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The influence of long range interparticle correlations on the magnetically induced optical anisotropy in magnetic colloids

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ABSTRACT

In this paper we develop a theoretical model for the magnetically induced optical anisotropy in dense magnetic colloids made of spherical and un-aggregated magnetic monodomain nanoparticles. Both dipole–field and dipole–dipole magnetic and electric interactions between the magnetic monodomain particles are taken into account in the Hamiltonian of the system. Using the pair correlation function in a colloidal suspension of magnetic nanoparticles developed by Ivanov and Kuznetsova (2001) [11], the complex dielectric constant of a magnetic colloid is modeled as a function of the light polarization direction, the magnetic field intensity and magnetic particle concentration and diameter. The two main features of the model are that, on the one hand, it predicts the possibility of magnetically induced optical anisotropy in dense magnetic colloids made of spherical and un-aggregated monodomain nanoparticles, and on the other hand, unlike the existing models for diluted samples, it predicts a non-linear dependence of dichroism and birefringence on magnetic particle concentration.

1. Introduction

The magnetic colloids, also known as magnetic fluids or ferrofluids, are colloidal suspensions of magnetic monodomain nanoparticles, sterically or electrostatically stabilized and dispersed in a carrier liquid [1]. Owing to the permanent magnetic dipoles of the magnetic nanoparticles, the magnetic colloids are susceptible to undergo a uniaxial structural anisotropy under the action of an external magnetic field. The magnetically induced structural anisotropy is responsible for the magneto-optical phenomena in magnetic colloids. Therefore, the theoretical models for the magneto-optical phenomena in magnetic colloids, among which the most extensively studied are the transverse birefringence and dichroism, must be founded on specific structural assumptions. The direct link between the structure and the optical properties of the magnetic colloids makes the experimental investigation of the magneto-optical phenomena a candidate of choice for testing the theoretical models aimed at describing the structure of the magnetic colloids.

In diluted magnetic colloids, where the magnetic interparticle interactions can be neglected, the magnetically induced optical anisotropy is caused by the orientation of prolate particles with their shape anisotropy axis along the field, due to

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the coupling between the particle magnetic moment and the particle easy magnetization axis, which for particles made of soft magnetic materials like magnetite coincides with the shape anisotropy axis [1]. When the magnetic moment is frozen along the shape anisotropy axis one speaks about Brown particles and when the magnetic moment is coupled to the shape anisotropy axis by means of a finite magnetic shape or crystalline anisotropy energy one speaks about Néel particles [1]. Theoretical models for magnetically induced optical anisotropy in diluted magnetic colloids with Brown and Néel particles were developed and published, among others, in Refs. [2–4] and in Ref. [5] respectively. Two common features of these models are that, on the one hand, they predict the possibility of the magnetically induced optical anisotropy only in magnetic colloids made of non-spherical particles (most often approximated as prolate rotation ellipsoids), and on the other hand, that the magnitude of the magnetically induced optical anisotropy is linearly dependent on the sample's particle concentration.

The theoretical models of the magnetically induced optical anisotropy in concentrated magnetic colloids must take into account the synergy between the dipole–field and dipole–dipole magnetic interactions which leads to two main, not necessarily disjoint, structural change hypotheses: particle agglomeration and long range interparticle correlation. The influence of the particle agglomeration on the magnetically induced dichroism and birefringence in magnetic colloids, proposed by Taketomi [6], was developed in Refs. [7,8], where the magnetically induced optical anisotropy originates from the formation of chain-like aggregates of magnetic nanoparticles that are large enough such that the magnetic dipole–dipole interaction overcomes the thermal energy. When the magnetic particles are small enough not to lead to thermodynamically stable aggregates, the coupling between the magnetic dipole–field and the long range magnetic dipole–dipole interactions lead to a magnetically induced anisotropy of the particle pair correlation function, due to the fact that the particles will attract and repel each other in the directions parallel and perpendicular to the external field respectively. The influence of the long range interparticle correlation on the magnetically induced dichroism and birefringence in magnetic colloids was theoretically described in Refs. [9,10] respectively. These two classes of models, based on particle agglomeration and long range interparticle correlation, predict the possibility of magnetically induced optical anisotropy in magnetic colloids made of spherical particles as well as a nonlinear dependence of dichroism and birefringence on the particle concentration.

The interest in the magneto-optical models based on the long range interparticle dipole–dipole interactions is twofold: the first is that they prove the possibility of the magneto-optical phenomena in magnetic colloids made of spherical and independent particles, and the second is that they allow for the estimation of the influence of the magnetic long-range correlations on the magnitude of the magneto-optical phenomena in concentrated magnetic colloids with nonspherical particles and/or particle clusters. Using the pair correlation function in a colloidal suspension of magnetic nanoparticles developed by Ivanov and Kuznetsova [11], Popescu and Socoliuc [9] derived the magnetic colloid absorption coefficient for light polarized parallel and perpendicular to the magnetic field, which allowed for the description of the magnetically induced linear dichroism dependence on magnetic particle size and concentration. It was found that the dichroism has a nonlinear dependence on the magnetic particle concentration. Zubarev and Iskakova [10] studied the magnetically induced linear birefringence. It was found that the influence on the magnetically induced linear birefringence of the long range dipole–dipole interactions is of the same order of magnitude as that of both rotation and particle agglomeration mechanisms.

In this paper we generalize the model published in Ref. [9], which was limited to parallel and perpendicular polarization states of the light as well as to the unrealistic hypothesis of neglecting the nonmagnetic and the surfactant layers at the surface of the coloidal magnetic nanoparticles, to the influence of the magnetic field on the complex refractive index of magnetic colloids for light polarized to an arbitrary angle with respect to the external magnetic field direction and also make the model explicitly dependent on the thickness of the surfactant layer. Numerical evaluations of the model are presented in order to investigate the influence of the magnetic field intensity, particle diameter and concentration, surfactant layer thickness and light polarization angle on the absorption coefficient of concentrated magnetic colloids made of spherical particles.

Particular attention is given to the dependence on the magnetic field intensity and particle concentration of the absorption coefficient of light polarized at $\theta_c = 54.75^\circ$ ($\cos(\theta_c)^2 = 1/3$) with respect to the external magnetic field direction. Kopcansky and co-workers [12] showed that the absorption of light polarized at θ_c is independent of the magnetic field intensity in magnetic colloids where the rotation mechanism is responsible for the magnetically induced optical anisotropy. On the other hand, we found experimental evidence that the absorption coefficient of light polarized at θ_c is diminishing with increasing both magnetic field intensity and sample particle volume fraction in an ultra-stable magnetic colloid with magnetite particles stabilized with oleic acid and dispersed in transformer oil (details on the experiment and the results will be published elsewhere). Therefore, in order to find the correct explanation for the experimental findings it is important to assess the influence of the long range interparticle correlations on the magnetic field dependence of the absorption coefficient of light polarized at θ_c.

2. Theoretical basis

In this section we derive the complex index of refraction of a magnetic colloid made of identical magnetic nanoparticles assumed to have a spherical shape and made of a material with isotropic dielectric properties.