10 fs ultrafast all-optical switching in polystyrene nonlinear photonic crystals

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An ultrafast all-optical switching with the response time on the order of 10 fs is demonstrated in a three-dimensional opal polystyrene nonlinear photonic crystal by means of precise femtosecond pump-probe technique. The switching is realized by the shift in the photonic band gap under external optical pumping of 8 fs Ti:sapphire pulse laser with a peak pump power of 20.6 GW/cm². The good performance of optical switching is attributed to the very strong and fast Kerr nonlinear optical response of the polystyrene material. © 2009 American Institute of Physics.


All-optical switching has received much attention for a long time due to its potential applications in integrated optics and ultrahigh-speed information processing. Photonic crystal all-optical switching is a promising scheme to realize switching with ultrafast response time.1–6 The refractive index contrast of photonic crystal made from Kerr nonlinear materials can change under optical pump of high-intensity light. The subsequent rapid shift in band gap or defect mode can be exploited as optical switching. Organic polymer materials such as polystyrene has a large third-order nonlinear susceptibility which is on the order of $10^{-12}$ cm²/W and an extremely fast response time down to several femtosecond as the third-order nonlinearity originates from the delocalization of the π-conjugated electrons along the polymer chains.7–10 In contrast, semiconductor materials have relatively small nonlinearity (on the order of $10^{-14}$ cm²/W) and slow response time due to the slow relaxation of carrier created by two-photon absorption, which is on the order of nanosecond in bulk silicon5 and 100 picosecond in bulk AlGaAs.11,12 However, the carrier lifetime can be shortened with the microstructures due to the increased surface area and strong nonradiative recombination of the carriers, leading to a response time of few tens of picoseconds in silicon photonic crystal,5,13,14 15 picoseconds in GaAs photonic crystal15 and several picoseconds in AlGaAs photonic crystal.12 Polymer-metal nanoparticle composite artificial nonlinear materials have their response time limited to subpicosecond16 although they can have large nonlinearity. In our previous works that demonstrated 120 (Ref. 17) and 20 fs10 response time of optical switching by means of 120 and 25 fs pump pulse, respectively, we have found that when the duration of pump pulse becomes shorter, faster response time of all-optical switching will be obtained. In this work we demonstrate that the response time of optical switching can be further shortened to an extremely fast level of 10 fs in three-dimensional polystyrene photonic crystal by implementing 8 fs pump pulse.

The principle of this ultrafast all-optical switching is very simple. Under strong pump pulse, the refractive index of polystyrene changes dynamically, which result in the shift in photonic band edge. When the wavelength of probe light is located at the band edge, the transmittivity will be changed accordingly, which will realize the “open” and “close” state of all-optical switching. In this experiment, the three-dimensional photonic crystal is made from monodispersed polystyrene spheres with a diameter of 350 nm (3350A, Duke Scientific Corporation, USA) on a substrate of glass slide. It was prepared by the pressure controlled isothermal heating vertical deposition method18 with the concentration of 1 wt%, heating temperature of 39.5 °C, and pressure of 12.0 ± 0.2 kPa. The scanning electron microscopy (SEM) picture of the synthesized polystyrene opal photonic crystal sample is displayed in Fig. 1(a). The sample has a face-centered cubic structure and its (111) plane is parallel to the surface of the substrate, namely, the Γ–L crystalline direction is perpendicular to the surface. Although the opal photonic crystal does not have a complete band gap due to low index contrast, it suffices to only consider a directional band gap along Γ–L direction for current purpose. The measured linear transmission spectrum along the Γ–L direction is shown in Fig. 1(b). The central wavelength of the band gap is located at 800 nm which agrees well with the theoretical result simulated by finite-difference time-domain (FDTD) method. The short-wavelength band gap edge appears steeper than the long-wavelength band gap edge, so the probe beam in this experiment was selected at the wavelength of 785 nm which is located at the short-wavelength band gap edge.

We characterize the performance of all-optical switching by means of the standard femtosecond pump-probe technique. The experiment setup is schematically shown in Fig. 2(a). The ultrafast laser output from a homemade chirped-mirrors Ti:sapphire laser (Horizon-10, CAS, Beijing) with

![FIG. 1. (a) SEM image and (b) measured transmission spectrum of the polystyrene opal sample with the sphere diameter of 350 nm.](image-url)
the duration of several femtosecond and the repetition rate of 80 MHz is adopted in our experiment. The experimental Fourier transform spectrum and the interferometric autocorrelation signal of the pulse laser, which shows a pulse duration of about 8 fs.

In the experimental process, we should first adjust the synchronization of the two ultrashort pulses at the position of the sample. As both the duration of pump and probe beam are only several femtosecond, the synchronization will be greatly degraded and even disappear when the deflection of the optical path difference by the piezoelectric controller, the time-delay curve for the wavelength of 785 nm, which is the dependence of the transmittance change in the probe light signal intensity on the time delay between the pump and probe light pulses, is obtained. The result is depicted in Fig. 3.

In Fig. 3(a), the dots are the experimental data, and the solid lines are the exponential fitting curves for rising and dropping process. Due to the positive third-order susceptibility of polystyrene, the band gap shifts to long-wavelength under strong pump pulse, and the transmittance at the wavelength of 785 nm, which is located at the short-wavelength band gap edge, increases at first, and then decreases when the pump pulse passes through. The full width at half maximum of the time-delay curve is around 11 fs, with the rise time of 3 fs and the drop time of 8 fs. The rise time of optical switching is almost the same with the half duration of pump pulse, while the drop time is much slower than the rise time. The asymmetric feature can be understood as follows. The spatial length of pulse with 10 fs is only 3 μm, and the sample length is about 15 μm. At early stage, the photonic crystal will respond to the pump pulse almost instantaneously, leading to rapid rise time. Due to the multiple scattering of photonic crystal, the ultrashort pulse will last longer within the photonic crystal and result in a longer drop time. On the other hand, the signal contrast of this optical switching is only 25% (from 25% to 50%) under the excitation of 20.6 GW/cm² peak pump power. Possible reasons for this relatively low contrast include not steep enough band gap edge of opal sample and large noise in the ultrafast pump-probe system, such as the instability of the ultrashort pulse laser, the dispersion of the optical elements, and the relatively large electronic noise by the PMT and lock-in amplifier.

To have a clarified understanding of the experimental result, we do numerical simulations on the optical switching problem by means of the nonlinear FDTD method. An ultrashort pulse with a Gaussian temporal profile and 8 fs duration is used in the computer pump-probe experiment. In our FDTD simulation, we consider instantaneous Kerr nonlinearity as the material response time is still much shorter than the pump pulse duration. We also neglect the material dispersion due to the weak dispersion of polystyrene in the wavelength range from 600 to 900 nm. The result is displayed in Fig. 3(b). The theoretical time-delay curve exhibits the same major features as the experimental data. It also has a faster rise time and a slower drop time, and the overall response time is also about 10 fs. The theoretical value of switching contrast is slightly higher with 40% (from 25% to

FIG. 2. (Color online) (a) Experimental setup for characterization of all-optical switching in three-dimensional polystyrene photonic crystals by means of femtosecond pump-probe technique. (b) Fourier transform spectrum and (c) corresponding interferometric autocorrelation signal of the pump pulse laser, which shows a pulse duration of about 8 fs.

FIG. 3. (Color online) (a) Measured and (b) simulated transmittance change with respect to the time delay between pump and probe light. The wavelength of the probe light is 785 nm and the peak pump power is 20.6 GW/cm².
65%). Both the experimental and theoretical delay curve show an oscillation at the delay time of \( t=20 \text{ fs} \), and this might be induced by the multiple scattering effect of pump pulse within the photonic crystal as mentioned above. The weak oscillation appearing before \( t=-15 \text{ fs} \) in the experimental data [see Fig. 3(a)] is not found in theory [see Fig. 3(b)], and it might be an experimental artifact induced by inaccuracy in measurement.

In our experiment, the average power of pump pulse is about 250 mW, the repetition rate is 80 MHz with the single pulse energy of 3.125 nJ, and the radius of focus point for pump laser is around 30 \( \mu \text{m} \), which gives the peak pump power of 20.6 GW/cm\(^2\). Although the peak intensity of pump pulse is quite high, the total energy that is involved in the laser pulse is in a relatively low level due to its extremely short duration. The pump power is comparable to the value used to trigger optical switching in a silicon photonic crystal with a 3-dimensional polystyrene nonlinear photonic crystal under optical pump of 8 fs laser pulse. This switching response time has been closing to the material response limit of polystyrene. The pump energy is in a practically accessible low level. A 10 fs optical switching can find many interesting applications in ultrafast laser technology and fundamental research on ultrafast physics and chemistry problems.

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