Phononic properties of opals

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Abstract. We study the vibrational properties of opals consisting of silica spheres stacked periodically. To analyze their characteristic features (frequency gaps etc.) we start from the calculation of eigenfrequencies of a single sphere and see how they are modulated when spheres are combined to form a single layer and then multiple layers stacked periodically. The theoretical band structure is compared with the vibrational spectra measured experimentally.

1. Introduction

We present the theoretical and experimental studies of the elastic vibrations in the films assembled from closely packed silica spheres, known as synthetic opals. Opals possess properties typical for photonic crystals, e. g., photonic stop band, and are prospective materials for nanophotonics.[1] In the present work we introduce opals as a new class of submicron three-dimensional (3D) phononic crystals and calculate the band structure of lattice vibrations by assuming a perfect 3D periodicity of silica spheres and also the vibrational spectra of the silica films consisting of finite number of stacked layers. We compare our theoretical results with the experimental vibrational spectra obtained by the picosecond optical pump-probe technique.

Here we note that he vibrations of an isotropic, elastic sphere was solved by Lamb many years ago.[2] He found two kinds of vibrational modes that are decoupled from each other, i. e., the spheroidal and torsional modes. We are interested in the former mode whose eigenfrequencies are given from

\[
\left\{ \frac{2l(l-1)-\kappa_{T}^{2}}{\kappa_{L}^{2} j_{l}(\kappa_{L}) + 4\kappa_{L} j_{l+1}(\kappa_{L})} \right\} \left\{ \frac{2(l^2-1)-\kappa_{T}^{2}}{\kappa_{T}^{2} j_{l}(\kappa_{T}) + 2\kappa_{T} j_{l+1}(\kappa_{T})} \right\} -4(l+1)\left\{ \frac{2(l-1)j_{l}(\kappa_{L}) - \kappa_{L} j_{l+1}(\kappa_{L})}{\kappa_{T}} \right\} \left\{ \frac{j_{l}(\kappa_{T}) - \kappa_{T} j_{l+1}(\kappa_{T})}{\kappa_{L}} \right\} = 0, \tag{1}
\]

where \( l \) is the order of the spherical Bessel function \( j_{l} \), \( \kappa_{L} = \omega r_{0}/v_{L} \) and \( \kappa_{T} = \omega r_{0}/v_{T} \) with \( r_{0} \) the sphere radius, \( v_{L} = \sqrt{\lambda+2\mu}/\rho \) and \( v_{T} = \sqrt{\mu}/\rho \) the velocities of the longitudinal (L) and transverse (T) waves, respectively ( \( \lambda \) and \( \mu \) are the Lame coefficients, and \( \rho \) is mass density).

2. Numerical study of the vibrational spectra of opals

We assume a sphere composed of silica (SiO\textsubscript{2}) glass with diameter \( D = 2r_{0} = 357 \) nm. The relevant parameters are \( \rho = 2.204 \) g/cm\textsuperscript{3}, and \( v_{T} = 3.766 \times 10^{5} \) cm/s and \( v_{L} = 5.97 \times 10^{5} \) cm/s. The lowest several frequencies obtained from Eq. (1) are \( \nu_{l=0} = 12.77 \) GHz, \( \nu_{l=1} = 10.88 \)
Figure 1. (color online) Theoretical spectra for $u_z$ versus frequency. Black lines are for a single (free) silica sphere calculated by dividing $D = 2r_0$ into 120 bins (dashed line) and with twice finer meshes in one direction (solid line). Blue lines are for the connected two spheres with silica substrate (solid line) and without substrate (dashed line). Red lines are for an infinite linear chain of spheres on the substrate (solid line) and without substrate (dashed line). The vertical arrows indicate the eigenfrequencies of spheroidal modes (the first number is the order of the spherical Bessel function and the second one is the mode number).

GHz, $\nu_l=2 = 8.824$ GHz, $\nu_{l=3} = 13.02$ GHz. These discrete frequencies should be broadened and are changed into frequency bands in the structures where the spheres are stacked periodically both vertically and laterally and form a 3D phononic crystal. For the calculation of vibrational spectra in phononic crystals we employ the finite-difference-time-domain (FDTD) scheme. In this scheme we give a very short Gaussian force (almost a delta-function and hence very broad in frequency) at several points of a sphere. The direction is along the $z$ axis parallel to the stacking direction of the opal layers. Then we detect the displacement component $u_z$ at several points inside the spheres and they are Fourier transformed to see the frequency distribution.

2.1. Vibrations of a single and coupled spheres without and with a substrate

First we have calculated the vibrational spectra of a free silica sphere and a chain of spheres connected slightly. The spectra of a single sphere at a low-frequency region are shown in Fig. 1. In the FDTD scheme the cubic region of $2r_0 \times 2r_0 \times 2r_0$ (involving a single sphere) is subdivided into either $120 \times 120 \times 120$ or $240 \times 240 \times 240$ small grids. The eigenfrequencies obtained from Eq. (1) have also been indicated. The deviation of the peak frequencies from the exact spheroidal-mode-frequencies is less than 3.5 % for the finer grids but with the larger grids it becomes 6 % at largest. To save computation time we assume the grids of $120 \times 120 \times 120$ unless otherwise stated. The signals of torsional vibrations are very small for our excitation scheme except for the lowest one close to the lowest spheroidal mode frequency.

When two spheres are connected each other (the distance $2a$ between the centers of two spheres assumed is $2a = 2r_0/1.01$) the splittings of each spectral peaks are seen. Also a new signal due to collective motions is found at a low frequency region. When the number of spheres is increased in a line, each peak becomes much broader and spectral bands are produced. An interesting result is the effect of the substrate on the vibrational spectra. Figure 1 also shows the frequency spectra detected inside spheres when they are on the silica substrate. Each spectrum becomes broader and at the same time additional spectra in the low-frequency range are induced by the interaction with the extended vibrational modes in the substrate. These effects become more remarkable as the number of spheres connected increases.

2.2. Vibrations of opal films

In order to see how the frequency band is created as the number of the layers are increased in the 3D periodic opals, we next calculate the vibrational spectra in opal films consisting of few layers of spheres. In each layer spheres of radius $r_0$ (with $r_0/a = 1.01$) are arranged periodically, making a triangular lattice. These layers are stacked vertically so that they may eventually make a FCC lattice. The resulting spectra obtained for one layer of the spheres to those for ten layers
on the silica substrate are illustrated in Fig. 2. In obtaining these spectra the stress-free and absorbing boundary conditions are assumed at the top of the first layer and at the bottom of the spheres in contact with the substrate, respectively. We see that even in the system composed of three layers (consisting of a unit period of the FCC structure), the essential features of the band structure of the 3D opals are realized. More precisely, we can recognize the existence of frequency ranges where the vibration is absent. We shall see that they become frequency gaps for the perfect, periodic lattice (next subsection).

2.3. Vibrations of 3D periodic arrays of spheres
Now we consider perfect 3D opals composed of silica spheres stacked periodically in the [111] direction with a FCC structure. The unit cell assumed is shown together with the band structure in Fig. 3. The calculated dispersion curves are folded many times in the Brillouin zone. We find for \( r_0/a = 1.01 \) the existence of two complete gaps in the frequency range (0 to 16 GHz) displayed. These frequency gaps become smaller as \( r_0/a \) increases as illustrated in Fig. 3(c) because the phonon scattering at the joints of the spheres reduces and they vanish at \( r_0/a = 1.025 \). (Our numerical scheme is not reliable for \( r_0/a < 1.01 \) owing to the magnitudes of grids assumed.)

3. Opal samples and experimental details of the vibrational spectra
The samples used were synthesized by liquid epitaxy colloidal method with surface tension self-organization on a silica substrate. The opal films were formed from \( (350 \pm 10) \) nm amorphous silica spheres. Ten layers of closely packed spheres have been grown on an area of 1 cm\(^2\). The grown films possess the optical properties typical for 3D photonic crystals with a lowest stopband around 800 nm. The optical reflectivity spectrum of the films shows the Bragg peak with the peak reflectivity close to unity and the width 75 nm.

The elastic vibrations in opals were injected using experimental methods of picosecond acoustics.[3] The 100 nm Al film, deposited on the surface of the opal sample was excited by the intense pump pulses from a femtosecond 800 nm laser which induced the heating of the metal film. The film expanded rapidly due to the thermoelastic effect and the picosecond strain pulse was injected to the opal layer. The elastic vibrations of spheres in the opal sample were monitored by measuring the intensity of the probe beam reflected from surface of opal sample through the silica substrate. The probe beam was formed from the same laser. In such experimental scheme the elastic vibrations modulate the photonic band gap due to the change of the sphere size and silica refractive index. The wavelength of the probe beam coincides with the edge of photonic band gap in our samples which provides a high sensitivity to elastic vibrations. The time evolution of the signal obtained by means of scanning the delay line showed a number of oscillations in the Fourier spectrum shown in Fig. 3(c).

We see the existence of the spectral peaks at about 2 GHz, 6.5 GHz and 11 GHz. [The hatched region is the experimentally unreliable range.] Comparing with the band structure and
Figure 3. (color online) (a) Calculated band structure of opals (with $r_0 = 1.01a$). The thickness of the unit cell in the [111] direction is $L_z = 2\sqrt{6}a$. (b) The spectral intensity for $u_z$ in the opal film (with ten layers) on a silica substrate. (c) Experimental vibrational spectra. Hatch shows the frequency range where the experimental data is not reliable. Upper inset shows the unit cell structure consisting of three layers of spheres (the top, second and bottom layers are shown in green, red and black, respectively). The lower inset shows the width of the lowest and the second lowest frequency gaps in (a) versus $r_0/a$.

the calculated spectral peaks for the film consisting of ten layers, the lowest peak is found in the lowest band and the second peak is close to the upper edge of the first band. The third peak is much larger and broader and covers the whole frequency range of the second band.

4. Summary and discussions
An interesting effect on the vibrational spectra in phononic crystals is often induced by the presence of defects embedded in the otherwise perfect, periodic ones. Those defects can be a free surface, a substrate, vacancies, unequal sphere radius and the wrong stacking order of layers and so on. They are expected to produce, in general, localized vibrations with eigenfrequencies inside the band gaps of the host crystals. The effects of the free surface and substrate have already been included in the present calculation. However, no spectral peak associated with them is seen inside the lowest two gaps. We also found that a few vacancies in a unit cell do not destroy the original frequency gaps in the 3D phononic crystals we study.

To summarize, we have made theoretical and experimental studies of the elastic vibrations of the opal films. Opals possess properties typical for photonic crystals, e.g., photonic stop band, and are prospective materials for micro-nanophotonics. We expect that the combination of the properties for phonons and photons specific for 3D periodic structures opens a new way for manipulations with optical beams in optoelectronic devices. We have calculated with the FDTD method the vibrational band structure of opals by assuming a perfect 3D periodicity and also the vibrational spectra of the opal films consisting of finite number of layers. The existence of large band gaps and the the frequency spectra insensitive to the presence of defects make the opals promising for the future application to micro-nano acousto-optic devices.

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