



The effect of suspended Fe_3O_4 nanoparticle size on magneto-optical properties of ferrofluids



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ABSTRACT

We investigate the effect of hydrodynamic particle size on the magnetic field induced light transmission and transmitted speckle pattern in water based ferrofluids containing functionalized Fe_3O_4 nanoparticles of size ranging from 15 to 46 nm. Three water-based magnetic nanofluids, containing Fe_3O_4 nanoparticles functionalized with poly-acrylic acid (PAA), tetra-methyl ammonium hydroxide (TMAOH) and phosphate, are used in the present study. In all three cases, the transmitted light intensity starts decreasing above a certain magnetic field (called first critical field) and becomes a minimum at another field (second critical field). These two critical fields signify the onset of linear aggregation process and zipper transitions between fully grown chains, respectively. Both these critical fields shift towards a lower magnetic field with increasing hydrodynamic diameter, due to stronger magnetic dipolar interactions. The first and the second critical fields showed a power law dependence on the hydrodynamic diameters. The dipolar resonances occurring at certain values of the scatterer size, leads to the field induced extinction of light. Both the onset of chaining and zipper transitions were clearly evident in the time dependent transmitted light intensity. Above the first critical field, the lobe part of the transmitted intensity and the lobe speckle contrast values increase with increasing external magnetic field due to reduced Brownian motion of the field induced aggregates. The speckle contrast was highest for nanoparticle with the largest hydrodynamic diameter, due to reduced Brownian motion. These results provide better insight into field dependent light control in magnetic colloids, which may find interesting applications in magneto-optical devices.

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1. Introduction

Magnetic colloids such as ferrofluids and ferro-emulsions are exciting systems for both fundamental studies and for practical applications, owing to their unique tunable optical properties in the presence of an external magnetic field [1–9]. The tunable optical properties originate from the external field induced structural reorganization of suspended magnetic colloidal particles [10–13,14]. The field dependent tunable optical properties of ferrofluids have been used in developing optical sensors [15,16], tunable optical filters [17], tunable photonic devices [18] etc. The effect of external magnetic field on aggregation of magnetic nanoparticles and its influence on propagation of light have been studied extensively [19]. An understanding of various parameters influencing the aggregation process in magnetic nano-colloids is

important from both fundamental and practical application point of view [20].

Recently, the role of the applied field exposure time and viscous force on the particle aggregation and de-aggregation kinetics in magnetic nanofluids was studied experimentally using light scattering techniques [21]. Also, the kinetics of particle aggregation in magnetic nanofluid have been studied using various techniques [22–25]. The functional groups adhered to the nanoparticles (stabilizers) can influence the kinetics of field induced chainlike formation in magnetic nanofluid [26]. Among other influencing parameters, the dipolar interaction among the magnetic nanoparticles is the main driving force for particle aggregation process, and the field induced structural transitions [1]. However, for a given condition (constant external magnetic field strength and particle volume fraction), the particle size has a strong influence on the dipolar interactions, that can affect the magneto-optical properties of nanofluids significantly. Studies show that the non-linear index of refraction and photon absorption in magnetic nanofluids are affected by the nanoparticles size and its surface coating agents [27]. To the best of our knowledge, no systematic experimental study has been reported on the role of suspended

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particle size on the external field induced particle aggregation process, and the resulting optical properties in magnetic nanofluids.

In this paper, we have systematically studied the variation of normalized transmitted light intensity and transmitted speckle patterns as a function of external magnetic field in three different water based magnetic nanofluids containing magnetic nanoparticles with three different hydrodynamic diameters. The primary objective of this study is to obtain insight into the effect of particle size on magnetic field induced aggregation and its kinetics.

2. Materials and experimental set-up

Three water-based stable magnetic nanofluids containing Fe_3O_4 nanoparticles coated with (i) poly-acrylic acid (PAA) (ii) tetramethyl ammonium hydroxide (TMAOH) and (iii) phosphate were used in our studies. The size distributions in dispersed medium were measured using dynamic light scattering (DLS). Fig. 1 shows the size distributions of the three magnetic nanofluids. The measured average hydrodynamic sizes of the PAA, TMAOH and phosphate coated nanoparticles were 46, 30 and 15 nm, respectively. These suspensions showed excellent long-term stability even after prolonged application of strong magnetic fields. The volume fractions (ϕ) for all the samples were kept constant at 0.00916 in all our experiments. In all the three magnetic nanofluid samples, the particle sizes were much less than the incident light wavelength i.e. $a \ll \lambda$. The forward transmitted light intensity was measured as a function of applied magnetic field, where the direction of the applied field is perpendicular to the direction of the incident light. Details of the experimental technique and schematic of the experimental set up are described elsewhere [28,29].

3. Results and discussions

Fig. 2 shows the transmitted light scattering pattern projected on a screen from three different magnetic nanofluids at the external magnetic fields 0, 100, 200, 300 and 400 G. Fig. 2(a–e) corresponds to PAA coated Fe_3O_4 nanofluid, Fig. 2(f–j) corresponds to TMAOH coated Fe_3O_4 nanofluid and Fig. 2(k–o) corresponds to phosphate coated Fe_3O_4 nanofluid. In all three cases $\phi = 0.00916$. The external magnetic field was increased at a constant ramp rate

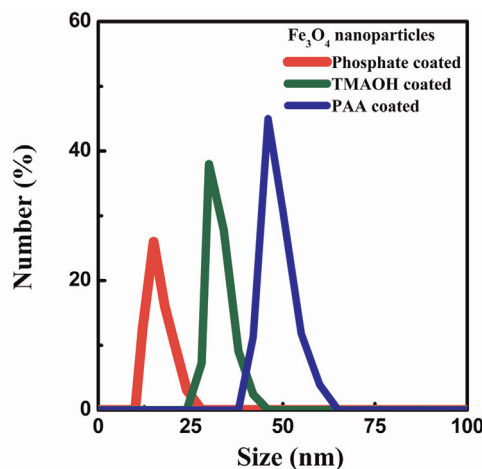


Fig. 1. Dynamic light scattering based characterization (size distribution) of three different magnetic nanofluids with PAA, TMAOH, Phosphate as nanoparticles surface coating agents.

of 2.5 G/s. In the absence of an external field ($B=0$ G) only a bright circular spot was observed. But on increasing the external magnetic field, a straight line like pattern is observed. Similar trend is observed in all three magnetic nanofluids. Nevertheless, features of the straight line pattern were slightly different for those three samples. For the PAA coated Fe_3O_4 nanofluid, the straight line pattern was formed within a magnetic field strength of 100 G, whereas, in the other two samples they were discernible above 100 G only. The straight line like pattern was observed at a lower external magnetic field for the TMAOH coated Fe_3O_4 nanofluid, compared to the phosphate coated nanofluid. What is the origin of these straight line patterns? The suspended magnetic nanoparticles acquire dipole moments ($m = (\pi/6)a^3\chi B$) in the presence of an external magnetic field [30,31]. Here, a is the diameter of the single bare magnetic nanoparticle, χ is the effective susceptibility of an individual nanoparticle, and B is the magnitude of external magnetic field. The anisotropic interaction energy U_{ij} between two identical, parallel, point dipoles is given by [21,31]

$$U_{ij}(r_{ij}, \theta_{ij}) = \frac{m^2\mu_0}{4\pi} \left(\frac{1 - 3\cos^2\theta_{ij}}{r_{ij}^3} \right) \quad (1)$$

where μ_0 is the magnetic permeability of free space, r_{ij} is the magnitude of the vector describing the distance between the centers of i th and j th nanoparticles, and θ_{ij} is the angle between the vector r_{ij} and the external field vector. The effective magnetic interaction between two magnetic nanoparticles is described by the coupling constant $L = -U/k_B T = \pi\mu_0 a^3 \chi^2 B^2 / 72 k_B T$ [21,31]. Here, k_B is the Boltzmann constant and T is the temperature. The magnetic nanoparticles in the dispersion self-assemble into structures that are aligned along the external magnetic field, when $L \gg 1$ [21,31]. Such field induced aggregation of magnetic nanoparticles into chainlike structures has been verified experimentally [32–34] and by computer simulation [35]. When light interacts with such chain like structures with their axis perpendicular to the direction of the incident light, a straight line pattern is formed [36].

The optical properties of particle aggregates in dispersion is different from that of solid particles, because the optical cross-sectional area of the particle aggregates in the former becomes larger than the bare solid particles of the same mass [37]. The external field induced evolution of transmitted light pattern (straight-line like), in magnetic nanofluid depends on the hydrodynamic diameter (d_h) of magnetic nanoparticles [36]. As the hydrodynamic diameter of the PAA coated nanoparticles was the highest, the straight line like pattern was fully evolved within an external magnetic field strength of 100 G. On the other hand, for the TMAOH and phosphate coated nanoparticles, straight line like patterns were observed at a higher external magnetic field. Because the coating layer thickness in all the three cases are nearly same (~ 1 – 1.5 nm) and the primary Fe_3O_4 nanoparticle size is ~ 10 nm, the observed hydrodynamic diameters indicates that 2–4 primary particles form aggregates upon coating with different functional groups.

Fig. 3 shows the normalized transmitted light intensity as a function of external magnetic field for different magnetic nanofluids. The inset of Fig. 3 shows the transmitted light pattern from the phosphate coated magnetic nanofluid ($d_h = 15$ nm) at an external magnetic field of 430 G. In general, the normalized transmitted light intensity as a function of external magnetic field shows similar behavior for all the three samples. Initially, the normalized transmitted light intensity increases and attains a maximum at a certain external magnetic field, called the first critical field (B_{C1}).

Beyond B_{C1} , the normalized transmitted light intensity decreases continuously and becomes a minimum at a second critical field (B_{C2}). The values of B_{C1} (and B_{C2}) for the magnetic nanofluids

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