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Direct observation of a hypersonic band gap in two-dimensional single crystalline phononic structures

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Brillouin light scattering is employed to record the phonon dispersion relation of two-dimensional (2D) hypersonic phononic crystals with square lattice plane group $p4mm$ symmetry. The samples are single crystalline arrays of cylindrical holes with a lattice constant of 750 nm and 35% porosity patterned in epoxy using interference lithography. The dispersion relation reveals the presence of a phononic band gap between 1.21 and 1.57 GHz at the edge of the first Brillouin zone for elastic waves propagating along the [10] direction and conclusively demonstrates a band gap in a single crystalline 2D polymer based phononic structure at hypersonic frequencies. © 2007 American Institute of Physics. [DOI: 10.1063/1.2786605]

The ability to engineer a dispersion relation of high frequency acoustic phonons opens up an exciting opportunity to influence acoustic and acousto-optical properties of materials.^{1–4} Hypersonic phononic crystals have been actively investigated over the past several years because of their ability to forbid propagation of acoustic phonons within certain frequency windows.^{5–8} As a result, hypersonic phononic crystals have been suggested to enhance acousto-optical interactions,^{3,9–11} to realize stimulated emission of acoustic phonons in superlattices,¹² and to increase the efficiency of light emission in silicon.¹³

Fabrication of hypersonic phononic crystals and mapping of their phonon dispersion present significant experimental challenges. Fabrication involves creating periodic patterns with submicron lattice constants, which can be achieved using self-assembly or lithographic approaches. Lithographic methods possess significant advantages because they allow fabrication of single crystalline structures, where direction-resolved measurements are possible. Experimental characterization of hypersonic crystals is done using various optical techniques. In particular, Brillouin light scattering (BLS) has been used due to its ability to measure the phonon dispersion relation $\omega = \omega(\mathbf{k})$ directly.^{6,8,14–16} The ability to measure the dispersion relationship along any direction in the Brillouin zone is valuable because, in addition to determining the location of band gaps, it allows us to study the detailed features of the propagation bands.

Until recently, most studies have focused either on one-dimensional systems,^{5,14,17} or on self-assembled poly(styrene-*b*-isoprene) gels with very low mechanical contrast.^{15,16} The first experimental observation of a hypersonic band gap was reported by Cheng *et al.*⁸ for a three-dimensional assembly of colloidal particles. However, due to the nature of the fabrication technique, their samples were polycrystalline making it difficult to control the measurement direction. Measurements of the dispersion relation of single crystalline phononic structures were previously done on two-dimensional (2D) single crystals consisting of hexagonal arrays of air holes in epoxy matrix.⁶ While multiple higher

phonon propagation bands were observed, the band gap between the first and the second propagation bands was at frequencies below the BLS detection limit due to the relatively large lattice constant of the crystals (1.36 μm).

Here we report experimental observation of a hypersonic band gap along the [10] direction in a single crystalline epoxy/air sample using BLS. The square lattice constant was $a=750$ nm and the radius of the holes was $r=0.33a=250$ nm resulting in $\phi=35\%$ air volume fraction. The samples were fabricated using multibeam interference lithography. Fabrication involves the sequential, double exposure with a 90° rotation of the sample on a rotation stage relative to the interference pattern of two equal intensity laser beams and the transfer of the resultant intensity pattern into a photoresist via cationic polymerization. The glass substrate was pretreated with a 700-nm-thick buffer layer of a cross-linked SU8 material to assure the firm adherence between the structured film and the substrate. The interference lithography (IL) exposure was done using a 532 nm cw (Nd doped yttrium aluminum garnet) laser with an intensity of 1.5 W for 10–15 s. After baking the 6- μm -thick film at 75 °C for 3 min, the uncured regions are developed away in propylene glycol monomethylether acetate followed by rinsing with isopropanol to yield the 2D photopatterned structure.

Scanning electron microscope (SEM) images of the top view and the cross section of the 6- μm -thick film are shown in Figs. 1(a) and 1(b), respectively. They reveal an excellent uniform, single crystalline sample over a large area.

BLS employs scattering of a laser beam by random thermal phonons in the sample. A schematic representation of our measurement geometry is shown in Fig. 2. The angle between the propagation direction of the incident light defined by \mathbf{q}_i and the normal to the sample plane \mathbf{n} is equal to the angle between the propagation direction of the scattered light defined by \mathbf{q}_s and the normal to the sample plane \mathbf{n} and is equal to the half of the scattering angle θ . This configuration ensures that the phonon wave vector \mathbf{k} is always parallel to the sample plane. In this geometry the amplitude of the phonon wave vector \mathbf{k} is determined entirely by the scattering angle θ . The direction of \mathbf{k} in the Brillouin zone is controlled by the orientation angle φ . The polarization of the incident and scattered light was maintained perpendicular to

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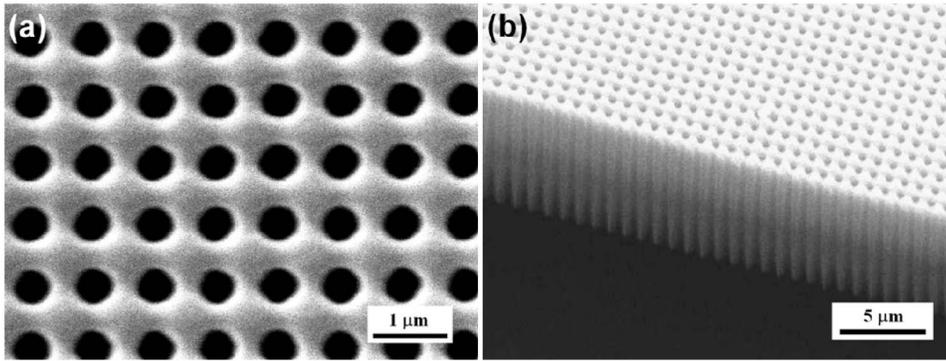


FIG. 1. SEM images of the top view (a) and the cross section (b) of the 2D crystals. The lattice constant is 750 nm, the hole size is 250 nm, and the film thickness is 6 μm .

the scattering plane [vertical-vertical (VV) geometry].

To eliminate the effects of optical diffraction and minimize the elastic light scattering, the samples were infiltrated with a refractive index matching fluid. We used phenylmethyl silicone with $n=1.62$ (at 532 nm), $\rho=1.097\text{ g/cm}^3$, and $c_L=2030\text{ m/s}$ for infiltration. The refractive index of SU8 epoxy is $n_{\text{epoxy}}=1.62$ (at 532 nm), while its density and sound velocities are $\rho_{\text{epoxy}}=1.19\text{ g/cm}^3$, $c_{L,\text{epoxy}}=2860\text{ m/s}$, and $c_{T,\text{epoxy}}=1800\text{ m/s}$, respectively. The index matching fluid eliminates undesirable optical effects, but at the same time, the mechanical contrast decreases considerably after fluid infiltration.

The BLS spectrum taken at $k=4.1\ \mu\text{m}^{-1}$ along the [10] direction, which corresponds to the magnitude of the wave vector at the edge of the first Brillouin zone, is shown in Fig. 3(a). There are three phonon peaks present in the spectrum. The highest intensity peak (g) at 3.72 GHz comes from the glass substrate quasilongitudinal phonons, while the weaker peaks (1) and (2) at 1.21 and 1.57 GHz, respectively, represent phonons propagating in the first and second band of the 2D polymer based phononic crystal. The peak frequencies were obtained by fitting the experimental spectrum with multiple Lorentzian oscillators. The overall fit is plotted in the lower part of the graph using a solid line, while two individual oscillators representing phonons from the first and the second propagation band of the phononic crystal are plotted with dashed lines. The split in the frequencies between modes (1) and (2) $\Delta f=1.57-1.21=0.36\text{ GHz}$ defines the size of the phononic band gap along the [10] direction. The ratio of the gap width to its midgap frequency is $\xi=0.36/0.5(1.21+1.57)\times 100\% \approx 26\%$. To confirm that there are no phonon states in the band gap at other values \mathbf{k} we did BLS measurements for a range of phonon wave vectors from $k=2$ to $k=10\ \mu\text{m}^{-1}$. The results are shown in Fig. 3(b), where the modes of the periodic pattern are plotted with circles and the modes of the glass substrate—with triangles. Furthermore, at higher wave vectors we resolve an additional

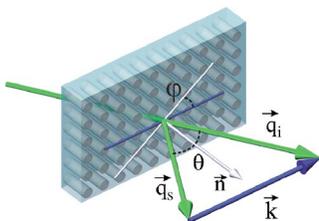


FIG. 2. (Color online) Geometry of BLS experiments: the amplitude of the phonon wave vector \mathbf{k} is determined by the scattering angle θ , while its direction in the Brillouin zone is controlled by the orientation angle φ ; note also that \mathbf{k} is always parallel to the sample plane.

mode plotted with diamonds, which represents the contribution from longitudinal phonons propagating in the unpatterned buffer layer.

The phonon dispersions of the glass substrate and the buffer layer are linear with the sound velocities of 5708 and 2860 m/s for glass and epoxy, respectively. In contrast, the phonon dispersion of the periodic pattern is nonlinear with significant amount of band bending, especially in the vicinity of the first Brillouin zone edge, which leads to the formation of the band gap for longitudinal phonons.

The theoretical phononic band diagram for 2D epoxy/air square crystal with 30% porosity computed using finite ele-

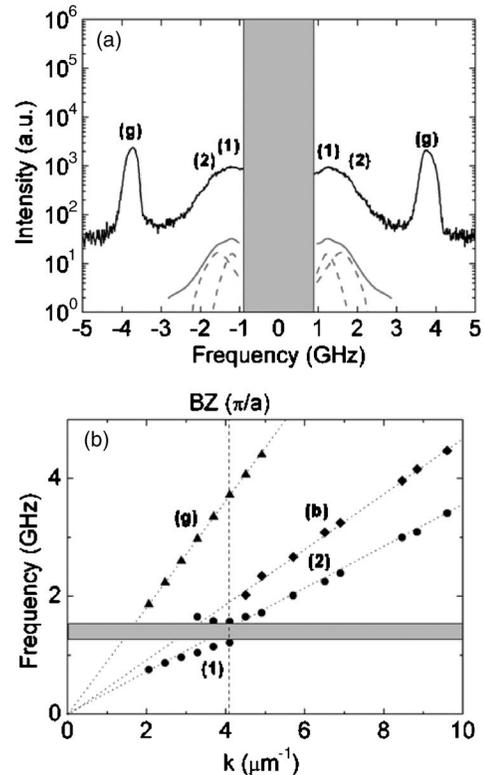


FIG. 3. (a) BLS spectrum taken at $k=4.1\ \mu\text{m}^{-1}$ that corresponds to the edge of the first Brillouin zone along the [10] direction. Peaks (1) and (2) represent phonon modes from the first and the second propagation bands and peak (g) comes from the longitudinal phonons of the glass substrate. To obtain the frequencies of phonon modes, the experimental data were fitted with a multiple Lorentz oscillator model. The resultant fit is plotted in solid line, while the oscillators representing phononic crystal modes are plotted in dashed lines. (b) Phononic dispersion relation along the [10] direction showing a partial band gap between 1.21 and 1.57 GHz (in gray). Circles represent the phononic modes of the pattern, diamonds the buffer layer, and triangles the glass substrate.

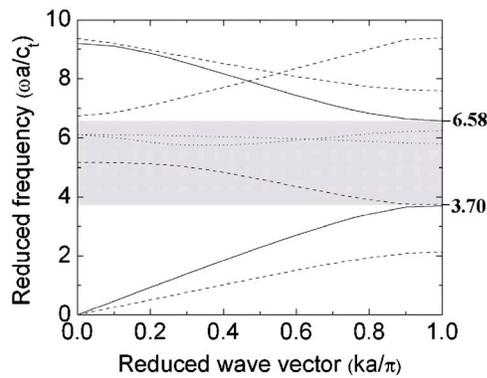


FIG. 4. Theoretical band diagram for a 2D square epoxy/air phononic crystal with 30% porosity for the phonon wave vector along the [10] direction. QL modes are plotted with solid lines, QT modes with dashed lines, and mixed modes with dotted lines, where c_t refers to the transverse velocity of the epoxy. The partial band gap for QL phonons is highlighted in gray.

ment analysis is shown in Fig. 4. Quasilongitudinal (QL), quasitransverse (QT), and mixed modes are plotted with solid, dash and dotted lines, respectively. In the VV geometry only the QL modes are detected experimentally. Therefore, the experimentally observed partial phononic band gap [Fig. 3(b)] corresponds to the gap between the first and the second propagation band for the QL modes. Theory predicts that the ratio of the gap width to its midgap frequency for the QL gap should be $\xi = 6.58 - 3.70 / 0.5(6.58 + 3.70) \times 100\% = 56\%$, which is more than twice the experimental ratio. This is expected since the contrast in density and sound velocities between epoxy and air is much larger than that between epoxy and the refractive index matching fluid.

In summary, we investigated the phonon dispersion relation of single crystalline two dimensional phononic crystals consisting of a square array of air holes in epoxy matrix. We observed significant bending of the first and second propaga-

tion bands in the vicinity of the edge of the first Brillouin zone along the [10] direction. This bending results in the formation of a phononic band gap between 1.21 and 1.57 GHz.

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