Observation of Photonic Magnetoresistance

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In this Letter, we report the observation of a quadratic magnetic-field dependence of diffuse light transmission through random media. This effect constitutes the photonic analogon of magnetoresistance known for diffusive electronic transport. The experiments are found to be in good agreement with recent theoretical predictions. [S0031-9007(97)03728-9]

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Recently, it was shown both theoretically [1] and experimentally [2] that light propagating in a scattering medium subject to a transverse magnetic field can be deflected in a direction perpendicular to both the incident light beam and the magnetic field. This new effect bears a strong phenomenological resemblance to the electronic Hall effect, where the Lorentz force causes a deflection of the electron trajectories. Basically, the "photonic Hall effect" finds its origin in the magnetically induced changes of the optical scattering properties of the individual particles in the disordered medium.

The mechanism causing the photonic Hall effect has also been predicted to generate photonic magnetoresistance [3]. This light transport phenomenon manifests itself as a dependence of the diffuse transmission coefficient on an externally applied homogeneous magnetic field, quite similar to the electronic magnetoresistance of disordered conductors [4]. For symmetry reasons, only even powers of the magnetic field can affect the light transmission. It is tempting to interpret the magnetoresistance as a magnetic-field dependence of the scattering mean free path. However, a simple analysis [3] shows that such a dependence averages out for random polarization and/or random wave vector, both intrinsic to multiple scattering. In contrast to our prediction in Ref. [3] there are at least two recent papers [5,6] that deal with optical diffusion in a magnetic field but do not predict photonic magnetoresistance. To our knowledge, such a photonic effect has never been observed before.

Maxwell's equations plus the known magneto-optical material parameters, in principle, offer the possibility to calculate magneto-optical diffusion. In practice, this is, however, completely impossible and approximations have to be made. In fact, the magneto-optical properties of even only one single Mie scatterer are not known, let alone the behavior of multiple scattering in a magnetic field. Therefore the theoretical simplification first made in Ref. [3] is valuable and a comparison of this theory with experiment, no doubt, represents an important test of our understanding of multiple light scattering, which is still progressing. The relevant transport quantity for light diffusion is the second-rank diffusion tensor relating the diffuse optical current density to the gradient of the optical energy density. This tensor can be expressed as [3]

$$D_{ij}(\mathbf{B}) = D_0\delta_{ij} + D_H\,\epsilon_{ijk}B_k + \Delta D_\perp(B^2\delta_{ij} - B_iB_j) + \Delta D_\parallel B_iB_j\,, \tag{1}$$

where D_0 describes conventional isotropic diffusion and D_H transverse diffusion, linear in the magnetic field. The existence of the latter, magneto-transverse, Hall-type diffusion term D_H has recently experimentally been verified [2]. The experimental investigation of the magnetoresistance term $\Delta D_\perp$, observable in transmission perpendicular to the magnetic field, is the subject of this Letter. Anisotropic light diffusion has recently also become important in nematic liquid crystals [7].

The effect of a static magnetic field $\mathbf{B}$ on the optical properties of an isotropic nonabsorbing medium is described by the refractive index tensor $n(\mathbf{B})$, given up to second order in $\mathbf{B}$ by [8]

$$n_{ij}(\mathbf{B}) = \left(n + \frac{M}{k}B^2\right)\delta_{ij} + i\frac{V}{k}\epsilon_{ijk}B_k + \frac{C}{k}B_iB_j\,, \tag{2}$$

where k is the vacuum wave vector, n is the refractive index of the medium, the Verdet constant V determines the strength of magnetic circular birefringence (the Faraday effect, a difference in refractive index for left- and right-handed circularly polarized light), C is that of magnetic linear birefringence (the Cotton-Mouton effect, a difference in refractive index for light polarized parallel and perpendicular to the magnetic field), and M reflects the effect of magnetostriction. Our experiment deals with small scatterers made of magneto-optically active material with refractive index n_{scat}. They are randomly distributed with a volume fraction f in an isotropic matrix with refractive index n_{med}. The latter has, to good approximation, no significant magnetic-field dependence. If the transport mean free path of the light ℓ^* is much smaller than the geometrical dimensions of the scattering ensemble, the propagation of light is diffusive and can be characterized by the second-rank diffusion tensor D_{ij} introduced

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in Eq. (1). The magnetic-field dependence of the refractive index results in the magnetic-field dependence of the diffusion tensor.

In Ref. [3] it is shown that the quadratic field terms in Eq. (1), $\Delta D_\perp$ and $\Delta D_\parallel$, contain terms proportional to V^2, C, and M. For Rayleigh scatterers, i.e., particles smaller than the wavelength of the light, one infers that the V^2 terms dominate and that essentially

$$\frac{B^2 \Delta D_{\perp,\parallel}}{D_0} \propto (fVB\ell^*)^2. \quad (3)$$

In large magnetic fields the parameter $fVB\ell^*$ can approach unity [9], but in the experiments that we present $fVB\ell^* \approx 10^{-2}$, so that terms beyond the perturbational quadratic ones can be neglected. Since fV can be identified as an effective medium estimate of the Verdet constant of the average medium, $fVB\ell^*$ determines the average number of (Faraday) rotations of the electric polarization vector between subsequent scattering events. The same parameter has been shown to be important for the suppression of coherent backscattering in magnetic fields, both experimentally [9,10] and numerically [5]. At first sight it may seem somewhat surprising that a linear magneto-optic effect results in a quadratic magnetic-field dependence of the diffusion tensor. However, 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 scattering events. Simple free-electron models show that the resulting longitudinal electronic magnetoresistance is proportional to $(\omega_c\tau)^2$ [4], i.e., analogous to Eq. (3). Also in the case of the Beenakker-Senftleben effect [11], i.e., the magnetic-field dependence of the thermal conductivity of gases, an analogous parameter has been established, namely, the number of spin precessions between molecular collisions.

For the experimental observation of the photonic magnetoresistance, Eq. (3) predicts it to be advantageous to have a long mean free path and a large volume fraction of magnetoactive scatterers with a large Verdet constant. Note that over a large extent of volume fractions, ℓ^* is inversely proportional to f, so that $f\ell^*$ is independent of the concentration of given scatterers. Still, at a large volume fraction, a large mean free path can be obtained by using a small, isotropic refractive index difference between scatterers and matrix. A large Verdet constant can be achieved by choosing paramagnetic scatterers, whose magneto-optical properties can be enhanced by cooling.

As scattering material we have selected EuF_2 in the form of two milled powders with different particle sizes. The EuF_2 particles have irregular shapes, and their average diameters are 0.4 and 2 μm, respectively. The magneto-optical properties of EuF_2 are well known [12]. Its low and isotropic refractive index ($n_{\rm scat} = 1.582$, determined by immersion) allows near index matching with a transparent UV curable optical adhesive ($n_{\rm med} = 1.505$). This enables the preparation of solid disks of scattering material with volume fractions of EuF_2 up to 0.4. The mean free path of the light in such disks has been determined by measuring their optical transmission T by means of an integrating sphere and application of the relation $T = 1.6\ell^*/L$ [13]. L is the sample thickness with typical values around 0.6 mm. The magnetic-field dependence of the transmission was measured in the setup shown in the inset of Fig. 1. An alternating magnetic field $B(t) = B\cos\Omega t$ was applied perpendicular to the illuminating and collecting light guides. Illumination was provided by an argon ion laser at a wavelength of 488 nm. At this wavelength, absorption of the scattering samples is negligible. The transmitted light was measured with a silicon photodiode outside the magnetic-field region. By comparing the transmission measured with an integrating sphere to the transmission measured with the setup in Fig. 1, we find that our setup captures approximately 60% of the forwardly scattered light. As we are interested in the quadratic magnetic-field response, we have detected the photodiode signal phase-sensitively with a lock-in amplifier at the second harmonic frequency 2Ω. It can be easily seen that this component represents the quadratic magnetic-field response. Great care was taken to eliminate spurious signals due to mechanical movements, as signals induced by vibration of the magnet may also occur at 2Ω.

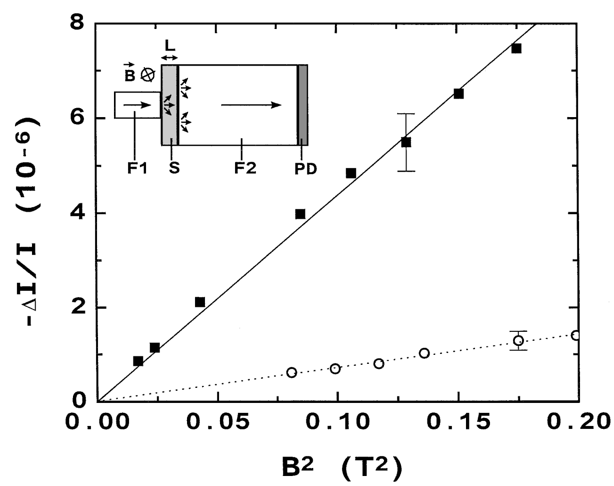

FIG. 1. Transmission modulation at 2Ω versus the square of the magnetic-field strength. Temperature is 105 K. Full symbols represent samples with an average particle diameter of 2 μm, with a volume fraction of scatterers of 0.17; open symbols are for an average particle diameter of 0.4 μm and a volume fraction of 0.1. Lines are linear fits to the data points. The inset shows a schematic setup. Light is guided through an optical fiber F1 [diam 1 mm, numerical aperture (NA) = 0.47] to the sample S, which consists of EuF_2 powder ($n_{\rm scat} = 1.58$) in a polymer disk ($n_{\rm med} = 1.505$). Forwardly scattered light is collected by a lightguide F2 (NA = 0.7) and detected by a silicon photodiode (PD). The magnetic field is applied perpendicular to the plane of the drawing.

Figure 1 shows our results for the relative transmission modulation at 2Ω as a function of the square of the magnetic-field amplitude. Both curves, measured for different particle sizes and volume fractions, show a clear quadratic field dependence. This proves that the response is proportional to $B(t)^2$ as predicted in Eq. (3). From the lock-in phase, we deduce that the relative transmission decreases with increasing magnetic field, i.e., $\Delta I/I \propto -B^2$.

For EuF_2 the Verdet constant V is inversely proportional to temperature. Since the other optical constants hardly depend on temperature, the explicit dependence of the transmission modulation on the Verdet constant can be conveniently studied by varying the temperature. Figure 2 shows the relative transmission modulation at 2Ω as a function of the square of the inverse temperature at constant magnetic field. The observed linear dependence confirms the quadratic dependence of the photonic magnetoresistance on V.

Figure 3 shows that the transport mean free path ℓ^* in our samples is inversely proportional to the volume fraction of scatterers f for both scatterer sizes. On the basis of Eq. (3), this implies that the magnetoresistance should become independent of f once we are in the diffusive regime. This is demonstrated in Fig. 4: for high volume fractions the observed relative transmission modulation is independent of f. For low volume fractions, the mean free path is no longer small compared to the sample thickness, and the diffusion theory approach in Eq. (3) should break down. Experimentally, we see that at low concentrations the relative transmission modulation is proportional to the concentration, which is consistent with what may be anticipated from low order scattering events. We further note that the ratio of the saturated transmission modulation of the 2 μm particles to that of the 0.4 μm particles is roughly 2.2, which equals the square of the ratio of the mean free paths, deduced from the slopes in Fig. 3. This suggests that the proportionality factor of Eq. (3) does not depend on particle size in this size regime.

In the ideal case that all the transmitted diffuse light would have been captured and that the incident light would have been a genuine point source near the boundary, the diffusion equation relates the transmission modulation directly to the diffusion modulation, according to $\Delta I/I = \Delta D_\perp B^2/D_0$. Although our experiment does not rigorously match these ideal conditions, we shall adopt $\Delta D_\perp/D_0 \approx -3 \times 10^{-5}\ \mathrm{T}^{-2}$ as a reasonable quantitative estimate for the 0.4 μm particles and $\Delta D_\perp/D_0 \approx -7 \times 10^{-5}\ \mathrm{T}^{-2}$ for the 2 μm particles at 105 K.

Having confirmed the basic features of the theory in Ref. [3], we now turn to a quantitative comparison. Unfortunately, such a comparison is hampered by several uncertainties. First, we do not have a reliable estimate of the systematic errors induced by the *idealization* of our optical configuration and that the transport mean free path can be deduced from $T_{\mathrm{all}} = 1.6\ell^*/L$. Furthermore, it is important to note that the theory in Ref. [3] is valid for monodisperse Rayleigh scatterers. Electron-microscope pictures revealed that the particles in our experiment are not Rayleigh scatterers, not monodisperse and nonspherical. The estimated (mean) radius of the samples with the small particles, $a \approx 0.2\ \mu$m, which puts them in the Rayleigh-Gans scattering regime [14], since $|n_{\mathrm{scat}} - n_{\mathrm{med}}| \ll 1$ and $2(m-1)x \approx 0.4$, where $x \equiv n_{\mathrm{med}}ka$ is the so-called size parameter and $m \equiv n_{\mathrm{scat}}/n_{\mathrm{med}}$. Rayleigh-Gans theory for spheres [14] gives for the transport mean free path $1/\ell^* = \frac{8}{9} f(m-1)^2\phi(x)/a$. Here $\phi(x)$ is function that can be approximated by x^4 for $x < 1$ and by $\frac{27}{16}\ln 2x$ for $x > 4$. Using the measured values $m = 1.051$ and

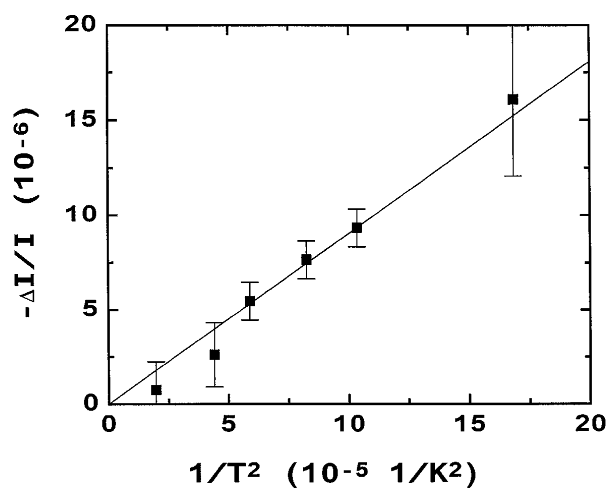

FIG. 2. Transmission modulation at 2Ω versus the inverse square of the temperature. Volume fraction of the scatterers with average particle diameter of 0.4 μm is 0.17. Magnetic field strength is 0.36 T. Solid line is a linear fit to the data points.

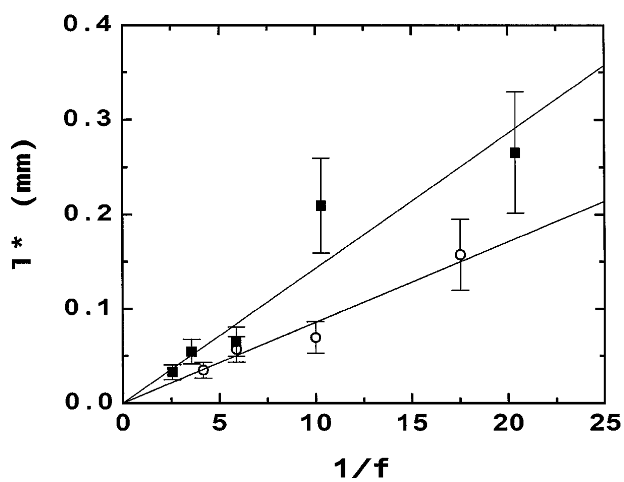

FIG. 3. Transport mean free path versus inverse volume fraction of scatterers. Full symbols denote samples with an average particle diameter of 2 μm and open symbols denote an average particle diameter of 0.4 μm.

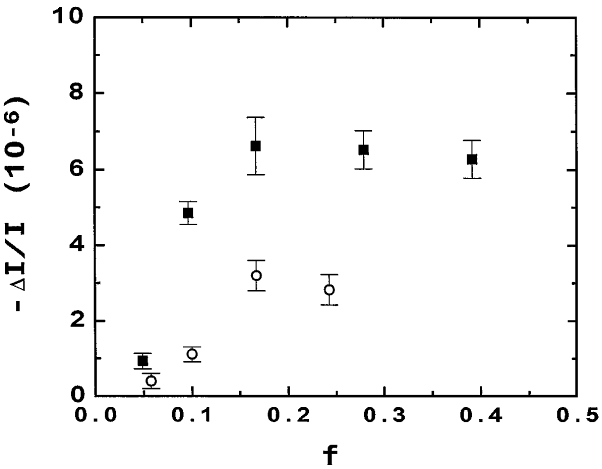

FIG. 4. Transmission modulation at 2Ω versus the volume fraction of scatterers. Temperature is 105 K, magnetic-field strength is 0.36 T. Full symbols represent samples with an average particle diameter of 2 μm and open symbols denote an average particle diameter of 0.4 μm.

$a = 0.2$ μm one predicts $f\ell^* = 25$ μm. The difference with our measurement $f\ell^* \approx 8.3$ μm (Fig. 2) is sufficiently small to be attributed to systematic errors and polydispersity. By the absence of a Rayleigh-Gans theory for magnetic field effects on light scattering, we shall compare our experimental results with the prediction for monodisperse Rayleigh particles,

$$\frac{\Delta D_\perp}{D_0} = -\frac{972}{5} m^2 \frac{(Va)^2}{(m^2 - 1)^4 x^8}\,. \tag{4}$$

For the observed range of radii $0.15 < a < 0.25$ μm, Eq. (4) provides the theoretical range -2×10^{-6} T$^{-2} > \Delta D_\perp / D_0 > -4 \times 10^{-5}$ T^{-2}, which agrees in sign and order of magnitude with the experimental value $\Delta D_\perp / D_0 \approx -3 \times 10^{-5}$ T^{-2} at 105 K.

The 2 μm scatterers are no longer in the Rayleigh-Gans regime and a prediction on the basis of Rayleigh scattering theory is even less justifiable. Empirically, the scaling relation (3) is found to apply. The appropriate theoretical investigation of the Mie problem in combination with an external magnetic field is now underway. This study should also be able to reveal the importance of Cotton-Mouton birefringence and magnetostriction in the magnetoresistance for samples with Mie scatterers.

In conclusion, we have for the first time observed photonic magnetoresistance in scattering media. The relative transmission modulation, measured perpendicular to the magnetic field, is found to be proportional to the square of the field strength and to the square of the Verdet constant of the scatterers. In the diffusive regime, it is independent of the concentration of the scatterers. All our results are in qualitative agreement with recent theoretical predictions. Quantitative agreement with the Rayleigh theory has been established for samples with Rayleigh-Gans scatterers.

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