Enhancement of optical and magneto-optical effects in three-dimensional opal/Fe_3O_4 magnetic photonic crystals

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Three-dimensional magnetic photonic crystals, based on an artificial opal matrix with embedded magnetite Fe_3O_4, were investigated in both transmission and reflection in the near-infrared and visible spectral range. A strong enhancement of the polar Kerr effect and a modification of the Faraday effect have been found near the photonic band gap at about 1.8 eV. Surprisingly the shapes of the loops of magnetic hysteresis measured by magnetic circular dichroism were found to depend on the wavelength of light. This observation has been explained using a model where two types of magnetite particles have different coercive fields. © *2008 American Institute of Physics*.
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Modern demands for the high speed technology for telecommunication and data processing motivate an intense search for advanced materials and nanostructures, allowing an effective control and manipulation of the electromagnetic radiation in the optical spectral range. In particular, this search has led to the invention of photonic crystals (PCs), i.e., *n*-dimensional (nD, $n=1-3$) space-modulated structures having a lattice parameter comparable with the light wavelength.[1] In such PCs, interference phenomena give rise to the formation of specific photonic bands having high-transparency and high-reflectance regions.

Although the overwhelming majority of studies of PCs is devoted to *nonmagnetic* structures,[1–3] a construction of a photonic structure from a magnetic material or filling of PCs with magnetic material can provide possibilities for the manipulation of light. Such *magnetic* PCs (MPCs) would give an extra degree of freedom in achieving enhanced reciprocal or nonreciprocal optical phenomena such as magneto-optical (MO) Faraday and Kerr effects, magnetic linear birefringence, magnetic-field-induced second-harmonic generation, and magnetic gyrotropic birefringence. Basic types of MPCs are reviewed in Refs. 4 and 5. Though linear optical and MO effects in two-dimensional MPCs have been experimentally and theoretically studied,[5–7] it seems that such phenomena in three-dimensional (3D) MPCs have not been studied yet.

In this paper we present experimental results on optical and MO phenomena in 3D MPCs based on opal PC with embedded magnetite Fe_3O_4. A formation of the photonic band gap (PBG) in the spectral range of 1.6–2.0 eV was proven by the optical transmission and reflection techniques. A strong enhancement of the polar MO Kerr effect and a modification of the Faraday effect have been found near the PBG due to localization of the light field. Surprisingly the shapes of the loops of magnetic hysteresis measured by magnetic circular dichroism (MCD) were found to depend on the wavelength of light. This unusual observation has been explained using a model with two types of magnetite particles inside the opal matrix having different coercive fields.

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3D MPCs studied in this work were designed to have a photonic gap around 1.8 eV. Artificial opal was chosen as a volumetric matrix formed from submicron silica spheres with effective diameter of 290 nm. SiO_2 spheres were synthesized on the basis of polycondensation of silica molecules. These spheres had the compact packing of face-centered-cubic type. A technology for opal pores filling with Fe_3O_4 will be published elsewhere. Using this technology, opal/Fe_3O_4 MPCs with the pore volume filling from 20% to 70% were prepared. Opal thin film (111) samples of artificial opal PC with embedded magnetite Fe_3O_4 had a thickness of 5 μm. A formation of the crystalline magnetite inside the opal matrix was confirmed by the x-ray analysis (see the inset of Fig. 1).

Optical and MO properties of 3D opal/Fe_3O_4 MPCs with the pore volume filling of 70% were studied in transmission and reflection. In particular, optical transmittance, reflectance, MCD, and MO Faraday and polar Kerr effects were measured. Figure 1 shows spectra of the transmission, Faraday rotation, and MCD in the photon energy range of 1.4–2.8 eV. The opal/Fe_3O_4 sample becomes highly absorbing at photon energies above 2.4 eV. A formation of the PBG is observed as a noticeable minimum in the transmission, Faraday rotation, and MCD spectra near 1.8 eV. Optical properties of the pure opal matrix and their changes at impregnation with magnetite were discussed in Refs. 8 and 9. Spectral behavior of the Faraday effect and MCD in MPCs are in agreement with literature data on MO Kerr effect in pure magnetite.[10–12] There is correspondence between spectral behavior of the Faraday rotation and Kerr ellipticity and MCD and Kerr rotation due to known MO relationship.[13] Several contributions to the MCD may result from the intervalence and intersublattice charge transfer transitions.[10–12]

Figure 2 shows spectra of reflection and MO polar Kerr effect in opal/Fe_3O_4 MPC with the pore volume filling of 70%. Optical reflection spectrum is given in the photon energy range of 1.0–2.8 eV. The spectrum has a deep minimum and a sharp maximum near the PBG. Such spectral behavior is usually observed in the opal photonic structures.[14,15] MO Kerr rotation and ellipticity in the photon energy range of 1.2–2.4 eV are shown in Fig. 2(b) at an incidence angle of

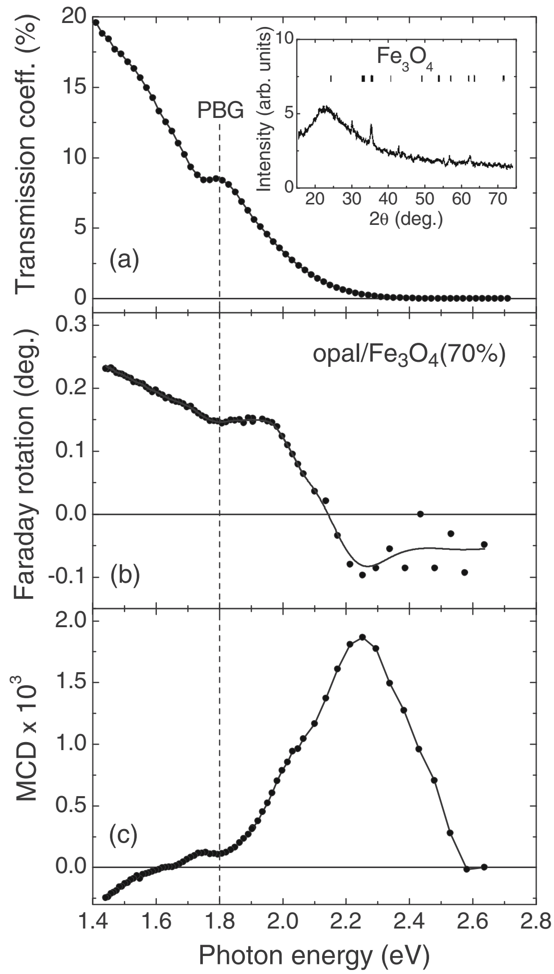

FIG. 1. Spectra of (a) transmission, (b) Faraday rotation, and (c) MCD for the opal/Fe_3O_4 sample with the pore volume filling of 70%. Measurements were done at normal incidence to the (111)-plane of the opal matrix. Inset shows x-ray diffraction pattern of opal/Fe_3O_4. Vertical dash line indicates the PBG spectral position.

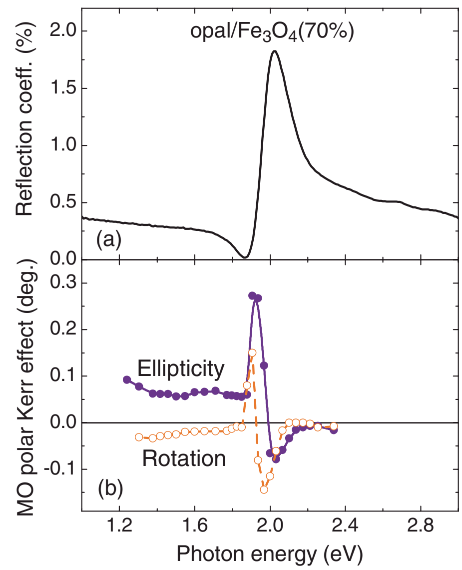

FIG. 2. (Color online) Spectra of (a) reflection and (b) polar Kerr effect for the opal/Fe_3O_4 sample with the pore volume filling of 70% in reflection from (111)-plane of the opal matrix. Angle of incidence is 45°.

45°. The Kerr ellipticity shows a strong maximum at photon energies where the reflection coefficient has a minimum, then the Kerr ellipticity reverses sign along with a strong increase in the reflection coefficient. The Kerr rotation reverses sign near the PBG, has a maximum at ~1.9 eV, and reaches a minimum at ~1.95 eV. This observation clearly demonstrates a strong enhancement of the polar Kerr effect in the spectral range near the PBG due to the strong light localization as a consequence of interference phenomena on the studied opal/Fe_3O_4 structure.

In Fig. 3 hysteresis curves measured by the MCD are shown. In the spectral region where the MCD reverses its sign, a surprising sensitivity of the shapes of hysteresis curves to photon energy has been observed. In order to explain this unusual spectral behavior of hysteresis curves we suggest the following model. If particles of magnetite Fe_3O_4 inside the opal matrix are assumed to be spherical, they are expected to be magnetically isotropic and thus characterized by a square hysteresis loop. It is known that magnetite particles with different sizes have noticeably different coercive fields.[16,17] The distribution of the coercive field of the magnetite particles in the matrix can be assumed as Gaussian with an expected value H_c and a variance H_v. We assume that inside the opal matrix there are two types of magnetite particles having coercive fields H_{c1} and H_{c2} with variances H_{v1} and H_{v2}. In addition, magnetite particles with different sizes must have somewhat different spectral behaviors of the MO effects, namely, the change in the MCD occurs at slightly different photon energies E_1 and E_2. If one considers a narrow spectral range close to the photon energies $E_{1(2)}$ where the MCD reverses sign, the MCD can be described as a linear function $\mathrm{MCD}(E,M) \propto (1 - E/E_{1(2)}) M_{1(2)}$, where $M_{1(2)}$ are total magnetizations of two types of magnetite particles. Then after integration the total magnetization $M_{1(2)}$ is

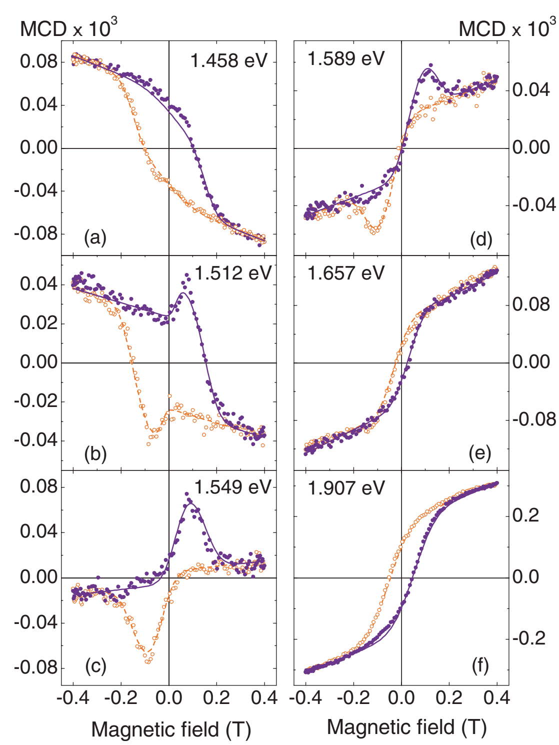

FIG. 3. (Color online) Hysteresis loops for the opal/Fe_3O_4 sample with the pore volume filling of 70% at different photon energies. Points are experimental data and lines are model calculations based on Eq. (1).

described by the function $M_{1(2)}(H) \propto \text{erf}[(H-H_{c1(2)})/\sqrt{2}H_{v1(2)}]$. Thus, the hysteresis behavior of opal/Fe_3O_4 MPC measured by the MCD at the photon energy E can be modeled by means of the following expression:

$$\text{MCD}(E,H) \propto A\left(1-\frac{E}{E_1}\right)\text{erf}\,\frac{H-H_{c1}}{\sqrt{2}H_{v1}} + B\left(1-\frac{E}{E_2}\right)\text{erf}\,\frac{H-H_{c2}}{\sqrt{2}H_{v2}} + DH, \quad (1)$$

where A and B are the coefficients obtained after the integration of the MCD over all the particles of the probed volume and D is the slope at saturation. All experimental hystereses in Fig. 3 are fitted by Eq. (1) giving coercive fields $H_{c1}=0.03$ T and $H_{c2}=0.15$ T. The fitted curves closely reproduce experimental data that prove fidelity of the model used.

In conclusion, we have designed and fabricated 3D MPCs, based on the opal PC with embedded magnetite Fe_3O_4 particles. Opal/Fe_3O_4 MPCs with the pore volume filling of up to 70% were studied by optical transmission, reflection, MCD, and MO Faraday and Kerr effects. Transmission and reflection spectroscopies have revealed a formation of the PBG in the range from 1.6 to 2.0 eV. A strong enhancement of the Kerr effect is found near the PBG. This phenomenon is explained by the strong light localization as a consequence of interference phenomena on the submicron structure. A modification of the Faraday effect is found near the PBG. Surprisingly, the shapes of the loops of magnetic hysteresis measured by MCD were found to depend on the wavelength of light. This unusual observation has been modeled by two types of magnetite particles having different coercive fields. Besides the interesting physics of 3D opal/Fe_3O_4 MPCs, the strong enhancement of the MO effects may open potentials for future integral optical devices.

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