Aircraft trapped colloidal crystals that are reconfigurable in real time

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Photonic and phononic crystals are metamaterials with repeating unit cells that result in internal resonances leading to a range of wave guiding and filtering properties and are opening up new applications such as hyperlenses and superabsorbers. Here we show the first, to our knowledge, 3D colloidal phononic crystal that is reconfigurable in real time and demonstrate its ability to rapidly alter its frequency filtering characteristics. Our reconfigurable material is assembled from microspheres in aqueous solution, trapped with acoustic radiation forces. The acoustic radiation force is governed by an energy landscape, determined by an applied high-amplitude acoustic standing wave field, in which particles move swiftly to energy minima. This creates a colloidal crystal of several milliliters in volume with spheres arranged in an orthorhombic lattice in which the acoustic wavelength is used to control the lattice spacing. Transmission acoustic spectroscopy shows that the new colloidal crystal behaves as a phononic metamaterial and exhibits clear band-pass and band-stop frequencies which are adjusted in real time.

Results

The metadevice used to form the 3D reconfigurable metamaterial is summarized in Fig. 1. To generate the desired lattice pattern, the metadevice creates a high-amplitude acoustic standing wave field using two planar piezoceramic transducers arranged as shown in Fig. 1A. The metadevice consists of three orthogonally arranged systems: the levitation system, arranged vertically, which lifts the particles and holds them in horizontal planes, and two orthogonal manipulation stages, which use the counterpropagating wave method to produce a grid of acoustic traps in the horizontal plane. The lattice spacing can be controlled by the frequencies of the standing waves and the form and location of the lattice by the phase differences (15). The

Significance

We have been working on metamaterials that are reconfigurable in real time with a view to creating genuinely active metamaterials. Such materials will allow researchers to gain unprecedented control over a range of optical and acoustic wave phenomena. To date, although numerous examples of metamaterials exist, none is reconfigurable in three dimensions. We have developed a method for creating three-dimensional colloidal crystals that are reconfigurable in real time. Our method uses acoustic assembly to trap a suspension of microspheres in patterns resembling crystal lattices. We show that it is possible to dynamically alter the geometry of the colloidal crystal and use our metamaterial as an ultrasonic filter that can be tuned in real time.

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resultant 3D standing wave exerts an acoustic radiation force on the particles (17). This force stems from a combination of the time-averaged acoustic pressure and inertial interaction between the particles and the acoustic field (18). Under Rayleigh scattering conditions, the wavelength is larger than the particle size ($\lambda >> a$) and acoustic radiation force due to a standing wave is governed by the spatial gradient of the energy landscape, $U$, i.e., $F = -\nabla U$ (17). Particles with densities greater than the fluid experience forces toward pressure nodes. However, whereas this model is helpful to understand the basic operating principles of our metadevice, the Rayleigh limit is not strictly satisfied in our current metamaterial design (see extended theory in the first section of SI Text). Fig. S1 illustrates the acoustic radiation force for a polystyrene sphere in a standing wave field.

The experiments were performed using 90-$\mu$m-diameter polystyrene spheres in aqueous suspension contained within a central cylindrical chamber of 0.33 mL in volume which becomes our reconfigurable colloidal metamaterial (Methods). Under the action of the levitation stage but without a signal applied to the manipulation transducers, the particles collect in horizontal planes one above the other separated by $a_z = \lambda_z/2$, where $\lambda_z$ is the wavelength in the Z direction, typically $\lambda_z = 215$ $\mu$m when excited at 3.45 MHz assuming the speed of sound in water, $c_0 = 1,480$ m/s. When the manipulation transducers were excited in-phase, the particles were further formed into a grid in the horizontal $X - Y$ plane with lattice spacing $a_x = \lambda_x/\sqrt{2}$ and $a_y = \lambda_y/\sqrt{2}$. Here the wavelengths in the $X$- and $Y$ directions, $\lambda_x$ and $\lambda_y$, were used to reconfigure the metamaterial (see the second section of SI Text). Typical acoustic landscape is depicted in Fig. S2, while the particle positions are shown in Fig. S3. The acoustic standing wave patterns were first characterized by a Schlieren optical system which maps the time-averaged intensity field distribution. Fig. 2 shows the intensity distributions formed when the manipulation transducers are excited at $f_x = f_y = 2.25$ MHz, 3.75 MHz, and 5.25 MHz which lead to $a_x = a_y = 465$ $\mu$m, 279 $\mu$m, and 199 $\mu$m, respectively. The acoustic radiation force associated with these distributions traps particles at the intensity minima which set the X- and Y-lattice dimensions to be equal, leading to simple tetragonal lattice arrangements. Fig. 3 A–C shows views of the $X - Y$ plane of the metamaterial assembled using the acoustic field distributions of Fig. 2. It is apparent that the lattice spacing depends on the frequency used and it becomes more densely spaced as the frequency increases. Note that the concentration of particles was $\phi \simeq 5\%$ by volume, chosen such that at 5.25 MHz most of the traps contained a single polystyrene sphere. Fig. 3D shows an $X - Z$ view of the reconfigurable metamaterial when the levitation stage was excited at $f_z = 3.45$ MHz, together with...
The colloidal metamatals were characterized by transmission acoustic spectroscopy \( \text{(Methods)} \), where the transmission magnitude \( T \) was obtained by normalizing the Fourier-transformed spectra of the signal transmitted through the cylindrically shaped metamaterial, \( A_1(f) \), with respect to the signal through the same chamber filled with deionized water, \( A_0(f) \): 
\[
T = \frac{A_1(f)}{A_0(f)}.
\]

With respect to the lattice geometry, the incident wavefront used for transmission lies in the \((100)\) plane of the lattice. We measured the acoustic transmission through the metamaterial patterns with parameters shown in Fig. 3 \((\text{see typical experimental transmitted signals in Fig. S6})\). An additional measurement was made through a random suspension of the same polystyrene particles. Fig. 4 illustrates the experimental transmission spectra demonstrating the reconfigurability of the system.

Also shown in Fig. 4A–C are the results of a finite-difference time-domain model of the acoustic propagation through an idealized version of the cylindrically shaped metamaterial \((\text{see the first part of the third section of SI Text})\). A generally good correspondence between measured and modeled data is observed. By using an effective field approach \((19)\), the effective mass density and bulk modulus of the corresponding acoustic metamaterial with random microstructure was calculated \((\text{see the second part of the third section of SI Text})\). Predicted values for the acoustic transmission magnitude for the random acoustic metamaterial confined within the central cylindrical chamber with \( \phi \approx 5\% \) volume fraction of polystyrene particles in water are displayed in Fig. 4D and show good agreement with experiment. Fig. S7 shows measured and predicted values for the transmission magnitude \( T \) with \( \phi = 2.5\% \) vol fraction of polystyrene particles in water. The agreement between experimental results and predictions using Eqs. S5–S8 is excellent over the entire frequency range displayed in Fig. S7.

By comparing Fig. 4A–C \((\text{lattice})\) with Fig. 4D \((\text{random})\), it can be seen that some of the most prominent band gaps occur in the vicinity of the resonant frequencies of individual particles, whereas others suggest a simultaneous occurrence of structure-directed and particle-resonance-induced phononic gaps. We believe that small imperfections in the symmetry of lattice we have created, such as missing or multiple particles at some nodes, result in an increase in the level of the observed transmission minima compared with simulation \((\text{which assumed perfect symmetry})\). This is in line with other findings \((20)\) where crystal imperfections caused deviations from the perfect symmetry case. However, it is clear that the measured minima in transmission are in reasonable agreement with simulations indicating that the reconfigurable metamaterial is behaving as expected.

Fig. 5 shows the transmission spectra measured with a scattering angle \( \theta_s \) varying from \(-6^\circ\) to \(6^\circ\) in the \(X-Y\) plane \((\text{Fig. S4})\) for the metamaterial patterns shown in Fig. 3A–C. An additional measurement was made through a random suspension of the same polystyrene particles. The transmission map is plotted in Fig. 5B as a function of both the frequency and the scattering angle. The transmission patterns are symmetrical with respect to the center of the array receiver, indicating good alignment, and considerable variation is seen with both angle and frequency suggesting anisotropy. Various marked differences can be seen between the different reconfigured metamaterials, particularly in the frequency ranges 5–6.5 MHz and 8.5–9.5 MHz, which were also seen in Fig. 4. Fig. S8 shows the effect of real-time reconfiguration \((\text{SI Text})\). Here a 30-cycle sinusoid, centered on 6 MHz, is transmitted through the metamaterial and the lattice reconfigured between \( a_x = a_y = 199 \mu m \) and \( a_x = a_y = 279 \mu m \). The amplitude of the transmitted signal is seen to change by a factor of 6 and the reconfiguration takes significantly less than 0.1 s \((\text{Movie S1})\).

**Discussion**

This realization of a 3D colloidal metamaterial that is reconfigurable in real time represents a new and versatile means by which wave propagation phenomena can be controlled. The use of acoustic assembly is attractive as it has advantages over other potential techniques, such as optical tweezers. Acoustic assembly is easily scalable with wavelength; here we use wavelengths of hundreds of micrometers and create a metamaterial of milliliters in volume. This could be scaled down to the order of micrometers before acoustic absorption and scattering become limiting \((21)\). Conversely, there is no obvious upper limit on the wavelength. The ability to trap particles in an acoustic field requires contrast between at least one of the key fluid and particle

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**Fig. 2.** Time-averaged intensity field distributions in the acoustic metadevice. (A–C) Schlieren images of the acoustic landscape in the center of the device obtained with the \(X-Y\) acoustic manipulator excited at 2.25, 3.75, and 5.25 MHz, respectively. The images are in a grayscale where black represents zero intensity and the maximum intensity amplitude is white. The central circular region is 5 mm in diameter.

**Fig. 3.** Assembled colloidal crystals with the acoustic metadevice. (A–C) Experimental micrographs of the \(X-Y\) plane of the assembled colloidal metamaterials using the same parameters as Fig. 2A–C, respectively. (D) Side view of the acoustic levitator excited at 3.45 MHz with the acoustic manipulator excited at 3.75 MHz.
In the more immediate future these concepts will lead to, for example, the construction of a variety of reconfigurable quasicrystalline structures (22, 23) which would be difficult by self-assembly techniques. Indeed, the range of structures that can be constructed is limited only by the ability to produce the appropriate acoustic energy landscape. Multielement array devices would further extend the versatility (24). Arrays with wide bandwidths could, in principle, be used to produce gradations in the lattice parameters which would lead to the ability to construct reconfigurable waveguides and acoustic cloaking devices. In addition, such arrays could actively control the detail of the lattice geometry to fabricate more perfectly symmetrical lattices than were possible here. Tunability of the band structure could also be achieved with constitutive media with mixed properties such as acousto-optic or acousto-magnetic properties.

**Methods**

**Device Fabrication.** The four manipulation transducers consist of 10 × 15 × 3-mm-thick piezoceramic plates of NCE51 material (a soft-doped lead zirconate titanate (PZT) material; Noliac group).

These PZT plates are fitted into a specially designed polymer chamber which is fabricated using fused deposition modeling, a method commonly used for prototyping and rapid manufacturing. The distance between opposing pairs of transducers is 38 mm.

The levitation transducer is a piezoceramic disk of NCE51 material of 5 mm diameter and is 2 mm thick. This PZT disk is fitted into one end of a round tube of Mylar material (a fairly rigid, transparent, heat-shrinkable polyester film, Precision Paper Tube Co.). The tube defines the central cylindrical chamber, within which the metamaterial is constructed; it is 17 mm in height and has an ∼50-μm wall thickness.

**Particle Suspension.** Monodisperse (90 ± 3)-μm-diameter polystyrene spheres (Polysciences Europe) were diluted in a solution of deuterium oxide (D_2O) and water (H_2O). The solvent densities (1.11 g/cm^3 for D_2O and 1 g/cm^3 for H_2O) were matched to keep the solid particles (1.05 g/cm^3 for polystyrene) approximately neutrally buoyant. A solvent ratio of ∼1:1 volume will match the particles’ density, however only over a narrow temperature range. The buoyancy was further improved once the fluid–particle mixture was released inside a thin-wall polyester tube acting as an acoustic levitator. The remainder of the metadevice chamber was filled with deionized water.

**Experimental Setup and Data Acquisition.** Fabrication of the colloidal metamaterial. Each of the five piezoceramic transducers was driven using a continuous sine wave by a signal generator. The four X–Y manipulation transducers (arranged in opposing pairs) require additional power amplification to provide voltages of 30 V_{pp}. The manipulation transducers were excited at three resonance frequencies (2.25, 3.75, and 5.25 MHz, related to the thickness of these plates) to gain additional control, and each pair was synchronized to allow the relative phase of the excitations of the opposing transducers to be adjusted. Synchronization between channels was achieved using two arbitrary waveform generators (33220A, Agilent) providing four outputs allowing independent control of the amplitude, phase, and frequency of the transducer pairs. The levitation and manipulation stages are not synchronized together. The resonant nature of the levitation stage ensured that the signal direct from the generator (10 V_{pp} at 3.45 MHz) was sufficient to trap 90-μm-diameter polystyrene spheres against gravity.

**Transmission acoustic spectroscopy.** The measurements of the transmission of the acoustic waves in the megahertz frequency range were performed in immersion. Two immersion transducers of nominal central frequency 15 MHz were fitted onto the metadevice in between two opposing manipulation transducers, along the Y direction. The distance between the two transducers is 35 mm. The excitation signal is a broadband pulse generated by a monolithic transducer (V313-SM, Panametrics) of 6.3-mm diameter. The receiver is a 2D matrix array probe transducer (CDCC662-1, Imasonic) and has 128 elements. The array has an element pitch of Δx = Δz = 300 μm and covers a total active area (L_x × L_z) of 3.85 × 3.55 mm^2. The transmitted field is averaged over 10–15 successive acquisitions on individual array elements. The measured time signals are then Fourier transformed to obtain the frequency dependence of the complex transmission, so that both amplitude and phase are determined. Transmission spectra in Fig. 4 were measured on a single centrally located element, and angular variations in Fig. 5 were averaged over four elements in the Z direction (i.e., 1,150 μm). We note this averaging procedure reduces the depth of the transmission minima in Fig. 5 compared to measured values.
with the results in Fig. 4, however, the main features of the different metamaterial lattices are preserved (see *SI Text*, Fig. S5 for a detail of these results in the [100] direction, i.e., \( \theta = 0 \).

Modulation Time. The modulation (or switching) time for the metadevice is determined by the time taken for particles to move between different patterns. If the acoustic radiation force on the polystyrene particles (*SI Text*, Eq. S1) is locally balanced by Stokes’ drag, the particle trajectories can be simulated. This reveals that the time taken for a single particle to reach a lattice location is governed by a combination of viscosity of the suspending fluid and the acoustic pressure amplitude and distribution, as well as the various parameters that govern the acousto-phoretic contrast. As in practice, the upper limit on acoustic pressure is limited by the disrupting effects of acoustic streaming to below \( p_0 \approx 1 \) MPa. If we use this limiting pressure and take the dynamic viscosity of the \( \text{D}_2\text{O} - \text{H}_2\text{O} \) mixture to be \( \eta = 0.9 \) kPa·s the time taken for a 90-μm polystyrene particle to move from antinode to node in a 1D standing wave at 5.25 MHz is \( t \approx 5 \) ms.