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Magneto-optics with diffuse light

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Abstract

Several novel magneto-optical effects will be summarized that show up for multiple scattering in a magnetic field. © 2000 Elsevier Science B.V. All rights reserved.

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1. Introduction

Analogies between light and electrons have obtained a lot of attention in the last 15 years of research. In particular, the regime of multiple scattering looks so universal that known differences between light and electrons, such as charge, spin and mass (i.e. different dispersion law) seem to disappear. The discovery of coherent backscattering of light in the 1980s (for a complete recent bibliography see Ref. [1]) was stimulated enormously by supposed analogies with weak localization of electrons [2,3]. In weak localization studies, interference is conveniently controlled externally by temperature or magnetic fields. Magneto-optics with diffuse light may thus provide deeper insight into how interference modifies radiative transfer.

Sometimes, the supposed analogies between the equations of motion for light and electrons turned out incomplete. One example is the exact description of the velocity of diffuse waves v_E for which a “simple” application of the analogy gave the wrong answer [4]. Sometimes, the analogies reach farther than one would have believed. The absence of “photon charge” seems to rule out phenomena such as Hall effect and magneto-resistance, well

established for electrons in an ohmic conductor. “photon-Hall effect” (PHE) [5,6] and “photon magneto-resistance” (PMR) [7,8] have been predicted and observed by us for diffuse waves, in a way surprisingly “analogous” to electrons.

It is well known that light propagation can be influenced by an external magnetic field. The effect of a static magnetic field \mathbf{B} on the optical properties of a homogeneous and isotropic medium is described by the refractive index tensor $n_{ij}(\mathbf{B})$ which can contain only three different terms [8],

$$n_{ij}(\mathbf{B}) = (m + MB^2)\delta_{ij} + i\frac{V}{k}\varepsilon_{ijk}B_k + CB_iB_j. \quad (1)$$

Here, k is the vacuum wave number, and ε_{ijk} the Lévi-Civita tensor density. The material constants M , V and C describe magnetostriction, magnetic circular birefringence (the Faraday effect) and magnetic linear birefringence (the Cotton-Mouton effect). Symmetry arguments impose the Faraday effect to be odd in the applied magnetic field. The Verdet constant V has the symmetry of “charge” in time-reversal, parity and charge conjugation operations [1], and could for that reason be called photon charge.

In a homogeneous medium, the Faraday effect generates different indices of refraction $n \pm V(\mathbf{B} \cdot \mathbf{k})/k^2$ for different circular polarizations. What is perhaps less well known is that magnetic fields actually deflect light. If V is real-valued, the group velocity can easily be

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seen to be,

$$\frac{\partial\omega}{\partial\mathbf{k}} = \frac{c_0}{m} \left(\hat{\mathbf{k}} \pm \frac{V}{m} \mathbf{B} \right). \quad (2)$$

Bending of light occurs in the direction of the magnetic field, and not in the magneto-transverse direction $\mathbf{k} \times \mathbf{B}$. Although implicitly predicted by Landau et al. [9], the observation of magneto-bending of light was reported by us only 2 years ago [10]. We explicitly checked the non-existence of magneto-transverse deflection in homogeneous media, in spite of other claims [11].

Calculations involving magneto-optical effects in inhomogeneous media rapidly become technical, even when the scatterers are assumed to be small [12,13]. Fortunately, qualitative results can be obtained from symmetry arguments. Well known is that \mathbf{B} is a pseudo-vector, changing sign under time-reversal but not under mirror reflection. As a result, the Faraday effect modifies reciprocity in light propagation. Reciprocity is at the basis of coherent backscattering, which should severely be affected by the presence of a magnetic field. This was shown experimentally by Erbacher et al. [14,15] using $B < 25T$. Broken time-reversal also affects transport coefficients. The coefficient relevant for multiple scattering of waves is the diffusion “constant”, relating macroscopic energy current to the gradient of wave energy. This coefficient is in general real-valued second-rank tensor, although until three years ago no anisotropic diffusion constants in light scattering had been reported. At the time of writing, only light in nematic liquid crystals [16] and light in a magnetic field [6–8] have been seen to diffuse anisotropically. Onsager’s relations impose that $D_{ij}(\mathbf{B}) = D_{ji}(-\mathbf{B})$. Hence,

$$D_{ij}(\mathbf{B}) = [D_0 + B^2 D_\perp] \delta_{ij} + D_H \epsilon_{ijk} B_k + [D_\parallel - D_\perp] B_i B_j. \quad (3)$$

The term involving D_H creates a current perpendicular to the original flow of energy, and will be called the *photon Hall effect* (PHE). Note that this contribution must be linear in \mathbf{B} , a crucial notion for its experimental verification. Parallel to electronic language, the diffusion constants $D_{\perp,\parallel}$ will be called photon magneto-resistance (PMR).

2. Experiments

The challenge is to observe the three diffusion constants, quantify them and to carry out a comparison to microscopic theories. Problem is that the effects are small, typically $D_H/D_0 \sim 10^{-5}/T$ and $D_{\perp,\parallel}/D_0 \sim 10^{-4}/T^2$. This required us to use oscillating magnetic fields (with frequencies typically around 10 Hz) and phase-sensitive detection. Detailed discussion of their experimental evidence can be found in Refs. [6–8]. Sam-

ples contain micron-sized magneto-active particles made of diamagnetic (Al_2O_3 , TiO_2) or paramagnetic materials (CeF_3 , EuF_2). Care was taken to avoid absorption bands so to favor multiple scattering. Controllable parameters are of average particle size, particle species and concentration. External variables are magnetic field and temperature. The magneto-optical effects occur inside the particle. The inverse case, as well the inclusion of absorption are now also under study.

Charge symmetry imposes the PHE to be proportional to the “photon charge” V just like the electronic Hall effect is proportional to the charge q of the current carriers. This leads to a first severe test for the PHE, ruling out most other mechanisms. As a rule of the thumb, diamagnetic materials have $V > 0$ and behave like “holes” whereas paramagnetic materials have a negative Faraday rotation ($V < 0$) and should behave like electrons. It has been demonstrated, both theoretically and experimentally [5,6] that D_H is proportional to V , including its sign. The PMR varies as $D_\perp \sim V^2$ and is independent of sign. For paramagnetic materials, V is known to be inversely proportional to the temperature, allowing a check of the relations $D_H \sim V$ and $D_\perp \sim V^2$ without changing the sample.

3. Magneto-Mie scattering

The first theoretical step to be taken is to relate the diffusion constants D_H and $D_{\perp,\parallel}$ to properties of one scatterer, just like the usual expressions $D_0 = \frac{1}{3} v_E \ell^*$ and $\ell^* = 1/n\sigma(1 - \langle \cos\theta \rangle)$ relate diffusion constant D_0 and transport mean free path ℓ^* to the differential cross-section of one particle. The last relation is expected to hold in the dilute regime only, in which case scattering from one particle is the building block for multiple scattering.

To this end, we must first understand how the optical cross-section of one particle is modified in the presence of a magnetic field. A long time ago already, Ford and Werner [17] made an extensive study of magneto-Mie scattering, but did not discuss PHE. In order to do this, we have concentrated on a perturbation theory linear in the magnetic field similar to the standard treatment of the Zeeman effect in atomic orbitals. The first order magneto-optical change in cross-section $d\sigma/d\Omega(\mathbf{k} \rightarrow \mathbf{k}')$ can be guessed from symmetry arguments. Let us consider a Mie sphere made of a dielectric constant given by Eq. (1). Being a scalar, the magneto cross-section linear in \mathbf{B} must be proportional to either $\mathbf{k} \cdot \mathbf{B}$, $\mathbf{k}' \cdot \mathbf{B}$ or $\det(\mathbf{k}, \mathbf{k}', \mathbf{B})$. The first two options are parity forbidden since they change sign under a parity operation. Thus,

$$\frac{1}{\sigma_{\text{tot}}} \frac{d\sigma}{d\Omega}(\mathbf{k} \rightarrow \mathbf{k}'; \mathbf{B}) = F_0(\theta) + F_1(\theta) \det(\mathbf{k}, \mathbf{k}', \mathbf{B}). \quad (4)$$

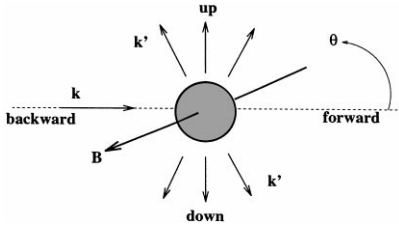


Fig. 1. Scattering geometry for magneto transverse scattering from a Mie sphere.

This cross-section also obeys the reciprocity principle $d\sigma/d\Omega(\mathbf{k} \rightarrow \mathbf{k}'; \mathbf{B}) = d\sigma/d\Omega(-\mathbf{k}' \rightarrow -\mathbf{k}; -\mathbf{B})$. $F_0(\theta)$ is the phase function of the conventional Mie problem [18] and — by rotational symmetry — only dependent on the angle θ between \mathbf{k} and \mathbf{k}' . For the same reason, F_1 depends on the angle θ only. In Ref. [20] we have developed a method to calculate $F_1(\theta)$. For a Rayleigh scatterer it can easily be deduced that $F_1(\theta) \sim (V/k)\cos\theta$.

It is well known that for applications in multiple scattering the anisotropy of the cross-section is important [19]. For Mie spheres this anisotropy is quantified by the “anisotropy factor” $\langle \cos\theta \rangle$, which is $\cos\theta$ averaged over $F_0(\theta)$. It discriminates forward ($\cos\theta > 0$) from backward ($\cos\theta < 0$) scattering. In magneto Mie scattering a second anisotropy shows up that discriminates “upward” from “downward” scattering (Fig. 1). If the magnetic field is perpendicular to both incident and outgoing wave vector, Eq. (4) predicts a difference between upward and downward flux, both defined with respect to the magneto-transverse direction $\mathbf{k} \times \mathbf{B}$. As is the case with $\langle \cos\theta \rangle$, this anisotropy does not survive the θ integral in the case of one Rayleigh scatterer. A Mie sphere with finite size is needed. The magneto-anisotropy η can be quantified exactly as the normalized difference between total flux upwards and total flux downwards. An easy calculation yields [20],

$$\eta \equiv \frac{2\pi}{\sigma_{\text{tot}}} \int_0^\pi d\theta \sin^3\theta F_1(\theta). \quad (5)$$

Note that the linear magneto-cross-section F_1 does not contribute to the total cross-section σ_{tot} . The PHE for one Mie sphere can be defined as a nonzero value for η .

4. Magneto-diffusion of light

With the scattering cross-section of one sphere at our disposal, we can now calculate the diffusion constants D_H , D_\perp and D_\parallel defined in Eq. (3) and associated to magneto-diffusion. For the magneto-resistance ($D_{\perp,\parallel}$) this has so far only been done using Rayleigh scatterers [13]. It was shown that the dominant contribution takes the

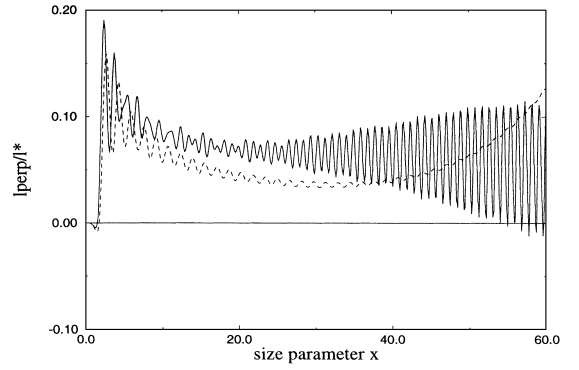


Fig. 2. Ratio D_H/D_0 of magneto transverse diffusion to isotropic diffusion, in units of the dimensionless parameter $2\pi V B \ell^*/k$, as a function of size parameter $x = ka$. Solid line corresponds to Mie spheres with index of refraction $m = 1.28$, dashed line is the Rayleigh-Gans approximation, valid as $m \rightarrow 1$ [20].

form

$$\frac{B^2 D_{\perp,\parallel}}{D_0} \propto (fV B \ell^*)^2. \quad (6)$$

Here, f is the volume fraction of the magneto-active scatterers. The product fV is identified as an “effective-medium” estimate for the Verdet constant, so that the product $fV B \ell^*$ denotes a typical Faraday rotation of the polarization vector accumulated between two collisions. The same parameter was seen to determine the suppression of coherent backscattering [14,15]. Eq. (6) is reminiscent of the classical formula $\Delta\sigma/\sigma \propto (\omega_c \tau)^2$ for the normal electronic magneto-conductance, with ω_c the cyclotron frequency and τ the mean free time [21]. In our experiments $fV B \ell^* \approx 10^{-2}$ which roughly explains the observed order of magnitude $D_{\perp,\parallel}/D_0 \approx -10^{-5}/T^2$ on a sample of EuF_2 scatterers in the Rayleigh-Gans regime [7,8]. The negative sign is consistent with the theory for Rayleigh scatterers. The theoretical prediction $D_\parallel/D_\perp = \frac{1}{2}$ for Rayleigh scatterers has so far not been verified.

The theory for the transverse diffusion constant D_H has recently been generalized to real Mie scatterers [22]. The outcome is a surprisingly simple relation between D_H and the magneto-transverse anisotropy parameter η defined in Eq. (5),

$$\frac{D_H}{D_0} = \frac{\eta}{1 - \langle \cos\theta \rangle}. \quad (7)$$

Here $\langle \cos\theta \rangle$ is the conventional anisotropy factor in Mie scattering. The ratio D_0/D_H is called the Hall coefficient R_H in metals, and is independent of the impurity concentration, here the concentration of Mie spheres. Eq. (7) states that the macroscopic PHE of the inhomogeneous medium is directly proportional to PHE η of one single

Mie scatterer in the medium. The factor of proportionality can become large for large spheres, and explains why the PHE is larger in multiple scattering than in single scattering, albeit with the same sign, as observed in the experiments [6]. Relation (7) reproduces sign and order of magnitude of the observed PHE. It is deduced from Fig. 2 that large size parameters can have an anomalous sign of the PHE, i.e. different from the sign of the “photon charge” V . As the mismatch $m \rightarrow 1$ — the so-called Rayleigh–Gans regime [18] — D_H/D_0 approaches a universal curve, only dependent on size parameter x .

5. What more?

Several phenomena will be investigated in the future. They all focus on the specific role a magnetic field can play to control interference phenomena.

Our first challenge is the observation of weak localization of light, i.e. the positive magneto-conductance [2]. It is caused by the suppression of destructive interference mechanism in the diffusion constant. A back-of-the-envelope calculation yields,

$$\frac{\Delta D}{D_0} \approx \frac{2\pi|fVB|}{k^2\ell}. \quad (8)$$

This means that a small mean free path ℓ favors weak localization, as opposed to the “normal” magneto-conductance given in Eq. (6). Observation has so far not been possible. One complication is the unexpectedly large normal magneto-conductance, which is also positive.

What about coherent backscattering in a magnetic field? Experiments have shown its suppression in a magnetic field [14,15], in agreement with theoretical predictions [12]. But there is more. The line shape of the cone is predicted to depend on the direction of the magnetic field [13], so far not investigated experimentally. Furthermore, triggered by recent observations [23], we have verified that the maximum of the cone is shifted away from exact backscattering. A displacement is seen in the helicity-conserving (+ +) channel along the magnetic field \mathbf{B} , and is of order $V B \ell$. Its origin is the bending of light inside the surface layer of depth ℓ , given in Eq. (2). A second displacement occurs in the magneto-transverse

direction $\mathbf{k} \times \mathbf{B}$. Its origin is again the magneto-transverse scattering from one sphere. Like the PHE, this shift is independent of polarization. Future work will address these effects in more detail.

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