Long-Living GHz Vibrations in Opal-Based Hypersonic Crystals

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We report experiments in which high-quality silica opal film is used as a three-dimensional hypersonic crystal in the GHz frequency range. We inject into the sample a broadband elastic wave packet by means of optical excitation of metal hypersound transducer and study the temporal evolution of GHz coherent vibrations monitoring the modulation of film reflectivity. Measurements carried out at different incident angles of probe beam allow us to determine that only coherent surface vibrations contribute to the measured modulation of reflectivity. This conclusion is supported by the long lifetime of surface vibrational modes with the frequency that matches a phononic band gap.

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I. INTRODUCTION

Phononic crystals are the structures with spatially periodic acoustic impedance, and they have obtained their name in analogy to photonic crystals, whose optical properties are governed by the periodicity of refractive index. Phononic crystals have attracted considerable interest due to their remarkable sonic properties and have been widely studied for the last ten years. The specific feature of phononic crystal is the appearance of band gaps in the phononic spectra. If the frequencies and wavevectors of the elastic wave correspond to the band gap, then this sound wave cannot propagate through the crystal and becomes localized due to destructive interference of acoustic waves scattered by the elastic periodicity. The existence of phononic band gap opens a way for manipulations with sound like sound filtering, wave guiding and focusing, which have been already demonstrated in the phononic crystals with the band gaps in the MHz (ultrasonic) or kHz ranges [1, 2].

Synthetic opals are artificial materials consisting of close-packed spheres of submicrometer diameter and are well known as three-dimensional (3D) photonic crystals [3]. Recently these artificial structures have also attracted great interest as phononic crystals [4–9]. The sub-micrometer spatial period of these materials in combination with 3D periodicity leads to the formation of complete phononic band gap in GHz and THz frequency range [4–6]. The phononic structures with such a high frequency band gap are called hypersonic crystals (HC), emphasizing the challenging step from ultrasonics to the elastic

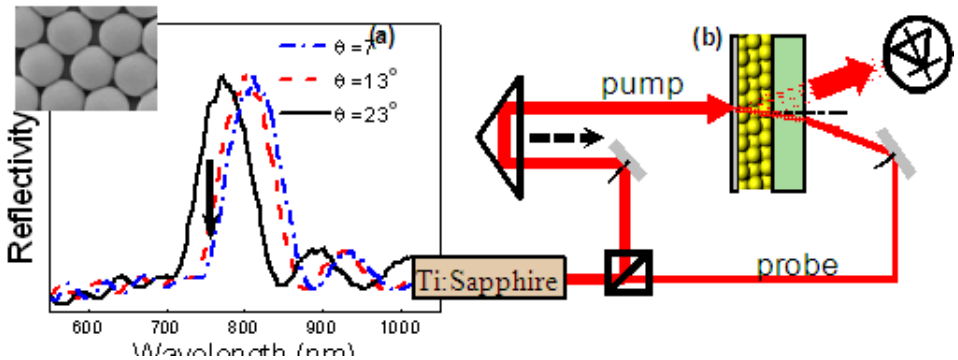

FIG. 1: (a) Reflectivity spectra of studied sample measured at different incident angles. Vertical arrow shows the wavelength of the probe laser. SEM image of top layer of that sample is presented in the inset. (b) Setup of the pump-probe experiments.

vibrations at the frequency limit for solids. It is predicted that the fabrication of HCs with a sufficiently large complete phononic band gap should open the way for realization of many attractive ideas [10]. However, in the GHz frequency range, only few studies have shown complete phononic band gaps [5, 6, 11].

Pioneering experiments on the direct optical detection of hypersonic coherent elastic vibrations in artificial opals have been published recently [4]. Later the anomalous long lifetime of vibrations with the frequency lying in the predicted complete phononic band gap of the studied structure have been reported in Ref. 5. One has been demonstrated in these studies that the interplay between bulk and surface acoustic waves is the very important factor, which determines the evolution of elastic modes with different frequencies. The localization of elastic vibrations at the surface or alternatively propagation to the bulk ultimately determines their lifetime.

The aim of the present work is to monitor the temporal evolution of GHz coherent acoustic vibrations excited in artificial opal films and measured at different experimental conditions. The signals measured at experiment are the temporal evolutions of reflectivity due to the modulation of photonic-crystalline properties of the studied film. We are focusing on the experimental evidence, which allows us to determine the nature of elastic vibrations contributing to the measured optical signals. The long lifetime of elastic modes with the frequency lying in the predicted phononic band confirms that the main contribution to the optical signal is provided by surface coherent vibrations.

II. STUDIED STRUCTURES

We study the coherent elastic vibrations in silica opal films consisting of 7 monolayers of close-packed in fcc lattice SiO_2 spheres with diameters $D \approx 350$ nm deposited on silica substrate. A scanning electron microscope (SEM) image of a studied film is shown in the inset of Fig. 1(a). Details of the film growth procedure can be found elsewhere [12]. It is

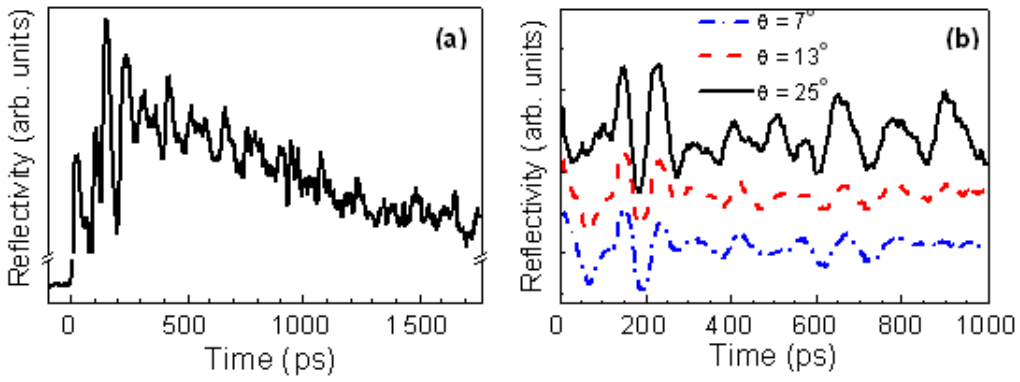

FIG. 2: (a) Temporal evolution of reflectivity induced by coherent elastic vibrations in the studied sample. Moment $t = 0$ corresponds to the pump excitation. (b) Temporal evolution of reflectivity after subtraction of "step-like" background for different incident angles of the probe beam.

seen that neighboring spheres are penetrating each other, which is a result of the sintering during the opal formation process. The sintering controls the elastic bonding between the spheres and therefore governs the spectral width of the phononic band gap [4]. The studied film is characterized by sintering coefficient $\chi = 4.6 \times 10^{-3}$ [13]. The theoretically predicted phononic band gap for the studied structure has the central frequency $\nu = 7.5$ GHz and spectral width of 2 GHz [4].

The typical reflectivity spectra of the studied opal film measured at room temperature by means of halogen lamp as a light source and spectrometer with a CCD camera are shown in Fig. 1(a). The Bragg reflectivity peaks with a maximum at $\lambda = 810$, 800 and 770 nm for the light incident angle $\theta = 7$, 13 and 25°, respectively, arise due to the photonic stop band of the opal film. The maximum reflectivity spectral position is given by $\lambda_m = 2d_0\sqrt{\varepsilon_0 - \sin^2\theta}$, where $\varepsilon_0 = 1.75$ is the mean permittivity of the studied opal structure, and $d_0 = 375$ nm is the spacing between adjacent (111) lattice planes. The measured spectra [Fig. 1(a)] show oscillations due to Fabry-Perot interferences in the opal film. The oscillation periods correspond to film thicknesses of 7 silica sphere layers.

III. TEMPORAL EVOLUTION OF REFLECTIVITY

The idea of the experiment [Fig. 1(b)] is to inject into the sample a picosecond coherent elastic wave packet from a hypersonic transducer and make time-resolved measurements of the corresponding changes in the Bragg reflectivity spectrum. This experimental technique was developed earlier and used for studying of the coherent vibrations in opal films and colloidal crystals [4, 5, 7–9, 14]. An aluminum film with a thickness of 100 nm was deposited on the free surface of the opal sample. This metal film played the role of a hyper-

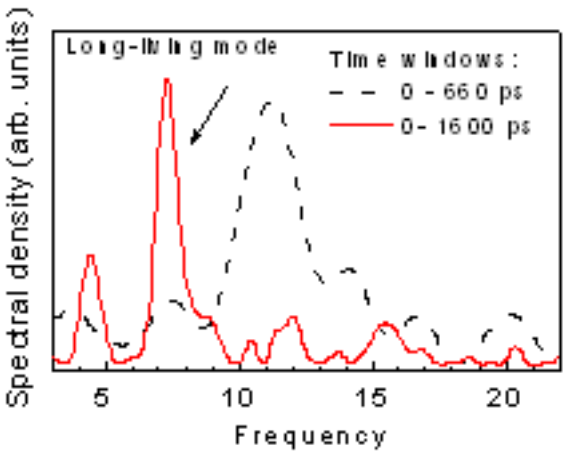

FIG. 3: Power spectral density obtained by FFT of the measured reflectivity $\Delta R(t)$ as a function of vibration frequency. The solid and dashed curves represent the spectra obtained for long ($\Delta t = 1600$ ps) and short ($\Delta t = 660$ ps) time windows, respectively.

sonic transducer for the generation of a picosecond strain pulse, which is injected into the opal film [15, 16]. For the strain pulse excitation, 300-fs pump pulses from a Ti:sapphire laser with a regenerative amplifier ($\lambda = 800$ nm, repetition rate 100 kHz, maximum energy per pulse 1 μJ) were used. The pump beam passed through a variable delay line and focused on the metal transducer and the diameter of the focus spot was 200 μm. The probe pulse was split from the same laser beam and focused from the silica substrate side at incidence angle θ exactly opposite to the pump spot. Probe spot diameter was order of 20 μm. The specularly reflected probe beam was detected by a photodiode. To increase the signal-to-noise ratio, the pump beam was modulated by a mechanical chopper with a frequency of 2 kHz.

Figure 2(a) shows the reflectivity changes $\Delta R(t)$ measured at $\theta = 23°$. After a sharp rise at $t = 0$ ps, pronounced oscillations are observed in $\Delta R(t)$. The oscillatory part of the measured signal cannot be described by a single period and depends on the sample. Figure 2(b) shows the temporal signals $\Delta R(t)$ measured at several incident angles θ. It is seen that the temporal evolution of the signal for various values of θ is almost identical: the maxima and minima in the oscillating signals take place at the same time at $t < 1000$ ps. At longer times, the comparison is not relevant due to higher noise levels for $\theta = 7°$ and 13°.

Figure 3 shows the power spectra obtained by fast Fourier transformation (FFT) of the measured temporal reflectivity signal. The solid line shows the spectrum obtained by

FFT over a wide time window $t = 0$–1600 ps. The spectrum shows a well-isolated peak centered at $\nu = 7.5$ GHz with a FFT limited spectral width of ~ 1 GHz. The dashed curve in Fig. 3 shows the FFTs of $\Delta R(t)$ over a shorter time window $t = 0$–600 ps. This spectrum demonstrates the high amplitude peak at frequency of 11 GHz only.

IV. DISCUSSION

Here we discuss the nature of the signal detected by the optical probe pulse. Obviously the detected signal is determined by the mechanisms of the interaction of light with coherent elastic vibrations in a film. It is well known that in *homogeneous media* (without periodic structure), the probe signal in the setup similar to the one shown in Fig. 1(b) is governed by the elasto-optical effect. In thin films, the signal includes two contributions [17]: (*i*) reflectivity changes governed by the modulation of the thickness of the film due to the interference of beams reflected from the top and bottom surfaces; and (*ii*) dynamical interference of the optical reflected beam on the propagating acoustic wave in the bulk of the film, also known as coherent Brillouin scattering phenomenon [18]. Thus, contribution (*i*) gives a signal which is caused only by the displacement of the surface in the direction perpendicular to the plane of the film, while contribution (*ii*) is sensitive only to the acoustic waves propagating in the bulk. The essential difference between (*i*) and (*ii*) is that (*i*) gives the real time evolution of the vibrations at the surface, while (*ii*) is governed by the energy and momentum conservation for light and sound and thus reflects only one spectral component $\nu = 2n \cos\theta/\lambda$ (here n — refractive index, θ — sound velocity, λ — wavelength of probe light) out of a broad spectrum of generated acoustic frequencies. Thus, in case of contribution (*i*), changing the incident angle θ should affect only the sensitivity of probing without changing the temporal evolution of the detected signal. Contrary, when contribution (*ii*) is dominant, changing θ obviously changes the active phonon wavevector and correspondingly the frequency of the detected acoustic waves.

In periodic structures like opals, the mechanisms, which govern the probing mechanisms, are the same. Indeed contribution (*i*) obviously is identical to the case of a homogenous film. The Brillouin scattering (contribution (*ii*)) is more sophisticated and depends on whether the elastic modes are localized or propagating [6]. For propagating modes, we can still expect the dependence of temporal evolution of $\Delta R(t)$ on θ. As shown in Figure 2(b), the temporal signals $\Delta R(t)$ measured in the sample for several incident angles θ are almost identical. The independence of $\Delta R(t)$ on θ lead us to the assumption that the coherent part of $\Delta R(t)$ is governed mostly by vibrations localized at the surface of the opal film (left side of the crystal in Fig. 1(b)), where they are initially generated by the pump laser pulse. Actually, this is not surprising because it is reasonable to expect that the coherence of the bulk hypersonic waves is lost already at the second layer of the opal film due to inhomogeneities of the elastic bonds between the sintered spheres.

The main contribution of surface waves to the detected signals is also supported by the spectra obtained by FFT (Fig. 3), which allow us to estimate the lifetime of elastic modes with difference frequencies. The Fourier spectrum calculated for the short time range $t = 0$–

600 ps shows that at early times, the mode with a frequency of 11 GHz dominates in $\Delta R(t)$. However, the spectra obtained for $t = 0$–1600 ps show relatively small contribution of these modes to the detected signal. On contrary, the mode with $\nu = 7.5$ GHz is well pronounced in the long time interval. The complete phononic band gap predicted theoretically for our structures [4] has the center frequency at $\nu \approx 7.5$ GHz and the spectral width of 2 GHz. The experimentally observed long-living mode at 7.5 GHz exactly matches the center frequency of the predicted band gap and therefore cannot propagate in the crystal [4, 5]. Thus, mode with this frequency cannot be detected in $\Delta R(t)$ if it is contributed by bulk modes. However, we see that being localized at the surface, the mode with frequency corresponding to the center of phononic band gap gives the contribution to the detected signal in the long time interval. Elastic modes with $\nu = 11$ GHz, which are excited in the same surface layer, do not match with the phononic band gap, and therefore, may effectively propagate to the bulk of the crystal, but their contribution to $\Delta R(t)$ decays fast. Therefore high-frequency components provide the contribution to $\Delta R(t)$ only at early times before their escape to the bulk. This observation confirms that in the detected signal the contribution of coherent surface vibrations is dominant.

V. CONCLUSIONS

We have carried out comprehensive studies on generation and propagation of GHz coherent elastic vibrations in opal-based 3D HC. We have shown that long-living (up to 1 ns) coherent GHz surface modes may be effectively generated in these structures and provide main contribution to the modulation of opal film reflectivity. The possibility to generate long-living coherent waves of such a high frequency is extremely important for the future possible applications of 3D HC. In combination with the control of the interaction between surface and bulk waves, integrated hypersonic devices combining the remarkable features of 2D phononic crystals [19] with a 3D band gap for bulk acoustic waves could be obtained. Further possibilities to exploit the observed long-living surface modes in real opal-like structures depends on whether these modes propagate in the surface plane possessing the features of 2D HCs or whether they are strongly localized due to the disorder. If the surface modes propagate without scattering on a distance essentially higher than the period of structure, then hypersonic waveguiding on the surface will be one of the most attractive tasks for further experimental studies.

Acknowledgments

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