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Ferromagnetic resonance in a dilute suspension of uniaxial superparamagnetic particles

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ABSTRACT

A consistent theory of ferromagnetic resonance in a dilute suspension of superparamagnetic particles with uniaxial anisotropy of arbitrary strength is presented. The developed approach is used for studying the high-frequency response of a magnetic fluid at different temperatures. It is shown that in a certain temperature interval the absorption line splits into two components. The width of this interval is essentially dependent on the magnitude of the particle anisotropy.

1. Introduction

In the last decade magnetic nanoparticles have found broad application in a variety of systems, from solar cells to living organisms [1]. Evidently, efficient use of nanomagnets is impossible without detailed knowledge of their properties. One of the most powerful tools to do that, is ferromagnetic resonance (FMR). With the burst out of nanotechnologies, the theory of FMR has got a boost, because the standard approach (the athermic Landau-Lifshitz-Gilbert equation) has proved to be inappropriate. The crucial issue is the superparamagnetism of those nanoparticles: the entrainment of their magnetic moment in thermofluctuation process due to which the response of such particles depends essentially on temperature. In the recent paper [2], we have presented a theory of FMR in uniaxial nanoparticles, embedded in a solid matrix (a polymer, for example). The FMR of the particles, dispersed in a liquid medium, is no less interesting though. The demand for a robust theory is especially high because of the progress in experimental work, see e.g. [3–8]. Meanwhile, up to now there exist only two approximate methods: the quantum theory by Noginova et al. [3] and the kinetic approach proposed by Raikher and Stepanov [9]. In methodical scheme [3], a nanoparticle is considered as a giant exchange cluster with a discrete set of states. The absorption spectrum is calculated by combining all the transitions between the allowed levels. It is clear that such a description is applicable only for the particles whose diameters do not exceed just few nanometers. The Raikher-Stepanov method is based on the Brown kinetic equation for the orientational distribution function of the magnetic moment [10]. The advantage of that approach is that it consistently takes into account the thermal fluctuations of the magnetic moment and the rotatory mobility of the particles. However, the use of the scheme [9] is justified only for the particles whose effective anisotropy field is much lower than the magnetizing field of a spectrometer. In the present paper, the kinetic approach is generalized for magnetic fluids (ferrocolloids) where the particles possess uniaxial magnetic anisotropy of arbitrary strength.

2. Ferromagnetic resonance

Consider a magnetic fluid based on single-domain particles with easy-axis (uniaxial) anisotropy under assumption that the volume fraction of magnetic material is low (dilute suspension), so that interactions between the dispersed particles might be neglected. In that case, the magnetic energy of each particle

$$U = -M_s v \boldsymbol{e}\cdot\boldsymbol{H} - Kv(\boldsymbol{e}\cdot\boldsymbol{n})^2, \tag{1}$$

includes only the energy of magnetic moment in external field $\boldsymbol{H}$ (first term) and the anisotropy energy (second term). In expression (1) the quantity M_s is the particle magnetization, v its volume, K anisotropy constant, while $\boldsymbol{e}$ and $\boldsymbol{n}$ are unit vectors of magnetic moment $\boldsymbol{\mu} = M_s v\boldsymbol{e}$ and easy magnetization axis, respectively.

To observe ferromagnetic resonance, one has to subject the suspension to a time-independent $\boldsymbol{H}_0$ and radio-frequency $\boldsymbol{h}(t)$ fields, whose directions are mutually orthogonal. If the suspension is frozen, the dispersed particles are immovable, and for each of them the angle ψ between the direction of the magnetizing field and the easy magnetization axis is fixed. In that situation, the response of the system to a probing field $\boldsymbol{h}(t)$ could be found by calculating the partial dynamic susceptibility for each ψ and averaging the result with a certain

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distribution function $f(\psi)$ that is independent of the applied field. For instance, in the isotropic case this function is constant $f(\psi) = 1/4\pi$. The corresponding problem was solved in Ref. [2].

A particle suspended in a liquid, as it is free to rotate, tends to reorient in such a way that its magnetic moment $\boldsymbol{e}$, anisotropy axis $\boldsymbol{n}$ and magnetizing field $\boldsymbol{H}_0$ would be co-aligned. Full collinearity of those vectors is not attainable however, because of thermal fluctuations, i.e., the rotatory Brownian motion. For such a situation, if to take into account the intraparticle magnetic interactions, it becomes clear that the degree of orientational order of easy magnetizaton axes of the particles—it is described by function $f(\psi)$—should depend on the ratio of the magnetic anisotropy energy to thermal one. In view of that, the theory of FMR in a magnetic fluid should combine solutions of two problems. First, it should describe the resonance response of the superparamagnetic particle with the given angle ψ between the anisotropy axis and the magnetizing field. Second, it should specify the orientational distribution function $f(\psi)$ at any external field. Then the averaging of the partial FMR spectra by means of that function must be done.

With respect to the first problem, it suffices to treat it in linear approximation. Indeed, the exciting field $\boldsymbol{h}(t)$ in FMR experiments has a circular frequency $\omega \sim 60{\cdot}10^9$ rad/s. Normally, it is not feasible to generate a field of large amplitude at ultrahigh frequencies, and that is why for most experiments $h(t) \ll H_0$. That condition allows one to present the magnetic moment of a particle as a sum of equilibrium value $\boldsymbol{e}_0$ and a small perturbation $|\delta\boldsymbol{e}| \ll |\boldsymbol{e}_0|$.

The theory of linear FMR in a superparamagnetic particle is given in [2]. As the magnetic moment of a single-domain particle undergoes thermal fluctuations, its state is described by the orientational distribution function $W(\boldsymbol{e}, \boldsymbol{n}, t)$ which obeys the Brown equation [10]:

$$2\tau_D \frac{\partial W}{\partial t} = \hat{\boldsymbol{J}} \cdot W \left(\hat{\boldsymbol{J}} + \frac{1}{\alpha} \nabla \right) \left(\ln W + \frac{U}{k_B T} \right); \tag{2}$$

here $\nabla = \frac{\partial}{\partial \boldsymbol{e}}$ is the gradient operator on the surface of a unit sphere, $\hat{\boldsymbol{J}} = \boldsymbol{e} \times \nabla$ the operator of infinitesimal rotation, U magnetic energy (1), α precession damping constant, γ gyromagnetic ratio, and $\tau_D = (1 + \alpha^2)\mu/(2\alpha\gamma k_B T)$ the reference time of rotary diffusion of the magnetic moment. Evidently, the equilibrium solution of that equation is a Boltzmann-type function.

In a typical FMR setup, the derivative with respect to the magnetizing field of the energy absorbed during a cycle of the probing field is measured. The theoretical analog for that quantity is found by solving the Brown Eq. (2). We introduce a spherical coordinate system with the polar axis along the magnetizing field and expand the distribution function in a series of normalized spherical harmonics:

$$W(\vartheta, \varphi, t) = \sum_{l=0}^{l=\infty} \sum_{k=-l}^{k=l} b_{l,k}(t) Y_{l,k}(\vartheta, \varphi), \tag{3}$$

defined as

$$Y_{l,k}(\vartheta, \varphi) = (-1)^k \sqrt{\frac{(2l+1)(l-k)!}{4\pi(l+k)!}} P_{l,k}(\cos\vartheta) e^{ik\varphi}, \; -l \le k \le l,$$
$$Y^*_{l,k} = (-1)^k Y_{l,-k}, \tag{4}$$

where $P_{l,k}$ are associated Legendre polynomials and (ϑ, φ) are the angle coordinates of vector $\boldsymbol{e}$. As the spherical functions are orthonormalized, the coefficients $b_{l,k}(t)$ in (3) turn out to be statistical moments of the distribution function:

$$b_{l,k} = \int W(\vartheta, \varphi) Y^*_{l,k} \sin\vartheta d\vartheta d\varphi = \langle Y^*_{l,k} \rangle; \tag{5}$$

as W is a real function, they satisfy the relation $b_{l,-k} = (-1)^k b^*_{l,k}$. Substitution of series (3) into (2) leads to a set of recurrence relations for the variables $b_{l,k}(t)$. Before solving this set, let us introduce nondimensional quantities. The strengths of the magnetizing and probing fields are rendered by parameters $\boldsymbol{Q} = \gamma \boldsymbol{H}_0/\omega$ and $\boldsymbol{q}(t) = \gamma \boldsymbol{h}(t)/\omega$, while the anisotropy and temperature are characterized by $\varepsilon = \gamma K/\omega M_s$ and $\xi_L = \mu\omega/\gamma k_B T$, respectively. Under condition $q \ll Q$, any moment $b_{l,k}(t)$ is presented as a sum of the equilibrium value and small non-equilibrium perturbation: $b_{l,k} = b^0_{l,k} + \delta b_{l,k}(t)$. Assuming $\delta b_{l,k} \ll b^0_{l,k}$, we linearize the derived set of equations and make its Fourier transform. That yields a set of linear algebraic equations for Fourier coefficients $\delta b^\omega_{l,k}$, whose combinations determine the dynamic magnetic moment of the particle as

$$\langle \delta e^\omega_x \rangle = \sqrt{\frac{2\pi}{3}} (\delta b^\omega_{1,-1} - \delta b^\omega_{1,1}); \langle \delta e^\omega_y \rangle = -i\sqrt{\frac{2\pi}{3}} (\delta b^\omega_{1,1} + \delta b^\omega_{1,-1}); \langle \delta e^\omega_z \rangle = \sqrt{\frac{4\pi}{3}} \delta b^\omega_{1,0}. \tag{6}$$

Let the harmonic exciting field be right-hand circularly polarized and perpendicular to the magnetizing one. In that case, full information about the resonance properties of the system is delivered by the dynamic susceptibility $\chi_+ = \delta e^\omega_+ / q^\omega_+$ that defines the relation between circular components of the magnetic moment $\delta e^\omega_+ = \delta e^\omega_x + i\delta e^\omega_y$ and the probing field $q^\omega_+ = q^\omega_x + iq^\omega_y$. The derivative of imaginary part of the susceptibility, $d\chi_{\prime\prime +}/dQ$, taken as a function of the field Q, is the theoretical analog of the FMR spectrum, recorded in an experiment.

In a magnetic fluid the distribution of anisotropy axes of the particles is a tunable characteristic. Since the probing field is weak, to a first approximation that distribution is determined only by the magnetizing field Q and, thus, can be regarded as stationary. This means that the probability density $f(\psi)$ of finding a particle whose anisotropy axis is inclined under angle ψ to the magnetizing field, could be found by integrating the equilibrium distribution function $W(\boldsymbol{e}, \boldsymbol{n}, t)$ over all possible directions of the magnetic moment $\boldsymbol{e}$. Such a calculation was accomplished in Ref. [9], where it was demonstrated that $f(\psi)$ could be taken in the form

$$f(\psi) = \frac{1}{4\pi} \left[1 + I^{-1}_{\frac{1}{2}}(\xi) \sum_{k=1}^{\infty} (4k+1) I_{2k+\frac{1}{2}}(\xi) R_{2k}(\sigma) P_{2k}(\cos\psi) \right]; \tag{7}$$

where parameters $\xi = \frac{\mu H}{k_B T}$ and $\sigma = \frac{Kv}{k_B T}$ are, respectively, the Zeeman and anisotropy energies measured in temperature units. The functions $P_{2k}(x)$ in (4) are Legendre polynomials of order $2k$, while $I_{2k+\frac{1}{2}}(\xi)$ are modified Bessel functions of semi-integer index; $R_{2k}(\sigma)$ defined as

$$R_{2k}(\sigma) = \frac{\int_0^1 P_{2k}(x) e^{\sigma x^2} dx}{\int_0^1 e^{\sigma x^2} dx}. \tag{8}$$

In Fig. 1, the orientational distribution function $f(\psi)$ of easy magnetization axes is shown for different values of temperature (a) and anisotropy (b). As seen from Fig. 1a, cooling of the system leads to orientational ordering (texturing) of the suspension, i.e., to growth of the number of particles, whose axes are aligned with the external field. At any fixed temperature, the same effect takes place if to increase the particles anisotropy, see Fig. 1b.

3. Results

The above-outlined method was used to model the FMR spectra of a dilute magnetic fluid. In Fig. 2 the absorption lines corresponding to different values of anisotropy parameter $\varepsilon = 0.1$ (a), 0.3 (b), 0.6 (c) and for different ξ_L's are shown. At high temperature ($\xi_L = 1$, dash-dot lines) the resonance lines are very much the same as those of an ensemble of randomly oriented particles, cf. Fig. 5 from Ref. [2]. That result is completely expectable, since at high temperature, as Fig. 1 shows, the distribution of anisotropy axes in a magnetic fluid is by large isotropic.

At small anisotropy ($\varepsilon = 0.1$, Fig. 2a), the temperature decrease, i.e., growth of parameter ξ_L, at first, results in a slight shift of the spectrum towards greater fields. However, starting with certain temperature, the