

# Tunable Hybrid Phononic Crystal Lens Using Thermo-Acoustic Polymers

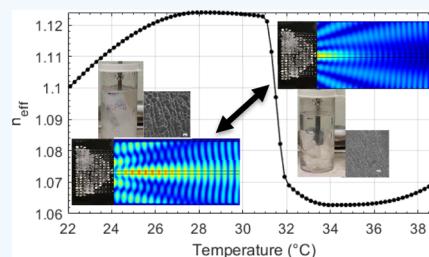
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**ABSTRACT:** Solid phononic crystal (PnC) lenses were made active on infiltration with thermosensitive polymers to produce a thermoactuated hybrid solid lens with variable focusing. Acoustic lenses, both solid state and PnC-based, are passive elements with a fixed focal length. Their focal characteristics are functions of the lens structure or the arrangement of the PnC unit cell. Dispersion effects, liquid-filled membranes, and phase delay in a multi-element emitter have been used for variable focusing. The high thermal, electric, and electromagnetic sensitivity of the elastic properties of poly(vinyl alcohol) (PVA) poly(*N*-isopropylacrylamide) (PNIPAm)-based hydrogels enable them to operate as tunable solids. However, these solids do not have strong enough contrast with water or well-controlled shape parameters to function as standalone lenses. Here, a tunable hybrid solid ultrasonic lens is realized by combining a PnC lens with PVA-PNIPAm thermoacoustic hydrogel to modify the transmission and dispersion properties of transient acoustic waves. Variable focusing is demonstrated from 40 to 50 mm using the anomalous thermosensitivity of the elasticity and speed of sound of the hydrogel.



## INTRODUCTION

Phononic or sonic crystals (PnCs, SCs) allow the fabrication of artificial structures from the composites of a periodically arranged material embedded within a medium with a large elastic contrast. The modification of the density, elastic parameters, and hylemorphic features, such as the size, shape, and arrangement of scatterers in the PnC strongly changes the behavior of transient mechanical or acoustic waves through the crystal. Controlling any of the properties enables functional material systems that may behave as filters,<sup>1</sup> cloaks,<sup>2</sup> negative index lenses,<sup>3</sup> and negative mass density<sup>4</sup> amongst other unique behaviors.<sup>5</sup>

The earliest demonstrations of phononic or sonic lenses utilized the linear dispersion region of the periodic band structure to create an effective medium to focus sound.<sup>6–8</sup> At low frequencies below the homogenization limit, the SC behaves as a homogenous object, where the filling fraction of the heterostructure scatterers combined with the contrast in elastic properties produces an effective index of refraction that results in the refraction sound. PnC lenses operating in the linear dispersion regime are governed by the same geometric optic laws of a traditionally homogeneous optical lens with the added advantage that the effective index of refraction can be readily controlled.<sup>9</sup> The ability to artificially control the bulk material properties makes the structures particularly exciting for their potential impacts in biomedical imaging and ultrasonic microscopy.

Focusing of mechanical waves, which include acoustic waves, is accomplished using either lenses, curved emission sources, or phased array elements. Lenses and curved emission sources

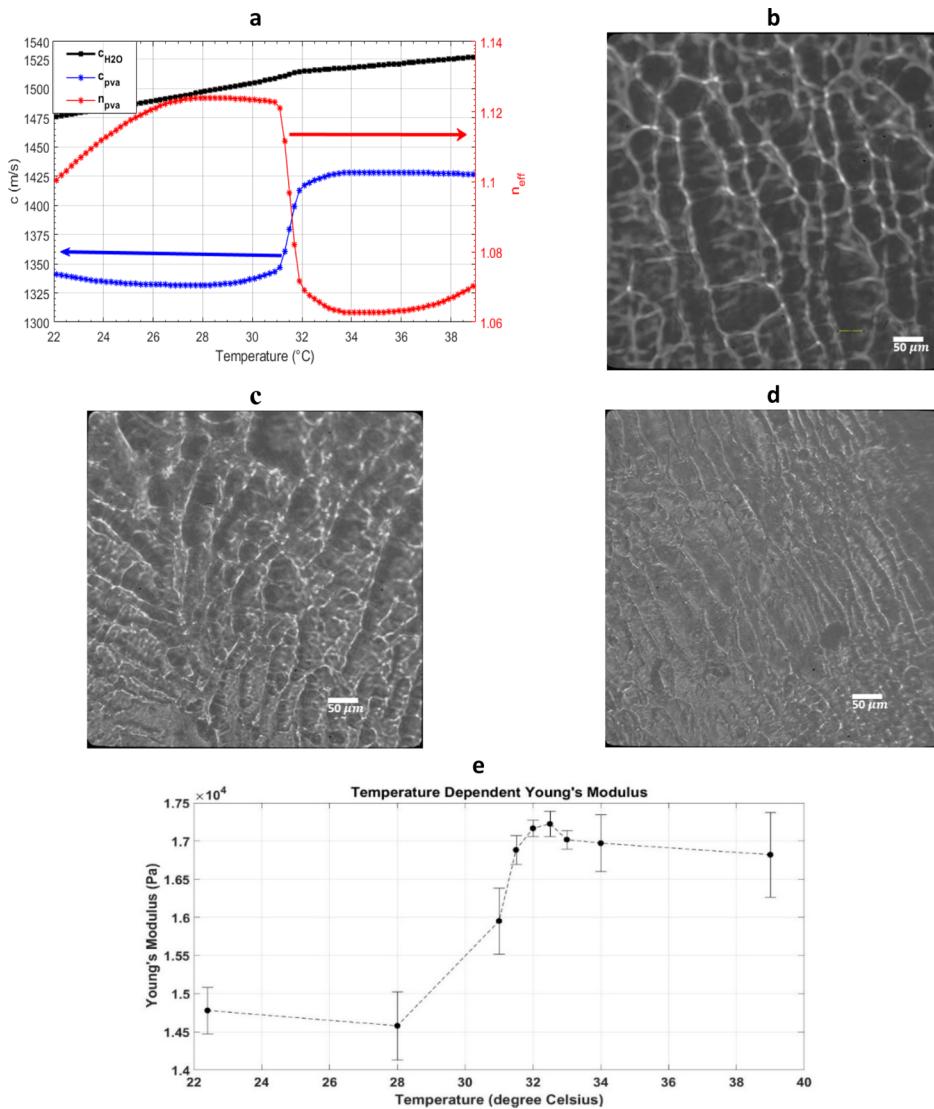
generally have a single focal point determined by their geometry (curvature, thickness) and mechanical characteristics. Moreover, the focal length of mechanical wave emission sources such as transducers is restricted to the Fresnel zone where the emitted waves behave as a planar. Ideally, however, the geometry of a lens allows for an arbitrarily long focal length, which extends the characterization capabilities of ultrasound in both organic and inorganic systems.

Whereas the focal length of solid lenses and curved emission sources is typically fixed, other mechanisms have been used to vary the focal length of an emission source. Phased array emission varies the focal length by dictating the location of the constructive phase interference between sound waves emitted from spatially separated elements. Yin et al. demonstrated dynamic focusing by obtaining the transfer function of a randomly scattering lens, then modulating an input pulse transient profile to shift its focal point.<sup>10</sup> Variable focusing has also been accomplished with fluidic lenses that pneumatically change the shape of a fluid-filled membrane.<sup>11,12</sup> Pneumatically changing the filling volume of the lens results in a lens with a dynamic radius, and thus a variable focal length.<sup>13</sup> For reflective focusing, metasurfaces have been used to induce a dispersive phase interference, resulting in a frequency dependent focal length.<sup>14</sup> Unlike both fluidic lenses and phased array sources, metasurfaces and transfer function-based tuning are frequency modulated.

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**Figure 1.** (a) Temperature dependent speed of sound for PVA-PNIPAm and water (left axis) and effective index of refraction for hydrogel derived from ref 17. PVA-PNIPAm undergoes a discontinuous volumetric phase transition at  $\sim 32$  °C resulting in an anomalous increase in the speed of sound, density, Poisson's ratio, and ultrasonic index of refraction. The (b–d) microscopy image of PVA-PNIPAm at room temperature (b), 31 (c), and 39 °C (d). The increasing density of the cross-link polymers is clearly seen with increasing temperature. (e) Young's modulus indicates the strong increase in stiffness above the LCST.

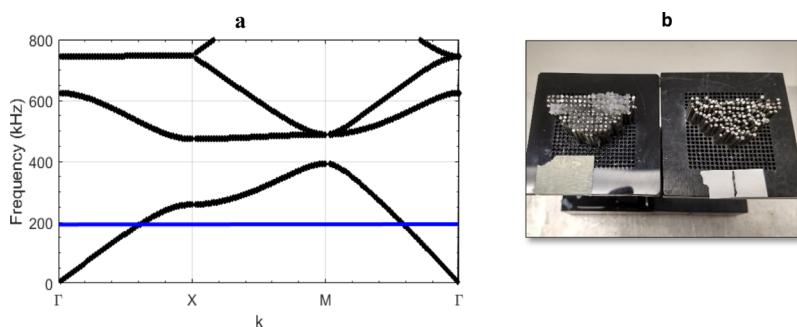
In prior works, we have demonstrated the control and modulation of acoustic waves through PnC by electromagnetic waves to realize active ultrasonic filters and modulators.<sup>15,16</sup> In this work, a new mechanism is presented to actively tune the focal point of a solid acoustic lens. The lens is comprised of a PnC lens embedded with a stimulus sensitive polymer, poly(vinyl alcohol) poly-N-isopropylacrylamide (PVA-PNIPAm) hydrogel. The PnC lens is solid, but the modulation of its focal length is achieved through thermally induced elastic property changes in the hydrogel component. Unlike prior approaches of tunable acoustic lens or sources, the variation in the focal length is not frequency modulated, a function of geometric optics, or phase coherence, but instead utilizes the modification in its intrinsic mechanical properties to realize a tunable solid lens.

## RESULTS

**Tunable Lens Design and Fabrication.** A PVA-PNIPAm hydrogel is a stimulus sensitive polymer with mechanical

properties that are anomalously dependent on temperature.<sup>17</sup> At the lower critical solution temperature (LCST) of  $\sim 32$  °C, the hydrogel undergoes a discontinuous volumetric phase transition as it goes from a hydrophilic to hydrophobic state. The result is mechanical properties that also demonstrate strong variance. The contrast in the change of speed of sound between water and hydrogel allows for the effects due to the hydrogel being easily distinguished. Based on the definition,  $n_{\text{eff}} = c/v$  where  $c$  is the speed of sound in the ambient medium, and  $v$  is the speed of sound in the hydrogel, the effective refractive index,  $n_{\text{eff}}$  ranges from 1.12 to 1.06 (Figure 1a). As water is the ambient medium in this work, the anomalous change in the speed of sound near the phase transition temperature is strongly represented in the effective index.

Ideally, PVA-PNIPAm and variants could serve as stand-alone acoustic lenses. Their tunable elastic characteristics, specifically speed of sound, make them materials with effective acoustic index of refraction that is variable under an external stimulus. However, the volumetric phase transition associated



**Figure 2.** (a) Band structure calculated using the RBZ for the base, square lattice PnC in PVA PNIPAm ambient. Tunable focusing is demonstrated in the first transmission band. (b) Top view of the PnC lens both with (left) and without (right).

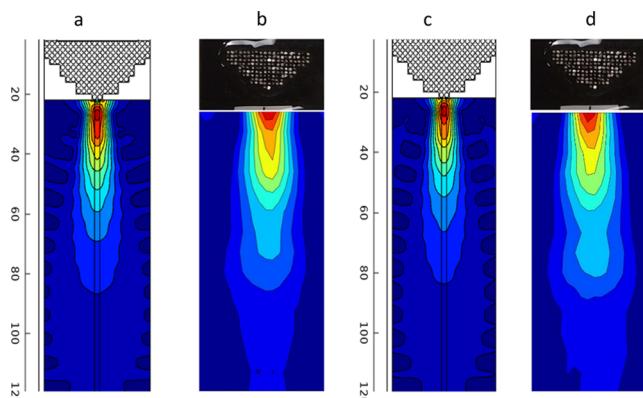
with the stimulus induced changes make maintaining a controlled shape for lensing challenging. Figure 1b–d show the compression of the hydrogel at below, near, and above the LCST. Shrinkage above the LCST occurs because of NIPA polymer chains shedding bonds with free water molecules, expelling them from the cross-linked network, to self-bond. The process is thermodynamically repeatable and reversible. However, additional time for equilibrium when going from a hydrophobic to hydrophilic state is necessary because the adsorption time of water molecules into the bulk gel network. The selection of PVA-PNIPAm over pure PNIPAm was due to the faster and more dramatic elastic and volumetric changes in the PVA-PNIPAm synthesized bulk hydrogels as investigated internally reported elsewhere.<sup>16</sup> The same work also details the reversibility of the process by turning on/off an electromagnetic stimulus and mapping the time-dependent dispersion. The synthesis procedures are given in the **Materials and Methods** section.

The challenges of maintaining a controlled shape were best addressed by combining the hydrogel with a more stable structure. A PnC lens, which maintains an effective index based on the periodicity, composition, and filling fraction of the lattice, was used to provide a stable framework for the hydrogels.<sup>9</sup> By filling the interstitial lattice spacing with hydrogel, the tunable index of refraction is readily leveraged for tunable focusing. The strongly anomalous in stiffness as indicated by the Young's modulus also affects the elastic contrast between the stainless steel scatterers and the hydrogel, further enabling the tuning of transmission (Figure 1e).

PNIPAm hydrogels occupy a unique physical state between liquid and solid due to their hydrophilic and hydrophobic nature.<sup>18</sup> At the LCST of  $\sim 32$  °C, PNIPAm-based hydrogels undergo a volumetric phase change caused by the transition between hydrophilicity below the LCST and hydrophobicity above the LCST.<sup>18</sup> In the hydrophilic state, hydrogels can readily be comprised of greater than 90 wt % water with a density nearly equal that of water, and a speed of sound equal to water at frequencies above 1 MHz. The center frequency of the emission source used in this work, however, is 0.5 MHz, and dispersion causes the hydrogels to have a lower speed of sound in this frequency range.<sup>17</sup>

The selection of a planoconvex design for the lens was based on the limitations of the fabrication techniques, available characterization resources, finite element modeling (FEM) software COMSOL, and the ability to utilize PVA-PNIPAm for tunable focusing. The band structure of the PnC was calculated and a frequency range selected that coincided both with the first transmission band and capabilities of utilized emission and

detection sources (Figure 2a). Within the first band, the PnC behaves as an effective medium, allowing for the basic principles of geometric optics to be used to guide the design of the lens. A plano-convex was selected, simulated in COMSOL to verify the existence of focusing within a measurable region, and experimentally verified at 215 kHz both below and above the phase transition temperature of the hydrogel. For water, the temperature does not discernably shift the focus of the lens either numerically or experimentally as shown in Figure 3.



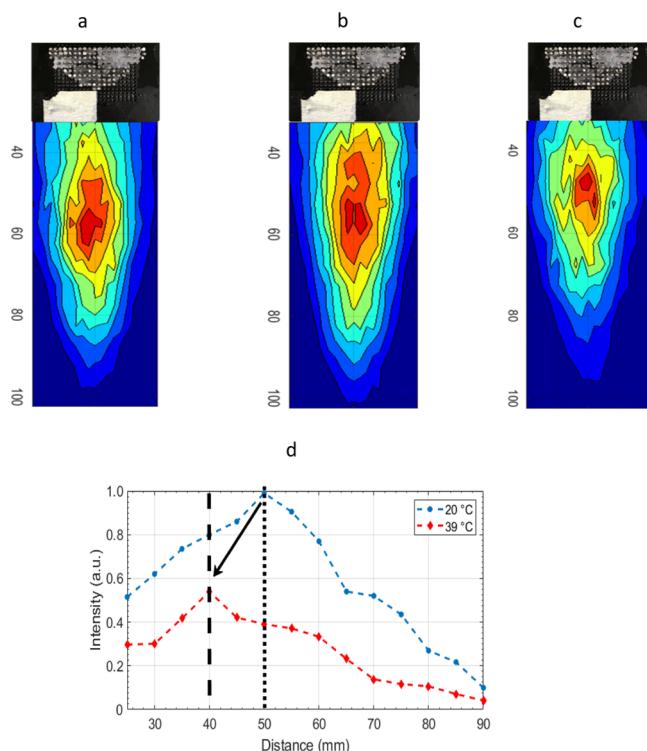
**Figure 3.** Modeled (a,c) and measured (b,d) sound fields of the PnC lens without hydrogel at room temperature (a,b) and 39 °C (c,d). The experimental measurements start about 20 mm from the surface of the lens in contrast to the modeled measurement which shows the entire sound field. The discrepancy is due to the overhang of the lens-stabilizing plate. The increasing temperature in water does not discernably shift the focus of the phononic lens.

The device is composed of a PnC lens operating in water ambient with the thermosensitive polymer, PVA PNIPAm interstitially filling the spacing between the PnC scatterers (Figure 2b). The lens was fabricated from a square lattice of stainless steel cylindrical rods of radius 0.80 mm and lattice constant,  $a$ , of 1.96 mm. PVA-PNIPAm was synthesized using free radical polymerization to form a bulk hydrogel, and filled into the PnC lattice spacing following the procedure from other works.<sup>16</sup> The lens was formed by first polymerizing the hydrogel in a  $10 \times 20$  period PnC, where the thickness is 10 periods along the direction of wave propagation, then rods and hydrogel removed to form a lens with a radius  $\sim 10a$ .

## ■ DISCUSSION

The ultrasound velocity of PVA-PNIPAm was measured to be 1341 m/s at 22 °C, 1415 at 34 °C, and 1426 m/s at 39 °C, respectively. As the speed of sound is indicative of the elastic properties of a material, the abrupt increase in the speed of sound (Figure 1a) has been related to the increased elastic stiffness and higher density.<sup>18,19</sup> In a hybrid PnC structure, modulating the phase of the hydrogel thus will impact the effective medium of the lens, leading to tuning of its behavior. From Figure 1a, it is apparent that though the speed of sound in water increases with temperature, the gradual increase is much less than that of PVA-PNIPAm. Moreover, the temperature-dependent volumetric phase transition of the polymer enables a unique way of modifying the focal length of the polymer-infiltrated lens reversibly.

Figures 3 and 4 show the temperature-dependent behavior of the phononic lens both with (Figure 4) and without (Figure



**Figure 4.** Experimentally measured sound field of the tunable lens at room temperature (a), 20 °C, the LCST at 31 °C (b), and (c) above the LCST at 39 °C. (d) Lens focuses to 50 mm at room temperature below the LCST to ~40 mm above it.

3) hydrogel. For clarity, the lens without hydrogel is termed the sonic lens (SL), and with hydrogel termed the tunable SL (TSL). At room temperature, the SL has a focal point of 25 mm. An increase of the water temperature from 20 to 39 °C has a negligible effect on focusing length as observed from Figure 3a–d. Over this temperature range, the speed of sound in water increases from 1484 m/s at 20 °C to 1524 m/s at 39 °C, an increase of roughly 2.6% while the hydrogel anomalously increases 6.3%. In addition to the change in the speed of sound, the density and bulk modulus also maintain a discontinuous dependence on temperature.<sup>17</sup> Prior works have shown that heating water does cause blueshift of the dispersion curve towards higher frequencies, but the shift does not

necessarily change the relative dispersion relation that strongly impacts the effective index of refraction of the lens.<sup>6,9,16</sup>

The acoustic wave propagation through the SL is significantly modified because of the infiltration of the hydrogel polymer within the matrix as shown in Figure 3. It can be observed that at 20 °C, which is far below the LCST, the lens has a significantly longer focal length of 50 mm compared to ambient water at the same temperature. The focal length undergoes shifting as the hydrogel undergoes its phase transition, reducing to ~46 mm as the temperature approaches the phase transition temperature around 31 °C. As the temperature is further increased above LCST to 39 °C, the focal length increases to 40 mm (Figure 4).

The temperature-dependent behavior of the focal length of the hybrid SL due to the modification of the phase of the polymer can be explained by studying the acoustic properties of the PVA-PNIPAm used as an active material of the lens. The speed of sound in PVA-PNIPAm and hydrogels can be highly dependent on the frequency of the acoustic wave.<sup>17</sup> Additionally, hydrogels not only undergo an anomalous change in the speed of sound with temperature, but also density, Poisson ratio, and other mechanical properties like stiffness.<sup>18</sup> As the dispersion relation is dependent on the contrast in the elastic parameters between the stainless steel scatterers and the hydrogel that interstitially fills the lattice spacing, the anomalous behavior of the elastic properties of the hydrogel with temperature more strongly impact the effective index of refraction and thus the focal length.

For the current work, the sound field was measured using a needle hydrophone that was raster scanned, requiring significant amounts of time for complete data acquisition. Resistive strips were used to heat the water in ambient temperature and maintain a constant temperature throughout the measurement. Neither the raster scanning nor temperature control mechanisms were adequate to test the rate at which the focus could be manipulated in situ. However, the ideal limit to the rate of tuning is a function of speed at which a hydrogel can undergo its phase transition and the penetration of the stimulus through the entire device. Changes of 75% swelling/deswelling have been achieved for PNIPAm-based hydrogels in nearly 1 s with an electrical stimulus.<sup>20</sup> Incorporation of fast-switching PNIPAm composites with an associated stimulus would result in tunable focusing on the scale of seconds. Further developments would be necessary in fast-switching polymers to approach tuning speeds of the current phased array scanning systems.

Tunable focusing for acoustic waves is currently achieved primarily through phase delay in phased array emitters and pneumatic, fluid-filled membrane lenses. The hydrogel-filled PnC lens differs from both in that it is completely solid. The discontinuous nature of the volumetric phase transition in PNIPAm-based hydrogels leads to variable focusing because of the strong dependence of the mechanical properties on temperature and frequency. Though temperature is used as the control in this work, hydrogels have been actuated electrically,<sup>21</sup> magnetically,<sup>21</sup> with pH,<sup>18</sup> and even through biomimetic stress.<sup>22</sup> The result is a solid lens that can be controlled using various stimuli.

## ■ CONCLUSIONS

The ability to control the band structure of PnCs has resulted in distinct advantages compared to traditional homogenous structures. For example, cloaking,<sup>23</sup> super resolution,<sup>3</sup> focusing

of elastic waves,<sup>24</sup> flat lenses,<sup>25</sup> and self-collimation<sup>26</sup> uniquely derive their properties from the wavevector manipulation only achievable due to wave propagation in periodic structures. Techniques dynamically changing the point of focus for an ultrasonic wave currently include pneumatically-actuated, liquid-filled membranes, or phase delay to achieve tunable focusing. Currently, a solid object cannot change the focal length of an ultrasonic wave without also changing the frequency. Here, we presented the first tunable hybrid ultrasonic solid lens that has a dynamic focal range of 25% when shifting from a focal length of 40 mm at 39 °C to 50 mm at 20 °C. The lens is comprised of a square lattice stainless steel PnC with the stimulus sensitive, thermoacoustic hydrogel, PVA-PNIPAm, interstitially filling the spacing between the scatterers, and functions in water ambient at ultrasonic frequencies.

## MATERIALS AND METHODS

**FEM Modeling.** FEM software COMSOL was utilized to model the PnC lens as pressure waves solved via the Helmholtz equation coupled to solid mechanics for numerical analysis. In addition to water, the physical properties of PVA-PNIPAm hydrogel such as density, speed of sound, bulk, and Young's modulus, and Poisson ratio were measured and previously reported,<sup>17</sup> and implemented into the COMSOL model. The emission source comprised an incident wave, perpendicular to the lens optical axis, and the simulations were performed at room temperature (20 °C) and 39 °C. As the filling fraction of the hydrogel is not 100% in either the hydrophilic or hydrophobic state experimentally, FEM modeling was utilized only to qualitatively determine if shifting would occur in the ideal case of 100% filling fraction.

**Hybrid Lens.** The device is composed of a PnC lens operating in water ambient with the thermosensitive polymer, PVA-PNIPAm interstitially filling the spacing between the PnC scatterers (Figure 2b). The lens was fabricated from a square lattice of stainless steel cylindrical rods of radius 0.80 mm and lattice constant,  $a$ , of 1.96 mm. PVA-PNIPAm was synthesized using free radical polymerization to form a bulk hydrogel. A 400 g tub of monomer solution of the PVA-PNIPAm monomer solution was made by magnetically stirring *N*-isopropylacrylamide (NIPA, Polysciences, Inc.), *N,N'*-methylenebisacrylamide (BIS, Polysciences, Inc.), and a 5% DI water diluted PVA (Polysciences, Inc.) solution in ratios of 0.10 (wt): 0.02 (mol. NIPA): 0.84 (wt) at 70 °C for >24 h.

To interstitially fill the spacing of the hybrid lens, a 10 × 20 period PnC was placed in the monomer solution in a sealed container. The entire container was then placed in an ice bath, with  $N_2$  continuously pumped into the monomer solution while being magnetically stirred on high speed for >30 min. The ammonium persulfate initiator and *N,N,N',N'*-tetramethyl-*ethyl*enediamine accelerator solutions were made by mixing with water in ratios of 0.1 (APS, TEMED)/0.9 (H<sub>2</sub>O). The initiator and accelerator were then injected into the container with the monomer solution and PnC. Injection occurred in a  $N_2$  atmosphere until the hydrogel polymerized. For final preparation, the impinging surfaces of the PnC were cleared of the PVA-hydrogel and rods and hydrogel removed to form a hybrid lens with a radius ~10 $a$ .

**Measurements.** Measurements were performed in a bistatic thru-transmission arrangement in a 51 cm × 51 cm × 46 cm acrylic tank. The emitting transducer was at the plane side of the plano-convex lens, and a needle hydrophone

detector scanned over the output area of the lens. Transmission was measured with both pulsed and continuous wave emission in separate experiments to evaluate qualitative reliability and reproducibility.

The emitting source was a Panametrics V301 0.5 MHz, 1.0" unfocused plane wave immersion transducer. For continuous wave emission, the transducer was connected to a Teledyne-LeCroy WaveStation 2012 wavefunction generator. The wavefunction generator was operated in a continuous wave mode, swept over the frequency range from 200 to 300 kHz with a sweep time of 300 s and resolution bandwidth of 600 Hz. The range was selected to contain frequencies in the linear portion of the first transmission band, that is, the homogenization limit (Figure 2a). For pulsing, the transducer was connected to an Imaginant DPR300 Pulser-Receiver set at 0 dB gain. A Müller-Platte 0.5 mm needle probe, connected to a Tektronix MDO3024b spectrum analyzer, was raster scanned over a 70 × 32 mm area starting 25 mm from the edge of the tunable lens in 5 mm increments along the propagation axis, and 2 mm increments along the axis perpendicular to propagation.

Temperature was controlled using an Omegalux resistive tape heater combined with a thermocouple placed adjacent to the emitting transducer. The water was heated at a rate of 0.3 °C per h with an equilibration time of 10 min once a target temperature was reached. The sound field was mapped at 20.5, 31.5, and 39.5 ± 0.5 °C for both a lens with and without the hydrogel to determine the impact of the hydrogel of focusing capabilities. 20.5, 31.5, and 39.5 °C serve to demonstrate effects below, near, and above the LCST.

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### Notes

The authors declare no competing financial interest.

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