

Measurement of the magneto-optical correlation length in turbid media

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In multiple light scattering media, magnetic field induced circular birefringence (Faraday effect) influences interference effects such as speckle pattern or coherent backscattering. It was predicted that in the diffusive regime the relevant correlation length with respect to the Faraday rotation ℓ_F^* differs, in general, from the transport mean free path ℓ^* . We have experimentally verified the prediction that the ratio ℓ_F^*/ℓ^* equals 2 for Rayleigh scattering and decreases to 1 with increasing scatterer size. We also discuss the influence of the structure factor on ℓ_F^* .

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INTRODUCTION

Magnetic field effects have significantly improved the understanding of electron transport in semiconductors and metals. For example, the observation of a magnetic field induced increase of the conductivity can be explained by a field induced destruction of weak localization. For a detailed description of this phenomenon which is based on the time-reversal symmetry of (quantum) wave paths the reader is referred to [1] and references there.

In the case of electromagnetic wave transport numerous magnetic field effects [2–5] have been discovered in recent years, in particular in the visible range: magnetic field dependent speckle correlation [3], destruction of the coherent backscattering (CB) cone, the so-called precursor of weak localization [2,6,7], and the magneto-optical Hall effect [8]. The observed destruction of weak localization of light suggests the relevance of magneto-optical effects in the investigations of strong localization of electromagnetic waves [9–14].

In some of those experiments [3,6] the magnetic field effect was stronger than initially [2] expected. Therefore it was suggested [6,15] that the correlation length of the magnetic field induced phase shift differs from the transport mean free path ℓ^* . In this work, we experimentally determine the “new” characteristic length ℓ_F^* . This is done in a quantitative way for different size parameters of polystyrene spheres in water. Finally, we discuss the effect of the structure factor on ℓ_F^* in the case of large volume fractions.

In order to fully understand the magnetic field effects mentioned above, one needs a quantitative comprehension of how Faraday rotation operates on the length scale ℓ^* . To this end, a very precise understanding of coherent backscattering in connection with the magnetic field effects is of general importance.

We have studied the length ℓ_F^* by exploiting CB not only because CB is the precursor to weak localization but also because it provides the most precise determination of ℓ_F^* . Nevertheless, the considerations are also valid for magnetic field dependent speckle correlation measurements.

THEORY

CB originates from the constructive interference between each light path and its reversed path. In the exact back-

scattering direction ($\vec{k}_f = -\vec{k}_i$ for final and incident wave vectors) both paths always have exactly the same length, thus leading to a constructive interference enhancement of a factor of 2 in the exact backscattering direction with respect to the “incoherent background.” For more details, see Ref. [16], for example. Out of exact backscattering a phase shift of $(\vec{k}_f + \vec{k}_i) \cdot \vec{\rho}$ is introduced, where $\vec{\rho}$ is the distance vector between the start and end points of a light path on the surface of the sample. This phase shift results in a decrease of the CB enhancement, which decays to the incoherent background at wide angles on average over all light paths. For simplicity, we neglect the vector character of the light as well as single scattering here. The shape of the so-called CB cone $E(\vec{q}_b)$ as a function of the “backscattering vector” $\vec{q}_b := \vec{k}_f + \vec{k}_i$ is given by the Fourier transform of the intensity distribution $I(\vec{\rho})$ [17,18]:

$$E(\vec{q}_b) = 1 + \int I(\vec{\rho}) \cos(\vec{q}_b \cdot \vec{\rho}) d\vec{\rho}. \quad (1)$$

In the diffusion approximation one obtains [17]

$$E(q_b) \approx 1 + \int p(s) \exp\left(-\frac{1}{3}s\ell^*q_b^2\right) ds, \quad (2)$$

where $p(s)$ is the path length distribution. Faraday rotation (FR) introduces a phase shift of $\pm V\vec{B} \cdot \vec{r}$ for circularly polarized light (handedness ± 1) propagating a distance \vec{r} parallel to a magnetic field \vec{B} . V is the specific Verdet constant. In the first models [2] describing the influence of FR on CB, the average distance between two scattering events ($\langle |\vec{r}| \rangle$) was approximated by ℓ^* and it was assumed that the handedness changes randomly after each scattering event. Then the phase of a light amplitude experiences a mean square deviation of $\frac{1}{3}s\ell^*(VB)^2$ along a path of length s . For small arguments, this phase shift can be included into Eq. (2) by an additional approximate factor $\exp[-\frac{1}{3}s\ell^*(2VB)^2]$. For a detailed description, see [6]. The factor 2 in front of VB results from the fact that in the case of CB the phase shifts of counterpropagating waves add up. Thus, in the diffusion limit, due to the replacement of q_b by $2VB$ in the exponents in Eq. (2), the

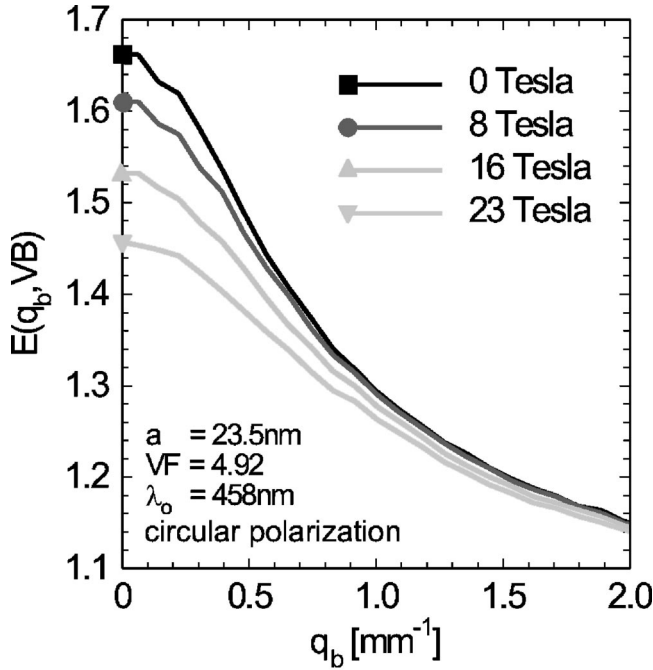


FIG. 1. (Color online only) Destruction of the CB cone with increasing magnetic field for a sample of polystyrene spheres (radius $a=23.5$ nm, VF 4.92%) in water.

cone shape as a function of q_b and VB is obtained by the replacement $q_b^2 \rightarrow q_b^2 + q_F^2$, with $q_F^2 := (2VB)^2$.

However, the approximation of a random helicity flip after each scattering event neglects correlations between depolarization and scattering angle. In fact, the circular depolarization is usually strongly correlated with the scattering angle θ . In the case of Rayleigh scattering, for example, there is a complete helicity flip only for $\theta = \pi$. Therefore, it was suggested [6] to introduce a different correlation length for the influence of FR according to a modified differential cross section $d\sigma_F(\theta) = d\sigma^{++}(\theta) + d\sigma^{\pm}(\theta - \pi)$, corresponding to the parts of the light that are scattered in the same $++$ and in the orthogonal \pm circular polarization states, respectively. Therefore the correlation length is obtained in analogy to the definition of ℓ^* in the case of nonisotropic scattering: $\ell_F^*/\ell = (1 - \langle \cos \theta \rangle)^{-1}$, where the average is taken according to $d\sigma_F$ now. Thus, for small arguments, in the diffusion approximation, the influence of FR on CB is given by the replacement $q_b^2 \rightarrow q_b^2 + (\ell_F^*/\ell^*)q_F^2$. Experimentally, the specific value ℓ_F^*/ℓ^* can be obtained by measuring the CB cone without FR, $E(q_b)$, and comparing it to the maximum enhancement factors $E(VB)$ at $q_b = 0$, as a function of VB . The rescaling factor ℓ_F^*/ℓ^* is obtained independently of $p(s)$ and ℓ^* .

EXPERIMENTS

The basic experimental setup has already been described elsewhere [6,19]. Here, we used a 23 T magnet with a vertical bore. The sample was placed at a depth of about 400 mm in the center of the field and illuminated from the top using a 90° deviating mirror. The rest of the CB setup (semitranspar-

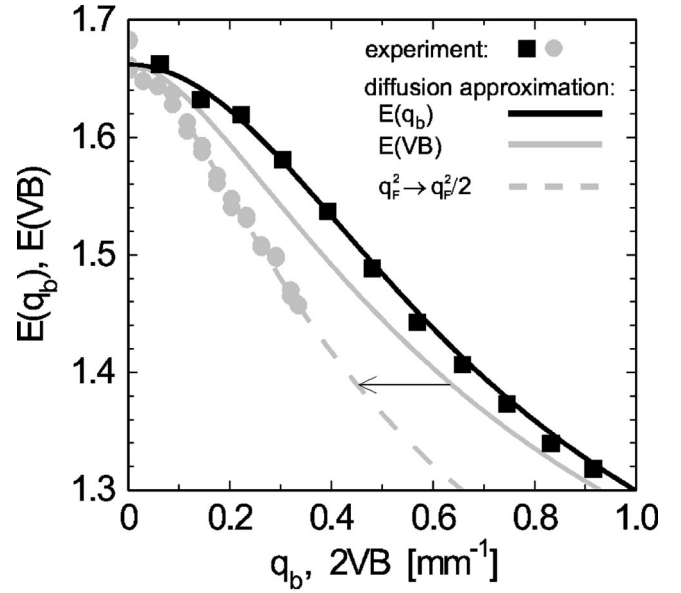


FIG. 2. (Color online only) Decrease of CB enhancement as a function of q_b and $2VB$. Marks, experiments; lines, theory. Same sample as in Fig. 1. The solid black line was fitted to the measured $E(q_b)$ (see text, fit parameters $dq_{res} = 0.24$ mm $^{-1}$, $\gamma = 2.3$). The same parameters have been used to calculate $E(2VB)$ (solid gray line). However, as $\ell_F^* > \ell^*$, the measured curve $E(2VB)$ decreases faster than expected. Rescaling in the argument (dashed line) gives the value $\ell_F^*/\ell^* = 2$ here. Note that in case of a “perfect” experimental setup the solid gray and black lines should be identical.

ent mirror, beam expander, polarizers, lens of focal length 1330 mm in front of a charge-coupled device camera, Ar $^+$ laser at $\lambda = 458$ nm) was placed at a distance of about 2 m from this mirror, where magnetic field effects on the components of the setup could be neglected. The stability of the setup was verified by measuring the cone of a white Teflon sample positioned at the end of the magnet bore. For a precise angular calibration we measured the backscattering profile of a well defined glass sphere (diameter 5 mm) [20,21]. The cell (diameter 40 mm, height 28 mm) containing the samples of polystyrene spheres suspended in water was open on top in order to avoid undesired effects (e.g., FR in glass windows, reflections).

In most samples the volume content of polystyrene beads was sufficiently small that the Faraday effect of pure water (0.379°/mm T at $\lambda = 458$ nm) could be used. At higher volume fractions we calculated the volume averaged Verdet constant ($V_{PS} = 1.093^\circ/\text{mm T}$ at 458 nm [22]). In order to obtain a significant magnetic field effect on the CB cone, the volume fraction (VF) of the polystyrene spheres was adjusted such that all samples had a transport mean free path of $\ell^* \approx 500$ μm . As an example, Fig. 1 shows the destruction of a CB cone with increasing magnetic field.

EVALUATION OF EXPERIMENTAL RESULTS

Due to the extreme experimental conditions (e.g., limited angular resolution of the setup, limited lateral intensity profile of the incident laser beam leading to slightly different

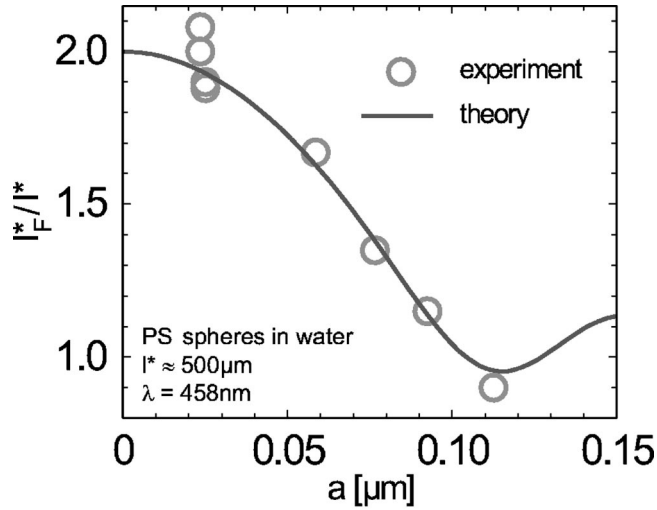


FIG. 3. (Color online only) Measured and calculated [6] values ℓ_F^*/ℓ^* as a function of the polystyrene sphere radius a .

amplitudes in the direct and reversed paths) the measured cone shapes could not be compared directly to the analytical curves given by the earlier analysis in. Both experimental artifacts reduce the enhancement factor below 2 as can be seen in Fig. 1. The limited resolution dq_{res} can be included approximately into Eq. (1) by a convolution in \vec{q}_b with $\exp[-(\vec{q}_b' - \vec{q}_b)^2/dq_{res}^2]$ (system response). The limited lateral intensity profile can be approximated by an additional convolution with $\exp[-\vec{R}^2/dR^2]\exp[-(\vec{R}' - \vec{\rho})^2/dR^2]$, where \vec{R} and $\vec{R}' - \vec{\rho}$ represent the start and end points of a random walk, and dR reflects the width of the Gaussian intensity distribution (in our case $dR = 6.1$ mm). Assuming a path length distribution $p(s)$ as suggested in [17,18], integration over \vec{q}_b and \vec{R} can be performed analytically; the integration over s was done numerically. The curve shape obtained was fitted to the experimental cone shape at zero field, $E(q_b)$, using dq_{res} and γ (see [17,18,25]) as free fit parameters. Then, the decrease of the cone tip $E(VB)$ as a function of the magnetic field was calculated using the same expression with the same parameters. Finally, the value ℓ_F^*/ℓ^* was obtained by rescaling the calculated curve in the argument until best coincidence with the measured curve $E(VB)$ was found. Figure 2 shows the result for the same sample as in Fig. 1. We found that in our experiments ℓ_F^*/ℓ^* is rather insensitive to the precise value of the fit parameters. The evaluation could be improved by simulating $p(s)$ in a scalar random walk model [19]. Figure 3 shows the result for different particle sizes. The coincidence with the theoretically predicted values of ℓ_F^*/ℓ^* is very good.

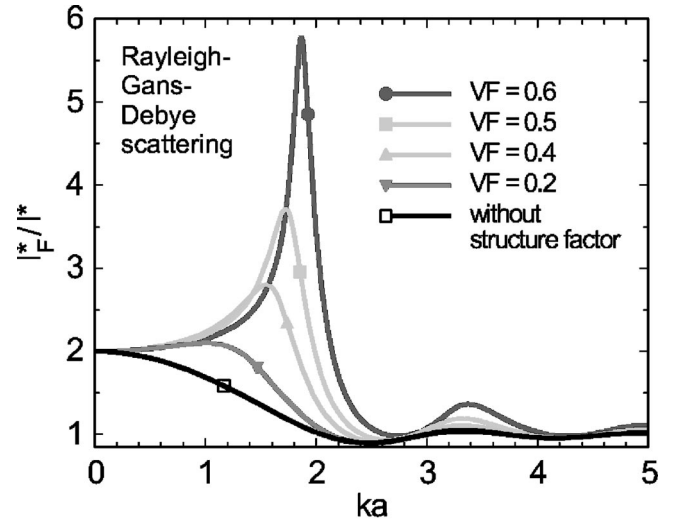


FIG. 4. (Color online only) ℓ_F^*/ℓ^* as a function of size parameter for different volume fractions calculated for Rayleigh-Gans-Debye scattering including a Percus-Yevick structure factor.

CONCLUSION AND OUTLOOK

In conclusion, we have shown the importance of correlations between depolarization and scattering angle for multiple light scattering. They give rise to a strong scatterer size dependence of the relevant correlation length ℓ_F^* divided by the transport mean free path ℓ^* . The suggested model [6] explains this dependence quite well and predicts the measured decrease of ℓ_F^*/ℓ^* from 2 to about 1 in a quantitatively correct way.

Another interesting question concerns the role of positional correlations of scatterers as expressed by the structure factor. In our case, the samples with the smallest radius had a volume fraction of about 5%, i.e., the structure factor may already be non-negligible (for the other samples the VF was always smaller than 1%). In Fig. 4 we have calculated ℓ_F^*/ℓ^* for Rayleigh-Gans-Debye scattering, including a Percus-Yevick structure factor [23,24] in the differential scattering cross section. As can be seen, for $1 \lesssim ka \lesssim 2$ the value ℓ_F^*/ℓ^* becomes larger than 2 and approaches nearly a value of 6.

Consequently, ℓ_F^* is a very useful parameter for studying light scattering in dense media by CB measurements or FR dependent speckle correlation measurements in transmission.

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