# Faraday effect of photonic crystals

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We measured Faraday rotation in three-dimensional photonic colloidal crystals impregnated with a Faraday active, transparent liquid. The Faraday effect was found to strongly increase inside the stop band, whereas outside it follows the normal spectral behavior of a paramagnetic dielectric with an effective Verdet constant equal to the product of the liquid's Verdet constant and its filling fraction. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1558954]

In recent years, photonic crystals [or photonic band gap materials (PBG)] have been the subject of intensive theoretical and experimental studies. Their periodic dielectric structure result in Bloch-like electromagnetic waves inside the crystal with a stop band in their frequency spectrum for certain propagation directions and sometimes even a genuine gap with a vanishing density of states. The properties of PBG have led to much excitement about possible new optical devices.[1–5] Faraday rotators, as they are used in optical isolators, are still rather bulky devices for integrated optics. This led to the recent exploration of alternatives.[6–8] One might expect that the Faraday effect in PBG is much stronger and that optical isolators based on PBG could be much smaller than bulk devices. In this letter we present measurements of the Faraday effect in a PBG.

The Faraday effect is fundamental in the study of the magneto-optical properties of matter.[9] The direction of polarization of light propagating parallel to a magnetic field is rotated. The rotation angle $\theta$ is given by $\theta = V\,Bl$, where $B$ is the magnetic field, $l$ the propagation distance, and the material parameter $V$ is called the Verdet constant. The effects of magnetic fields on the propagation of light in PBG are still unresolved. The usual methods to calculate dispersion relations and transmission[10,11] fail when time symmetry is broken by an applied magnetic field. Even the simple case of light scattering by single spheres in magnetic fields has only recently been calculated.[12] Related problems are optical activity in PBG[13] and the experimental investigations of the Faraday effect in one dimensional PBG.[14]

For our experiment we used colloidal crystals consisting of a fcc packing of silica spheres.[15] The crystals are 1 mm in thickness and are several square millimeters large. Two different crystal batches, differing in sphere size with stop bands around 573 and 630 nm along the [111] direction, respectively, have been examined. Figure 1 depicts the typical transmission spectrum. We have filled the voids between the spheres with a saturated glycerol solution of dysprosium nitrate. This transparent liquid has a relatively high Verdet constant of $V = -22$ rad/Tm at 573 nm and a refractive index $n = 1.484$ which makes our samples more sensitive to the applied magnetic fields. In comparison, fused silica has a $V$ of $+3.48$ rad/Tm at 632.8 nm.[16] The beads that are used in our PBG are made up of silica clusters of 10–30 nm forming spheres of less than 300 nm in size. The lower refractive index of our beads, $n = 1.415$ to be compared to $n = 1.460$ for fused silica, means that these spheres are porous so that small amounts of liquid can penetrate. This will result in an even lower $V$, since the $V$ of the liquid has the opposite sign and the $V$ of air is practically zero. The wavelength dependence of $V$ is given by $V = V_0/(\lambda^2 - \lambda_0^2)$, $\lambda_0 = 171$ nm, $V_0 = 6.6\times10^6$ rad/Tm.[17] The silica is assumed to be only slightly influenced by the magnetic field. With the only off-

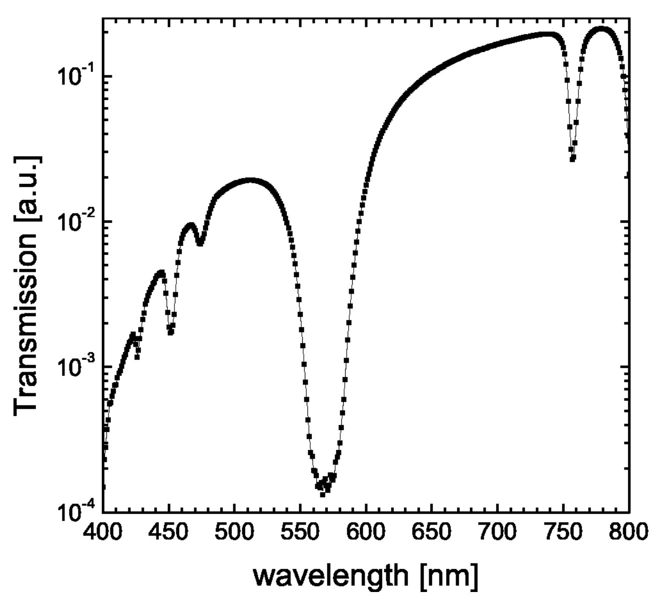


FIG. 1. Transmission spectrum of our impregnated silica photonic crystal measured with a commercial spectrometer showing a stop band around 570 nm. The impregnating liquid is a glycerol solution saturated with dysprosium nitrate. Several dysprosium absorption peaks to the right and left of the stop band can be found as well ($d = 260$ nm, $l = 1$ mm, $\lambda_B = 573$ nm, $n_{liquid} = 1.484$, $\Delta n = 0.070$).

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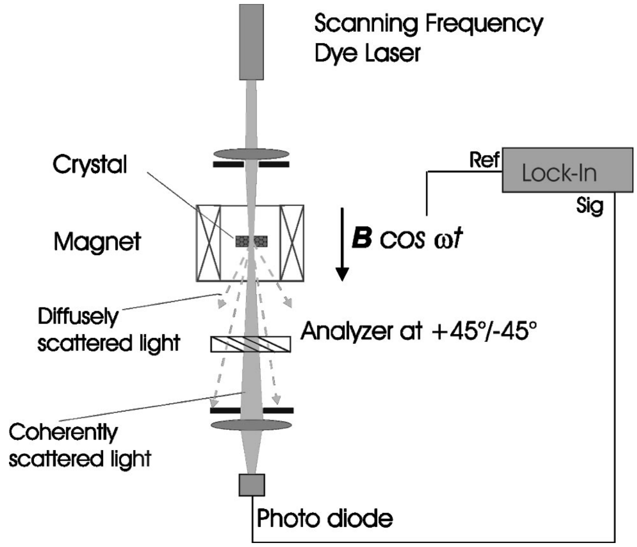


FIG. 2. Experimental setup to measure Faraday effect in photonic crystals.

diagonal elements in the dielectric constant occurring in the liquid due to the external magnetic field, we get:[9] $\hat{\epsilon}_{ik} = \epsilon(\delta_{ik} - i e_{ikl} Q_l)$, ($e_{ikl}$ Levi–Civita tensor, magnetic susceptibility $\mu \approx 1$) with the magneto-optic parameter $Q$ proportional to the magnetic field: $Q_z = \lambda \pi^{-1} \epsilon_l^{-1/2} V B_z$. It is important to note that despite the advances made in recent years in fabricating PBG, defects—especially stacking faults—still put certain limits on the optical quality.[18–20] Nevertheless, by choosing only a small dielectric contrast between the liquid and the silica spheres, scattering is less pronounced and measurements in transmission become feasible.

The experimental setup to measure the Faraday effect in our samples is shown in Fig. 2. A linearly polarized light beam enters the sample parallel to the crystal's [111] direction, which is also parallel to the magnetic field. We use an alternating magnetic field of 70 Hz to allow for phase sensitive detection. A tunable dye laser serves as a coherent light source and supplies us with sufficient power near the stop band where the transmission is low. Care must be taken to find a position on the crystal with few defects, so that there is a clear coherent transmission dominating the diffusely scattered background. An analyzer at 45° translates the rotation angle variations in intensity variation.

Figure 3 shows the Faraday effect of the impregnated crystal. The transmission of laser light through the crystal is likewise depicted. Since the laser has a rather small aperture the measured stop band is narrower and deeper than in Fig. 1, for which we used a commercial spectrometer. The PBG is characterized by its Bragg wavelength of $\lambda_B = 573$ nm when filled with glycerol solution. From this we can calculate the diameter of the beads to be around 245 nm, which is fairly consistent with scanning electron microscopy (SEM) measurements showing $d \approx 260$ nm. The refractive index contrast is $\Delta n = +0.070$. A value $Q \approx 1 \times 10^{-7}$ is produced by a magnetic field of $B = 33.5$ mT. The value for the Verdet constant outside the stop band corresponds to that of the liquid corrected for the volume fraction $f$ of the liquid in the crystal. Inside the stop band however, the Verdet constant increases drastically. Note that there is hardly any change observed in the Faraday effect near the stop band when transmission already falls several orders of magnitude.

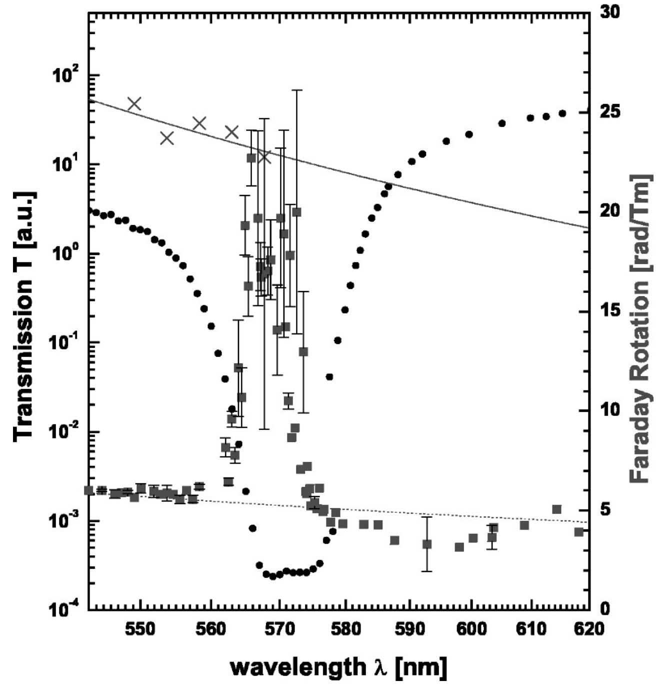


FIG. 3. Transmission of laser light (circles) and Faraday rotation (squares) spectra of an impregnated silica photonic crystal [$d = 260$ nm, $l = 1$ mm, $\lambda_B = 573$ nm, $V_{\text{liquid}}(\lambda_B) = -22$ rad/Tm, $n_{\text{liquid}} = 1.484$, $B = 33.5$ mT , $Q \approx 1 \times 10^{-7}$, $\Delta n = 0.070$]. The upper line indicates the corresponding Faraday rotation of the pure liquid (crosses are measurements) and results in the lower dotted line when corrected with the filling fraction inside the PBG.

The results of a second sample [$d = 270$ nm (SEM:275 nm), $l = 1$ mm, $\lambda_B = 630$ nm, $V_{\text{liquid}}(\lambda_B) = -12.5$ rad/Tm, $n_{\text{liquid}} = 1.489$, $B = 36.4$ mT, $Q \approx 6 \times 10^{-8}$, $\Delta n = 0.075$] are plotted in Fig. 4. Both types of our PBG exhibit the same characteristic spectral dependence.

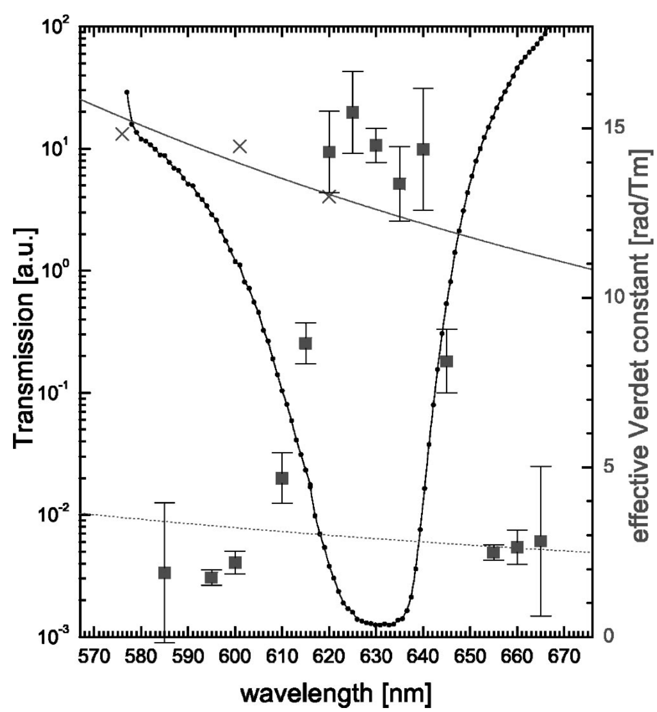


FIG. 4. Same as in Fig. 3, except for $d \approx 275$ nm, $\lambda_B = 630$ nm, $V_{\text{liquid}}(\lambda_B) = -12.5$ rad/Tm, $n_{\text{liquid}} = 1.489$, $B = 36.4$ mT, $Q \approx 6 \times 10^{-8}$, $\Delta n = 0.075$.

The Verdet constant outside the stop band takes the values of a simple effective medium associated with the two composites: $V_{\mathrm{eff}}=V_a f_a+V_b(1-f_a)$. Effective medium theories exist for periodic media[21,22] and for magneto-optical quantities,[23] but they fail to show such a simple relation. It is remarkable that in opal-based photonic crystal structures the same simple effective medium approximation, although it should not necessarily be applicable to such a densely packed system of scatterers, proves to be valid. It describes in a correct manner the effective refractive index $n_{\mathrm{eff}}$ appearing in the expression for the Bragg wavelength $\lambda_B$ under perpendicular incidence, as determined from optical reflectance or transmission measurements: $\lambda_B=2cn_{\mathrm{eff}}$, where $c$ is the distance between (111) planes of the lattice. Here we take the effective refractive index as $n_{\mathrm{eff}}=n_a f_a+n_b(1-f_a)$.[24] This relationship indeed describes the Bragg wavelength rather well, as is shown by experiments with silica-based opals[25,26] and solid-state opals built of polymer beads.[27] If not exactly the above simple approximation, then some close (for the given conditions) as shown in Refs. 28 and 29 for colloidal crystals of polymer particles in aqueous medium.

The mechanism responsible for the enhanced Faraday rotation is the occurrence of multiple internal Bragg-like reflections inside the stop band as in a Fabry–Pérot interferometer.[12] As is well known,[30] the Faraday rotation is cumulative for waves propagating back and forth. It is remarkable that in the Faraday rotation spectra for both crystals, we find the maximum effective Verdet constant is equal to the one measured for the pure liquid. This might still be purely coincidental, but could just as well point to an underlying cause that remains to be identified. The question naturally arises if a similar effect occurs in the reflected wave. As most light is reflected in the uppermost layers of the PBG, it cannot see much of the Faraday-active liquid, therefore no large effect is expected.

We also performed measurements on magnetic circular dichroism (MCD), which required only small changes to the Faraday rotation setup. In order to get circularly polarized light we placed a $\lambda/4$-plate between the laser and the sample and by removing the analyzer the photodiode signal directly reflects transmission changes. Since MCD is a difference in absorption for the two circularly polarized states, we have to deal with an imaginary part in the refractive index: $n\rightarrow\hat{n}=n+i\kappa$, leading to two different values: $n_{\pm}=n(1\pm\frac{1}{2}\frac{\delta n}{n})+i\kappa(1\pm\frac{1}{2}\frac{\delta\kappa}{\kappa})$. The measurement showed MCD to be smaller than the detection limits of $\delta\kappa/\kappa=10^{-6}\ \mathrm{T}^{-1}$ outside and $\delta\kappa/\kappa=10^{-2}\ \mathrm{T}^{-1}$ in the center of the stop band taken at the characteristic points of the spectrum.

In summary, we measured the Faraday rotation of photonic crystals, which were impregnated with a Faraday-active liquid. The Faraday rotation outside the stop band follows the spectral behavior of the pure liquid multiplied by its volume fraction. Inside the stop band, the Faraday rotation is enhanced by up to a factor of five. The origin of this remains to be identified since no existing theory explains this behavior.

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