Waves Random Media **10** (2000) 367–372. Printed in the UK PII: S0959-7174(00)11429-6

Transport mean free path for magneto-transverse light diffusion: an alternative approach

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Received 31 January 2000, in final form 11 April 2000

Abstract. This paper presents a derivation of the transport mean free path for magneto-transverse light diffusion, $\ell_{\perp}^{*}$, in an arbitrary random mixture of Faraday-active and non-Faraday-active Mie scatterers. This derivation is based on the standard radiative transfer equation. The expression of the transport mean free path obtained previously from the Bethe–Salpeter equation, for the case where only Faraday-active scatterers are present, is recovered. This simpler formulation can include the case of homogeneous mixtures of Faraday-active and non-Faraday-active scatterers.

1. Introduction

Magneto-transverse light diffusion—also known as the 'photonic Hall effect' (PHE)—was predicted theoretically five years ago by Van Tiggelen [1], and was confirmed experimentally one year later by Rikken [2]. This effect is analogous to the electronic Hall effect, well known in semiconductor physics. The evident driver behind the electronic Hall effect is the Lorentz force acting on charged particles. The PHE finds its origin in the Faraday effect, present inside dielectric scatterers, which changes their scattering amplitude slightly. This is the reason for the similarities between the PHE and the so-called Beenakker–Senftleben effect, which concerns the transport coefficients of dilute paramagnetic gases [3].

Motivated by the experimental observation of the PHE, theoretical work was first started on the single scattering of spherical magneto-optical particles. The scattering matrix and the scattering cross section were calculated exactly for a single Faraday-active dielectric sphere using perturbation theory [4]. From this solution, the Stokes parameters, which completely describe the intensity and polarization of the scattered light, were derived [5]. Perturbational methods for the scattering by weakly anisotropic particles were first used by Kuz'min and Babenko for the single scattering case [6].

The PHE in multiple scattering is controlled by a length, which has been defined as the mean free path for magneto-transverse light diffusion, $\ell_{\perp}^{*}$. A simple expression for this length was obtained and successfully compared with experiments using a formulation based on the ladder approximation of the Bethe–Salpeter equation [7]. The experiments investigated the dependence of the PHE on the volume fraction of the scatterers, first on the real part of the dielectric constant of the scatterers [2], and more recently on their imaginary part [8]. However, some parts of the derivation of [7] are rather technical and fail to give a satisfying explanation of the origin of the dependence of $\ell_{\perp}^{*}$ on the anisotropy of the scattering. This paper presents a simpler derivation of $\ell_{\perp}^{*}$, based on the radiative transfer equation, which should clarify this point. The radiative transfer equation is a Boltzmann-type equation which describes the

0959-7174/00/030367+06$30.00

transport of light in multiple light scattering [9, 10]. From the radiative transfer equation, the diffusion equation is derived in an infinite medium when the scattering is not highly anisotropic. In other types of anisotropic materials, such as single-domain nematic liquid crystals, similar methods were developed, using either the Bethe–Salpeter equation [11, 12] or the radiative transfer equation [13].

2. Single scattering

The single scattering of light by one dielectric sphere made of a Faraday-active material embedded in an isotropic medium with no magneto-optical properties is first considered. In a magnetic field, the dielectric constant of the sphere, ϵ_B, is a tensor of rank two. It depends on the distance to the centre of the sphere of radius, R, via the Heaviside function, $\Theta(|\boldsymbol{r}| - R)$, which equals one inside the sphere and zero outside [4],

$$\epsilon_B(\boldsymbol{B}, \boldsymbol{r})_{ij} = \left[(\epsilon_0 - 1)\delta_{ij} + \mathrm{i}\epsilon_F \epsilon_{ijk} \hat{B}_k\right]\Theta(|\boldsymbol{r}| - R) \tag{1}$$

where ϵ_0 is the value of the normal isotropic dielectric constant of the sphere, $\epsilon_F = 2{\epsilon_0}^{1/2} V_0 B/\omega$ is the coupling constant of the Faraday effect, δ and ϵ are, respectively, the Kronecker and Levi-Cevita tensors. The Verdet constant of the Faraday effect is denoted by V_0, B is the amplitude of the magnetic field and ω is the frequency. The intensity of the scattered light, in single scattering, is characterized by the phase function which is also the essential ingredient for the transport theory of light. The phase function, $F(\hat{\boldsymbol{k}}, \hat{\boldsymbol{k}}', \boldsymbol{B})$, is proportional to the differential scattering cross section averaged with respect to incoming and outgoing polarizations. It depends on the direction of the incoming plane wave, $\hat{\boldsymbol{k}}$, on the direction of the scattered wave, $\hat{\boldsymbol{k}}'$, and on the direction of the magnetic field, $\boldsymbol{B}$. The hat above the vectors denotes normalized vectors. For spherical magneto-optical particles and to linear order in the applied magnetic field, this phase function can be written as [4]

$$F(\hat{\boldsymbol{k}}, \hat{\boldsymbol{k}}', \boldsymbol{B}) = F_0(\hat{\boldsymbol{k}}, \hat{\boldsymbol{k}}') + \det(\hat{\boldsymbol{k}}, \hat{\boldsymbol{k}}', \hat{\boldsymbol{B}})\, F_1(\hat{\boldsymbol{k}}, \hat{\boldsymbol{k}}') \tag{2}$$

where $\det(\boldsymbol{A}, \boldsymbol{B}, \boldsymbol{C}) = \boldsymbol{A} \cdot (\boldsymbol{B} \times \boldsymbol{C})$ denotes the scalar determinant. Due to the rotational symmetry of the scatterer, the functions, F_0 and F_1, only depend on the scattering angle, θ, which is the angle between $\hat{\boldsymbol{k}}$ and $\hat{\boldsymbol{k}}'$. The phase function F_1 only depends on the difference in the azimuthal angles associated with $\hat{\boldsymbol{k}}$ and $\hat{\boldsymbol{k}}'$, because of the axial symmetry of the scatterer around the direction of the magnetic field. These symmetry properties simplify considerably the use of the radiative transfer equation [14]. The amplitude of F_1 was found to be proportional to the dimensionless parameter, ϵ_F [4].

The albedo is introduced as the ratio of the scattering cross section over the extinction cross section [15]

$$a = \frac{Q_{\text{scatt}}}{Q_{\text{ext}}}. \tag{3}$$

It is related to the phase function defined in equation (2),

$$a = \int \mathrm{d}\hat{\boldsymbol{k}}\, F(\hat{\boldsymbol{k}}, \hat{\boldsymbol{k}}', \hat{\boldsymbol{B}}) = \int \mathrm{d}\hat{\boldsymbol{k}}\, F_0(\hat{\boldsymbol{k}}, \hat{\boldsymbol{k}}') \tag{4}$$

where the last equality follows from equation (2). Thus in the absence of absorption the phase function is normalized ($a = 1$).

3. Transport in magneto-optical diffuse media

In an isotropic medium much larger than the transport mean free path, ℓ^*, Fick's law relates the diffusion current, $\boldsymbol{J}$, to the energy-density gradient, ∇I, by $\boldsymbol{J} = -D_0 \nabla I$. The conventional diffusion constant for the radiative transfer equation is denoted by D_0 and is usually related to the transport mean free path, ℓ^*, and the transport velocity, v_E, by $D_0 = \frac{1}{3} v_E \ell^*$. In the presence of a magnetic field, the diffusion constant is a second-rank tensor. The part of this tensor linear in the magnetic field is responsible for the PHE, whereas the part quadratic in the magnetic field generates a photonic magneto-resistance, which could also be observed experimentally [16]. By Onsager's relation, $D_{ij}(\boldsymbol{B}) = D_{ji}(-\boldsymbol{B})$, the part linear in the external magnetic field must be an antisymmetric tensor, and Fick's law becomes,

$$\boldsymbol{J} = -\boldsymbol{D}(\boldsymbol{B}) \cdot \nabla I = -D_0 \nabla I - D_\perp \hat{\boldsymbol{B}} \times \nabla I. \tag{5}$$

The term containing $D_\perp$ describes the magneto-transverse diffusion current responsible for the PHE. In analogy to the definition of ℓ^*, the transport mean free path, $\ell_\perp^*$, for magneto-transverse diffusion is defined as $D_\perp = \frac{1}{3} v_E \ell_\perp^*$. For the electronic Hall effect, $\ell_\perp^*$ is proportional to the Hall conductivity, σ_{xy}, the sign of which is determined by the charge of the current carriers. Similarly, a positive $\ell_\perp^*$ means that the PHE has the same sign as the Verdet constant of the scatterers, and a negative $\ell_\perp^*$ means that the PHE has an opposite sign, which is also possible depending on the scattering. This point has been carefully checked experimentally [2].

The phase function introduced above can be used to describe not only the single scattering of light by independently scattering particles but also light scattering by a large collection of such particles in the regime of multiple light scattering. It is assumed that only magneto-optical Mie scatterers are present, and included in a matrix with no magneto-optical properties. A transport equation for a medium comprising randomly distributed spherical particles embedded in an isotropic medium can be written as follows [17]:

$$\frac{\ell}{v_E} \partial_t \mathcal{I}(\boldsymbol{r}, \hat{\boldsymbol{k}}, t) + \ell \, \hat{\boldsymbol{k}} \cdot \nabla \mathcal{I}(\boldsymbol{r}, \hat{\boldsymbol{k}}, t) + \mathcal{I}(\boldsymbol{r}, \hat{\boldsymbol{k}}, t) = \int \mathrm{d}\hat{\boldsymbol{k}}' \, F(\hat{\boldsymbol{k}}, \hat{\boldsymbol{k}}', \boldsymbol{B}) \mathcal{I}(\boldsymbol{r}, \hat{\boldsymbol{k}}', t) \tag{6}$$

where ℓ denotes the elastic mean free path, which is the average distance between two subsequent scattering events. The specific intensity $\mathcal{I}(\boldsymbol{r}, \hat{\boldsymbol{k}}, t)$ is defined as the density of radiation at position $\boldsymbol{r}$, time t, in the direction $\hat{\boldsymbol{k}}$. The specific intensity has the dimension of a monochromatic radiance: energy per unit solid angle, time, wavelength and surface area [18]. When supplemented by appropriate boundary conditions, the radiative transfer equation can be solved for a particular problem. It is useful to introduce the average specific intensity, $I(\boldsymbol{r}, t)$, and the current, $\boldsymbol{J}(\boldsymbol{r}, t)$, which are defined as

$$I(\boldsymbol{r}, t) = \int \mathcal{I}(\boldsymbol{r}, \hat{\boldsymbol{k}}, t) \, \mathrm{d}\hat{\boldsymbol{k}} \qquad \boldsymbol{J}(\boldsymbol{r}, t) = v_E \int \hat{\boldsymbol{k}} \, \mathrm{d}\hat{\boldsymbol{k}} \, \mathcal{I}(\boldsymbol{r}, \hat{\boldsymbol{k}}, t). \tag{7}$$

To simplify the notation, the dependence of the specific intensity or of related quantities upon the magnetic field, $\boldsymbol{B}$, is not explicitly indicated. The integration of equation (6) with respect to $\hat{\boldsymbol{k}}$ leads to the continuity equation

$$\partial_t I(\boldsymbol{r}, t) + \nabla \cdot \boldsymbol{J}(\boldsymbol{r}, t) = -\frac{(1-a) v_E}{\ell} I(\boldsymbol{r}, t) \tag{8}$$

where a denotes the albedo defined in equation (4).

If equation (6) is multiplied by $\hat{\boldsymbol{k}}$ and integrated over the direction $\hat{\boldsymbol{k}}$, the right-hand side of equation (6) will contain two integrals which depend on F_0 and F_1. These integrals are the two dimensionless quantities

$$\langle \cos\theta \rangle = \int \mathrm{d}\hat{\boldsymbol{k}} \, F_0(\hat{\boldsymbol{k}}, \hat{\boldsymbol{k}}') \, \hat{\boldsymbol{k}} \cdot \hat{\boldsymbol{k}}' \tag{9}$$

and

$$A_1 = \int \mathrm{d}\hat{\boldsymbol{k}}\, F_1(\hat{\boldsymbol{k}}, \hat{\boldsymbol{k}}') \det(\hat{\boldsymbol{k}}, \hat{\boldsymbol{k}}', \hat{\boldsymbol{B}})^2. \tag{10}$$

By choosing a system of coordinates linked with $\hat{\boldsymbol{k}}'$, it can be proved quite generally that

$$\int \mathrm{d}\hat{\boldsymbol{k}}\, F(\hat{\boldsymbol{k}}, \hat{\boldsymbol{k}}', \hat{\boldsymbol{B}})\, \hat{\boldsymbol{k}} = \langle\cos\theta\rangle\hat{\boldsymbol{k}}' - A_1\hat{\boldsymbol{B}} \times \hat{\boldsymbol{k}}'. \tag{11}$$

After the integration over $\hat{\boldsymbol{k}}'$ the following equation is obtained:

$$\frac{\ell}{v_E}\,\partial_t \boldsymbol{J} + \boldsymbol{J}\,\left(1 - \langle\cos\theta\rangle\right) + A_1\hat{\boldsymbol{B}} \times \boldsymbol{J} = -\ell v_E\,\nabla\cdot \int \mathrm{d}\hat{\boldsymbol{k}}\, \mathcal{I}(\boldsymbol{r}, \hat{\boldsymbol{k}}, t)\, \hat{\boldsymbol{k}}\hat{\boldsymbol{k}}. \tag{12}$$

If it is assumed that the angular distribution of the specific intensity is almost isotropic, the current is much smaller than the average specific intensity. In that case the following approximation is valid [19]:

$$\mathcal{I}(\boldsymbol{r}, \hat{\boldsymbol{k}}, t) \approx I(\boldsymbol{r}, t) + \frac{3}{v_E}\boldsymbol{J}(\boldsymbol{r}, t)\cdot\hat{\boldsymbol{k}} + \cdots. \tag{13}$$

Substituting this relation into equation (12) and neglecting $\partial_t \boldsymbol{J}$ for processes which are slow with respect to the characteristic time between two scatterings, ℓ/v_E, gives Fick's law [17]:

$$\boldsymbol{J}(\boldsymbol{r}, t) = -\boldsymbol{D}\cdot\nabla I(\boldsymbol{r}, t). \tag{14}$$

The diffusion tensor is given by

$$D(\boldsymbol{B})_{ij} = \tfrac{1}{3} v_E \ell \left[\left(1 - \langle\cos\theta\rangle\right)\delta_{ij} - A_1 \varepsilon_{ijk}\hat{B}_k \right]^{-1}. \tag{15}$$

It is important to note that the magnetic correction to the scattering cross section plays a role similar to the role of the asymmetry factor, $\langle\cos\theta\rangle$, present when no magnetic field is applied. The dependence of $\langle\cos\theta\rangle$ can be obtained from standard Mie theory [15]. Both anisotropy factors, $\langle\cos\theta\rangle$ and A_1, are non-zero provided that the symmetry between forward and backward scattering is broken, which is the case for a finite-size scatterer. This is the reason why the PHE of a Rayleigh scatterer vanishes, whereas the PHE of a Mie scatterer does not [4].

In the case where $A_1 \ll (1 - \langle\cos\theta\rangle)$, an expansion of the bracket gives

$$D_{ij}(\boldsymbol{B}) = \frac{1}{3} v_E \frac{\ell}{1 - \langle\cos\theta\rangle}\delta_{ij} + \frac{1}{3} v_E A_1 \frac{\ell}{(1 - \langle\cos\theta\rangle)^2}\varepsilon_{ijk}\hat{B}_k. \tag{16}$$

This equation identifies $\ell^* = \ell/(1 - \langle\cos\theta\rangle)$ with the transport mean free path, and $\ell^*_\perp = A_1\ell^*/(1 - \langle\cos\theta\rangle)$ as the transport mean free path for magneto-transverse diffusion. The final result of [7] is thus recovered with little effort. The order of magnitude of the ratio $\ell^*_\perp/\ell^*$ is 10^{-5} for an applied magnetic field of 1 T and for samples made of rare-earth materials [2]. The presence in equation (16) of the coefficient, A_1, which is directly connected with the PHE of a single scatterer [4], states that the PHE in multiple scattering is directly proportional to the normalized PHE of a single Mie sphere, is of the same sign and amplified by the factor, $1/(1 - \langle\cos\theta\rangle)^2$. The expansion to first order in the magnetic field involved in equation (16), clarifies the origin of the factor, $1/(1 - \langle\cos\theta\rangle)^2$, which makes $\ell^*_\perp$ even more dependent on the asymmetry factor, $\langle\cos\theta\rangle$ than the transport mean free path, ℓ^*.

Substituting equation (14) into the continuity equation (8) yields the diffusion equation for the density of radiation

$$\partial_t I(\boldsymbol{r}, t) = D_0 \nabla^2 I(\boldsymbol{r}, t) - \frac{(1-a)v_E}{\ell} I(\boldsymbol{r}, t) = D_0 \left[\nabla^2 I(\boldsymbol{r}, t) - \frac{1}{L_a^2} I(\boldsymbol{r}, t) \right]. \tag{17}$$

As expected, the part of the diffusion tensor which is linear in the magnetic field and therefore antisymmetric does not enter the diffusion equation which depends only on the symmetric part, D_0. In addition to the transport mean free path, ℓ^*, equation (17) defines the characteristic length for the absorption in multiple light scattering, L_a

$$L_a = \sqrt{\frac{\ell^* \cdot \ell}{3(1-a)}} = \sqrt{\frac{\ell^* \cdot \ell_{\text{abs}}}{3}} \tag{18}$$

as a function of the characteristic length for the absorption of the coherent beam ℓ_{abs}. Therefore, there is no dependence of L_a on the magnetic field to linear order. As discussed in [20], the diffusion coefficient D_0 does not depend on the absorption cross section, which is proportional to $1 - a$. Equation (16) shows that this property also holds for $\ell_\perp^*$ when the medium is dissipative.

4. Transport in mixtures of Faraday-active and non-Faraday-active spheres

The main result of equation (16) can be extended to the case of a homogeneous mixture of Faraday-active spheres with volume fraction, n_0, and non-Faraday-active spheres with volume fraction, n_1. The phase function of equation (2) is now modified for the case of the mixtures as follows:

$$\begin{aligned} F(\hat{\boldsymbol{k}}, \hat{\boldsymbol{k}}', \hat{\boldsymbol{B}})_{mix} &= \frac{n_0}{n_0 + n_1} F_0(\hat{\boldsymbol{k}}, \hat{\boldsymbol{k}}') + \frac{n_1}{n_0 + n_1} \left[F_0(\hat{\boldsymbol{k}}, \hat{\boldsymbol{k}}') + \det(\hat{\boldsymbol{k}}, \hat{\boldsymbol{k}}', \hat{\boldsymbol{B}})\, F_1(\hat{\boldsymbol{k}}, \hat{\boldsymbol{k}}') \right] \\ &= F_0(\hat{\boldsymbol{k}}, \hat{\boldsymbol{k}}') + \frac{n_1}{n_0 + n_1} \det(\hat{\boldsymbol{k}}, \hat{\boldsymbol{k}}', \hat{\boldsymbol{B}})\, F_1(\hat{\boldsymbol{k}}, \hat{\boldsymbol{k}}'). \end{aligned} \tag{19}$$

Equation (15) can be generalized to

$$D(\boldsymbol{B})_{ij} = \tfrac{1}{3} v_E \ell \left[(1 - \langle \cos\theta \rangle)\delta_{ij} - \frac{n_1}{n_0 + n_1} A_1 \varepsilon_{ijk} \hat{B}_k \right]^{-1}. \tag{20}$$

In the case when the last term of the bracket is small with respect to $1 - \langle \cos\theta \rangle$, the following expression for the ratio of the transport mean free path for magneto-transverse diffusion over the transport mean free path is obtained:

$$\frac{\ell_\perp^*}{\ell^*} = \frac{n_1}{n_0 + n_1} \frac{A_1}{1 - \langle \cos\theta \rangle}. \tag{21}$$

5. Conclusion

In conclusion, the present transport theory for magneto-optical spheres of arbitrary size confirms the results of [7] concerning the magneto-transverse light diffusion obtained from the Bethe–Salpeter equation. This new approach clarifies the origin of the dependence of the transport mean free path for magneto-transverse diffusion on the anisotropy of the scattering. It also shows that the magnetic anisotropy in the scattering cross section plays a similar role as $\langle \cos\theta \rangle$ in the transport mean free path. This derivation can be generalized to the case of mixtures of Faraday-active and non-Faraday-active scatterers.

Acknowledgments

I would like to thank B Van Tiggelen, G Rikken, S Wiebel and J-J Greffet for the many stimulating discussions. This work has been made possible by the Groupement de Recherches POAN.

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