Brillouin-Mandelstam Light Scattering Spectroscopy of the Nanoscale Phononic Superlattice Arrays

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Abstract: We used the Brillouin-Mandelstam light scattering spectroscopy to demonstrate strong modification of the phonon dispersion in two-dimensional alumina superlattices with nanometer feature sizes. Non-linearity of phonon dispersion in the phononic superlattice arrays creates new possibilities for controlling the optical, thermal and electronic properties of such structures.

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Photonic band gap materials made profound impact on the photonics and non-linear optics fields. Photonic band gap materials involve structures with periodic modulation in the dielectric constant on the appropriate length scale as compared to the light wavelength [1-2]. Photonic band gap materials allow for control of the light wave propagation. The photonic band gap materials, to a large degree, emerged owing to the analogy between electromagnetic wave propagation in multi-dimensionally periodic structures and electron wave propagation in semiconductor crystals [1]. In recent years the problem of heat removal became crucial for many electronic and optoelectronic technologies [3]. The latter motivated the search for new methods of tuning phonon propagation at nanometer scale. Phonons are quanta of crystal lattice vibrations, which are the main heat carriers in semiconductors and dielectric materials. Using optics analogy, the phononic band gap materials can be formed by periodic modulation of the acoustic impedance $\zeta=\rho\times v$ on the length scale below the phonon mean free path (ρ is the mass density and v is the group velocity of the acoustic phonons). The acoustic phonons, which carry most of heat, have linear energy dispersion relation near the Brillouin zone (BZ) center. Theoretical considerations show that periodic modulation of the acoustic mismatch can strongly change the phonon dispersion by inducing strongly non-linear dispersion branches, changing the group velocity of the main "true acoustic" phonon branch, and creating stop bands in certain energy ranges [3-4]. However, direct investigation of the modification of the phonon dispersion in the relevant frequency range and correlating it with the changes in the phonon heat transport has been challenging.

In this work we used the Brillouin-Mandelstam light scattering (BMS) spectroscopy to directly monitor the phonon dispersion in two-dimensional (2D) alumina (Al_2O_3) arrays with the hexagonal periodic arrangement of nano-pores. The in-house assembled instrumentation allowed us to investigate the phonon dispersion in the vicinity of the BZ center and correlate it with the measured thermal diffusivity. The alumina samples used for the study had cylindrical pores with the diameters of 25 nm and 40 nm. The inter-pore distances were 65 nm and 105 nm, respectively. The BMS spectrometer had the spectral resolution close to ~0.1 GHz. We were able to tune the wave vector of a phonon participating in the light scattering by changing the instrument geometry. The light scattering spectra were excited with the laser operating at λ=532 nm. Figure 1 shows the instrumentation, microscopy of the sample, Raman spectrum and thermal diffusivity data. The BMS light scattering measurements were performed using the six-pass tandem Fabry-Perot interferometer. In phononic periodic arrays, the scattered light can originate from bulk scattering or surface ripple scattering mechanisms. Figure 2 shows BMS spectrum from the samples with 25 nm diameter pores and 65 nm inter-pore distance. The Lorentzian fit was used to find the peak positions associated with the acoustic phonons at specific wave vector q. In the shown spectra, the peaks located at ~+/-42 and ~+/-22.5 GHz correspond to the bulk *LA* and *TA* modes of alumina, respectively. As the incident angles changed from 0 to 50 degrees with respect to the normal to the sample surface, the phonon wave vector associated with surface ripple mechanism changed from q = 0 to q = 0.018 nm^{-1}. As a result, we observed additional phonon branches with strongly non-linear dispersion, which appeared owing to the periodic boundary conditions of the 2D array. The strength of the modification of the phonon spectra, revealed with the inelastic light scattering, was correlated with the changes in the phonon heat conduction. Based on the results, we argue that the phonon band gap materials offer a potential way for controlling the heat transport at nanometer scale similar to the light propagation in the photonic band gap materials.

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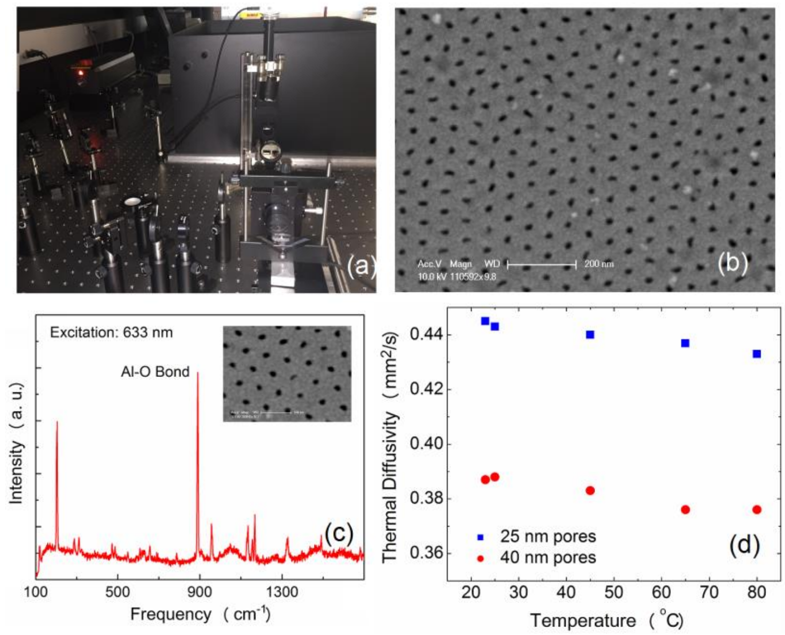

Figure 1: (a) Brillouin-Mandelstam light scattering experimental setup; (b) scanning electron microscopy image of the phononic crystal array; (c) Raman spectrum confirming the material composition; (d) thermal diffusivity data for two periodic arrays with different diameters of the pores and inter-pore distances.

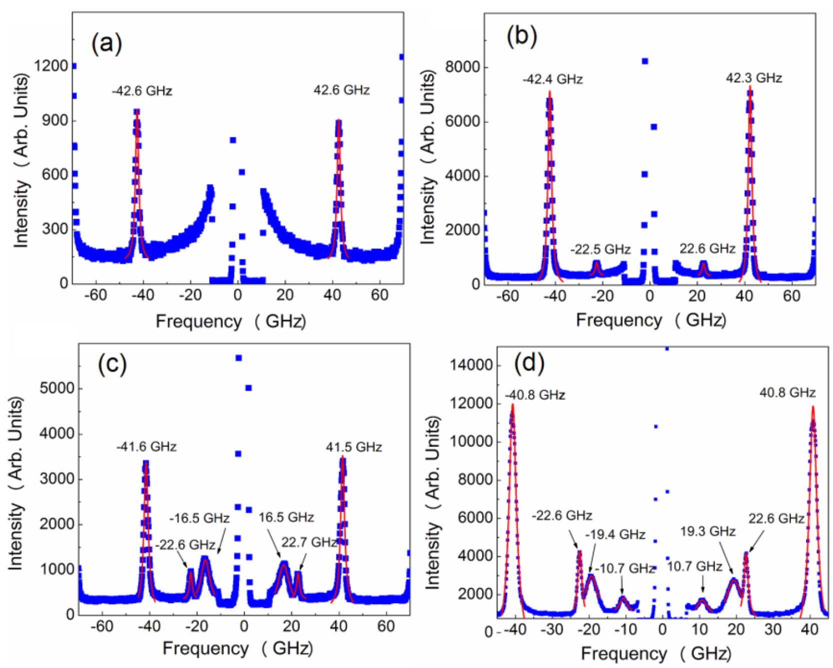

Figure 2: Brillouin-Mandelstam light scattering spectrum from the phononic crystal implemented with nano-porous alumina shown for the phonon wave vector (a) $q = 0\ nm^{-1}$ (b) $q = 0.008\ nm^{-1}$ (c) $q = 0.015\ nm^{-1}$ and (d) $q = 0.018\ nm^{-1}$. Periodic change in the acoustic impedance results in strongly non-linear phonon dispersion near BZ center.

References

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