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A tunable phononic crystal based ultrasonic filter was designed by stimulating the phase of the polymeric material embedded in a periodic structure using infrared radiation. The acoustic filter can be tuned remotely using thermal stimulation induced by the infrared radiation. The filter is composed of steel cylinder scatterers arranged periodically in a background of bulk poly (N-isopropylacrylamide) polymer hydrogel. The lattice structure creates forbidden bands for certain sets of mechanical waves that cause it to behave as an ultrasonic filter. Since the bandstructure is determined by not only the arrangement of the scatterers but also the physical properties of the materials composing the scatterers and background, modulating either the arrangement or physical properties will alter the effect of the crystal on propagating mechanical waves. Here, the physical properties of the filter are varied by inducing changes in the polymer hydrogel using an electromagnetic thermal stimulus. With particular focus on the k_{00}-wave, the transmission of ultrasonic wave changes by as much as 20 dBm, and band widths by 22% for select bands. © 2014 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4894489]

Phononic crystals are artificially fabricated periodic structures that can predictably affect sound waves that propagate through it. The physical properties of the materials composing the structure, termed the material parameters, and the arrangement of the materials, termed the hylemorphic properties, strongly affect the characteristics of the waves that propagate through the structure. The material parameters and hylemorphic properties can be combined to form structures that behave as filters, waveguides, and lenses amongst others things. The material parameters for sonic structures are primarily the density, sound velocity, and attenuation; essentially the analog of dielectric properties in structures that manipulate light. Tuning of the phononic structure can be accomplished by the manipulation of either the material parameters, the hylemorphic parameters, or a combination of the two. Compared to optical waves, mechanical waves, or sound, are more complicated to manipulate because they possess both longitudinal and transverse components.

The propagation of a mechanical wave through a phononic crystal (SC) is primarily controlled by the contrast in mechanical properties of the scatterers, background and ambient materials; the shape, size, and arrangement of the scatterers; and the wavevector of the impinging acoustic wave. Once a SC has been fabricated, its properties are fixed, and can only be modulated to the degree in which any of the above variables can be changed.[1] The first tunable phononic crystals were modulated through their hylemorphic properties by either physically changing the size of the scatterers in the crystal or rotating the crystal entirely.[1] Many methods achieve tuning by changing the hylemorphic properties of the structure through manipulations of the lattice parameters. Manipulation of the material parameters, and even

manipulation of combinations of hylemorphic and material parameters in phononic crystals has more recently been demonstrated through the implementation of piezoelectrics and dielectric elastomers in sonic structures that include phononic crystals. Tuning of sonic structures has been either demonstrated or theoretically explored by mechanical modulation,[2] applied static electric fields,[3–5] magnetically,[6–8] control of the depth of scatterers,[9] and rotation of the crystal.[10] Nearly all these techniques require physical interaction with the sonic structures and do not operate remotely. In this work, we propose a non-contact mode of remotely tuning a phononic crystal by using infrared electromagnetic radiation.

The speed of sound can vary considerably in periodic elastic or polymer composites.[11,12] Bulk poly (N-isopropylacrylamide) (PNIPAm) is a viscoelastic polymer hydrogel that is composed of cross-linked polymer chains in an aqueous solution. Bonding between the chains and water can be rearranged with external stimuli such as heat, light, and electric fields. Rearrangement results in a gel that can undergo a reversible volumetric phase change as induced by the external stimulus. The sound velocity, Young's modulus, attenuation, and other material parameters of bulk PNIPAm vary depending on the state of the gel.[12] Variations in the material parameters have been measured as high as 800 m/s for the sound velocity, 1.60×10^5 Pa for the Young's modulus, and 1000% for the attenuation due to the phase transition, making PNIPAm an ideal material for a tunable phononic crystal.[13] The range of variation in the material properties is heavily dependent on the amount of cross-linking in the gel and the frequency of sound used, but is not restricted to only these two parameters.

In this work, we demonstrate the tuning of ultrasonic wave propagation through a phononic crystal infiltrated with PNIPAm which undergoes volumetric phase transition due to the absorption of infrared electromagnetic waves. The

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unique mode of electromagnetically induced volumetric phase transition of the polymer filled periodic structure forms a non-contact mode of thermally tuned ultrasonic filter. The structure was designed for the ultrasonic wavelength and the filter resulting from this design has a band frequency between 350 kHz and 750 kHz.

The phononic structure is composed of a 10×10 square lattice of 3.2 mm diameter, 18 cm long stainless steel cylinder scatterers spaced 4.0 mm apart. The square lattice matrix was milled into two: Separate acrylic slabs and the stainless steel rods were placed into the slabs such that 15 cm of the rods were exposed to comprise the phononic crystal. The final filling fraction for the phononic crystal was 50.2% (Fig. 1(a)).

Bulk PNIPAm hydrogel was polymerized into the interstitial spacing between the rods using free radical polymerization to form the hydrogel component. N-Isopropylacrylamide monomer (NIPA, TCI Chemicals), N,N′-Methylene-bisacrylamide crosslinker (BIS, Polysciences Inc.), and DI water were mixed together in a ratio of 0.10 (wt.): 0.02 (mol. NIPA): 0.84 (wt.) to make a monomer solution. The structure was then placed into the solution, and the combination was set in an ice bath and pumped with N_2 while being stirred to remove oxygen from the solution. Ammonium Persulfate (98+%, Sigma-Aldrich) initiator and N,N,N′,N′-Tetramethylethylenediamine (99%, Sigma Aldrich, TEMED) accelerator were then added and mixed for final polymerization. Removal of excess reagents and unreacted chemicals from the polymer in the hydrogel-phononic crystal structure was achieved by dilution; immersing the entire structure in eight, separate, 12 h. long fresh DI water baths. (Fig. 1(b))

The setup was placed in the center of a large tank filled with deionized water, and temperatures measured using an epoxy encased Fluke 51 K/J thermocouple accurate to 0.1 °C. The thermocouple was placed at the outer edge of the emitting transducer midway between its upper and lower boundaries. Electromagnetic actuation of the filter was accomplished using four GE 37771, 250 W, unfocused, infrared light sources targeted towards the phononic filter.

Transmission measurements were performed using two Panameterics V301, unfocused, planar, immersion transducers centered at 0.5 MHz. One transducer operated as an emitter, and the other as a receiver with the transducers spaced 3.81 cm apart on opposite sides of the phononic filter. The emitter was arranged such that the faces of the

transducers were within 2 mm of the surfaces of the filter, and the emitted wave was a characteristic k_{00} plane wave with respect to the phononic crystal. The emitting transducer was driven by Tektronix Type 191 Constant Amplitude Signal Generator, and dually connected to a Tektronix TDS 2024B oscilloscope. The receiver was connected to an HP3585A Spectrum Analyzer, and dually to a separate channel on the same oscilloscope. The function generator was swept over the range 350–750 kHz at a constant power, and the maximum corresponding signal at each frequency was recorded on the HP3585A.

Phononic crystals can be operated as ultrasonic pass band filters that derive their properties from the phononic crystal's bandstructure. The bandstructure is dependent on the hylemorphic and elastic material properties of the crystal, with special focus on the filling fraction and the contrast in sound velocity between the scatterers and background materials. Tuning is accomplished in this work by modulating the physical property contrast between the scatterers and background material. The lower critical solution temperature (LCST) is roughly 33 °C. Around this temperature, under the influence of infrared radiation, PNIPAm undergoes a relatively discontinuous volumetric phase transition that results in strong variations in the density, sound velocity, and attenuation amongst other physical parameters that will present themselves in the changing band structure.[14] However, since PNIPAm contains upwards of 90 wt. % water in the hydrophillic state, and the sound velocity of water also varies with temperature, it is critical to establish that filtering is accomplished due to changes in the hydrogel, and not purely from the increased sound velocity in water.

Figure 1(c) contains the transmission spectrum of the filter without gel in water ambient at 21 °C and 38 °C. Estimates for the sound velocity of the water based on work by Al-Nassar *et al.* gives values of 1476 m/s at 21 °C and 1513 at 38 °C.[15] The effect on the tranmission of the phononic structure acting as the ultrasonic filter is apparent as the bands shift by roughly 13 kHz with the higher sound velocity at increased temperatures.

Figure 2 shows the transmission bands of the hydrogel dispersed sonic structure before and after the continuous radiation of the IR lamp. The radiation of the infrared waves initiates the volume phase transition in the hydrogel. Below the LCST, the central band is centered at 527 kHz with a width of 60.8 kHz. Above the LCST, the center frequency
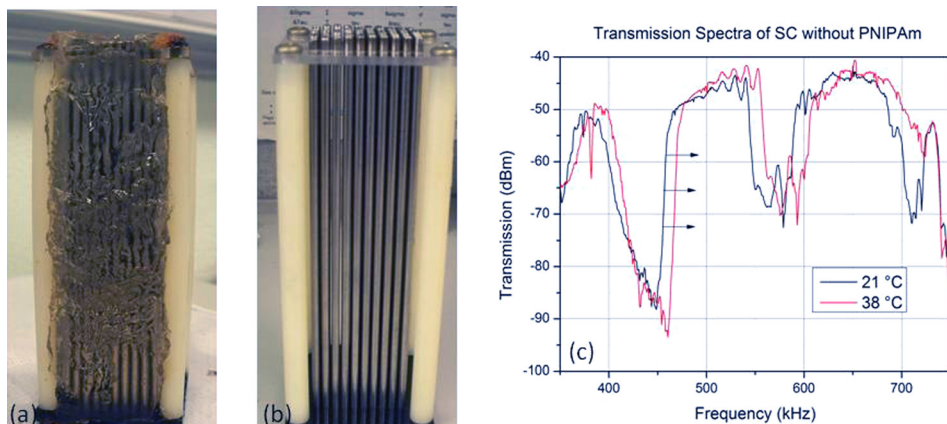

FIG. 1. (a) A phononic crystal fabricated with periodicity designed to have transmission in the ultrasonic region. (b) Phononic crystal filled with PNIPAm Hydrogel at room temperature. (c) Transmission spectrum of SC without PNIPAm at 21 °C and 38 °C. The blueshifting of the transmission bands is due to the increased ultrasonic sound velocity of water at increased temperatures.
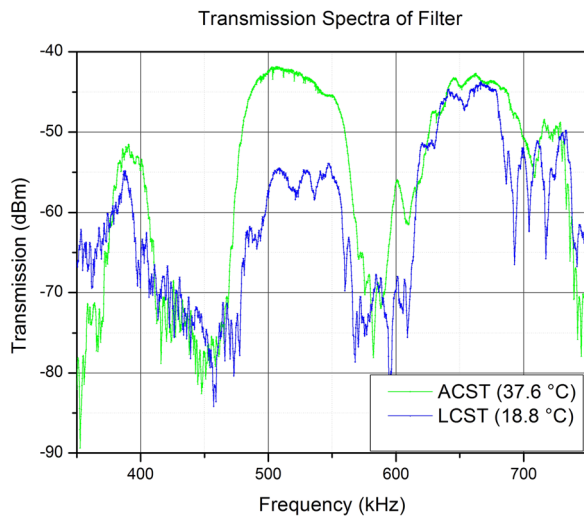
FIG. 2. Transmission spectrum of the filter above and below the LCST. Significant changes in the bandstructure can be readily observed across the bands, with the strongest changes apparent between 475–565 kHz.

decreases 1.6% to 518.5 kHz and increases 22.0% in width to 73.4 kHz. Variations in the behavior of the band can be easily seen as transmission at certain frequencies shows minimal changes between the two temperatures, while select frequencies show transmission increases of up to 19.7 dBm above the LCST. Examples of these frequencies are exhibited in Figure 3 where the 569 kHz and 381 kHz frequencies in the transmission spectrum show large scale variability, and 737 kHz and 470 kHz small variation over the same range.

The effects of heat on the water filled phononic crystal were almost exclusively a shift in the transmission band, with minimal effects on the shape. This is expected because the sound velocity of water can be considered uniform in the ultrasonic frequency range studied in this work. Indeed, examination of prior art reveals a lack of anomalous behavior in the velocity of sound over multiple decades of frequencies.
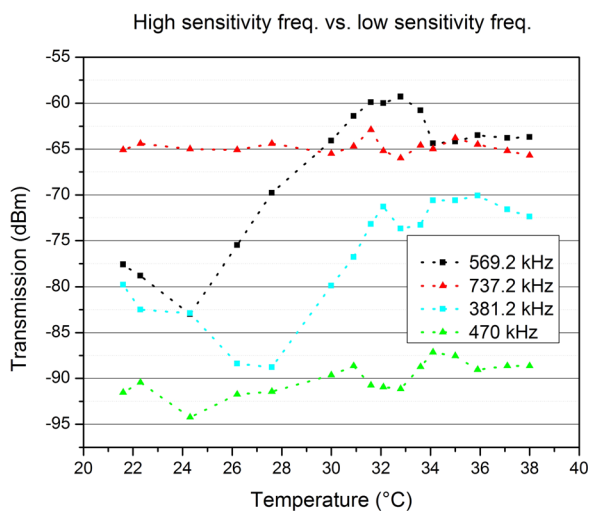

FIG. 3. Chart to illustrate the non-uniform frequency behavior of the filter. 569 kHz and 381 kHz both show significant variation over the temperature range examined. 737 kHz and 470 kHz maintain relative stability over the same range. Significant portions of the transmission bands exhibit the same characteristics.

Hydrogel, however, is subject to strong anomalous frequency and temperature dependence in the sound velocity. Using Brillouin scattering, which concerns the hypersonic GHz sound frequency region, Hirotsu *et al.* found the velocity of sound in bulk PNIPAm hydrogel to be roughly 1500 m/s below the LCST, and 2200 m/s above the LCST.[12] Other works focusing on the MHz region have found the velocity of sound to be very similar to water at roughly 1550 m/s both below and above the LCST. The authors' prior work on the same hydrogel, in the 200–800 kHz range, found the sound velocity to actually be lower than water both above and below the LCST, roughly 1350 m/s (below) – 1400 m/s (above). Additionally, strong indications of irregular frequency dependence of the sound velocity were revealed around and above the LCST from 26 °C to 35 °C.[13] For water, the increase in sound velocity versus temperature is not frequency dispersive. The resulting effect is primarily a shifting of the transmission bandstructure with increases in temperature. However, the expansion and minimal shifting of the central transmission band and the overall reshaping of the bandstructure above the LCST is indicative of the frequency dependence of the elastic properties of the PNIPAm hydrogel having an effect separate from water.

Further investigations were performed by modeling of the structure using an FDTD simulation software program to produce a transmission spectrum. PNIPAm hydrogel is a complex viscoelastic, semi-solid structure over 90 wt. % water in the hydrophilic state that increases in rigidity, dispersiveness, and density to varying degrees above the LCST.[12,17,19] In this case, even with gel shrinkage, the hylemorphic properties are maintained as the crystalline structure is not affected. Thus, the changing physical properties due to the changing state of the hydrogel are the largest contributors to the band reshaping that is seen in Figure 2.

For the modeling, the hylemorphic properties were set to the exact specifications of the phononic crystal. The attenuation constant, bulk and shear modulus, and viscoelastic properties of PNIPAm vary amongst cited literature. The probable frequency dependence of the speed of sound in the hydrogel was previously discussed. With special regard to the attenuation, Yuan *et al.* studied the attenuation properties of bulk form PNIPam hydrogels and found strong frequency dependence between 3 and 15 MHz.[16] Unfortunately, the lower limit of their work was 3 MHz, and the variation of damping between the highest frequency and the lowest ranged from roughly 25 dB/cm at 15 MHz to 4 dB/cm at the same temperature at 3 MHz. The behavior did not adequately provide an indication of what to expect at frequencies at order of magnitude lower than 3 MHz in the ~500 kHz range. The attenuation constant was also not able to be directly derived from the Fabry-Pérot resonances that were used to determine the sound velocity in the authors' prior work.[13]

Since the gel maintains properties as a semi-solid due to the large proportion of water that interstitially fills the polymer network, two models were simulated to qualitatively examine the bandstructure behavior. The same hydrogel used in the phononic crystal was also previously investigated by the authors, so the bulk and shear modulus parameters were derived directly.[13] The viscoelastic and attenuation

properties were estimated from multiple sources,[12,16,17,19] and the simulated results used only as a qualitative measure for the behavior of the resultant transmission spectra above/below the LCST. The simulation did not incorporate a frequency dependent speed of sound or attenuation as available literature was inadequate to produce a reliable model. The non-solid model (NS-Model) approximates the hydrogel as a non-solid with all of its derived elastic parameters, and attenuation equivalent to water (Figure 4, top). The solid model (S-Model) approximates the hydrogel as a solid with all of its derived elastic properties and attenuation constants as estimated from multiple sources.

Figure 4 is the resultant transmission band structure for a k_{00} propagating plane wave in comparison with the experimental results. Values for the physical parameters were either derived directly[13] or estimated from other works.[12,16–20] The discrepancy between the transmission band shape of the modeled and experimental results is apparent from Figure 4. However, close inspection of the simulated values corroborates the behavior of the measured transmission band.

The NS-Model predicts a blue-shifting and overall broadening of the transmission band between the LCST and ACST due, in part, to the increase in sound velocity of the hydrogel above the LCST. This shifting behavior is corroborated in the experimental results of water shown in Figure 1(c). Between 21 °C and 38 °C, the increase in sound velocity of pure water is nearly 35 m/s.[15,21] Though the effect of air bubbles trapped between the rods cannot be neglected, the increase in sound velocity of water significantly contributes to the blue-shifting in the transmission spectrum as shown in Figure 1(c). The NS-Model shows some significant discrepancies from the experimental measurements. The stop-bands are red-shifted from measured

values by nearly 50 kHz, the increasing transmission above/below the LCST is the reverse of experimental observation, and the overall blue-shifting of the transmission spectrum above the LCST does not occur in the experimentally measured samples. The transmission bands in both the LCST and ACST spectrum do, however, maintain similar breadth to the experimental results.

The S-Model modeled the PNIPAm hydrogel as a solid with the same elastic parameters as the NS-model and the addition of estimated attenuation. The behavior of the transmission spectrum qualitatively agrees with S-Model with these assumptions (Figure 4, center). The leading stop band edges are within 15 kHz of the experimental results. Though the attenuation increases above the LCST, the significant transmission increase above the LCST that is observed experimentally in the band centered around 530 kHz also occurs in the S-Model simulation. Additionally, the same band broadens by roughly 27% at the −6 dB points, with the center of the same band red-shifting above the LCST. The significant discrepancies between the S-Model and the experimental results are the lack in similarity in the shape of the transmission spectrum, the disagreement in the breadth of the ∼530 kHz transmission band, and the occurrence of partial stop gaps at various frequencies.

The variation between S-Model, NS-Model, and the experimental transmission spectra illustrate that further investigations are required into the physical properties of the bulk form of PNIPAm hydrogel. The solid/non-solid properties of the hydrophilic and hydrophobic hydrogel states are of special importance for accurate modeling in phononic crystal structures. Despite the discrepancies in both the S- and NS- Models, the qualitative behavior of the models and experiment indicates that the peculiar effects in the transmission spectra are primarily the result of the changing elastic properties of the hydrogel, and not of the speed of sound of the water.

In conclusion, we thereby demonstrate an optically responsive sonic structure that can modulate the filtering or transmission of ultrasonic waves through it. The modulation of the ultrasonic waves occurs due the electromagnetically induced volume phase transition of the PNIPAm hydrogel infiltrated within the periodic phononic structure.
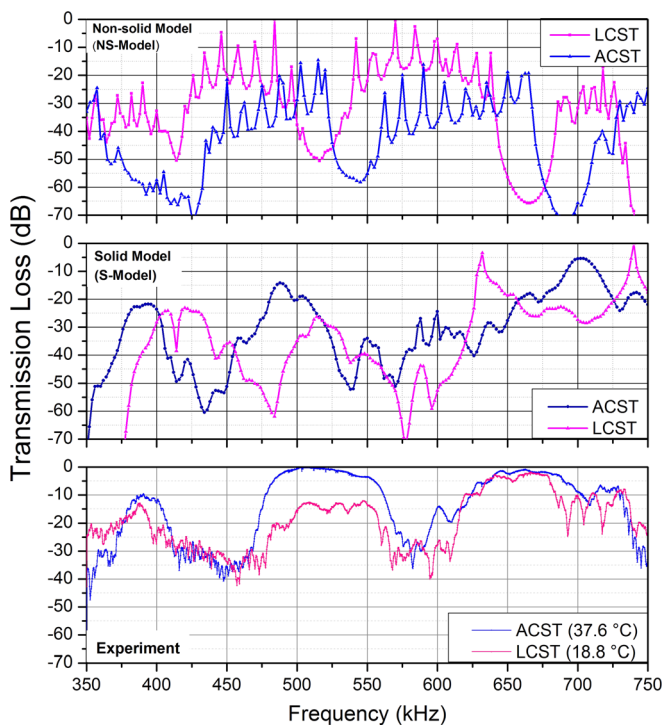

FIG. 4. Top: Transmission spectrum with parameters as non-solid. Middle: Transmission spectrum with hydrogel as solid. Bottom: Experimentally measured transmission spectrum.

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