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Fabrication and Modelling of Three-Dimensional Sub-kelvin Phononic Crystals

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Abstract. We have investigated the fabrication and computational modelling of three-dimensional phononic crystals for the observation of full band gaps for thermal phonons at sub-kelvin temperatures. Self-assembled arrays of monodisperse polystyrene nano-spheres have been fabricated using a vertical deposition technique. Optimal conditions for increasing crystal domain size and crystalline quality have been studied. In addition, the phononic band structure has been computed using the finite element method for the simple cubic lattice. The dependence of band structure on contact area between spheres has also been studied. For small enough contact area a large band gap is observed, predicting a strong influence on sub-Kelvin thermal transport.

1. Introduction

Arrays of monodisperse spherical particles have been used extensively to make photonic crystals for various applications. Such artificial opals, as they are commonly known, should also exhibit a phononic band gap depending on the particle diameter. The properties of three-dimensional phononic crystals have been studied extensively by various groups in the millimeter and micrometer scale at acoustic frequencies [1]. The behavior of thermal phonons, however, has not yet been properly studied in phononic crystals. We propose to study nano-scale phononic crystals at sub-kelvin temperatures to investigate their influence on thermal transport, in particular the effects of the appearance of full band gaps for thermal phonons. At this temperature range, the dominant thermal phonon wavelength is on the order of 100 nm, which is a typical diameter of readily available monodisperse nano-spheres. This would allow us to probe the thermal response of the crystal within the band gap region. Potential applications for such structures include thermal isolation for ultra-sensitive radiation detectors and phononic wave guides.

2. Fabrication of 3D Phononic Crystals

We fabricated three dimensional nano-sphere crystals using the vertical deposition technique [2,3]. Substrates were dipped into a solution of monodisperse polystyrene (PS) spheres (diameter 260 nm) obtained from Duke Scientific Inc. The concentration of the solution was varied from 10% to 0.02% and the dipping speed from 0.01 mm/min to 0.05 mm/min. All substrates were oxidized silicon chips with a thin film of titanium deposited by e-beam evaporation. Immediately following the metal evaporation, the chips were exposed to an oxygen atmosphere to form a TiO_x layer. This oxide layer increases the hydrophilicity of the substrates [4] and thus improves the surface wetting by the nano-sphere solution.

Characterization of the self-assembled arrays of nano-spheres was carried out to determine the effects of dipping speed and solution concentration on the size and quality of the crystal domains. Samples were imaged using scanning electron microscopy (SEM).

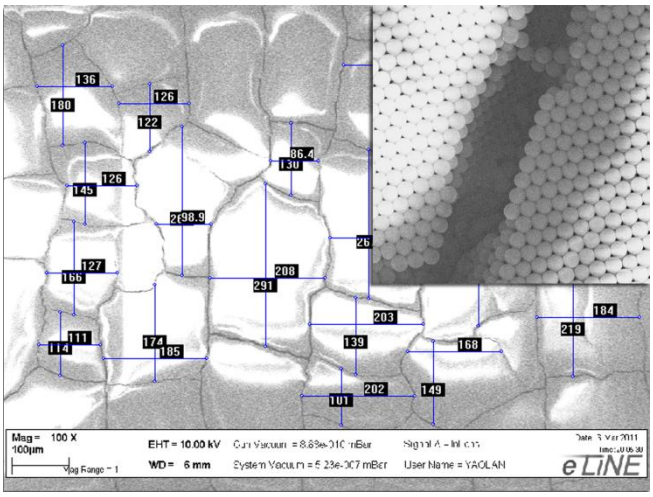

Figure 1. A typical SEM image used for domain size measurements, where the values shown are in µm. Vertical length is measured along the dipping direction. The inset shows a close-up of a large domain boundary with good hexagonal order in the domain region. Polystyrene spheres are 260 nm in diameter.

A typical SEM image for domain size measurements is shown in Figure 1. The measurement of the vertical (along the dipping direction) and horizontal lengths was done at several locations on each sample, and a large data set was collected in order to improve the statistics. The inset shows a closer view of the crystalline structure of the domains. Though the self-assembly process tends to favor the hexagonal close-packed (HCP), or face-centered cubic (FCC) lattices, we have found that the boundaries of sample chips, impurities on the substrate surface and other structures which constrain the self-assembly area can induce ordering into the body-centered cubic (BCC) or simple cubic (SC) structures.

We have found that larger domains are formed using lower dipping speeds and higher concentration solutions as seen before [3]. However, the size distribution is inversely proportional to the dipping speed. For example, while domains of up to 400 µm in vertical length were formed at 0.01 mm/min for 10 % solutions, the mean vertical length was approximately 130 µm. Both concentration and dipping speed were found to have a domain forming threshold value. At dipping speeds above 0.04 mm/min, no domains were formed even with the highest concentration. Similarly, the 0.02 and 0.2% concentrations formed no domains even at the lowest dipping speeds. Additionally, we found that elongation of the crystal domains occurs along the dipping direction and increases with dipping speed. An estimate of the number of layers deposited was obtained using a profilometer. As expected, we found that lower speeds and higher concentrations resulted in greater thickness. Using the 10% solution and 0.01 mm/min, we were able to obtain crystals with up to 30 layers. Investigating order in the thickness direction has so far been a challenge for the lack of a reliable method to image the side face without significantly changing the self-assembly conditions or altering the structure in post-processing. Ideally, thermal transport measurements of the structures should give information about the crystalline quality.

3. Finite Element Method Modelling of 3-D Phononic Crystals

Starting from the Wigner-Seitz unit cell, we developed computer models of the self-assembled nano-sphere arrays using the finite element method (FEM) with a commercial code (COMSOL). A Matlab script was used to generate dispersion relations for the geometry. The code was then verified by comparison with previous studies [5] done with the finite difference time domain (FDTD) method. Material properties (density, speeds of sound) for polystyrene were obtained from the Comsol material library for the sphere-vacuum geometries.

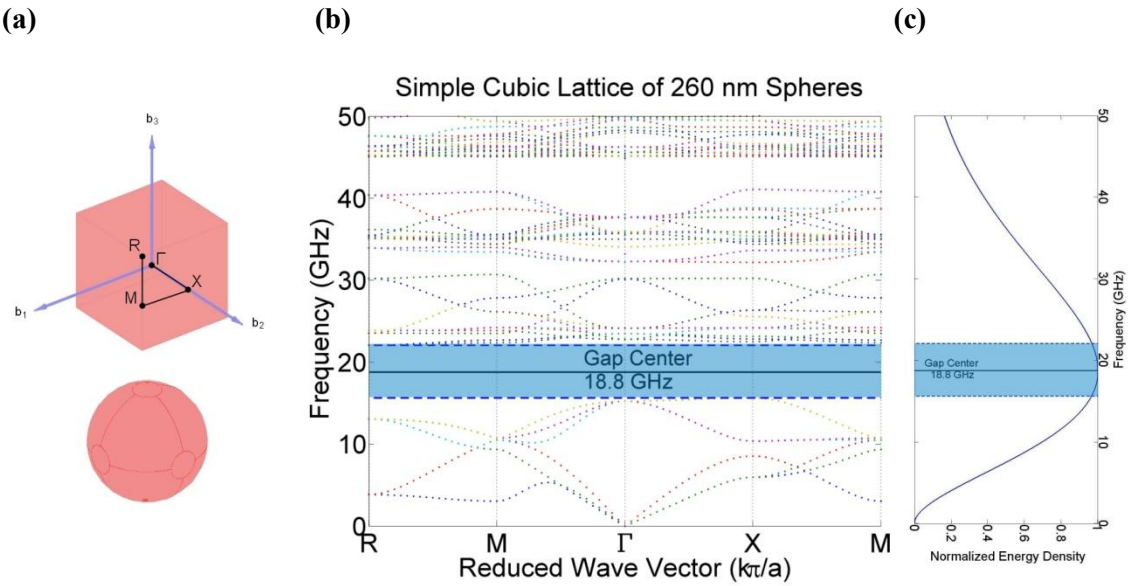

Figure 2. (a) First Brillouin zone for the SC lattice with high symmetry points indicated (Γ,X,M,R), along with the corresponding unit sphere geometry. **(b)** A representative dispersion relation calculation for a SC lattice of 260 nm spheres, exhibiting its lowest band gap (blue shaded region) centred at 18.8 GHz. **(c)** The peak of the Debye spectral energy density curve at 50 mK falls in the middle of this lowest band gap.

The first Brillouin zone and high symmetry points for the SC lattice are shown in Fig. 2(a) with the respective contacting sphere model geometry. Periodic boundary conditions satisfying Bloch's theorem are applied to the contact surfaces between neighboring spheres. The finite contact area avoids numerical problems with infinitesimal points, and also attempts to take into account the non-ideal contact between fabricated sphere arrays. A representative dispersion relation for the SC lattice with a sphere diameter 260 nm is shown in Figure 2(b). A full phononic band gap is clearly seen centered at 18.8 GHz. Also shown is the Debye spectral energy density, $g(\omega) = (3V\hbar/2\pi^2 v^3)\left[\omega^3\left(e^{\hbar\omega/k_B T} - 1\right)^{-1}\right]$ at 50 mK in Fig. 2(c). It can be seen that for the SC lattice of spheres with diameter 260 nm, the central gap frequency corresponds to the peak in the Debye spectrum peak at 50 mK. This illustrates that by choosing the nano-sphere diameter correctly, it is possible to tailor the band gap to coincide with the desired phonon energies and thus design structures to restrict thermal transport at chosen temperatures. Calculations for BCC and FCC lattices are also in progress.

During fabrication, soft spheres can undergo squeezing of varying degrees depending on the self-assembly conditions, while hard spheres are often sintered [6,7] to fix them in place. Such scenarios result in spheres whose contacts deviate significantly from points. To further investigate the phononic band gap behavior of these non-ideal crystals, we varied the contact area between the spheres. This was done by either increasing the size of the sphere while maintaining the bounding unit cell or by maintaining the sphere size while reducing the bounding cell. Both methods gave qualitatively identical dispersion relations. The general trend observed was that an increased contact area resulted in a reduction of the band gap. As the contact area increases, the structure approaches "bulk" geometry and the effects of periodicity are diminished. A comparison of phononic band structures with varying contact area size for the SC lattice is shown in Figure 3. We found that the gap closes rapidly; in fact, it is closed already at a contact

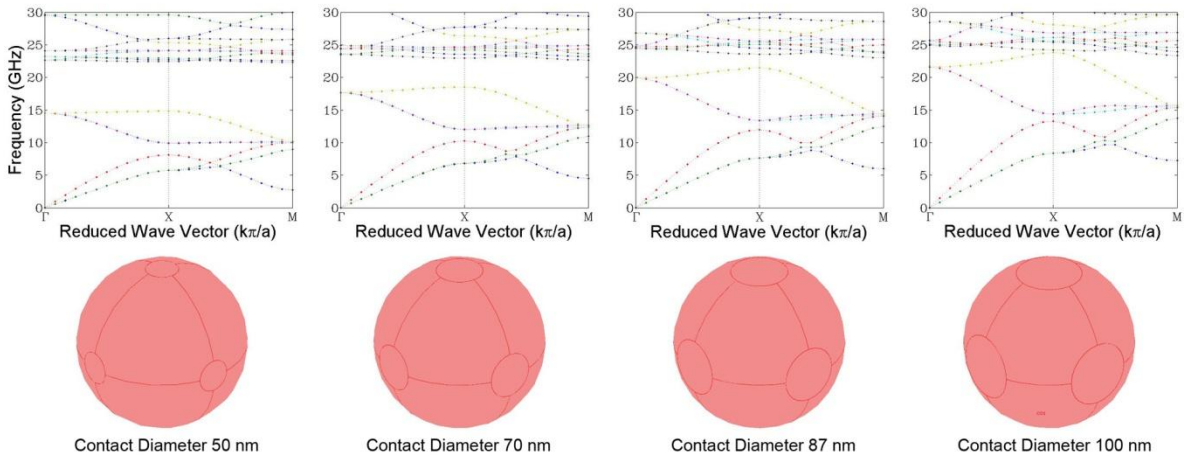

Figure 3. Comparison of dispersion relations in the Γ-X-M directions for varying contact area between spheres in a simple cubic lattice. Here contact diameter ranges from 50 to 100 nm. As the contact area increases, the band gap disappears. It can be seen that the band gap closes already when the diameter of the contact area is less than half of the sphere diameter.

diameter less than half of the sphere diameter. It seems that bulk-like dispersion behavior occurs long before the structure physically resembles a bulk.

4. Conclusions

We have characterized a vertical deposition technique for fabricating self-assembled arrays of nano-spheres. Slower dipping speeds and higher nano-sphere solution concentrations result in larger crystal domains. Using a dipping speed of 0.01 mm/min with a 10% concentration, we were able to produce crystal domains with average vertical lengths of approximately 130 μm. Elongation of the domains was found to follow dipping speed.

A computer model of a nanosphere array in the simple cubic lattice was constructed and dispersion relations calculated using FEM. The SC lattice of 260 nm polystyrene spheres has a gap of approximately 6.4 GHz centered at 18.8 GHz. This corresponds to the Debye energy density spectrum peak at 50 mK. By changing the contact area between spheres, we have seen how it affects the phononic behavior of such crystals. Increased contact area reduces the band gap as the periodic structure approaches the bulk geometry. These initial computational results are encouraging for the eventual measurement and detection of complete band gaps in three dimensional phononic crystals at sub-Kelvin temperatures.

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