

Transport mean free path for magneto-transverse light diffusion

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Abstract. – We derive an expression for the transport mean free path ℓ_{\perp}^* associated with magneto-transverse light diffusion for a random collection of Faraday-active Mie scatterers. This expression relates the magneto-transverse diffusion in multiple scattering directly to the magneto-transverse scattering of a single scatterer.

Magneto-transverse light diffusion—more popularly known as the “Photonic Hall Effect” (PHE)—has been predicted theoretically some three years ago [1], and has been confirmed experimentally one year later [2]. Phenomenologically, the effect has many similarities to the well-known electronic Hall effect: given a diffusion current \mathbf{J} , the presence of an external and constant magnetic field creates a flow in the “magneto-transverse” direction $q\mathbf{B} \times \mathbf{J}$, with q the charge of the current carriers. By the non-existence of photon charge, the physics of the PHE seems different, and compares perhaps better to the so-called Beenakker-Senftleben effect in dilute gases [3]. The evident driver behind the electronic Hall effect is the Lorentz force acting on a charged particle while colliding with the impurities. According to ref. [1] the PHE finds its origin in the Faraday effect for dielectric scatterers, that slightly changes their scattering amplitude. The charge q is replaced by a material parameter V with the symmetry of charge, quantifying the Faraday effect of the particle’s material. In a homogeneous medium the Faraday effect implies a rotation VB per unit length of the polarization vector of linearly polarized light. Two other magneto-optical effects in multiple scattering, such as the suppression of coherent backscattering in a magnetic field [4, 5], and “Photonic Magneto-Resistance” [6] are known to exist, and basically originate from the same Faraday effect.

In an isotropic medium, Fick’s phenomenological law relates the diffusion current to the energy-density gradient $\nabla\rho$ according to $\mathbf{J} = -D_0\nabla\rho$ [7]. D_0 is the conventional diffusion constant for radiative transfer and is usually related to the transport mean free path ℓ^* and the transport velocity v_E according to $D = \frac{1}{3}v_E\ell^*$. The velocity is relevant only for dynamical experiments. Fick’s law applies to media much larger than ℓ^* and, when supplied by boundary conditions involving the incident flux, can be solved for the emerging current. In a magnetic field, the diffusion constant must be replaced by a second-rank tensor. By Onsager’s relation $D_{ij}(\mathbf{B}) = D_{ji}(-\mathbf{B})$, the part linear in the external magnetic field must be an antisymmetric

tensor, and Fick's law becomes

$$\mathbf{J} = -\mathbf{D}(\mathbf{B}) \cdot \nabla \rho = -D_0 \nabla \rho - D_{\perp} \mathbf{B} \times \nabla \rho. \quad (1)$$

The term containing D_{\perp} describes a magneto-transverse diffusion current. In analogy to D_0 , we shall define the transport mean free path ℓ_{\perp}^* for magneto-transverse diffusion as $D_{\perp} = \frac{1}{3} v_E \ell_{\perp}^*$. For the electronic Hall effect, ℓ_{\perp}^* would be proportional to the Hall conductivity σ_{xy} , whose sign is determined by the charge of the current carriers. Similarly, ℓ_{\perp}^* of the PHE can have both signs depending on the scattering.

Anisotropy in the scattering cross-section—quantified by the familiar anisotropy factor $\langle \cos \theta \rangle$ [8]—is well known to make the transport mean free path ℓ^* different from the extinction length ℓ [9]. The latter is the average distance between two subsequent scattering events. In this letter we will show that the PHE can be understood as a generalization of this anisotropy factor to magneto light diffusion, which establishes a difference in scattering between “upward” and “downward” directions (with respect to the plane of incident light and magnetic field). To this end we use our solution for the Faraday-active dielectric sphere [10] to relate the PHE in multiple scattering, quantified by ℓ_{\perp}^* , directly to the PHE of one single particle. Although such a link may be physically clear, it is not evident from previous work [1]. A microscopic approach provides both the exact sign as well as the role of anisotropy, which will enable us to conclude this letter with a realistic comparison to reported experiments [2].

The magneto-active dielectric sphere has been discussed by Ford *et al.* [11]. Experiments and symmetry arguments show the PHE to be linear in B . Therefore we have derived a linear perturbation formula for the scattering amplitude $\mathbf{T}_{\mathbf{p}\mathbf{p}'}(\mathbf{B})$ of one magneto-active dielectric sphere [10]. This amplitude relates the scattered field in direction \mathbf{p}' to the incoming plane wave with wave vector \mathbf{p} . The differential cross-section, proportional to the modulus squared of the scattering amplitude, must satisfy the reciprocity relation $d\sigma/d\Omega(\mathbf{p} \rightarrow \mathbf{p}', \mathbf{B}) = d\sigma/d\Omega(-\mathbf{p}' \rightarrow -\mathbf{p}, -\mathbf{B})$. A magneto-cross-section proportional to $(\hat{\mathbf{p}} \cdot \hat{\mathbf{B}})$ or $(\hat{\mathbf{p}}' \cdot \hat{\mathbf{B}})$ is parity-forbidden since \mathbf{B} is a pseudo-vector and \mathbf{p} a vector. Together with the rotational symmetry of the sphere it must have the form

$$\frac{1}{\sigma_{\text{tot}}} \frac{d\sigma}{d\Omega}(\mathbf{p} \rightarrow \mathbf{p}', \mathbf{B}) = F_0(\theta) + \det(\hat{\mathbf{p}}, \hat{\mathbf{p}}', \hat{\mathbf{B}}) F_1(\theta), \quad (2)$$

where $\cos \theta = \hat{\mathbf{p}} \cdot \hat{\mathbf{p}}'$ defines the scattering angle θ , σ_{tot} is the total cross-section and $\det(\mathbf{A}, \mathbf{B}, \mathbf{C}) = \mathbf{A} \cdot (\mathbf{B} \times \mathbf{C})$ the scalar determinant that can be constructed from three vectors. The second term in eq. (2) will be called the magneto-cross-section. For a small, Rayleigh scatterer one finds $F_1(\theta) \sim VB \cos \theta c_0 / \omega$. The antisymmetry of this magneto-cross-section between forward scattering and backscattering causes the PHE to vanish. The Mie solution breaks this symmetry and a PHE was seen to emerge [10].

We will use field techniques developed in refs. [9, 12] to calculate the magneto-transverse transport mean free path for a random collection of identical Faraday-active dielectric spheres. The four-rank tensor $L_{ijkl, \mathbf{p}\mathbf{p}'}(\mathbf{q})$ linearly connects field correlations $\langle E_i(\omega, \mathbf{p} + \mathbf{q}/2) \bar{E}_j(\omega, \mathbf{p} - \mathbf{q}/2) \rangle$ of incident and outgoing fields in space; \mathbf{q} is the Fourier variable of space and \mathbf{p} is the optical wave vector. On long length scales ($\mathbf{q} \rightarrow 0$) and without absorption it takes the diffuse form

$$L_{ijkl, \mathbf{p}\mathbf{p}'}(\mathbf{q}, \mathbf{B}) = \frac{l_{ik}(\mathbf{p}, \mathbf{q}, \mathbf{B}) l_{lj}(-\mathbf{p}', -\mathbf{q}, -\mathbf{B})}{\mathbf{q} \cdot \mathbf{D}(\mathbf{B}) \cdot \mathbf{q}}. \quad (3)$$

The symmetric form of the numerator is imposed by the reciprocity principle. Rigorous transport theory yields [12]

$$\mathbf{l}(\mathbf{p}, \mathbf{q}, \mathbf{B}) = \mathbf{i}[\mathbf{G}(\mathbf{p}, \mathbf{B}) - \mathbf{G}^*(\mathbf{p}, \mathbf{B})] - \mathbf{i}\mathbf{G}(\mathbf{p}, \mathbf{B}) \cdot \mathbf{\Gamma}(\mathbf{p}, \mathbf{q}, \mathbf{B}) \cdot \mathbf{G}^*(\mathbf{p}, \mathbf{B}). \quad (4)$$

$\mathbf{G}(\mathbf{p}, \mathbf{B})$ denotes the Dyson Green's tensor of the ensemble-averaged electric field to be specified later; the asterisk denotes Hermitian conjugation in polarization space. We left out explicit reference to the optical frequency ω .

The tensor $\mathbf{\Gamma}(\mathbf{p}, \mathbf{q}, \mathbf{B})$ is linear in \mathbf{q} . In real space, the wave number \mathbf{q} corresponds to the gradient in Fick's law (1). The exact relation between $\mathbf{\Gamma}$ and the diffusion tensor is [1, 12]

$$\mathbf{D}(\mathbf{B}) \cdot \mathbf{q} = v_E \frac{\pi c_0}{\omega^2} \sum_{\mathbf{p}} \mathbf{p} \text{Tr} \mathbf{G}(\mathbf{p}, \mathbf{B}) \cdot \mathbf{\Gamma}(\mathbf{p}, \mathbf{q}, \mathbf{B}) \cdot \mathbf{G}^*(\mathbf{p}, \mathbf{B}) = v_E \ell \int \frac{d^2 \hat{\mathbf{p}}}{4\pi} \hat{\mathbf{p}} \text{Tr} \mathbf{\Gamma}(\hat{\mathbf{p}}, \mathbf{q}, \mathbf{B}). \quad (5)$$

This shows that only the trace of $\mathbf{\Gamma}$ comes in. The term $\text{Tr} \delta \mathbf{\Gamma}$ linear in \mathbf{B} provides the PHE.

For a low-density n of particles with T-matrix $\mathbf{T}_{\mathbf{p}\mathbf{p}'}(\mathbf{B})$, $\mathbf{\Gamma}$ obeys the Bethe-Salpeter equation [12]

$$\mathbf{\Gamma}(\mathbf{p}, \mathbf{q}, \mathbf{B}) = 2(\mathbf{p} \cdot \mathbf{q}) + n \sum_{\mathbf{p}'} \mathbf{T}_{\mathbf{p}\mathbf{p}'}(\mathbf{B}) \cdot \mathbf{G}(\mathbf{p}', \mathbf{B}) \cdot \mathbf{\Gamma}(\mathbf{p}', \mathbf{q}, \mathbf{B}) \cdot \mathbf{G}^*(\mathbf{p}', \mathbf{B}) \cdot \mathbf{T}_{\mathbf{p}\mathbf{p}'}^*(\mathbf{B}). \quad (6)$$

We shall solve this equation for the magneto-active Mie particle, up to linear contributions in \mathbf{B} . $\mathbf{\Gamma}$ determines the anisotropy in scattering; without magnetic field it can be ascertained that $\mathbf{\Gamma}^0(\mathbf{p}, \mathbf{q}) = 2(\mathbf{p} \cdot \mathbf{q}) / (1 - \langle \cos \theta \rangle)$, with $\langle \cos \theta \rangle$ the anisotropy factor in scattering [8].

Equation (6) shows that $\mathbf{\Gamma}$ is a Hermitian tensor, linear in \mathbf{q} . We will ignore the longitudinal field in eq. (6) that turned out to be of higher order. This makes $\mathbf{\Gamma}$ as well as \mathbf{G} effectively a Hermitian 2×2 matrix. It is convenient to express all such matrices with respect to a helicity base, where $\sigma(\mathbf{p}) = -\sigma(-\mathbf{p}) = \pm 1$ the helicity of a plane wave with wave vector \mathbf{p} . They must be a linear combination of the identity U and the three Pauli spin matrices $\sigma_{x,y,z}$ [13]. If the z -axis is taken along the wave vector \mathbf{p} , the Green's tensor can be written as $G_{\sigma\sigma'}(\mathbf{p}, \mathbf{B}) = G_0(p)U + G_1(p)(\mathbf{B} \cdot \mathbf{p})\sigma_z$, with $G_0(p) = 1/[\omega^2/c_0^2 - p^2 + i\omega/c_0\ell]$ the Dyson Green's function in terms of the extinction length ℓ ; furthermore $G_1(p) \sim G_0^2$.

We separate the magneto-optical term as $\mathbf{\Gamma}(\mathbf{B}) = \mathbf{\Gamma}^0 + \delta \mathbf{\Gamma}(\mathbf{B})$. Mirror-symmetry imposes that $\mathbf{T}_{\mathbf{p}\mathbf{p}'}(\mathbf{B}) = \mathbf{T}_{-\mathbf{p}-\mathbf{p}'}(\mathbf{B})$ so that, by eq. (6), $\mathbf{\Gamma}(\mathbf{p}, \mathbf{q}, \mathbf{B}) = -\mathbf{\Gamma}(-\mathbf{p}, \mathbf{q}, \mathbf{B})$. Finally, this implies

$$\begin{aligned} \delta \Gamma_{\sigma\sigma'}(\mathbf{p}, \mathbf{q}, \hat{\mathbf{B}}) &= a_1(\mathbf{p} \cdot \mathbf{q} \times \hat{\mathbf{B}})U + \sigma_z \left[a_2(\hat{\mathbf{B}} \cdot \mathbf{q}) + a_3(\hat{\mathbf{B}} \cdot \mathbf{p})(\mathbf{p} \cdot \mathbf{q}) \right] + \\ &+ a_4 \left[(\hat{B}_y q_x + \hat{B}_x q_y) \sigma_x + (\hat{B}_y q_y - \hat{B}_x q_x) \sigma_y \right] \end{aligned} \quad (7)$$

in terms of four real-valued coefficients a_n to be determined. They correspond to the four Stokes parameters I, Q, U and V of the diffuse light. The presence of matrices other than the unit matrix in eq. (7) implies that the diffuse radiation (4) becomes polarized in the presence of a magnetic field. By eq. (5), the PHE is determined by $\text{Tr} \delta \mathbf{\Gamma} \sim a_1$, for which eq. (6) provides a closed equation, as we will show now.

The linearization of eq. (6) in the magnetic field generates three coupled contributions: $\delta \mathbf{\Gamma} = \delta \mathbf{\Gamma}_1 + \delta \mathbf{\Gamma}_2 + \delta \mathbf{\Gamma}_3$. The first comes from $\delta \mathbf{\Gamma}(\mathbf{B})$ itself, the second from $\delta \mathbf{T}_{\mathbf{p}\mathbf{p}'}(\mathbf{B})$ and the third from $\delta \mathbf{G}(\mathbf{B})$. The T -matrix of a Mie sphere without magnetic field depends on the scattering angle θ and the azimuthal angle ϕ in a frame where the incident wave vector is directed along the z -axis [8]. With respect to a helicity base, it takes the form

$$\begin{aligned} T_{\sigma\mathbf{p}\sigma'\mathbf{p}'}^0 &= -\frac{2\pi i}{p} e^{-i\sigma\phi} [\bar{S}_1(\theta) + \bar{S}_2(\theta)\sigma(\mathbf{p})\sigma'(\mathbf{p}')] \\ &= -\frac{2\pi i}{p} e^{-i\sigma\phi} [(\bar{S}_1 + \bar{S}_2)U + (\bar{S}_1 - \bar{S}_2)\sigma_x]. \end{aligned} \quad (8)$$

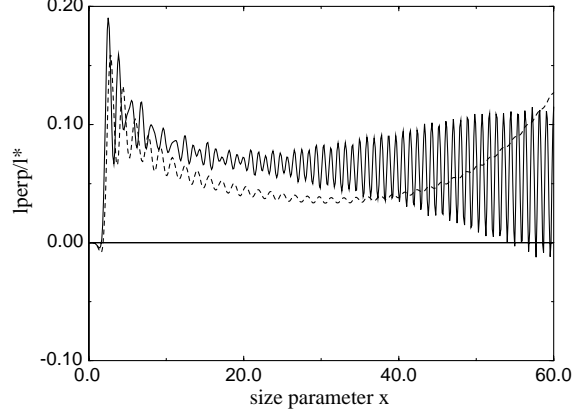


Fig. 1. – Ratio of magneto-transverse transport mean free path and isotropic transport mean free path $\ell_{\perp}^*/\ell^* = (1 - \langle \cos \theta \rangle) a_1$, as a function of the size parameter $x = 2\pi a/\lambda$ of the Mie spheres. The index of refraction $m = 1.128$ of the spheres corresponds to CeF_3 in glycerol. The ripple structure is attributed to resonances in the various partial waves that built up the scattering amplitude for large x . For size parameters $x < 1$ (Rayleigh regime) we find that $\ell_{\perp}/\ell^* \sim x^5$ vanishes rapidly. Dashed line: the same for $m \rightarrow 1$ (Rayleigh-Gans regime), for which a finite value is seen to survive.

The bars denote complex conjugation and stem from our different convention than the one used in ref. [8]. Since $\sigma_x \sigma_y = i\sigma_z$ and $\sigma_z \sigma_x = i\sigma_y$, many terms generated by $\delta\mathbf{\Gamma}$ in eq. (6) contain one of the traceless Pauli spin matrices. Since $\delta G \sim \sigma_z$ it follows that $\text{Tr } \delta\mathbf{\Gamma}_3 = \mathbf{0}$. Since $\sigma_x^2 = U$, one term involving a_4 survives in $\text{Tr } \delta\mathbf{\Gamma}_1$ but is eliminated by the integral over the azimuthal angle ϕ . The first contribution simplifies to

$$\text{Tr } \delta\mathbf{\Gamma}_1(\mathbf{p}, \mathbf{q}, \mathbf{B}) = \int d^2\hat{\mathbf{p}}' F_0(\theta) \text{Tr } \delta\mathbf{\Gamma}(\mathbf{p}', \mathbf{q}, \mathbf{B}) = 2a_1 \langle \cos \theta \rangle (\mathbf{p} \cdot \mathbf{q} \times \hat{\mathbf{B}}) \quad (9)$$

with $F_0(\theta)$ defined in eq. (2). The second equality follows by substituting $\text{Tr } \delta\mathbf{\Gamma}(\mathbf{p}', \mathbf{B}, \mathbf{q}) = 2a_1(\mathbf{p}' \cdot \mathbf{q} \times \hat{\mathbf{B}})$. The contribution $\text{Tr } \delta\mathbf{\Gamma}_2$ is obtained by substituting $\mathbf{\Gamma}^0 = 2(\mathbf{p} \cdot \mathbf{q})/(1 - \langle \cos \theta \rangle)$,

$$\text{Tr } \delta\mathbf{\Gamma}_2(\mathbf{p}, \mathbf{q}, \mathbf{B}) = \frac{2}{1 - \langle \cos \theta \rangle} \int d^2\hat{\mathbf{p}}' (\mathbf{p}' \cdot \mathbf{q}) \det(\hat{\mathbf{p}}, \hat{\mathbf{p}}', \hat{\mathbf{B}}) F_1(\theta), \quad (10)$$

and involves the magneto-cross-section introduced in eq. (2). From $\text{Tr } \delta\mathbf{\Gamma}_3 = 0$, and eq. (7) we have $\text{Tr } \delta\mathbf{\Gamma}_1 = 2a_1(\mathbf{p} \cdot \mathbf{q} \times \hat{\mathbf{B}}) - \text{Tr } \delta\mathbf{\Gamma}_2$. Equations (9) and (10) hold for any \mathbf{q} and it is convenient to choose $\mathbf{q} = \hat{\mathbf{p}} \times \hat{\mathbf{B}}$. This yields the desired equation

$$a_1 = \frac{1}{(1 - \langle \cos \theta \rangle)^2} \int d^2\hat{\mathbf{p}}' \frac{\det^2(\hat{\mathbf{p}}, \hat{\mathbf{p}}', \hat{\mathbf{B}})}{|\hat{\mathbf{p}} \times \hat{\mathbf{B}}|^2} F_1(\theta) = \frac{\pi}{(1 - \langle \cos \theta \rangle)^2} \int_0^\pi d\theta \sin^3 \theta F_1(\theta). \quad (11)$$

The last equality follows upon integration over the azimuthal angle ϕ .

We can now insert our result for $\text{Tr } \mathbf{\Gamma}$ into formula (5) for the diffusion tensor. Since $\text{Tr } \delta\mathbf{\Gamma} = 2a_1 p_i \epsilon_{ijk} q_j \hat{B}_k$, the final result reads

$$D_{ij}(\mathbf{B}) = \frac{1}{3} v_E \frac{\ell}{1 - \langle \cos \theta \rangle} \delta_{ij} + \frac{1}{3} v_E a_1 \ell \epsilon_{ijk} \hat{B}_k. \quad (12)$$

This equation identifies $\ell^* = \ell/(1 - \langle \cos \theta \rangle)$ as the transport mean free path of light in multiple scattering, and $\ell_{\perp}^* \equiv a_1 \ell$ as the transport mean free path for magneto-transverse

light scattering. In ref. [10] we have shown that the integral in eq. (11) equals exactly the normalized PHE of one magneto-active Mie sphere. Equation (12) thus states that the PHE in multiple scattering is directly proportional to the normalized PHE of one single Mie sphere, including the same sign, and amplified by the factor $1/(1 - \langle \cos \theta \rangle)^2$. Note that the magneto-transverse transport mean free path depends even more on the anisotropy factor $\langle \cos \theta \rangle$ than the conventional transport mean free path ℓ^* . Unfortunately, we have no simple explanation for this.

In fig. 1 we show ℓ_{\perp}^*/ℓ^* as a function of the size parameter, for an index of refraction $m = 1.128$, corresponding to CeF_3 in glycerol. Around $x \approx 40$ (radius $2 \mu\text{m}$), we calculate $\ell_{\perp}^*/\ell^* = +0.06 VB\lambda$ which, for $V = -1100 \text{ rad/mT}$ (at temperature $T = 77 \text{ K}$) and vacuum wavelength $\lambda_0 = 0.457 \mu\text{m}$ yields $\ell_{\perp}^*/\ell^* = -2 \cdot 10^{-5}/T$. The experimental value is $\ell_{\perp}^*/\ell^* \approx -1.1 \pm 0.3 \cdot 10^{-5}/T$ for a 10 vol-% suspension [2]. We estimate a systematic error of at least a factor of two in *attributing* values to ℓ_{\perp} and ℓ^* on the basis of Fick's law. Another uncertain factor is the broad size distribution in this sample, which probably washes out the oscillations of ℓ_{\perp}^*/ℓ^* as a function of size parameter x that are seen in fig. 1. Besides CeF_3 , the present theory is also able to reproduce the measured sign and magnitude for the PHE of polydisperse samples containing ZnS , Al_2O_3 , TiO_2 and EuF_2 . The sign can change as a function of x and m and cannot be predicted by a simple argument known to us. The estimated anisotropy factor for our sample equals $\langle \cos \theta \rangle \approx 0.9$. The amplification factor $1/(1 - \langle \cos \theta \rangle)^2$ for the PHE is therefore significant, and improves theoretical predictions made for Rayleigh scatterers considerably [2].

We conclude that the present theory for spheres of arbitrary size is a significant step forward in the qualitative and quantitative understanding of the magneto-transverse light diffusion.

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