

Birefringence of maghemite-based magnetic fluid in the highly concentrated regime

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Abstract

The transmitted light intensity of a series of ionic magnetic fluid samples based on maghemite nanoparticles was investigated using the typical birefringence experimental setup. The field dependence of the transmitted light intensity evolves from a peak-like profile to a typical second-order Langevin profile as the nanoparticle volume fraction reduces from 2.4% to 0.9% volume fraction. The onset of the peak-like profile as the nanoparticle volume fraction increases is accounted for by two points, namely the transmitted light first scales with a sine function and second the birefringence signal appears in the argument of the sine function. The observed negative differential transmitted light is explained as one brings the above-mentioned sine dependence into the nowadays used model for the analysis of birefringence in magnetic fluids.

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1. Introduction

Magnetic fluids (MFs) are ultra-stable colloidal systems consisting of monodomain magnetic nanoparticles suspended in organic or inorganic liquid carrier, while the nanoparticle concentration may go up to 10% volume fraction [1]. In ionic MFs the colloidal stability is mainly achieved through the electrostatic repulsion among the positively or negatively charged nanoparticles [2]. In that case the surface charge density and the ionic strength of the medium need to be precisely controlled in such a way that electrostatic repulsion does overcome the magnetic dipolar attraction that favors nanoparticle aggregation. In the usual ionic ferrite-based MF low pH aqueous medium sets in a positive charge density, due to proton transfer from the medium to the nanoparticle surface. By contrast, negative surface charge density is obtained at high pH values as a result of the proton transfer from the nanoparticle surface out to the aqueous medium, thus leaving behind negatively charged oxygen atoms at the nanoparticle surface. A microscopic model for the pH-dependent surface charge–discharge process in metal oxide-based ionic MFs

involves the proton tunneling across a strongly bonded water layer around the nanoparticle surface [3]. In surface-coated MFs, however, molecular species attached to the nanoparticle surface maintain colloidal stability, once steric repulsion among the coated magnetic nanoparticles overcomes the magnetic dipolar attraction [4]. The outer ends of the surface molecular coating in surface-coated MFs may be either polar or non-polar, thus resulting for instance in stable aqueous-based MFs or stable hydrocarbon-based MFs, respectively. In addition to the nanoparticle electrostatic repulsion and dipolar attraction competition between thermal energy and Van der Waals interaction plays an important role in the magnetic colloidal stability as well. While thermal energy favors colloidal stability by taking magnetic nanoparticles apart Van der Waals interaction tend to stick nanoparticles together favoring aggregation.

A wide variety of experimental techniques has been used to investigate the fundamental properties of ionic and surface-coated MFs. The traditional techniques used in the investigations are the high-resolution microscopy [5], X-ray diffraction [6], static magnetic birefringence [7], and magnetometry [8]. Since the last decade magnetic resonance has been systematically used as a valuable tool in the investigation of several properties of MFs [9]. The magnetic resonant center used in the

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investigation of MFs is either the magnetic nanoparticle itself [10] or a paramagnetic probe intentionally introduced as a dopant in the carrier fluid [11]. Effects of particle concentration [12], ionic strength [13], particle size and temperature [14] have been investigated using the nanoparticle itself as the resonant probe. More recently Raman spectroscopy emerged as a powerful technique to investigate qualitatively as well as quantitatively nanoparticle surface chemisorbed species. Raman spectroscopy has been used to investigate the interactions of the molecular species chemisorbed at the nanoparticle surface of copper and zinc-ferrite ionic MFs [15]. Also, the single surface-coated MF based on dodecanoic acid (first coating) and diluted in hydrocarbon and the double surface-coated MF based on dodecanoic acid (first coating) following an ethoxylated polyalcohol (second coating) and peptized in water were investigated using Raman spectroscopy [16]. Even more recently, photoacoustic spectroscopy has been introduced as a very promising experimental technique for investigation of MF samples [17]. Despite the simplicity of the photoacoustic spectroscopy the experimental data taken from thin MF films revealed important details regarding the core nanoparticle properties [18], the surface nanoparticle structure [19], and the nanoparticle coating layer characteristics [20].

Static magnetic birefringence (SMB) measurements have been long used as a standard way to investigate magnetic fluid samples [21–25]. This is simply because when polarized light is shining normal to a thin film of magnetic fluid with its electric polarization parallel to the magnetic field one observes a higher refractive index (positive birefringence) than the one experienced by light polarized perpendicular to the field. Though static magnetic birefringence in MFs has been described by many authors over the last five decades there are still many open points regarding the origin and the physical description of the effect. Yet, the observation of the effect in the limit of very concentrated MF samples has been made possible only recently after the synthesis of highly transparent samples [26]. In this study we present static magnetic birefringence measurements as a function of the nanoparticle concentration close to the upper limit of concentrations. The first approach for the interpretation of the SMB experimental data is presented as well.

2. Static magnetic birefringence

Orientation of a slightly non-spherical isolated nanoparticle accounts for very small SMB signal only, while large SMB signal experimentally observed in MFs is more likely due to field orientation of preexisting aggregates [24]. Though nanoparticle aggregation also influences the dichroism of MF samples the effect is typically one order of magnitude smaller than birefringence [24]. This means that dichroism usually adds little deviations in the birefringence data. Evidences that nanoparticle aggregation (chain-like structure) respond for the optical birefringence in MFs have been long available both from theoretical considerations [27] as well as from experimental observations [28,29]. Monte Carlo simulation showed that the large SMB signal is mainly caused by the combined

effects of field-dependent chain-like formation and field-induced orientation of chains [30]. Xu and Ridler [31] have succeeded in explaining the SMB data obtained by Sakhnini and Popplewell [32] based on the oscillating dipole-interaction anisotropy concept, which includes contributions of isolated nanoparticles (monomers) and nanoparticle aggregates (dimers, trimers, ...). The temperature, particle size, and concentration dependence of the zero-field birefringence observed in ionic MFs were explained assuming preexisting dimers in the sample [33]. Irreversibility of the zero-field birefringence in MFs upon temperature reversal was also explained through the picture of a thermal disruption of dimers [34]. In fact, earlier SMB experiments performed in the low-field region have showed the signature of the zero-field birefringence [35].

On the other hand evidences that the field (H) dependence of the magnetic permeability (μ) in a multi-domain ferrimagnetic-based material follows a sort of asymmetric bell-shaped curve have been long available in the literature [36]. Likewise, the field dependence of the magnetic susceptibility (χ) follows an asymmetric bell-shaped diagram [36]. The permeability curve peaks at the field value corresponding to the knee of the $B-H$ diagram, i.e., at the field where magnetization reaches its maximum value. In fact, the linear relationship between magnetic permeability and magnetic field, in the very low-field regime, is the well-known Rayleigh law [37]. From the microscopic viewpoint, however, the field dependence of the magnetic permeability depends upon two distinct mechanisms, namely domain wall motion and domain magnetic moment rotation [38]. Nevertheless, it has been found experimentally that a more generalized dependence of the relative magnetic permeability upon the magnetic field would be written as $\mu_r = \alpha + f(H)$ [39]. Note that domain magnetic moment rotation does take place in a chain-like structure containing single-domain ferrimagnetic nanoparticles. In fact, chain-like structures can be seen as extrinsic multi-domain effective particles, i.e., interaction domains. Therefore, it is quite obvious that rotational magnetic permeability (μ_Q^r) as well as rotational magnetic susceptibility (χ_Q^r) may be associated to a linear chain of monodomain magnetic nanoparticles consisting of Q single units ($Q > 1$). In addition, it is quite reasonable to assume that a bell-shaped function would describe the rotational magnetic susceptibility of the chain-like structure, i.e., $\chi_Q^r = M_Q / H_A^Q$, where M_Q and H_A^Q are the magnetization and anisotropy field associated to the chain-like structure consisting of Q single units (Q -chain), respectively. Among the asymmetric functions used to describe the field dependence of the reduced rotational susceptibility the log-normal one is a good candidate. Note that the log-normal function has been successfully used to describe the anisotropy field distribution associated to uniaxial nanomagnetic particles [40]. Indeed, the magnetization associated to the Q -chain will be taken as $M_Q = [1 + \beta C_Q P_Q(H)] M_S$, with $P_Q(H)$ given by the log-normal function, C_Q representing the fraction related to each Q component, and β a constant obtained from the fitting of the SMB data. Complete determination of $P_Q(H)$ is obtained from the modal field ($\langle H_Q \rangle$) and zeroth-order logarithmic standard deviation (σ_Q) associated to the particular Q -chain.

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