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Varying Refractive Index Induced Intensity Correlations in Magnetic Colloids

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One crucial metric for optical transports in heterogeneous media is intensity correlation. Here, intensity correlation in magnetic colloids as a function of magnetically fluctuating refractive index is calculated using an extended transport theory. The normalized transmitted intensity experimental results as a function of applied magnetic field in transverse configuration are compared with the same. Both exhibit comparable dependencies, indicating that refractive index control is crucial for optical transmission in magnetic colloids. The sample utilized in this experiment, which is made up of magnetic spheres the size of microns suspended in a ferrofluid, was deliberately and precisely developed with the goal of changing the relative refractive index of the scatterer via magnetic tuning of the ferrofluid refractive index.
Keywords: varying refractive index, light scattering, magnetic colloids, correlations.

1. Introduction

Recent research has shown several intriguing photonic features of magnetically adjustable light scattering by magnetic colloids[1–7]. One of the key parameters to examine when examining the photonic properties of this material is the tuneable refractive index-mediated intensity correlation[8]. The refractive index fluctuation in ferrofluids that is dependent on the magnetic field was studied using techniques for retroreflection and light refraction[9–11]. It was found that the ferrofluid's refractive index varies when an external magnetic field is applied; the greater the magnetic field, the higher the refractive index. The magnetically generated structure in ferrofluids was blamed for this behaviour[10]. Magneto chromatics is another amazing magneto-optical effect of magnetic fluid in which light is scattered into different colours as it travels through a ferrofluid with an ordered structure when an external magnetic field is applied[12]. Similarly, magnetically tuneable structural lithographic printing is demonstrated by employing a magnetic field to modulate the periodicity of structures[3]; a basic technique is used to illustrate all the colours of the spectrum, from violet to red, at varying field strengths[1]. Ferrofluid should be diluted for magneto-optical studies for two reasons: (a) to permit light transmission; and (b) to decrease dipolar energy more than thermal energy. Langevin's theory of paramagnetism, which describes noninteracting single domain magnetic particles scattered in an appropriate liquid carrier, provides an excellent description of the magnetization of magnetic fluid[13]. However, when tiny magnetic particles are distributed throughout a ferrofluid, the optical characteristics of the magnetic fluid are significantly altered. The magnetically induced Mie resonance and scattering by magnetic scatterers provide the explanation for the magnetically induced zero forward scattering, enhanced back scattering, light storage, and retrieval that our group has recently proven[5–7]. The transmitted intensity is also observed to increase with an externally applied magnetic field for small particle sizes (usually less than 10 nm), but to decrease for larger particle sizes. This phenomenon is explained by the competition between van der Waals and dipole-dipole interactions[14]. Mie resonance within the magnetic scatterers provides an explanation for the observed substantial drop in light transmission at critical magnetic field, which follows a power law dependency with volume fraction of nanoparticles[5–7, 15]. Most of the above-mentioned references provide a macroscopic explanation for the magneto optical phenomenon, either on the magnetically induced structures of magnetic nanoparticles in ferrofluid or on the magnetic particles' light scattering. Tuneable refractive index allows for the study of induced intensity correlations under the microscope, providing a more comprehensive understanding of the origin of optical transport in magnetic colloids. As a material's intrinsic feature, correlations offer crucial details regarding transport parameters. Correlations have previously been measured in frequency [16] and duration [17] to find the light's diffusion constant. By altering the effective refractive index in heavily scattering material, a novel method for determining the effective medium and the transport characteristics of light propagation in heterogeneous media has recently been investigated[8].

In this study, we use Green's theorem to examine intensity correlation as a function of effective refractive index that can be magnetically tuned. In a transverse configuration, the same is compared with normalized transmitted intensity as a function of applied magnetic field. The theoretical predictions and experimental results correspond well. The purpose of the sample utilized in this experiment is to magnetically tune the refractive index of the ferrofluid, a magnetically active medium, in order to alter the relative refractive index of the micron size scatterer (3-5 μm) suspended in it. Based on Mie resonance within the scatterer, which is backed by resonance in the forward-backward anisotropy factor <cos θ> as a function of applied magnetic field, the observed decrease in magnetic field dependent optical transport is explained.

In Ref.[18, 19], the relationships employing generalized transport are clearly discussed. Early on, the effective refractive index is visible, and the average amplitude Green's function is expressed as[8].

$$G(r;\, n_{scatt}, l_s) = -\frac{e^{\left[\left(\frac{i n_{scatt}\omega}{c}\right)-\left(\frac{1}{2 l_s}\right)\right] r}}{4\pi r} \quad (1)$$

Here, the average length of scattering free path is given as l_s which is function of the refractive index (n_{scatt}) of micron sized magnetic scatterers suspended in the magnetic fluid and it is expressed as $l_s = \frac{l}{(1-\langle\cos\theta\rangle)}$. a_n and b_n, the Mie coefficients, can be used to define the anisotropy factor $\langle\cos\theta\rangle$ and $l = \frac{1}{\Phi\sigma}$[20]. Here, σ represents the total scattering cross section and Φ represents the number density of the scatterer. Considering this the refractive index tuning is[8].

$$\delta = \frac{1}{n_f}\frac{\partial n_{scatt}}{\partial n_f} \quad (2)$$

This δ shows the relative change in refractive index of scatterer with respect to the variable refractive index of the magnetic fluid, controlled by an external magnetic field. Here, n_f is the refractive index of the fluid, $n_f = n_0 + n_\infty F(\alpha)$; n_0 is the refractive index of the ferro nanofluid when magnetic field is not applied externally, when external magnetic field is applied the refractive index of the magnetic fluid increases initially and eventually reaches to its saturation value (n_∞) for fixed magnitude of applied field, $F(\alpha) = \frac{\alpha L^2(\alpha)}{\alpha - L(\alpha)}$, $L(\alpha) = \coth\alpha - \frac{1}{\alpha}$, $\alpha = \frac{\mu H}{k_B T}$, μ is magnetic moment of the scatterer and, H is external magnetic field, k_B is Boltzmann constant and T is absolute temperature. Here, $F(\alpha)$ is obtained from effective magnetic field of medium[21]. Here, δ can be easily controlled by the external magnetic field as $L(\alpha)$ and consequently $F(\alpha)$ is a function of applied magnetic field. In this case the intensity correlation function can be used as[8],

$$C\left(n_f, n_f + \Delta n_f\right) = \frac{\tau_\delta \Delta n_f}{\cosh\sqrt{\tau_\delta \Delta n_f} - \cos\sqrt{\tau_\delta \Delta n_f}} \quad (3)$$

where, $\tau_\delta = \frac{2\omega\delta L^2}{D}\tau$, ω is the frequency, L is the optical path length, D is the diffusion constant. We have also considered the effective wave number $k = \frac{2\pi}{\lambda_e} + i\left(\frac{1}{2l_s}\right)$, λ_e is the effective wavelength, magnetically modulated by the ferrofluid's change in refractive index. Further, l_s is determined by $< \cos\theta >$, is a function of Mie scattering parameters a_n and b_n, with field dependent refractive index. Neglecting higher order terms the anisotropy factor (where, a is radius of the scatterer) is,

$$\langle\cos\theta\rangle = \frac{Re(a_1 b_1^*)}{(|a_1|^2 + |b_1|^2)} \quad (4)$$

The function $p(n, ka) = ka\tan(nka)$ is periodic, where $n = \frac{n_{scatt}}{n_f}$ and n_f depend on magnetic field and consequently $< \cos\theta >$ is field dependent. This theoretical prediction is used to analyze the field dependent light transmission in magnetic colloids. This concept is not exploited much because of the experimental difficulties[8], but we have made it simple by designing the sample accordingly and with moderate magnetic field tunability.

Fe_3O_4 magnetic nanocrystals were created using the traditional co-precipitation technique. Alkaline solution was mixed with a solution containing ferrous (Fe^{2+}) sulphate and ferric (Fe^{3+}) chloride. We replaced ferrous chloride (Fe^{2+}) with ferrous sulphate to maintain the stoichiometric proportion of $\frac{Fe^{2+}}{Fe^{3+}} = \frac{1}{2}$ since ferrous chloride degrades and generates hydroxide of ferrous in humid environments in the present synthesis.

To obtain crystallite size ~ 10nm, an alkaline source consisting of an 8M solution of ammonium hydroxide was utilized. When ammonia and salt solutions of iron were combined, black precipitates of Fe_3O_4 were produced at pH 10.5 instantaneously. To enable the nano crystallites to enlarge, the mixture was constantly agitated for 20 minutes at a pH of 10.5. To get rid of contaminants that were soluble in water, nano crystallites were magnetically decanted and repeatedly cleaned with double-distilled water. These nano crystallites were dissolved in kerosene after being treated with oleic acid to create a stable ferrofluid. For 20 minutes, the fluid was centrifuged at 12,000 rpm in order to get rid of any aggregates.

Magnetite powder that is available for purchase was acquired at the Alchemie Research Centre located in Mumbai, India.

In a planetary monomill (Pulverisette 6, Fritch GMbH), magnetite powder was ball milled while kerosene and oleic acid were present. The weight ratio of Fe_3O_4 to the oleic acid is 1:5. For 36 hours, the particles are ground at 300 rpm. The ratio of charge to ball was maintained at 1/3. Particles ranging in size from 3 to 5 µm were suspended in solutions using fractional sedimentation.

To obtain bidispersed magnetic dipolar fluid, magnetic nano fluid of desired volume fraction (10^{15} $\frac{1}{m^3}$) is mixed with a suspension containing micron sized magnetic particles of known volume fraction (10^5 $\frac{1}{m^3}$) homogenized by ultrasonication for 10 minutes.

The He-Ne laser (uniphase) with a wavelength of 633 nm and an output power of 10 mW was used to illuminate the experiment for three distinct optical path lengths, L = 40, 60, and 80 µm. An electromagnetic coil was used to create the magnetic field, which could reach 0.1 T. Light travels in a direction opposite to that of the magnetic field. The photo detector on the storage oscilloscope (Tektronix-TDS-2024) was used to record the transmitted intensity (see Fig. 1).

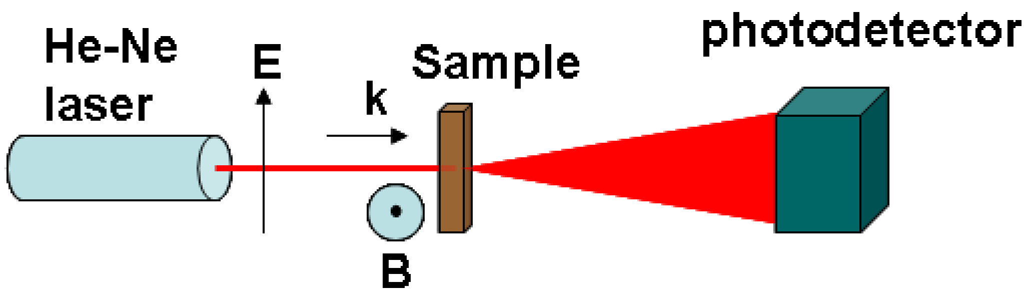

Fig.1. (Color online) Schematic of the experimental set up. E is the electric field vector, k is the wave vector and B is the magnetic field vector. The sample is illuminated by HeNe laser. The light transmission is detected by the photodetector.

A appropriate carrier liquid's magnetic particles develop dipole moments when an external magnetic field is present. The magnitude of the dipole moment induced is given as, $m = \left(\frac{\pi}{6}\right) d^3 \chi H$, here the particle diameter is d, χ is the effective susceptibility, H is the value of the applied external magnetic field. The anisotropic energy between the particles due to their interaction is given as,

$$U_{ij} = \frac{1}{4\pi\mu_0}\left[\frac{m_i m_j}{r_{ij}^3} - \frac{3(m_i r_{ij})(m_j r_{ij})}{r_{ij}^3}\right] \quad (5)$$

where m denotes the corresponding dipole moments, r_{ij} is the displacement vector of the two particles, and μ_0 is the vacuum permeability. The coupling constant can be expressed as follows: it is the ratio of the maximum interaction energy magnitude to the thermal energy.

$$x = \frac{m^2}{4\pi d^3 \mu_0 k_B T} \quad (6)$$

where, Boltzmann constant is k_B and T is absolute temperature. If $x >> 1$, the magnetic particles brought together by a magnetic field to form a structure like a chain. In such magnetic suspension (ferrofluid) in kerosene, the absorption study in visible range (wavelength 400nm - 800nm) exhibits absence of a discernible absorption peak around λ = 633 nm for a typical 10nm particle diameter[15]. Earlier, it was shown that the optical transmission in ferrofluid is due to magnetic field dependent structures. The struggle between the van der Waal and dipole-dipole interactions of the particles determines this structural behaviour. Van der Waal is more significant for particles less than 10 nm, while magnetic interaction is more significant for particles larger than 10 nm. Thus, it was demonstrated that light transmission rises with increasing magnetic field for 7 and 9 nm particles in ferrofluid and decreases with increasing magnetic field for 12 nm particles in ferrofluid[14]. It was explained in terms of a magnetically induced structural transition, without considering how an external magnetic field could alter the medium's effective refractive index.

Magnetically controlled refractive index of the ferrofluid sample is displayed in Fig. 2. The line can be interpreted as follows: $n_f = n_0 + n_\infty F(\alpha)$. A tiny electromagnet and a sample cell were mounted on one arm of the Michelson Interferometer to measure the refractive index. It is noted that the ferrofluid's refractive index rises with increasing field value and tends to saturate at higher fields. This is comparable to what has been shown with different methods[9–11]. This was previously described in terms of structure development triggered by magnetic fields. The ferrofluid's magnetically modulated dielectric constant ε_f, and the refractive index, $n_f = \sqrt{\varepsilon_f}$, $\mu_f \sim 1$ at optical frequency, can also be used to describe it. It is established that an external magnetic field can adjust ε_f of the ferrofluid[10, 22]. The scattering property of a sample changes significantly when micron-sized magnetic spheres (~3-5 μm) belonging to the same family—iron oxide—are scattered in a ferrofluid [5–7, 23]. When $d > \lambda$, the micron-sized particles will dominate the light scattering; therefore, the Mie scattering will be relevant.

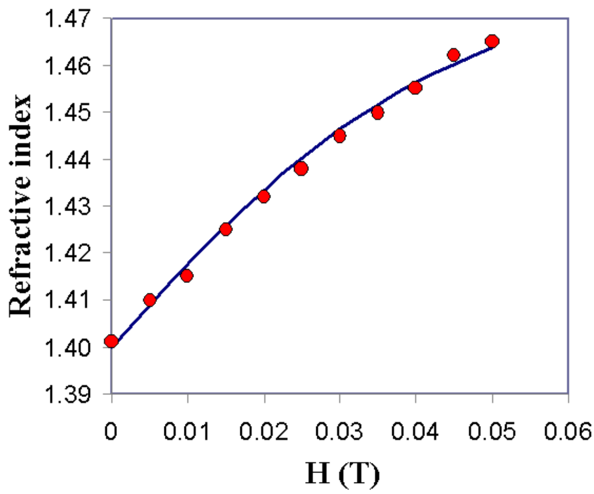

Fig.2. (color online) Variation of refractive index as a function of externally applied magnetic field in ferrofluid sample. The zero field refractive index n0 is 1.401 and at 0.05 T field it reaches 1.465.

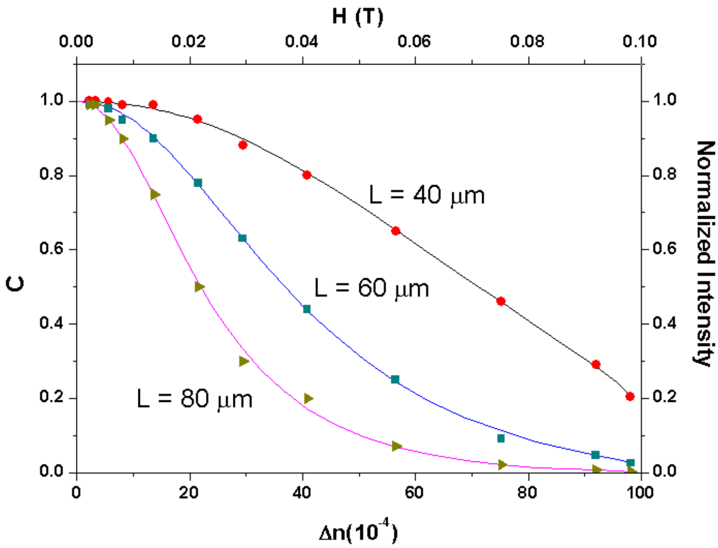

Fig.3. (Color online) Intensity correlation as a function of magnetically varying refractive index is plotted for magnetic spheres (3-5□m) suspended in a ferrofluid (optical path length L = 40, 60 and 80 □m). Solid line is fit to the Eq. (3) is compared with the magnetic field dependent normalized transmitted intensity for transverse magnetic field direction. Suggesting, importance of refractive index tuning in optical transport through magnetic colloids.

Fig. 3 shows a typical correlation function versus Δn_f Using Eq. (3), the solid line illustrates theory. The same graph displays the experimental data points for the normalized transmitted intensity for the transverse configuration as a function of applied magnetic field. Normalized transmitted intensity declines with increasing field strength and behaves similarly to $C \rightarrow \Delta n_f$. This implies that the light transport through magnetic colloids as a function of applied magnetic field is significantly influenced by the refractive index tuning of intensity correlation that is dependent on the magnetic field. Dipolar interactions (Eq. (5)) of the particles and Brownian rotation of micron-sized particles in a magnetically interacting medium account for the non-exponential behaviour. In such a magnetically interacting medium, the sluggish response to the micron-sized magnetic spheres is caused, at first, by the dipolar interaction energy [24] being greater than the externally supplied magnetic field. The explanation for the decrease in light transmission and intensity correlation as a function of applied magnetic field and the corresponding change in refractive index can be found in the morphology-dependent Mie resonance that the micron-sized magnetic spheres experience due to refractive index mismatch. Mie resonance is the reason behind theoretical and experimental observations of magnetic field-induced oscillations in light transport and transmission decrease[23]. Resonance in the anisotropy factor $< \cos\theta >$, is also produced by this, and it is dependent on the Mie scattering coefficients a_n and b_n. This leads to resonance in the scattering mean free path l_s. Figure 4 displays the 3-5 μm magnetic sphere oscillations in as a function of the external magnetic field suspended in a ferrofluid. This validates the scatterer's internal magnetic field-dependent Mie resonance. The Mie resonance that occurs inside the scatterer causes standing waves to accumulate, which in turn lowers the amount of light transfer.

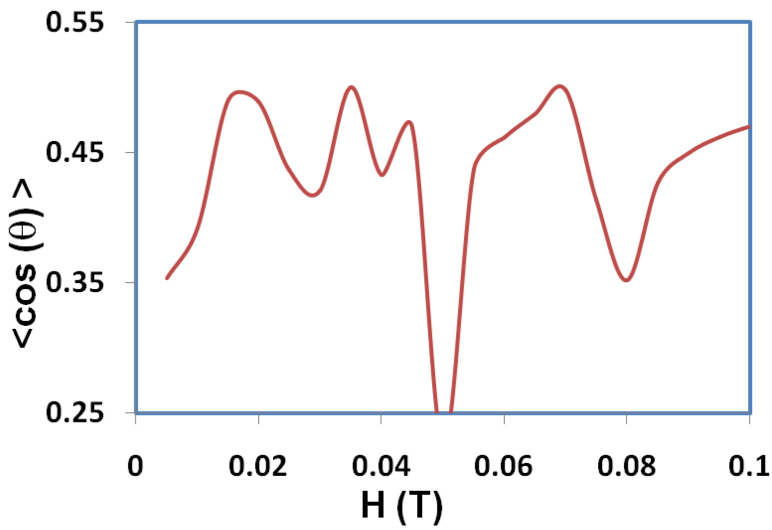

FIG.4. (Color online) The scattering anisotropy factor <cos□>, plotted as a function of magnetic field for ka = 30, where k is a wave vector and a is the radius of the scatterer. Here □ = 1. The oscillations in the anisotropy factor, suggests magnetic field dependent Mie resonance within the scatterer.

The intensity correlation function of a specially designed magnetic colloid, which is made up of magnetic spheres the size of microns suspended in a ferrofluid, and that obtained using generalized transport theory are interestingly compared in this study. This article discusses the significance of magnetic refractive index tuning for optical transport in magnetic colloids. As an external magnetic field is introduced, a drop in normalized transmission intensity is seen. The scatterer's morphology-dependent Mie resonance, which generates a standing wave and lowers light transport, provides an explanation for the fall in transmittance. The forward-backward anisotropy factor oscillations that are dependent on the magnetic field validate the presence of Mie resonance within the scatterer. The research contributes to the construction of new magneto optical and photonic systems where the magnetically fluctuating refractive index may be used to regulate the transmission of light.

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