Transverse Diffusion of Light in Faraday-Active Media

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A new effect is considered in multiple scattering of light in a disordered dielectric medium exposed to an external magnetic field: a transverse diffusive current. This current is proportional to the imaginary, antisymmetric part of the dielectric function of the effective medium.

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In this Letter I address the possibility of a transverse diffusion flow of light propagating in a random dielectric medium which is Faraday active. A transverse current is directed perpendicular to both an externally applied magnetic field and the electromagnetic energy density gradient. A transverse Hall current is well understood in the solid state and magnetized plasmas [1] where it is due to the Lorentz force (the normal Hall effect). It can also show up without such a clear physical origin, as in the Beenakker-Senftleben (BS) effect [2]. In the BS effect one measures the thermal conductivity of a paramagnetic or even diamagnetic gas exposed to a magnetic field and a current perpendicular to both temperature gradient and magnetic field is known to exist [3]. In this case it originates from the anisotropic nature of the scattering cross section of the colliding molecules. Whereas the Hall effect is the most important physical tool to determine the effective charge of the current carriers, the BS effect is a unique tool to deduce the Landé g factor of the molecules.

The combination of a magnetic field and multiple light scattering has achieved a lot of attention in recent years, theoretically [4,5], experimentally [6,7], and numerically [8]. From this work it has become clear that the breaking of time-reversal symmetry induced by the Faraday rotation in a magnetic field destroys coherent backscattering of light. This phenomenon is quite similar to the weak localization of electrons giving rise to a negative magnetoresistance [9]. These recent advancements pose the more general question whether (the direction of) the magnetic field plays any role in the diffusive domain of multiple light scattering.

In what follows I consider a rather simplistic model for light scattering from dielectric objects exposed to a magnetic field $\mathbf{B}$. In this model light is scattered from a collection of pointlike particles with dielectric tensor [10]

$$\boldsymbol{\varepsilon}(\mathbf{B}) = m^2\mathbf{I} + \frac{2mV_0}{k}\boldsymbol{\Phi}, \qquad (1)$$

consisting of the normal isotropic part and an antisymmetric part signifying Faraday rotation. The second-rank tensor $\Phi_{ij} \equiv i\epsilon_{ijk}B_k$ is Hermitian. This guarantees long-range diffusion in multiple scattering. As $\boldsymbol{\Phi} \neq \overline{\boldsymbol{\Phi}} = -\boldsymbol{\Phi}$ time-reversal symmetry is broken; V_0 is the Verdet constant determining the Faraday rotation per unit length per unit of magnetic field; $\omega \equiv kc_0$ is the frequency of the light. Alternatively, one can make the medium Faraday active, and not the scatterers. Both cases are relevant experimentally [6].

The first part of the calculation concerns the effective medium represented by the Dyson Green function $\mathbf{G}(\omega, \mathbf{p}, \mathbf{B})$ at momentum $\mathbf{p}$ and the Dyson self-energy $\boldsymbol{\Sigma}(\omega, \mathbf{B})$. The latter—here also a second-rank tensor—is known to be given by $n\mathbf{t}(\omega, \mathbf{B})$ [11] in the dilute regime, with n the number density of the particles and $\mathbf{t}(\omega, \mathbf{B})$ their scattering amplitude given by the Born series. For pointlike objects these can be summed analytically [12], also for the anisotropic dielectric tensor (1). The result is that

$$\boldsymbol{\Sigma}(\omega, \mathbf{B}) = n(t_0\mathbf{I} - \mu t_0^2\boldsymbol{\Phi}) + \mathcal{O}(\mathbf{B}^2, n^2). \qquad (2)$$

I introduced a material parameter $\mu = 2mV_0/vk^3(m^2 - 1)^2$ in which v is a volume associated with our scatterer. The symmetric part (ε_s) and antisymmetric part (ε_a) of the dielectric constant associated with the effective medium are defined as $\varepsilon_s\mathbf{I} + \varepsilon_a\boldsymbol{\Phi} \equiv \mathbf{I} - \boldsymbol{\Sigma}/k^2$. The parameter t_0 is the scattering amplitude of a radiating dipole with eigenfrequency ω_0,

$$t_0(\omega) = \frac{-4\pi\Gamma\omega^2}{\omega_0^2 - \omega^2 - \frac{2}{3}i\Gamma\omega^3/c_0}, \qquad (3)$$

and here to be regarded as a simplistic model for a Mie sphere. At low frequencies the model coincides with the exact Mie solution. Perhaps somewhat surprisingly we infer that the effective medium is Faraday active despite the fact that the rotation occurs in pointlike scatterers. Furthermore, I would like to draw attention to the fact that the antisymmetric part ε_a of the dielectric constant has also achieved an imaginary component. In view of the common definition of "complex index of refraction" [13] this imaginary part can be interpreted as a Hall conductivity at optical frequencies. In optical language it mimics an "antisymmetric" extinction mean free path.

The Dyson Green function is $\mathbf{G}(\omega, \mathbf{p}, \mathbf{B}) = [k^2 - \mathbf{p}^2\boldsymbol{\Delta}_\mathbf{p} - \boldsymbol{\Sigma}(\omega, \%\mathbf{B})]^{-1}$ and can be expanded as

$$\mathbf{G}(\mathbf{p}, \mathbf{B}) = \frac{1}{\varepsilon_s k^2 - p^2}\left(1 - \frac{\mathbf{pp}}{\varepsilon_s k^2}\right) + \frac{\varepsilon_a k^2}{(\varepsilon_s k^2 - p^2)^2}\mathbf{L}(\omega, \mathbf{p}, \mathbf{B}) + \mathcal{O}(\mathbf{B}^2), \qquad (4)$$

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in which $\mathbf{L}(\omega, \mathbf{p}, \mathbf{B}) \equiv -\mathbf{\Phi} + \{(\mathbf{\Phi}\cdot\mathbf{p})\mathbf{p} - \mathbf{p}(\mathbf{\Phi}\cdot\mathbf{p})\}/\varepsilon_s k^2$ is an antisymmetric transverse tensor taking care of the Faraday rotation.

The diffusion tensor is a property of the average intensity. We search for the familiar constitutive equation between current and density gradient, which in Fourier space reads

$$\mathbf{J}(\mathbf{q},\mathbf{B}) = -\mathbf{D}(\mathbf{B})\cdot i\mathbf{q}\rho(\mathbf{q},\mathbf{B}). \quad (5)$$

Here $\mathbf{D}(\mathbf{B})$ is the diffusion tensor of rank 2. By the Onsager relation its transverse component must be an antisymmetric tensor, odd in the field. I shall focus on terms linear in the field. The influence of a magnetic field on the symmetric part of $\mathbf{D}(\mathbf{B})$ must be of order $\mathbf{B}^2$ and will be obtained elsewhere [14].

The average Poynting vector is given by $\mathbf{J} = c_0\langle \mathbf{E}\times\overline{\mathbf{H}} + \mathbf{H}\times\overline{\mathbf{E}}\rangle$. ($\mathbf{H}$ is the magnetic induction to be distinguished from the externally applied magnetic field $\mathbf{B}$.) By a Maxwell equation for stationary harmonic fields we find $i\omega\mathbf{H} = c_0\nabla\times\mathbf{E}$ so that $H_i = c_0\epsilon_{ijk}p_jE_k/\omega$. The current, averaged over disorder and in Fourier space, can therefore be found according to [15]

$$J_i(\mathbf{q},\mathbf{B}) = \frac{c_0^2}{\omega}\sum_{\mathbf{p}}(2p_i\delta_{nk} - p_n\delta_{ik} - p_k\delta_{in}) \times \langle E_n(\mathbf{p}^+)\overline{E}_k(\mathbf{p}^-)\rangle . \quad (6)$$

Here $\mathbf{p}^{\pm} = \mathbf{p} \pm \frac{1}{2}\mathbf{q}$ and $\sum_{\mathbf{p}} \equiv \int d^3\mathbf{p}/(2\pi)^3$. The radiation energy density is given by [16]

$$\rho(\mathbf{q},\mathbf{B}) = 2\sum_{\mathbf{p}}\delta_{nk}\langle E_n(\mathbf{p}^+)\overline{E}_k(\mathbf{p}^-)\rangle . \quad (7)$$

I adopted equipartition between magnetic and electric energy.

The correlation function $\langle E_n\overline{E}_k\rangle$ can be obtained by adding outgoing Dyson Green's functions and a source $\mathbf{S}$ to the four-rank Ladder tensor $L_{ijkl}(\omega,\mathbf{q},\mathbf{B})$ according to [5]

$$\langle E_n(\mathbf{p}^+,\mathbf{B})\overline{E}_m(\mathbf{p}^-,\mathbf{B})\rangle = G_{ni}(\mathbf{p}^+,\mathbf{B})G^*_{km}(\mathbf{p}^-,\mathbf{B}) \times L_{ijkl}(\omega,\mathbf{q},\mathbf{B})S_{jl}.$$

In order to calculate the diffusion tensor it suffices to consider the hydrodynamic limit $\mathbf{q}\to\mathbf{0}$. It can be shown from the optical theorem that the eigenfunction of $L_{ijkl}(\mathbf{q},\mathbf{B})$ corresponding to the hydrodynamic eigenvalue proportional to $1/\mathbf{q}^2$ is $\mathbf{\Sigma}(\mathbf{B}) - \mathbf{\Sigma}^*(\mathbf{B}) \equiv \Delta\mathbf{\Sigma}(\mathbf{B})$ [14]. The diffusion tensor therefore follows from the current associated with the diffusive mode $\langle\mathbf{E}\overline{\mathbf{E}}\rangle = (\pi i/2k)\mathbf{G}(\mathbf{B},\mathbf{p}^+)\mathbf{G}^*(\mathbf{B},\mathbf{p}^-)\cdot\Delta\mathbf{\Sigma}(\mathbf{B})$, which conveniently has $\rho(\mathbf{q}) = 1$ (again by application of the optical theorem). Equations (2), (5), and (6) now combine to

$$[\mathbf{D}(\mathbf{B})\cdot\mathbf{q}]_i = c_0\frac{\pi i}{k\ell}\sum_{\mathbf{p}}(2p_i\delta_{nm} - p_n\delta_{im} - p_m\delta_{in}) \times G_{nj}(\mathbf{p}^+,\mathbf{B})G^*_{hm}(\mathbf{p}^-,\mathbf{B})(\delta_{jh} + k\ell\varepsilon_a\Phi_{jh}). \quad (8)$$

Here ℓ is the scattering mean free path in the absence of a magnetic field and given by $\ell = (-n\,\mathrm{Im}\,t_0/k)^{-1}$. The transverse current can be found by expanding the right hand side of Eq. (8) linearly in $\mathbf{q}$ and $\mathbf{B}$. Introducing

$$\delta\mathbf{L}(\mathbf{p},\mathbf{q},\mathbf{B}) \equiv \{(\mathbf{\Phi}\cdot\mathbf{q})\mathbf{p} + (\mathbf{\Phi}\cdot\mathbf{p})\mathbf{q} - \mathbf{p}(\mathbf{\Phi}\cdot\mathbf{q}) - \mathbf{q}(\mathbf{\Phi}\cdot\mathbf{p})\}/\varepsilon_s k^2,$$

we can—using Eq. (4)—collect all terms in the integrand of Eq. (8) linear in the field $\mathbf{B}$. A little algebra gives six terms accompanied by their Hermitian conjugates. Their momentum integrals can be calculated straightforwardly using an integral familiar from speckle calculations [17]. Collecting the various terms yields

$$\mathbf{D}_\perp(\mathbf{B}) = c_0\ell\,\mathrm{Im}\,\varepsilon_a(\boldsymbol{\epsilon}\cdot\mathbf{B}) \times\left[-\frac{1}{6} + \frac{5}{12} + \frac{1}{12} - \frac{1}{24} + \frac{1}{6} - \frac{1}{8}\right] = \frac{1}{3}c_0\ell\,\mathrm{Im}\,\varepsilon_a(\boldsymbol{\epsilon}\cdot\mathbf{B}). \quad (9)$$

This is the final result of this paper, which I shall now discuss.

The transverse diffusion tensor in Eq. (9) does not depend on the number density of the scatterers. Physically, this happens because we have let the Faraday rotation occur inside the scatterers, so that $\mathrm{Im}\,\varepsilon_a \sim n$. This conclusion might not be valid when the rotation occurs in the environment. The conclusion does *not* imply that the transverse current persists for very small number densities: In a finite medium with typical size L the diffusion approximation adopted here is valid only when $L \gg \ell \sim 1/n$, that is in the multiple scattering domain. Furthermore, the value of $D_\perp$ relative to the normal Boltzmann diffusion constant $D_{\mathrm{B}} = c_0\ell/3$ equals the *imaginary part of the antisymmetric* component of the dielectric constant in the effective medium. In this calculation it turns out that the "Hall conductance" at optical frequencies—identified earlier for the effective medium—induces a transverse current in multiple light scattering. This is an important new feature, that can so far only be demonstrated explicitly with our simple model. Hopefully, future experiments and more sophisticated (finite-size) models will establish a broader validity. Concerning the *real* part of the antisymmetric dielectric function, recent experimental work [6,7] has already demonstrated that theoretical predictions with Rayleigh scattering models [5] work qualitatively very well. The real part of ε_a gives rise to Faraday rotation, and suppresses coherent backscattering. In this way, both real and imaginary parts of the antisymmetric dielectric tensor ε_a find their place in the transport of the average intensity.

At low frequencies (the genuine Rayleigh scattering regime) the transport mean free path $\ell_\perp$ associated with transverse diffusion takes the form

$$\ell_\perp \equiv \frac{3|\mathbf{D}_\perp|}{c_0} = k^{-1}\frac{12m}{(m^2-1)(m^2+2)}\frac{V_0B}{k}. \quad (10)$$

Inserting $\mathbf{B} = 20$ T, $m^2 = 1.15$, and $V_0 = 500°/\text{mm T}$ (achievable at low temperatures) provides us with $\ell_\perp \approx 0.03\ \mu$m at optical frequencies. This is small, but not too small to be detectable, for instance, by photon counting experiments on bottom and top sides of a sample [18].

The direction of the transverse current depends on the sign of the Verdet constant. This parameter is positive for diamagnetic, and negative for paramagnetic materials. An anomalous sign of $\mathbf{D}_\perp$ can arise from multiple scattering, since $\text{Im}\,\varepsilon_a$ changes sign at the resonant frequency ω_0.

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[1] D.B. Melrose, *Plasma Astrophysics: Nonthermal Processes in Diffuse Magnetized Plasmas* (Gordon and Breach, New York, 1980), Vols. 1&2, Chap. 6.

[2] H. Senftleben, Phys. Z. **XXXI**, 961 (1930); J.J.M. Beenakker, G. Scoles, H.F.P. Knaap, and R.M. Jonkman, Phys. Lett. **2**, 5 (1962); Yu. Kagan and L. Maksimov, Sov. Phys. JETP **14**, 604 (1962); F.R. McCourt and R.F. Snider, J. Chem. Phys. **33**, 643 (1967).

[3] H.F.P. Knaap and J.J.M. Beenakker, Physica (Utrecht) **33**, 643 (1967); J. Korving, H. Hulsman, H.F.P. Knaap, and J.J.M. Beenakker, Phys. Lett. **21**, 5 (1966); L.J.F. Hermans, P.H. Fortuin, H.F.P. Knaap, and J.J.M. Beenakker, Phys. Lett. **25A**, 81 (1967).

[4] A.A. Golubenstev, Sov. Phys. JETP **59**, 26 (1984).

[5] F.C. MacKintosh and S. John, Phys. Rev. B **37**, 1884 (1988).

[6] F. Erbacher, R. Lenke, and G. Maret, Europhys. Lett. **21**, 551 (1993).

[7] R. Lenke, Ph.D thesis, Université Joseph Fourier, Grenoble, 1994.

[8] A.S. Martinez and R. Maynard, Phys. Rev. B **50**, 3714 (1994).

[9] S. Hikami, A.I. Larkin, and T. Nagaoka, Prog. Theor. Phys. **63**, 707 (1980).

[10] L.D. Landau, E.M. Lifshits, and L.P. Pitaevskii, *Electrodynamics of Continuous Media* (Pergamon, Oxford, 1984), Sec. 97.

[11] U. Frisch, *Probabilistic Methods in Applied Mathematics,* edited by A.T. Bharucha-Reid (Academic, New York, 1968), Vols. I&II.

[12] Th.M. Nieuwenhuizen, A. Lagendijk, and B.A. van Tiggelen, Phys. Lett. A **169**, 191 (1992).

[13] H.C. van de Hulst, *Light Scattering by Small Particles* (Dover, New York, 1981).

[14] B.A. van Tiggelen, R. Maynard, and Th.M. Nieuwenhuizen (to be published).

[15] See, e.g., for the electron case D. Vollhardt and P. Wölfle, Phys. Rev. B **22**, 4666 (1980).

[16] B.A. van Tiggelen and A. Lagendijk, Europhys. Lett. **23**, 311 (1993). In the present paper we assume that $v_E = c_0$.

[17] S. Feng and R. Berkovits, Phys. Rep. **238**, 135 (1994), see Eq. (85).

[18] R. Lenke and G. Maret (private communication).