



# Investigations on magnetic field induced optical transparency in magnetic nanofluids

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## ABSTRACT

We study the magnetic field induced optical transparency and its origin in magnetic nanoemulsion of droplets of average size  $\sim 200$  nm containing superparamagnetic iron oxide nanoparticles. Beyond a certain volume fraction ( $\Phi > 0.0021$ ) of magnetic nanoemulsion and a critical magnetic field ( $H_{c1}$ ), the transmitted light intensity increases drastically and reaches a maximum at another critical magnetic field ( $H_{c2}$ ), beyond which the transmitted light intensity decreases and reaches a plateau. Interestingly, the transmitted light intensity at  $H_{c2}$  is found to increase linearly with  $\Phi$  and the critical magnetic fields  $H_{c1}$  and  $H_{c2}$  follow power law decay with  $\Phi$  (i.e.  $H_c \sim \Phi^{-x}$ ), with exponents 0.48 and 0.27, respectively. The light intensity recovers to its initial value when the magnetic field is switched off, indicating the perfect reversibility of the field induced transparency process. The observed straight line scattered patterns above  $H_{c2}$ , on a screen placed perpendicular to the incident beam, confirms the formation of rod like anisotropic nanostructures perpendicular to the direction of light propagation. The magneto-optical measurements in the emulsion confirm that the observed field induced transparency in magnetic emulsions for  $\Phi > 0.0021$  is due to the optical birefringence caused by the rod like nanostructures. The reduced birefringence is found to be proportional to the square of the applied magnetic field. This finding offers several possibilities in using magnetic nanofluids in tunable optical devices.

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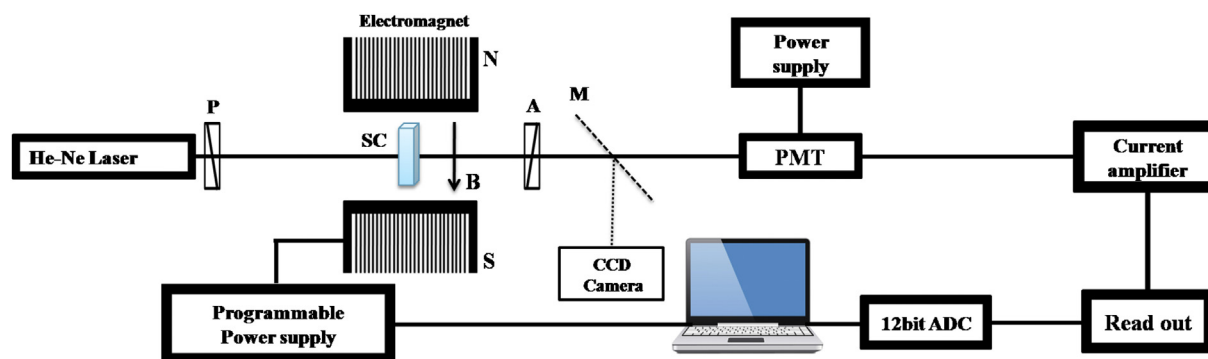
## 1. Introduction

Magnetic nanofluids are very interesting stimulus responsive materials whose optical properties can be controlled by an external magnetic field [1]. The magnetic field assisted assembly of magnetic nanoparticles show interesting optical phenomena, which is dictated by the competition between hydrodynamic and magnetic forces [2–5]. These magneto-optofluidic phenomena are being used in various applications like optical grating [6], sensors [7–9], optical fiber modulator [10,11], magneto-optical waveguide [12], holographic optical tweezers [13], tunable photonic devices [14,15], magneto-optical wavelength filter [16], optical switches [17], photonic switching by controlling the aspect ratio of the nanostructures [18] etc. They have also been used as model systems for probing fundamental phenomena [19–21] and have a wide range of applications in medicine too [22]. Magnetic nanoemulsions are another category of magnetic fluids, where

ferrofluid oil, containing single domain ferromagnetic or ferri-magnetic nanoparticles of typical size  $< 10$  nm, coated with a surfactant or polymer, are dispersed in water. A birefringence phenomenon, in general, is widely exploited in optical devices such as light modulator, quarter wave plate, half wave plate etc [23,24]. Magneto optical properties like dichroism and birefringence in pure ferrofluids and ferroemulsions have been studied in the past [25–34]. The initial thinking about the origin of birefringence in ferrofluid was due to the magnetisation of the particles caused by Neel rotation (free rotation of magnetic moment inside the particles) [31]. Subsequent studies considered nano-sized linear aggregates [29,35], spatial anisotropy [27] and shape anisotropy [31] as the cause for magnetic birefringence in ferrofluids. Pan et al. [36], considered the combined effects of field-induced orientation of shape-anisotropic scatterers and the chain-like aggregation of magnetic particles in an external magnetic field as the cause for the magnetic field-induced optical anisotropy and used Monte-Carlo technique to simulate the field dependence of magneto-birefringence. Di et al. [37] studied magnetic birefringence in magnetic fluids as a function of volume

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**Fig. 1.** Schematic representation of the light scattering experimental set up. The applied magnetic field is perpendicular to the direction of propagation of light. P- polarizer, A-analyzer, SC- sample cuvette, NS- pole pieces of electromagnet, M-mirror, PMT-photomultiplier tube.

fraction and wavelength and showed that the birefringence increases with concentration of particles.

The phenomena of scattering and diffraction of light in magnetic suspension and mixtures of magnetic and nonmagnetic scatterers were also investigated in the past [38–42]. Recently, variation of transmitted light intensity in a magnetic nanofluid with different wavelength in the presence of magnetic field was studied experimentally using light scattering technique. The relaxation of magnetic nanofluids increase as a power law of wavelength of incident light [43]. Also, the optical transmittance for a polarized light passing through the magnetic fluid was investigated in presence of magnetic field [21]. Ivey et al. [44] demonstrated that different structural transitions are possible in magnetic emulsions depending on the applied magnetic field. It is known that at lower magnetic field, individual particles start to interact with each other to form dimers, trimers and short chains in the direction of the magnetic field. As field strength increases, the gas like structure undergoes a nematic liquid like structure and then it becomes a rigid solid like structure. It was also reported that the magnetic permeability of magnetic nanoemulsion increases with magnetic field due to droplet elongation in the direction of magnetic field [45]. Shape anisotropy due to elongation of droplet in the presence of a small field is considered to be one of the reasons for field induced birefringence. In the earlier studies, though field induced transparency in magnetic fluids [46,47] was observed in some cases, the conditions for the formation of such field induced transparency and reasons for such changes in the transmitted light intensity was not fully understood. In this paper, we study the field induced optical transparency and magneto-optical birefringence in magnetic nanoemulsion as a function of volume fraction. We observed that such field induced transparency is seen only above a certain volume fraction and magnetic birefringence is the cause of the observed transparency, which was further confirmed from the fact that the transmitted intensity (reduced birefringence) is proportional to the square of the magnetic field in the low field region.

## 2. Materials and methods

Magnetic nanoparticles used are  $\text{Fe}_3\text{O}_4$  particles of average size ~10 nm. The details of co-precipitation technique to prepare magnetic nanoparticles are discussed elsewhere [48]. The particles were capped with oleic acid to prevent aggregation and dispersed in oil (octane). The surfactant used in this study, sodium dodecyl sulphate ( $\text{C}_{12}\text{H}_{25}\text{SO}_4\text{Na}$ ), was obtained from Sigma Aldrich, and used without further purification. Milli Q water, filtered with 0.22  $\mu\text{m}$  Millipore filter, was used to make surfactant solutions. The nanoemulsion was prepared by emulsifying octane containing

$\text{Fe}_3\text{O}_4$  magnetic nanoparticles with water containing sodium dodecyl sulphate, at ~1500 rpm, using a IKA homogenizer [49]. Emulsification results in formation of polydisperse emulsion, where droplet size varies between 0.05 and 1  $\mu\text{m}$ . Highly monodisperse emulsion of size 200 nm was obtained using fractionation approach [50].

Fig. 1 shows the schematic diagram of the experimental setup for measuring field induced light transmission and optical birefringence in magnetic emulsion. The magnetic emulsion was taken in a quartz cuvette having path length of 1 mm. An amplitude and frequency stabilized He–Ne laser (spectra physics), with a wavelength of 632.8 nm and an output power of 1 mW was used as the light source. The light beam first goes through a polarizer (P) and then through the sample placed in between the poles of an electromagnet. The magnetic field generated by the electromagnet was controlled by changing the current using programmable DC power supply. The direction of the applied magnetic field was perpendicular to the direction of propagation light. Finally, the transmitted light passed through an analyzer. The axes of polarizer and analyzer were set perpendicular to each other for the birefringence measurement, whereas they were parallel during the transmittance measurement. The forward transmitted intensity was measured by a photomultiplier tube (PMT). The output of PMT was fed to a readout through a current amplifier with variable gain. The analog output from the readout was connected to a 12 bit analog to digital converter (ADC) that was interfaced with a computer. For recording transmitted intensity patterns, the light scattered from the sample was projected on a screen and were recorded using a charge coupled device (CCD) camera.

## 3. Results and discussion

### 3.1. Dynamic light scattering measurement

In dynamic light scattering, the hydrodynamic size of suspended particles is measured from the time dependent fluctuations in the intensity of the scattered light. The intensity autocorrelation functions is given by  $g^2(\tau) = \frac{\langle I(t)I(t+\tau) \rangle}{\langle I(t) \rangle^2}$ , where  $I(t)$  is the scattered intensity at an arbitrary time,  $t$ , and  $I(t + \tau)$  is the scattered intensity after a delay time,  $\tau$ . The intensity autocorrelation function decays exponentially with time since the particles in the suspension are in Brownian motion. The intensity autocorrelation function,  $g^2(\tau)$ , is related to the electric field autocorrelation function ( $g^1(\tau)$ ) by Siegert's relation,  $g^1(\tau) = [g^2(\tau) - 1]^{0.5}$ . For a suspension of monodisperse spherical particles, the autocorrelation function decays exponentially with delay time  $\tau$  and is given as [51].

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