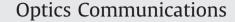
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Temperature dependent light transmission in ferrofluids

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ARTICLE INFO

Article history: Received 11 November 2014 Received in revised form 11 December 2014 Accepted 28 December 2014 Available online 30 December 2014

Keywords: Ferrofluid Nanofluid Rayleigh scattering Mie scattering Aggregation kinetics

ABSTRACT

We investigate the influence of temperature on the magnetic field induced light transmission in a kerosene based ferrofluid containing oleic acid coated Fe_3O_4 nanoparticles, where the direction of propagation of light is parallel to the direction of the external magnetic field. At a fixed temperature the transmitted light intensity is found to monotonically increase with incident wavelength due to reduced extinction efficiency at higher wavelength. The transmitted intensity decreases with external magnetic field due to enhanced scattering from the field induced linear chain like structures along the direction of the external magnetic field and due to the build-up of standing waves inside the scattering medium. The extinction of the field induced transmitted light intensity is found to occur at a lower external field as the sample temperature is lowered. The rate of extinction of normalized transmitted light intensity decreased linearly with increasing sample temperature due to slower field induced aggregation kinetics because of an increased Brownian motion of the suspended nanoparticles and a reduced coupling constant. The observed temperature dependent magneto-optical properties of magnetic nanofluids can be exploited for applications in optical devices.

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1. Introduction

Scattering and propagation of electromagnetic waves in magnetic colloidal dispersion have been an active research topic for the last few decades [1–5]. Magnetic colloidal dispersions such as ferrofluids and ferroemulsions are of immense importance both from fundamental and application point of view due to their distinct tunable properties in presence of an external magnetic field due to field induced structural ordering [6–12]. It has been known that the external magnetic field induced aggregation of magnetic nanoparticles in such dispersion can strongly affect the propagating light and gives rise to interesting optical effects [12,13]. These systems have found wide spread applications in optical sensors [14,15], smart cooling devices[16], tunable optical filters [17], tunable photonic devices [18–20], optical limiters [21] and magnetic field sensors [22,23].

The fundamental understanding of various parameters influencing the field induced aggregation in ferrofluid and ferroemulsion is important from practical applications point of view [2]. Field induced particle aggregation in magnetic nano-colloidal systems has been studied using numerous experimental techniques [24–26]. Recently the influence of surfactants on aggregation kinetics has been studied [27]. Using light scattering techniques it

http://dx.doi.org/10.1016/j.optcom.2014.12.073 0030-4018/© 2014 Elsevier B.V. All rights reserved.

has been shown that base fluid viscosity and field exposure time can also influence the field induced aggregation dynamics [28]. The field induced aggregation in magnetic nano-colloidal systems is mainly due to dipolar interaction between the dispersed magnetic nanoparticles which primarily depends on the applied magnetic field strength and particle diameters. Recently the authors have systematically investigated the effect of suspended particle size on field induced aggregation and reported an enhancement of dipolar interaction with increasing particles size [29]. Though numerous studies have considered the effects of dipolar interaction on field induced aggregation in magnetic nanocolloidal systems and Pu et al. studied the effect of temperature on the band gap properties of magnetic fluid based photonic crystals [30], to the best of our knowledge the effect of temperature on field induced aggregation and related magneto-optical properties has not been systematically studied earlier. In this paper, we have systematically studied the effect of temperature on the external field induced light transmission in kerosene based magnetic nanofluid.

2. Materials and experimental set-up

The experiments were performed using a stable colloidal system of magnetite (Fe_3O_4) particles coated with oleic acid and dispersed in kerosene [31]. Average diameter of particles and layer

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thickness of the organic coating were 6.5 nm and 1.5 nm, respectively. The suspensions used in our experiments had excellent long terms stability with no visible agglomerations even after prolonged application of very strong magnetic fields. The volume fractions (ϕ) was kept constant at 0.00155 throughout the experiments. The ferrofluid was taken in a cuvette of path length, L=10 mm and kept inside a Peltier based temperature controlled cuvette holder. The specimen temperature was varied using a programmable temperature controller. Experiments were performed at five different specimen temperatures, viz. 278, 288, 298, 303and 318 K. The cuvette holder was placed inside a solenoid in such a way that the direction of the incident light passing through the specimen is along the direction of the external magnetic field. A DC programmable power supply is used to power the solenoid. To record the transmission spectrum a standard fiber optic spectrometer (AvaSpec-2048, Avantes), with a tungsten halogen light source (usable wavelength range of 200-1100 nm) was used. The transmission spectrum was recorded in the wavelength range of 675-750 nm using a CCD linear array detector with 2048 pixels. The integration time and ramp rates were suitably optimized at each external magnetic field to record the spectrum. Typical schematic of the experimental setup is shown elsewhere.

3. Results and discussions

Fig. 1 shows light transmission through the ferrofluid as a function of incident wavelength at five different external magnetic fields B=0, 62, 130, 196, 264 G (\pm 2 G). The specimen temperature was kept constant at 278 K (\pm 0.5 K). The light transmission increases with incident wavelength for all the magnetic fields. As the Fe₃O₄ nanoparticle sizes are much lower than the incident wavelength ($\alpha \ll \lambda$) the light scattering can be described using Rayleigh's theory, where the scattering efficiency is inversely proportional to the fourth power of incident wavelength, i.e. $Q_{sca} \propto 1/\lambda^4$ [32]. Hence, the observed decrease in the scattering intensity with increasing wavelength is in good agreement with Rayleigh's theory, which explains the increase in the transmission of incident light with increasing wavelength in the ferrofluid. It can be further observed from Fig. 1 that the transmission of incident light is highest at B=0 G and it decreases with increase in

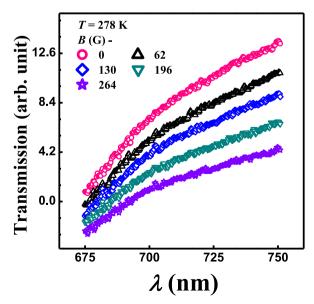


Fig. 1. Light transmission as a function of incident wavelength (λ) at different external magnetic fields (B=0, 62, 130, 196, 264 G) in magnetic nanofluid. Range of λ is 675–750 nm and specimen temperature was kept constant at 278 K.

the applied magnetic field. In the absence of an external magnetic field the oleic acid coated Fe_3O_4 nanoparticles are randomly dispersed in the carrier liquid due to Brownian motion of particles. On application of an external magnetic field the magnetic moments of the individual nanoparticles orient themselves along the direction of the external magnetic field and form linear chain like structures [6,33]. In the presence of an external magnetic field the magnetic field the magnetic coupling constant (Λ) between two nanoparticles can be described by the following equation [28,34]:

$$\Lambda = \frac{\pi\mu_0 d^3 \chi^2 H^2}{72k_B T}$$
(1)

Here, d, χ , H and k_BT are diameter of the nanoparticle, effective magnetic susceptibility, magnitude of the applied external magnetic field and thermal energy, respectively. The magnitude of Λ determines the dipolar attraction between the two nanoparticles and when $\Lambda \gg 1$, the dispersed magnetic nanoparticles undergo a disorder to order transition leading to the formation of linear chain like structures due to head-on aggregation of the dispersed nanoparticles along the direction of the external magnetic field. Formation of such linear chain-like structures under the influence of external magnetic field has been verified experimentally [26,35,36] and using numerical simulations [32]. The interaction of incident light with such linear chain like structures with their axis parallel to the direction of propagation of light gives rise to a ring like transmission spectrum [37]. The aggregation of nanoparticles increases with external magnetic field due to increase in the dipolar interaction leading to a larger scatterer size [38]. It has been earlier reported that on application of external magnetic field the magnetic nanoparticles form single chain like structures along the direction of external magnetic field up to a first critical field and beyond that zippering of chains takes place due to lateral aggregation and form bundles of nanochains [34]. The length of these chains increases with external magnetic field and the aggregation process depends on the strength of the field and exposure time. When the scatterer sizes are comparable with the incident light wavelength the scattering regime changes from Ravleigh and falls in Mie.

The transmission, absorption and distribution of the scattered light, during passage through a magnetic nanofluid depend on the nature of dispersed scatterers [39,40]. The total extinction efficiency (Q_{ext}) is the sum of scattering efficiency (Q_{sca}) and absorption efficiency (Q_{abs}) and can be expressed by the following equation [40]:

$$Q_{ext} = Q_{sca} + Q_{abs} \tag{2}$$

According to Mie scattering theory, Q_{ext} , Q_{sca} and Q_{abs} can be expressed by the following equations [32,40]:

$$Q_{ext} = \frac{2}{x^2} \sum_{n=1}^{\infty} (2n+1) \operatorname{Re} (a_n + b_n)$$
(3)

$$Q_{sca} = \frac{2}{x^2} \sum_{n=1}^{\infty} (2n+1)(|a_n|^2 + |b_n|^2)$$
(4)

$$Q_{abs} = \frac{4\varepsilon''}{x^2} \int_0^x \left\langle |\mathsf{E}|^2 \right\rangle x'^2 dx' \tag{5}$$

Here *E* is the electric field component of the incident light, ε'' is the imaginary part of the dielectric constant of Fe₃O₄ nanoparticles, a_n and b_n are the Mie scattering parameters which depend on the scatterer size parameter (x = ka, where *a* is the radius of the nanoparticle and $k = 2\pi/\lambda$).

The magnetic permeability (μ) increases with increasing magnetic field for ferromagnetic scatterers [41]. The enhancement of

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