shielding acoustic sound, because the structure would have to be the size of outdoor sculptures in order to shield environmental noises (*5*).

We present a class of sonic crystals that exhibit spectral gaps with lattice constants two orders of magnitude smaller than the relevant sonic wavelength. Our materials are based on the simple realization that composites with locally resonant structural units can exhibit effective negative elastic constants at certain frequency ranges. If a wave with angular frequency ω interacts with a medium carrying a localized excitation with frequency ω_o, the linear response functions will be proportional to $1/(\omega_o^2 - \omega^2)$. Such an effect is manifest in the electromagnetic frequency response of materials with optical resonances, where a negative dielectric constant ε (generally on the higher frequency side of the resonance) implies a purely imaginary wave vector $k = n\omega/c$ (where n is the index of refraction and c is the speed of light) and hence exponential attenuation of the electromagnetic wave (*6*). Here, we implement this idea in the context of elastic composites at sonic frequencies. By varying the size and geometry of the structural unit, we can tune the frequency ranges over which the effective elastic constants are negative.

Our composites have a simple microstructure unit consisting of a solid core material with relatively high density and a coating of elastically soft material. In the experiments described below, we used centimeter-sized lead balls as the core material, coated with a 2.5-mm layer of silicone rubber (Fig. 1A). The coated spheres are arranged in an 8 × 8 × 8 simple cubic crystal with a lattice constant of 1.55 cm (Fig. 1B), with epoxy as the hard matrix material. Sonic transmission was measured using a modified Bruel & Kjaer (B&K) two-microphone impedance measurement tube, type 4206. The sound source was mounted at one end of the tube. The sample was placed at the other end of the tube, with one microphone detector mounted on the surface of the sonic crystal facing the sound source and another a few centimeters toward the sound source. A small hole was drilled from the rear of the sample, along the centerline of the sonic crystal to its center. A detector was placed inside the hole, with the sensitive part approximately located at the center of the sonic crystal. Transmission was measured as a function of frequency from 250 Hz to >1600 Hz for effectively a four-layer sonic crystal. The sound source intensity was adjusted so as to maintain a nearly frequency-independent measured amplitude at the front of the crystal. The ratio of the amplitude measured at the center to the incident wave shows two dips, with a peak after each dip (Fig. 1C).

To understand the experimental results,

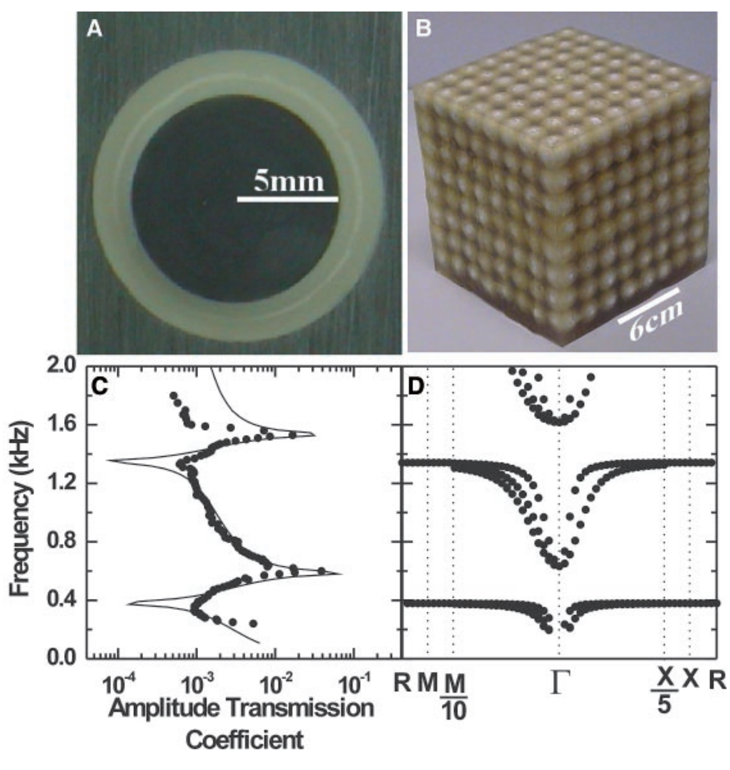

Fig. 1. (**A**) Cross section of a coated lead sphere that forms the basic structure unit (**B**) for an 8 × 8 × 8 sonic crystal. (**C**) Calculated (solid line) and measured (circles) amplitude transmission coefficient along the [100] direction are plotted as a function of frequency. The calculation is for a four-layer slab of simple cubic arrangement of coated spheres, periodic parallel to the slab. The observed transmission characteristics correspond well with the calculated band structure (**D**), from 200 to 2000 Hz, of a simple cubic structure of coated spheres. Three modes (two transverse and one longitudinal) are distinguishable in the [110] direction, to the left of the Γ point. The two transverse modes are degenerate along the [100] direction, to the right of the Γ point. Note the expanded scale near the Γ point.

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we have implemented a rigorous multiple-scattering theory (*7*) for the calculation of elastic wave propagation and scattering in a composite medium with coated spherical inclusions. The band structure for an infinite periodic structure with a simple cubic arrangement of coated spheres was calculated (Fig. 1D). The concentration, size of the core sphere, and coating thickness are fixed at the experimental values (*8*). The most notable predictions of the theory are the two large band gaps with flat lower edges, which indicate the existence of localized modes (*4*). The band structure (Fig. 1D) is quite different from conventional photonic or acoustic-elastic band structures, where the midgap frequency and the size of the spectral gap are dependent on direction and a complete band gap appears only when the spectral gaps in different directions happen to overlap in all 4π radians. Here, the gap is essentially the same at any *k*-point inside the Brillouin zone (Fig. 1D). Note that at 500 Hz, the center of the lower frequency gap, the lattice constant of our sonic crystal is 300 times smaller than that of longitudinal wavelength in epoxy.

The band structure (Fig. 1D) is for an infinite periodic system. Because measurement can only be done on finite structures, only partially developed gaps can be detected. For a better comparison, we have also calculated the amplitude transmission through a four-layer structure that is periodic along the two directions parallel to the slab (solid line in Fig. 1C). The theoretical predictions, with no adjustable parameters, are in good accord with the experimental results, in terms of the frequency positions of the dips (located at 380 and 1350 Hz) as well as the qualitative resonance features of a dip in transmission followed by a peak. Although transmission measurement in one direction alone does not establish the existence of a complete band gap, the agreement between theory and experiment lends support to the existence of a complete band gap in an infinite crystal of locally resonant structures (see below and Fig. 2), with subwavelength lattice constants. The discrepancy between the measured and predicted dip magnitudes is due to the sensitivity limit of the detector. The issues of absorption and phase are examined independently, as described below.

At frequencies away from the resonances, the composite system behaves as an effective medium in which the elastic waves scatter weakly from the subwavelength scatterers and have linear dispersion (ω versus *k*) relations. However, the built-in localized resonances due to the coated spheres give rise to flat dispersions that are nearly *k*-independent. Coupling with the otherwise linearly dispersed elastic waves opens spectral gaps in the band structure (Fig. 1D) due to the level-repulsion effect. For a finite sample, the transmission will have dips in the spectral gaps. At the first dip frequency, the lead particle is seen to move as a whole along the direction of wave propagation, with large strain at the lead particle–silicone rubber interface (Fig. 2A). This low-frequency resonance may be understood as an oscillation, in which the inner core provides the heavy mass and the silicone rubber provides the soft spring. At the second dip, the maximum displacement occurs inside the silicone rubber (Fig. 2B). The displacement of the lead particle is small but nonzero. This is analogous to the "optical mode" in molecular crystals with two atoms per unit cell, where one of the atoms is much heavier than the other. Around the resonance frequencies, the response function has large dispersion, leading to a dip in the region of negative elastic constants and hence exponential wave attenuation, followed by "resonant transmission" when the effective elastic constants satisfy the required condition. As the number of layers increases, the dip is seen to define the position of the lower edge of the band gap, whereas the peak defines the upper edge. Consistent with the large magnitude of the response function close to the resonances, the effective wavelength in our composite is substantially reduced in those frequency ranges (relative to that in air or the epoxy host), thus leading to coupling with the coated spheres and strong scattering.

Because of the localized nature of the resonances, sonic attenuation should be apparent even for one monolayer of coated spheres in the absence of periodicity. This may be demonstrated by measuring the amplitude transmission and reflection for a circular plate 2.1 cm thick and 9.8 cm in diameter, containing 48 volume % of randomly dispersed 10-mm lead spheres, each coated with a 3.5-mm layer of silicone rubber. Measurements were done using the same B&K system. The sample was tightly mounted at the end of the tube, with another tube directly mounted behind the sample. Four microphones were put in the couplers located symmetrically on both sides of the sample. The signals were measured using lock-in amplifiers. The transmission coefficient of the composite sample (Fig. 3) exhibits two significant dips centered at 400 and 1100 Hz, where the lower frequency minimum is at least one order of magnitude smaller than the epoxy reference (Fig. 3). These dips may be regarded as partially developed spectral gaps caused by negative effective elastic constants (see below and Fig. 4). Reflection measurements were carried out. For both the pure epoxy sample and the composite sample, reflection coefficient as a function of frequency varies between 0.98 and 1, that is, within the measurement error. We conclude that absorption is negligible for our samples. As a sound shield, our material thus differs from those commercial sound insulation materials that rely on absorption (*9*).

The amplitude transmission coefficient T at normal incidence through a slab of homogeneous material with thickness d (*10*) is

$$T = \frac{4\nu \exp(ik_2 d)}{(1+\nu)^2 - (1-\nu)^2 \exp(2ik_2 d)} \qquad (1)$$

where $\nu = (\kappa_2\rho_2/\kappa_1\rho_1)^{1/2}$, $k_2 = \omega/(\kappa_2/\rho_2)^{1/2}$, ω denotes angular frequency, κ_1 and κ_2 denote the longitudinal wave modulus for air and solid, respectively, and ρ_1 and ρ_2 are the mass densities. At sonic frequencies, where the slab thickness d is much smaller than the longitudinal sound wavelength in solid (so that $k_2 d \ll 1$ and $\nu \gg 1$), an accurate approximation to Eq. 1 is given by

$$T \cong i\,\frac{2\sqrt{\rho_1\kappa_1}}{\omega\rho_2 d} \qquad (2)$$

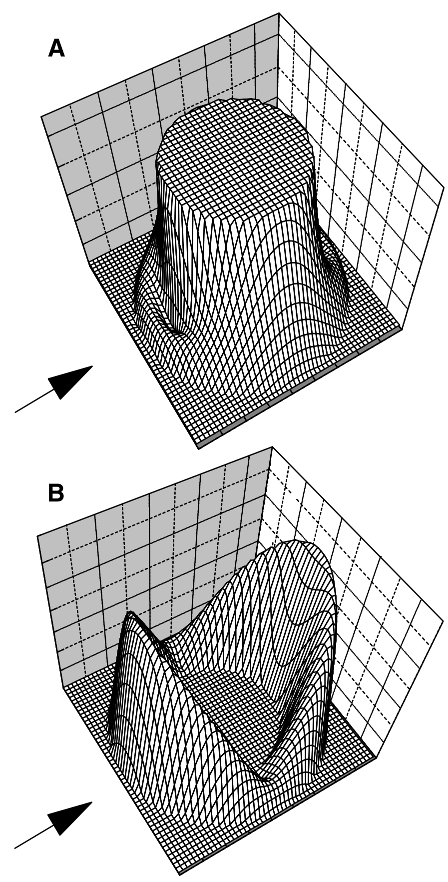

Fig. 2. Calculated displacement configurations at the first (**A**) and second (**B**) dip frequencies. The displacement shown is for a cross section through the center of one coated sphere, located at the front surface. The arrows indicate the direction of the incident wave.

Equation 2 is independent of κ_2, implying that the value of T given by the mass-density law is essentially the limit for $\kappa_2 \rightarrow \infty$. This equation is conventionally known as the mass (area) density law for sound shielding/transmission in the absence of absorption. It states that for a given level of sound transmission amplitude, the required mass area density is inversely proportional to the frequency.

Equation 2 was used to calculate the transmission coefficient for a 2.1-cm layer of epoxy (Fig. 3, dashed line). Using Eq. 1 for the same calculation leads to essentially the same result. We have also calculated the mass-density law for the composite sample by using the average density, which is somewhat higher than that of epoxy (Fig. 3, dot-dashed line). Thus, our composite sample, with one layer of the coated spheres, breaks the mass-density law by at least one order of magnitude at the first dip frequency. For comparison we have also calculated, using an equation slightly more complicated than Eq. 1 (*10*), sound transmission through a layered epoxy–silicone rubber–lead–silicone rubber–epoxy medium, with the same volume fractions of the three components as in the 2.1-cm random composite sample. The results show large transmission peaks due to the soft rubber layers, but the transmission minima are always equal to or larger than that given by the mass-density law. These comparisons show that the locally resonant microstructure is crucial for the observed dip characteristic.

We have also calculated the frequency positions of the transmission dips, using our multiple-scattering code, for a monolayer of coated lead spheres arranged in a two-dimensional hexagonal lattice, with the concentration and other parameters the same as the random composite used in the experiment. The results (Fig. 3, arrows) are in reasonable agreement with the experiment. The most notable difference between the ordered and random arrangements of coated spheres is that in the random case, the resonant transmission peaks are absent.

Equation 1 can be used in conjunction with the transmission data (Fig. 3) to do inversion for the effective κ_2 of a slab of homogeneous medium with the same transmission characteristics. By letting $\rho_2 = \rho_e$ be the average mass density of the sample, an effective κ_2, here denoted by κ_e, was obtained. Away from the resonances, κ_e has a value very similar to that of epoxy (and hence $1/\kappa_e$ is fairly small) (Fig. 4). However, close to the resonances the modulus actually turns negative (with $1/\kappa_e = 0$ defined by the amplitude transmission value given by the mass-density law). It is thus not surprising that the composite sample can break the mass-density law, because the material behaves like a total reflector. This is analogous to the reflection of electromagnetic waves (*11*) by a material having a dielectric constant that is real and negative.

Although the static elastic constant must be positive for maintaining structural stability, resonance-induced negative elastic constants should be possible, as demonstrated here at low sonic frequencies. This conclusion is further supported by the measured phase of the transmission signal (Fig. 4, open circles). In the region of positive κ_e, the phase is relatively constant because away from the resonance, the wavelength is much larger than the sample thickness. Coinciding with the frequencies where the computed $1/\kappa_e$ crosses zero, there are observed 180° phase jumps, giving direct evidence for the underlying resonance mechanism. Extension to lower and higher frequency elastic wave systems may lead to applications in seismic wave reflection and ultrasonics.

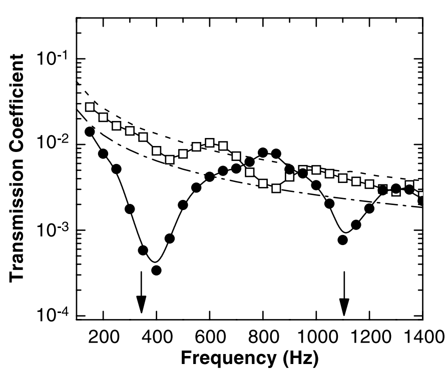

Fig. 3. Measured amplitude transmission (solid circles; the solid line is a guide to the eye) through a 2.1-cm slab of composite material containing 48 volume % of randomly dispersed coated lead spheres in an epoxy matrix. As a reference, the measured amplitude transmission through a 2.1-cm slab of epoxy is also plotted (open squares connected by a thin solid line). The dashed and dot-dashed lines, respectively, show the calculated transmission amplitudes of a 2.1-cm epoxy slab and a 2.1-cm homogeneous slab of the same density as that of the composite material containing the coated spheres. The two arrows indicate the dip frequency positions predicted by the multiple-scattering calculation for a monolayer of hexagonally arranged coated spheres in an epoxy matrix. For reference, at 400 Hz the sonic wavelength in air is about 80 cm.

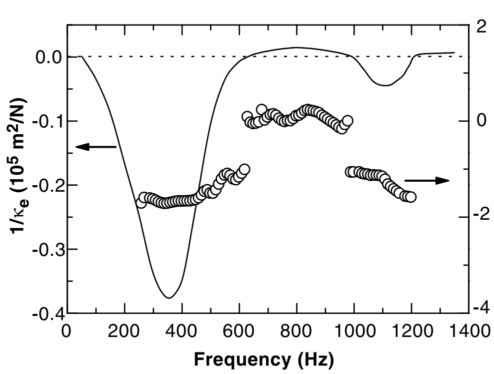

Fig. 4. The frequency-dependent effective longitudinal elastic modulus κ_e inverted from the amplitude transmission data shown in Fig. 3 (solid line) using Eq. 1. Open circles denote the measured phase of the transmission coefficient, with zero phase set arbitrarily. Undulations in the measured phase are due to noisy data.

References and Notes

1. See, e.g., J. Joannoupoulous, R. Meade, J. Winn, *Photonic Crystals* (Princeton Univ. Press, New York, 1995), and references therein.
2. M. Kushwaha *et al.*, *Phys. Rev. Lett.* **71**, 2022 (1993).
3. M. Kafesaki, M. Sigalas, E. N. Economou, *Solid State Commun.* **96**, 285 (1995).
4. M. Kafesaki and E. N. Economou, *Phys. Rev. B* **52**, 13317 (1995).
5. R. Martinez-Sala *et al.*, *Nature* **378**, 241 (1995).
6. See, e.g., J. D. Jackson, *Classical Electrodynamics* (Wiley, New York, ed. 2, 1975).
7. Z. Liu, C. T. Chan, P. Sheng, A. L. Goertzen, J. H. Page, *Phys. Rev. B* **62**, 2446 (2000). Similar formalisms can be found in published literature [see, e.g., M. Kafesaki and E. N. Economou, *Phys. Rev. B* **60**, 11993 (1999); I. E. Psarobas, N. Stefanou, A. Modinos, *Phys. Rev. B* **62**, 278 (2000)].
8. The material parameters used are $\rho = 11.6 \times 10^3$ kg/m^3, $\lambda = 4.23 \times 10^{10}$ N/m^2, $\mu = 1.49 \times 10^{10}$ N/m^2 for lead [C. Kittel, *Introduction to Solid State Physics* (Wiley, New York, ed. 3, 1966), p. 122]; $\rho = 1.3 \times 10^3$ kg/m^3, $\lambda = 6 \times 10^5$ N/m^2, $\mu = 4 \times 10^4$ N/m^2 for silicone rubber (L. Bousse, E. Dijkstra, O. Guenat, *Technical Digest, Solid State Sensor and Actuator Workshop*, Hilton Head Island, SC, 1996, p. 272); $\rho = 1.23$ kg/m^3, $\kappa = \lambda = 1.42 \times 10^5$ N/m^2 for air; and $\rho = 1.18 \times 10^3$ kg/m^3, $\kappa = \lambda + 2\mu = 7.61 \times 10^9$ N/m^2, $\mu = 1.59 \times 10^9$ N/m^2 for epoxy (*4*). Here, λ and μ are the Lamé constants, and κ is the longitudinal wave modulus.
9. Commercial products like acoustic foam and fibrous insulation rely on sound absorption. There are also sound barriers that work within the limit of mass-density law, typically containing lead sheets or using impedance mismatch, such as introducing an air gap, or alternating layers of materials with high and low acoustic impedance.
10. See, e.g., L. M. Brekhovskikh, *Waves in Layered Media* (Academic Press, New York, ed. 2, 1980).
11. However, in the case of an optical polariton there is almost always strong absorption, so even though a polariton will cause a spectral gap for electromagnetic waves, the resultant spectral gap is not quite a "photonic band gap" because the mode energy will be absorbed rather than reflected. In our system, the constituent materials have little absorption in the sonic frequency regime, hence the resonance reflects almost all the incoming wave energy for frequencies inside the spectral gap, as verified experimentally through the reflection measurement.
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