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HTML ABSTRACT + LINKS

Subpicosecond shifting of the photonic band gap in a three-dimensional photonic crystal

Dmitry A. Mazurenko,[a] Robert Kerst, and Jaap I. Dijkhuis
Atom Optics and Ultrafast Dynamics, Department of Physics and Astronomy and Debye Institute, University of Utrecht, P. O. Box 80000, 3508 TA Utrecht, The Netherlands

Andrey V. Akimov, Valery G. Golubev, Alexander A. Kaplyanskii, Dmitry A. Kurdyukov, and Alexander B. Pevtsov
Ioffe Physico-Technical Institute, Russian Academy of Sciences, Politechnicheskaya 26, St. Petersburg, 194021, Russia

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We demonstrate spectral shifting of the photonic band gap in a three-dimensional photonic crystal within a time of less than 350 fs. Single 120 fs high-power optical pulses are capable to induce the transition from the semiconductor to the metallic phase of VO_2 in the pores of our artificial silica opal. The phase transition produces a substantial decrease of the real part of the effective refractive index of the photonic crystal and shifts the spectral position of the photonic band gap. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1856687]

The main goal of photonics is to design optical devices working similarly but without the need of electrical contacts and much faster than traditional electronics. Photonic crystals, materials with a periodically modulated refractive index on the scale of the optical wavelength, are promising objects for designing integrated optical circuits. In a photonic crystal a forbidden gap for photons may arise, implying that light with a wavelength close to the period of modulation cannot propagate in certain directions.[1] The spectral range which corresponds to nonpropagating photons is called a photonic bandgap (PBG). In case of sufficient dielectric contrast, a complete PBG may be realized in three-dimensional (3D) photonic crystals where light in a range of wavelengths cannot propagate in *any* direction.[2–4] The spectral position and width of such a PBG depend on the complex refractive index, which can be modified by some external impact. To date, numerous schemes have been tested experimentally that allow for external control over the PBG spectral position in 3D photonic crystals on a time scale up to nanoseconds, like applying an external electric field,[5–9] optical excitation,[10] and temperature variation.[11–15] In practice, optical excitation is the fastest way to control the PBG properties. Recently, ultrafast switching of one-,[16] two-,[17,18] and three-dimensional[19,20] photonic crystals via photoexcited free-carriers was demonstrated on the femtosecond time scale. The fastest switching, as short as <30 fs, was reported by us in a 3D-photonic crystal based on an opal-Si composite.[20] Unfortunately, a large increase of the imaginary part of the refractive index occurred in these samples that masked the effect of the PBG shifting. Thus, demonstration of optical *shifting* of the PBG spectral position on the femtosecond time scale remains a challenge in 3D photonic crystals.[21,22]

In this letter, we demonstrate femtosecond shifting of the PBG in a synthetic opal filled with VO_2. Our sample was fabricated from an opal template composed of 0.24-μm-diam mono-dispersed (±5%) silica spheres. The voids of the opal were impregnated with VO_2 with filling factor 8%. The details of the fabrication method and the linear optical properties can be found in our recent publication.[13] We will take advantage of the structural phase transition[23] in VO_2 at $T_c=67$ °C that can take place on the subpicosecond time scale.[24] In the spectral region of red light, the transition is accompanied by changes in the real part of the refractive index[25,26] from $n(VO_2)\approx 2.9$ in the "cold" semiconductor phase to $n(VO_2)\approx 2.3$ in the "hot" metallic phase.

PBGs originate from multiple Bragg diffraction on the periodic planes of the photonic crystal lattice,[1] and produce a band of high reflectivity. Generally, the spectral position, λ,

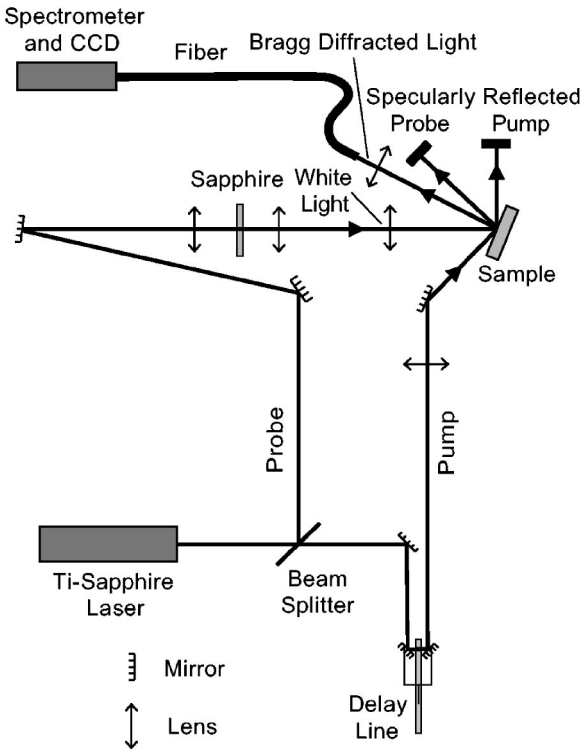

FIG. 1. Experimental setup.

[a]Present address: Solid State Physics Laboratory, University of Groningen, Nijenborg 4, 9747 AG Groningen, The Netherlands. Electronic mail: D.A.Mazurenko@rug.nl

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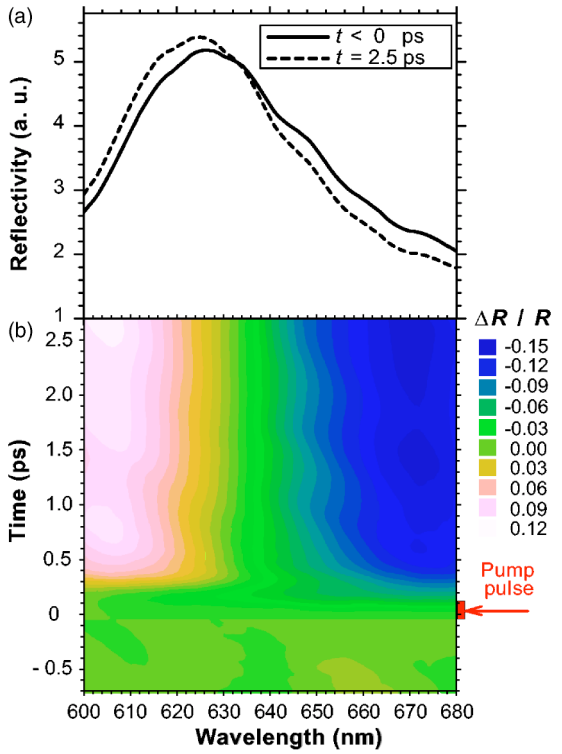

FIG. 2. (Color online) (a) Bragg reflection spectra of opal-VO_2 before (solid line) and after (dashed line) the optical excitation; (b) Spectral-temporal evolution of the relative changes of the Bragg reflectivity. Color scale indicates the signal amplitude. Red rectangle on the right indicates the arrival time and duration of the pump pulse.

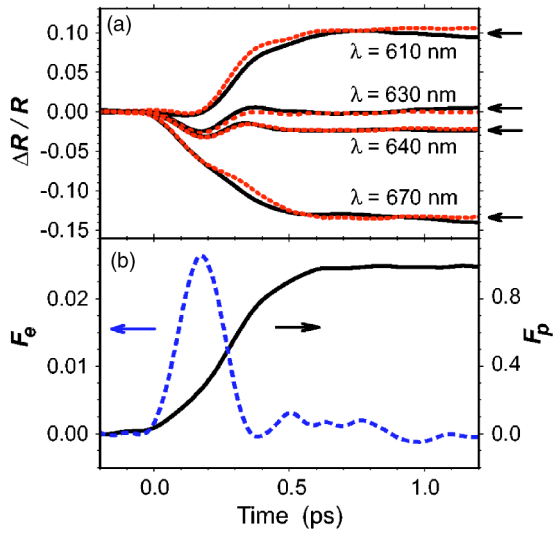

FIG. 3. (Color online) (a) Transient Bragg reflection signal at 610, 630, 640, and 670 nm. Solid lines show the experimental data, dotted lines the calculated $F_e(t)+A(\lambda)F_p(t)$; (b) $F_e(t)$ and $F_p(t)$ depicted by dashed and sold lines, respectively.

of the maximum of this band is close to the value corresponding to the Bragg condition computed for the effective refractive index $\overline{n}$. As soon as the temperature reaches T_c, the value of $n(VO_2)$, and thus that of $\overline{n}$, decreases, modifying the Bragg condition. As a result, the PBG shifts toward short wavelengths. In stationary[13] and nanosecond pulsed[27] experiments the shift of the PBG in opal-VO_2 composites has already been clearly demonstrated.

The scheme of our experimental setup is presented in Fig. 1. All experiments were carried out at room temperature. The opal-VO_2 photonic crystal was excited by an 800 nm 120 fs pump pulse from an amplified femtosecond Ti-sapphire laser operating at a pulse repetition rate of 1 kHz. The pump beam was focused to a 400 μm spot at the sample surface and partly absorbed in VO_2. A weak white-light probe beam, generated in a sapphire crystal plate by the same Ti-sapphire laser pulse, was focused to a 25 μm spot in the center of the pump spot. The temporal delay between pump and probe pulses was scanned by an optical delay line with 10 μm precision. The time-dependent reflectivity spectrum of the opal was measured by a spectrometer equipped with a CCD connected with a computer. The (111) plane of our photonic crystal makes an angle of a few degrees with the polished sample surface. This enables us to spatially separate the first-order Bragg diffracted beam and the specularly reflected beam.[20] We corrected all spectral-temporal dependencies of the reflectivity of the opal for the chirp in the optical probe pulse, using the instantaneous response of an amorphous silicon film[28] and obtained an overall temporal resolution of $\tau_p \approx 170$ fs.

Experimental results are presented in Fig. 2. Figure 2(a) shows the Bragg diffracted spectra measured just *before* (solid line) and 2.5 ps *after* the optical excitation (dashed line) in the region of the PBG. A clear shift of the peak associated with the PBG is observed. We measured identical values for the shifts in the stationary spectrum going through the phase transition. We note that the observed shift is smaller than the maximum expected value of ~70 nm because of the small (8%) filling factor of VO_2 in the voids in our sample.

Figure 2(b) shows the spectral-temporal evolution of the Bragg reflectivity. Colors indicate relative changes of the reflectivity, $\Delta R/R$: green no changes of the intensity, blue a decrease, and pink an increase. Of course, no differences in reflectivity are observed prior to the arrival of the pump pulse. At $t=0$, when the pump pulse arrives, the intensity rapidly increases on the *short-wavelength* side and decreases on the *long-wavelength* side. This signifies rapid shifting of the PBG. Within half a picosecond, the transition is completed and the reflectivity remains virtually constant over at least hundreds of picoseconds. This time is determined by the cooling down of the excited volume and takes as long as 100 μs.[27] No significant transient changes in the reflectivity spectra were observed in case the sample was held above the transition temperature, confirming that the rapid shifting is induced by the phase transition. Further, the shifts of the PBG were exclusively[24] observed when the pump power density exceeded a threshold of ~5–10 mJ/cm^2. Above 50 mJ/cm^2, irreversible changes in the reflectivity spectra are observed presumably due to chemical reactions and surface damage.

Figure 3(a) displays as solid lines the observed switching dynamics at the short-wavelength (610 nm) wing, at the long-wavelength (670 nm) wing, near the maximum (630 nm) of the PBG, and at an intermediate spectral position (640 nm). Beyond delays of ~500 fs, $\Delta R/R$ reaches a stationary value $A(\lambda)$, indicated as horizontal arrows. Further, the leading edge at 610 nm is delayed with respect to the laser pulse by a few hundred femtoseconds. We analyze our results by taking into account two independent contributions to $\Delta R/R$: (i) the phase transition in VO_2, responsible for a fast shift of the PBG characterized by $F_p(t)$ and a maxi-

mum effect $A(\lambda)$, and (ii) hot carriers, producing a small global change, $F_e(t)$. The latter contribution has the same origin as in our earlier experiments with opal-Si composites.[20] This assumption is reasonable because the experimentally observed changes in the spectrum are relatively small. Then $\Delta R/R$ as a function of time t and wavelength λ may be written as

$$\Delta R/R \approx A(\lambda)F_p(t) - F_e(t). \quad (1)$$

The empirical phenomenological functions $F_e(t)$ and $F_p(t)$ [dashed and solid lines in Fig. 3(b), respectively], were extracted from experiment by subtracting and summing $\Delta R(t)/R$ measured at two different wavelengths, for all combinations of curves in Fig. 3(a). Using these functions, the dotted lines of Fig. 3(a) represent the calculated curves according to Eq. (1) and excellently track the data. Apparently, at the short-wavelength side of the spectrum, the two contributions for $t<0.4$ ps partly compensate while at the long-wavelength side they add, explaining the fact that the switching speed depends on wavelengths. From the curve $F_p(t)$, we conclude that the optically induced phase transition takes only $\sim$250 fs, and from $F_e(t)$ we conclude that hot-electron-induced effects virtually track the laser pulse. Unfortunately, our setup does not allow for resolving the ultrafast electron dynamics. The measured speed of the phase transition exceeds the phonon equilibration speed by far signifying that the phase transition must be a nonequilibrium process and cannot be described by conventional heating.[29] Presumably, the phase transition is governed by nonequilibrium LO phonons directly created by hot carriers.[30] In semiconductors, namely, the characteristic emission time of LO phonons by hot electrons can be as short as a few hundred femtoseconds.

In conclusion, we demonstrate that a 3D photonic crystal based on an opal-vanadium dioxide composite is a promising system for ultrafast manipulation of the PBG position. The observed shift of the PBG is demonstrated to take place on the subpicosecond time scale. Further, the high refractive index of VO_2 and the possibility of fabrication of inverted VO_2 opals[27] make this material a candidate for obtaining a 3D tunable photonic crystal with a complete PBG. Already at the low concentration of VO_2 in the opal voids in our sample of $\sim$8%, substantial ultrafast shifts ($\sim$5 nm) of the PBG were reached. In nanosecond experiments and in other opal samples with a higher concentration of VO_2 in the pores, spectral shifts up to 40 nm have recently been observed, quite sizable relative to the PBG bandwidth.[31] This suggests that ultrafast control of light in VO_2-based photonic crystals is relevant for applications.

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