Photonic Hall effect in absorbing media

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We describe an experimental and theoretical study of the effect of optical absorption on the photonic Hall effect in a passive matrix containing magnetoactive scatterers. We find that for the case of absorbing scatterers, the magnetotransverse light current changes sign and increases with increasing absorption. Good agreement is obtained with numerical calculations. For the case of an absorbing matrix, no effect was observed.

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Recently it was shown both theoretically [1,2] and experimentally [3,4] that light diffusing in a disordered medium subject to a magnetic field can show behavior that bears a strong phenomenological resemblance to well-known electronic magnetotransport effects. In particular, the electronic Hall effect and magnetoresistance were found to have photonic analogues. This may be surprising at first sight, as photons do not carry electric charge and therefore should not couple to a magnetic field. However, such a coupling is indirectly provided through the induced polarization at optical frequencies. Nevertheless, conceptual differences between the two cases do exist. In the electronic case, the number of diffusing particles—the electrons or holes—is usually conserved. This only holds true for the photonic Hall effect, if optical absorption is negligible. Here we will show that introducing optical absorption drastically affects the photonic Hall effect.

The effect of a static magnetic field **B** on the optical properties of an isotropic medium is described by the refractive index tensor $\mathbf{n}(\mathbf{B})$, given up to first order in **B** by [5]

$$n_{ij}(\mathbf{B}) = (n + i\kappa)\,\delta_{ij} + i\frac{V}{k}\epsilon_{ijl}B_l, \tag{1}$$

where k is the vacuum wave vector, $n+i\kappa$ is the complex refractive index of the medium, and V the complex Verdet constant, Re V determining the strength of magnetic circular birefringence (the Faraday effect) and Im V determining the strength of magnetic circular dichroism.

In principle, Maxwell's equations plus the known (magneto-)optical material parameters as expressed in Eq. (1), enable us to exactly calculate the effect of a magnetic field on the diffusion of light in strongly disordered media. In practice however, this is an insurmountable task. Theoretical simplifications have to be made, as was first done in Refs. [1] and [2]. The relevant transport quantity for light diffusion is the second-rank diffusion tensor relating the diffuse photon flux density to the gradient of the photon density. The off-diagonal elements of this tensor represent the photonic Hall effect (PHE). This effect, which is linear in **B** for $VBl^* \ll 1$, is most conveniently characterized by the normalized magnetotransverse light current η, given by

$$\eta(B) \equiv \frac{1}{B}\frac{I_L(B) - I_R(B)}{I_L(B) + I_R(B)}. \tag{2}$$

For the definition of I_L and I_R, see Fig. 1.

Recently, a method has been developed to calculate η numerically for scattering spheres of arbitrary size and arbitrary refractive index. The magnetic-field dependent scattering cross section from a single dielectric Faraday-active sphere was obtained to first order in the field [6]. With this solution for the single-scattering problem, the PHE in multiple light scattering was calculated using transport theory for light in nonabsorbing media [7]. Below we will compare the results of an extension of such a calculation with our experimental results.

Our present experiment deals with scatterers made of magneto-optically active material with refractive index $n_s + i\kappa_s$ and Verdet constant V_s. They are randomly distributed with a volume fraction f in an isotropic matrix with refractive index n_m and negligible Verdet constant V_m. The case of a (nonabsorbing) magnetoactive matrix containing inactive scatterers was discussed in Ref. [8]. If the transport mean-free path l^* of the light is much smaller than the geometrical dimensions of the sample, the propagation of light is diffusive. The effect of the magnetic field on the diffusion can be described by the dimensionless parameter VBl^* [9]. The experiments we present here are in the range $VBl^* \ll 1$. The quantity VBl^* determines the average number of Faraday rotations of the electric polarization vector of the light between subsequent scattering events. The same parameter has been shown to be important for the suppression of coherent backscattering in magnetic fields, both experimentally [10,11] and numerically [12]. A similar situation occurs in diffusive electronic magnetotransport. There, the magnetic field effect is determined by the dimensionless parameter $\omega_c\tau$, being the average number of cyclotron orbits an electron completes between subsequent scattering events. Simple free-electron models show that the resulting Hall angle is proportional to $\omega_c\tau$ and that the longitudinal electronic magnetoresistance is proportional to $(\omega_c\tau)^2$ [13]. Also in the

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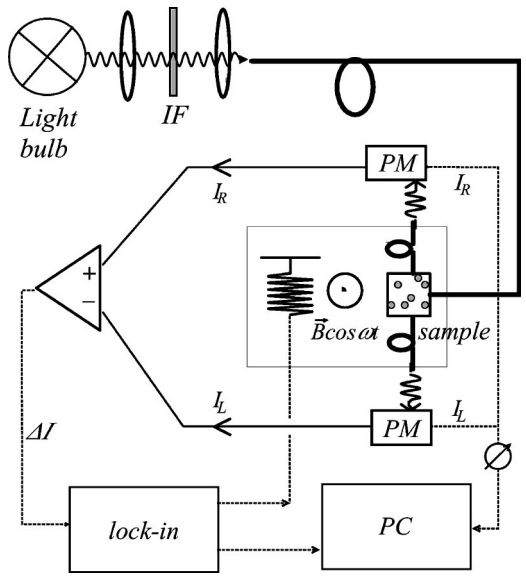

FIG. 1. Schematic setup of the photonic Hall effect measurement. IF denotes the narrow band interference filter and PM the photomultipliers.

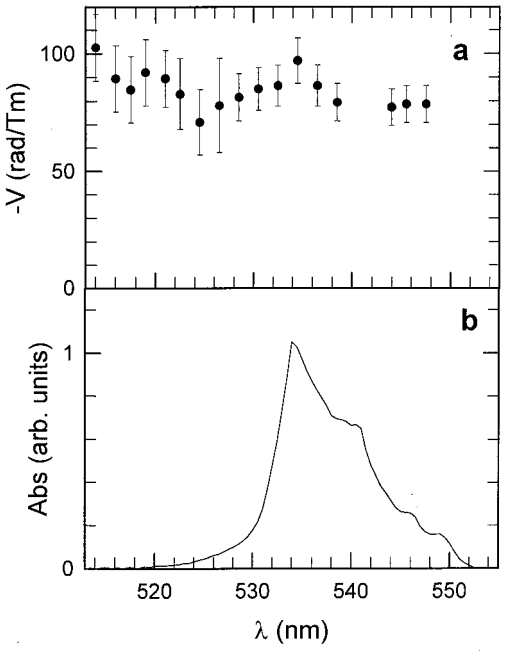

FIG. 2. (a) Verdet constant of Ho^{3+} ions in aqueous solution at 300 K. The result is scaled to a concentration of Ho^{3+} ions that is equal to that in HoF_3 powder. (b) Absorption spectrum of HoF_3 powder in resin at $T=300$ K. The scattering background has been subtracted.

case of the Beenakker-Senftleben effect [14], i.e., the magnetic-field dependence of the thermal conductivity of gases, an analogous parameter has been established, namely, the number of molecular spin precessions between molecular collisions. In the diffusive electronic magnetotransport and in the Beenakker-Senftleben effect, the number of the diffusing particles does not change during the diffusion process. However, in light diffusion, this does occur in absorbing media. The effect of absorption in multiple-scattering media is usually described by means of the absorption length $L_a = \sqrt{l^* l_a/3}$, l_a being the absorption length of the coherent beam in the medium (without scattering). It is related to the imaginary part of the refractive index by $l_a = 1/(2\kappa k)$. Thus, in the presence of absorption, the absorption length L_a has to be taken into account as an additional length scale, together with the diffusion length l^* and the sample length.

For the experimental observation of the photonic Hall effect, scatterers with a large Verdet constant are required. This can be found in materials containing large concentrations of rare-earth ions like Ce^{3+}, Ho^{3+}, or Dy^{3+}. In these paramagnetic materials, the Verdet constant is inversely proportional to temperature and can thus be further enhanced by cooling. In order to investigate the effect of absorption on the PHE, we have used scatterers containing Ho^{3+} ions, taking advantage of the narrow $4f$-$4f$ transition $^5I_8 \rightarrow {}^5F_4$ of this ion, around $\lambda \approx 534$ nm. By varying the wavelength of the diffusing light over a few tens of nanometers, we were able to scan across the absorption band, thereby strongly varying κ_s, whereas all other optical parameters remain practically constant. The scatterers were particles of HoF_3, obtained by chemical precipitation. They had an average radius r of 0.5 μm and a broad size distribution between $r=0.2$ μm and $r=5$ μm, which was determined using scanning electron microscopy. Their refractive index is 1.6 outside the absorption peak, and varies up to ± 0.004 around the maximum of absorption. The absorption spectrum in Fig. 2(b) was measured at room temperature from a thin slab of HoF_3 powder dispersed in an index matched resin at 23 vol % with a uv/vis spectrometer. The absorption spectrum of HoF_3, taken by means of a tunable interference filter and optical fibers, showed that the absorption maximum blueshifts from $\lambda = 534$ nm at $T=300$ K to $\lambda = 529$ nm at $T=85$ K. From the measurements carried out at $T=300$ K on samples of different thickness, we determine the imaginary part of the refractive index to be $\kappa_s \approx 0.0012$ at $\lambda = 535$ nm and $T=300$ K, and the absorption length in the multiple-scattering samples to be $L_a \approx 31 \pm 6$ μm at $\lambda = 535$ nm. We assume that these values are the same at $T=85$ K when corrected for the blueshift. Other optical parameters undergo only minor changes: $\mathrm{Re}\, V_s$ varies in a range of $\Delta V_s/V_s \approx 0.2$ as can be seen in Fig. 2(a), and $\mathrm{Im}\, V_s$ is estimated to be smaller than 1 rad/Tm [15] in this spectral range. For l^* we estimate a variation $\Delta l^*/l^* \leqslant 0.25$ across the absorption band due to the change of n_s with λ.

Figure 2(a) shows the Verdet constant of Ho^{3+} ions in water as a function of wavelength. A resonant behavior in the same region as the absorption peak at $T=300$ K is weakly visible. This curve has been obtained by measuring V of a paramagnetic aqueous $Ho(NO_3)_3 \cdot 5H_2O$ solution, and by correcting these results with those for a diamagnetic $La(NO_3)_3 \cdot 5H_2O$ solution. We deduce the Verdet constant for HoF_3 to be 400 rad/Tm at a temperature of 85 K. As matrix material, we used a thermally curable resin, which is amorphous, isotropic, and nonabsorbing. It has a negligible Verdet constant and a refractive index $n_m \approx 1.566$ at 589 nm. Samples were prepared by mixing HoF_3 powder at a volume fraction of $f=23\%$ with the liquid resin, followed by thermal curing in a mold. The transport mean-free path l^* of the light in these scattering media was determined by measuring their optical transmission T by means of an integrating

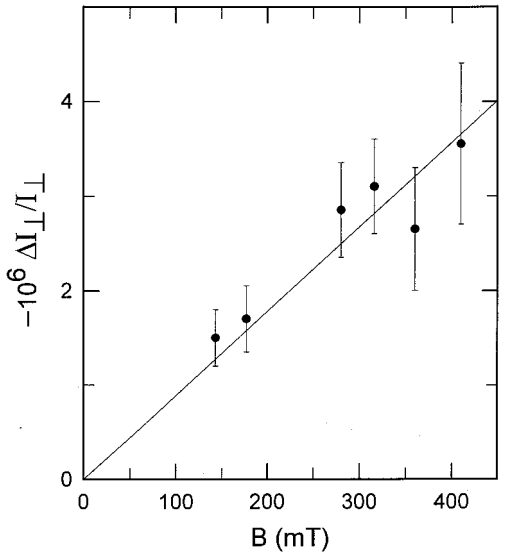

FIG. 3. Magnetotransverse photon flux at $T=85$ K and $\lambda=547$ nm as a function of the magnetic field. The sample contained HoF_3 powder at a volume fraction of $f=23\%$ in resin. The straight line is fitted through zero.

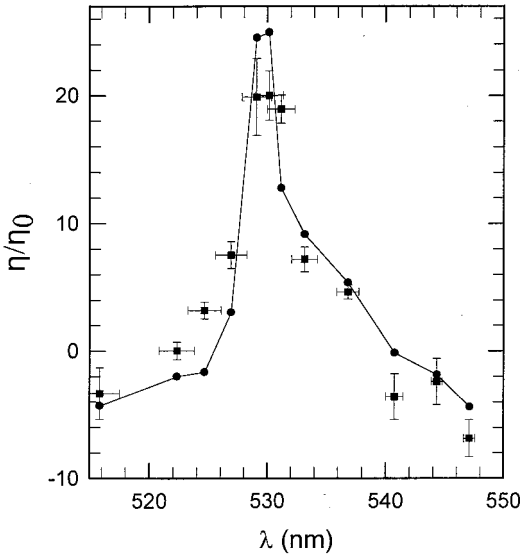

FIG. 4. Normalized magnetotransverse photon flux as a function of wavelength at $T=85$ K. The normalization constant $\eta_0=10^{-6}$ T^{-1}. Samples are made of HoF_3 powder at a volume fraction of $f=23\%$ in resin. Shown are experimental (squares) and numerical results (balls). Solid line is only meant to guide the eye.

sphere and using $T=l^*\cdot 1.6/L$ where L is the sample thickness [16]. We have found $l^*\approx 70\pm 26$ μm at a wavelength of $\lambda=548$ nm, which is in the transparent spectral region of HoF_3.

The PHE was measured by phase-sensitive detection of the magnetically induced changes in the difference of the intensities scattered to the left and the right (see Fig. 1). Narrow bandwidth illumination around 535 nm was provided by an incandescent lamp in combination with a narrow-band interference filter [full width at half maximum (FWHM) 0.5 nm]. Wavelength tuning was achieved by tilting the filter. The filtered light was guided to the sample by an optical fiber (diameter 1 mm, numerical aperture 0.47). The scattered light was collected by optical fibers of the same type and detected by means of photomultipliers. An alternating magnetic field $B(t)=B\cos\omega t$ with $B\approx 0.5$ T and $\omega\approx 30$ Hz was applied perpendicularly to the illuminating and collecting light guides. The samples were of cylindrical geometry with a diameter of 1 mm and a length of 1.5 mm. The axis was aligned parallel to the illuminating fiber. We measured the magnetotransverse photon flux $\Delta I_\perp=I_L-I_R$, which was then normalized by the transversely scattered intensity $I_\perp=I_L+I_R$. This ratio $\Delta I_\perp/I_\perp$ depends on the magnetotransverse and the normal diffusion coefficient. It is used to describe the magnetotransverse light current in a quantitative way and thus plays the same role as the Hall angle in electronic magnetotransport.

In Fig. 3, we show the observed linear magnetic-field dependence of $\Delta I_\perp/I_\perp$. Such a linear behavior was found independent of the amount of absorption. Normalizing this quantity by the magnetic field, we obtain the normalized Hall slope η [see Eq. (2)] which is characteristic for a given scattering medium, apart from a geometry factor of order unity. This geometry factor can be calculated from the solution of the diffusion equation in a cylindrical geometry. Figure 4 shows η as a function of wavelength around the absorption maximum. Outside the absorption band, we obtain negative values for η. These agree in sign and magnitude with the results obtained in Refs. [3] and [4] for similar paramagnetic scatterers. With increasing absorption, η decreases to zero and then strongly increases with opposite sign around the absorption band center. The sign change of η occurs at the wavelength where L_a roughly equals l^*. At the wavelength of maximum absorption, we obtain values for $|\eta|$, which are

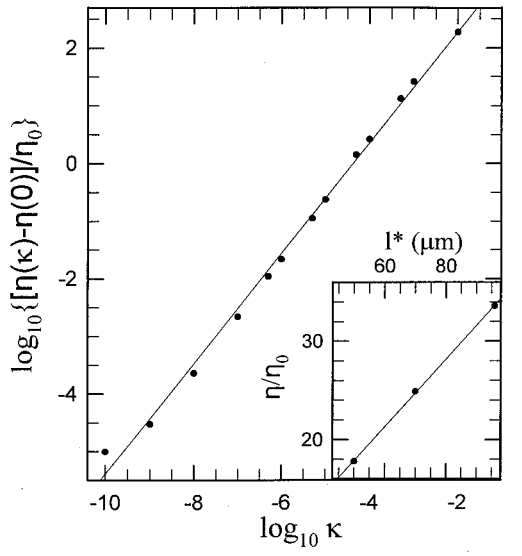

FIG. 5. Calculated change in the normalized magnetotransverse photon flux as a function of the imaginary part of the dielectric constant κ_s for $l^*=70$ μm. The normalization constant $\eta_0=10^{-6}$ T^{-1}. The straight line is a fit, which corresponds to an exponent of 0.96. Inset shows the calculated normalized magnetotransverse photon flux as a function of l^*, obtained by varying the scatterer density, at $\kappa_s=0.0012$. Straight line is a fit.

approximately 4 times larger than the ones measured in the transparent region of the HoF_3. At this wavelength, the absorption length $L_a \approx 31$ μm is smaller than l^* and much smaller than the sample dimensions. As the maximum of $|\eta|$ exactly corresponds to the wavelength of maximum absorption, we conclude that the observed changes in the photonic Hall effect are dominated by absorption effects, since the variations of the other relevant optical parameters would have led to only minor changes in η.

We have also prepared samples of similar, but transparent CeF_3 scatterers in the same resin matrix that was made absorbing by dissolving an organic dye into it. Values up to $\kappa_m \approx 0.001$ were obtained this way, i.e., comparable to the values of κ_s for the absorbing scatterers in the transparent matrix. We have found no significant variation of η with increasing κ_m. The role of absorption in the matrix is therefore clearly very different of that of absorption in the (magnetoactive) scatterers.

The method developed by us to calculate numerically the photonic Hall effect of spherical scatterers of arbitrary size [6] allows the inclusion of absorption inside the scattering particles. It does not allow for absorption in the matrix. In the calculation, we assume a complex refractive index $n_s + i\kappa_s(\lambda)$, and use $\kappa_s(\lambda)$ as obtained from the absorption spectra in Fig. 2(a), corrected for the low-temperature blueshift. We have taken $\mathrm{Re}\, V_s = 400$ rad/Tm as input for the calculations, assuming l^* to have the constant value of 70 μm, which was measured outside the absorption band. We have completely disregarded Im V_s. The geometry factor is kept constant as well, although we estimate theoretically that this factor will increase by about a factor of 2 in the range of absorption covered in this experiment. The broad size distribution of the scatterers was taken into account by averaging our numerical results over particle sizes between 0.2 and 5 μm. Our results were found to be quite independent of the exact choice of the particle size distribution. As shown in Fig. 4, they show good agreement with the experimental results. Note that no adjustable parameters are used, only experimentally determined ones. The uncertainty in these experimental values is estimated to impart a systematic uncertainty of 30% to our numerical results. The agreement confirms that the observed wavelength dependence of η is mainly due to absorption. In particular it is found to be due to the effect of absorption on the magnetic-field dependent part of the single Mie particle scattering cross section. We have explicitly calculated the effect of κ_s on η, assuming all other parameters to be constant, as shown in Fig. 5. The observed dependence is found to be linear. The inset shows the calculated effect on η of varying l^* through a variation of the scatterer density, for the κ_s that corresponds to the absorption maximum in Fig. 4. Again a linear dependence is observed. Such a dependence is found independent of the value of κ_s in the range of absorption in our experiment, Furthermore, it is consistent with earlier experimental results [3,9]. Our numerical results for the parameter ranges studied can be summarized by the relation

$$\eta = (\alpha + \beta\kappa_s) V_s l^*, \tag{3}$$

where α and β are parameters that may depend among others on sample geometry, refractive index contrast, size and shape distribution of the scatterers, etc., but not on Verdet constant, absorption, or magnetic field.

In conclusion, we have investigated the effect of optical absorption on the photonic Hall effect in multiple-scattering media with magneto-optically active scatterers. In such media, the number of the diffusing photons is not conserved, in contrast to the magnetotransport effects investigated up to now. We have found that optical absorption in the scatterers can strongly influence photonic magnetotransport. Within the absorption band of the magnetoactive scattering material, the magnetotransverse light current can change sign and become much larger. We find good agreement between our experimental results and our numerical calculations. For nonabsorbing scatterers in an absorbing matrix, no effect of absorption on the photonic Hall effect was experimentally observed. For this case, no theoretical description is available.

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