Optoacoustic characterization of synthetic opals

C Mecheri¹, P Ruello¹, D Mounier¹, J M Breteau¹, I Povey², M Pemble², S G Romanov² and V Gusev¹

¹Laboratoire de Physique de l’État Condensé, UMR CNRS 6087, Université du Maine, Avenue Olivier Messiaen, 72085 France.
²Tyndall National Institute, University College Cork, Prospect Row, Cork, Ireland.

E-mail: charfeddine.mechri.etu@univ-lemans.fr

Abstract. The development of laser-based ultrafast acoustic techniques allowed the investigation of the vibrations of nanostructures. In this communication, we report the results of the characterization of crystals composed of silica nanospheres (opals) by pump-probe setup. In our study, the excitation and detection of the vibrations of nanospheres by femtosecond laser pulses was facilitated by filling the pores in the opals by GaAs semiconducting material.

1. Introduction
Recently the vibrational eigenmodes of optically transparent nanospheres in synthetic opals has been investigated by Brillouin light scattering [1,2]. There are also multiple reports on fs optical pump probe investigation of vibrations of light absorbing nanoparticles [3,4]. In these experiments the vibrations are stimulated by optical excitation. Our experiments presented below demonstrate that generation and detection of vibrational eigenmodes of spheres in opals by femtosecond pump-probe reflectivity technique could be facilitated by filling the pores in opals by a light absorbing material.

2. Photonic Properties and Acoustic Modes in synthetics opals
Synthetic opals are spatially periodic structures self-assembled from silica nanospheres. These materials have proven to be useful because of their possible applications in photonics, phononics and optoelectronics. Most of the expected applications could be based on the ability of the structure to filter and localize acoustic and/or electromagnetic waves. Opals from few hundreds of nanometers spheres were prepared with a GaAs filling by Chemical Vapor Deposition [2] as sketched on Fig. 1

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2.1. Optical Study
First, an optical study of the GaAs-filled SiO$_2$ photonic crystals (PCs) was carried out in order to reveal the photonic band gaps. In Fig. 2, we report the results of these experiments which consist in focusing a non collimated continuous white light on the sample and measuring the spectrum of the reflected beam. Two Bragg peaks attributed to reflection onto (111) and (222) planes are observed with widths of about 150 nm and 100 nm, respectively. For the estimations we have considered in the first approximation that the effective refractive index of opal is a combination of indices of silica and of gallium arsenide. Considering the (fcc) close-packing lattice of spheres we have

$$n_{eff}^2 = 0.26n_{GaAs}^2 + 0.74n_{SiO2}^2 \approx 2.26 \quad (n_{SiO2} \approx 1.45 \text{ and } n_{GaAs} \approx 3.7 \text{ in infrared and } 4.6 \text{ in green were chosen}).$$

Using the well known Bragg formula, we found for the incident angle of $\theta = 0$ and for (111) and (222) planes the resonance wavelength $\lambda_{hkl} = 2n_{eff}d_{hkl}$ where $d_{hkl} = \sqrt{2D((h^2 + k^2 + l^2))^{0.5}}$ is the interplane distance and $D=250$ nm is the sphere diameter, applies, providing $\lambda_{111} = 922$ nm and $\lambda_{222} = 461$ nm. The “red” shift of the (222) band compared to the Bragg estimated occurs due to contribution to the effective index of refraction from the imaginary part of the $n_{GaAs}$ at the wavelengths shorter than the electron absorption edge of GaAs.

These physical parameters of opals can be precisely estimated thanks to a fitting procedure of the resonance wavelength $\lambda_c$ dependence on the angle of incidence. A typical mean least square fitting is given in Fig. 3 for the (111) Bragg reflection assuming the following law [2]:

$$\lambda_c^2 = 2d_{hkl}\sqrt{n_{eff}^2 - \sin^2(\theta)} \Leftrightarrow \lambda_c^2 = [2D^2n_{eff}^2 \quad ((h^2 + k^2 + l^2)) - [2D^2 \quad ((h^2 + k^2 + l^2))]\sin^2(\theta)$$

Where (h, k, l) are the Miller indexes. Figure 3 shows the linear correlation between $\lambda_c^2$ and $\sin^2(\theta)$. The slope provides a precise estimation of D while the zero intercept provides $n_{eff}$. The fit gives a
The diameter of D=253 nm (± 3%) and an effective refractive index of 2.24, which leads to an optical index of the filling material of 3.47, which are pretty reasonable values.

2.2. Picosecond study
The photo-induced dynamics was studied by traditional pump-probe fs time-resolved reflectivity. The sample was excited by a 150 fs pulse of a Ti-Sapphire. The beam was focused onto a 30μm diameter spot with energy of 0.03 mJ/cm², which allows operating in a linear regime. Different experiments have been conducted with a pump beam wavelength of 730 nm, 760 nm, 780 nm, 800 nm and 840 nm (Figure 4). It appears that good oscillations are observed with wavelength excitation of 730 nm and 760 nm. The larger magnitude is observed for 760 nm. On the other hand, for radiation of 780 nm, 800 nm and 840nm no oscillations are detected (Figure 5). Following our knowledge of the photonic bandgap properties described above, it appears that the 780–840 nm pump radiation is within the bandwidth covered by the dispersive (111) bandgap, if its angle dependence is taken into account (Fig.2b). The radiation in this range is stronger reflected compared that of the 760nm radiation. In others words, the optoacoustic conversion becomes more efficient out-of the forbidden band since light penetrates deeper and can consequently interact with larger volume of absorbing interstitial GaAs material. However, the wavelength dependence of the detection process also influences the magnitude of the detected oscillations but no definite optimisation of detection process has been established yet. The transient reflectivity diagram of Figure 4 shows oscillations of 8.7 GHz (115 ps). These oscillations are long-living oscillations coherently with recent observations in Au-coated silica sphere opals crystal. That characteristic frequency is close to the frequency of the first spherical mode of a sphere vibration. A numerical analysis for the solving of the following eigenvalue equation of the spherical mode gives indeed f_{10} \approx 9.7 GHz, if we consider the diameter of the sphere found in the previous section, i.e. D=260 nm.

\[\frac{2j_{L+1}(x)}{\alpha x} \left[1 + \frac{(L-1)(L+2)}{\alpha x} \left(\frac{j_{L+1}(\alpha x)}{j_L(\alpha x)} - \frac{L+1}{\alpha x}\right)\right] + j_L(x)^2 \left[-\frac{1}{2} \left(\frac{(L-1)(2L+1)}{(\alpha x)^2}\right) + \frac{1}{\alpha x} \left(1 - \frac{2L(L-1)(L+2)}{(\alpha x)^2}\right) \frac{j_{L+1}(\alpha x)}{j_L(\alpha x)}\right] = 0\]

where \(j_L\) is the Bessel spherical function, \(\alpha = V_L/V_T\), \(V_L\) (5.05 Km/s) and \(V_T\) (2.1 Km/s) are the transversal and longitudinal sound velocities respectively and the eigenvalue \(f = V_L x / \pi D\).
Figure 4. Transient reflectivity of opal/GaAs structure

Figure 5. No oscillation observed at 840nm (inside the forbidden band)

3. Conclusion

Femtosecond optical pump-probe technique has been applied for the investigation of acoustic vibrations of synthetic opals filled by GaAs semiconductor. The observed oscillation has been identified with the first breathing mode of the individual silica spheres composing the opal skeleton. The preliminary results demonstrate that the sensitivity of the technique to the generation and detection of the vibrational eigenmodes importantly depends on the choice of the photon energies of the pump and probe laser pulses relative to the photonic bandgap of the opal and the electron energy gap of the semiconductor.

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