Temperature-dependent dynamical behavior of nanoparticles as probed by ferromagnetic resonance using Landau-Lifshitz-Gilbert dynamics in a classical spin model

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The dynamical response of nanoparticles as probed by ferromagnetic resonance (FMR) is studied within a classical spin model using Landau-Lifshitz-Gilbert dynamics. The FMR signal is calculated numerically for different temperatures. The dependence of both the shift of the resonance signal and the linewidth on temperature is obtained, yielding information about the microscopic parameters of the particles.

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I. INTRODUCTION

Magnetic colloidal nanoparticles are currently of great interest both from a fundamental point of view and from expected technological perspectives.[1–3] Of particular interest are the material parameters of these particles which may be dramatically different from the bulk parameters. The reduced symmetry of the surface atoms, for instance, is expected to lead to a strong enhancement of the anisotropy energy. Another source of an enhancement of the anisotropy energy leading to an enhanced coercivity of nanoparticles is exchange coupling to an antiferromagnetic substrate.[4] An experimental tool for measuring anisotropy energies is ferromagnetic resonance (FMR), which has been applied with great success in the past, in particular to thin ferromagnetic films.[5]

For these ferromagnetic films bulk and surface contributions can be separated since they contribute differently to the elements of the susceptibility tensor which are measured in FMR. For zero temperatures exact results are available for the susceptibility within a model in which the ground-state magnetization is treated as a single "macrospin."[6] From these results the different contributions to the anisotropy energy can be obtained. An naive extension of these results to finite temperatures consists of a replacement of the energy by a mean-field-type free energy leading to temperature-dependent anisotropy energies from which the microscopic parameters can be obtained by extrapolating to zero temperature. Thus this approach again leads to a "macrospin" with a reduced magnetization due to thermal fluctuations. Note that more sophisticated results for the dynamics of ferromagnetic systems have been obtained by Garanin[7] for systems without anisotropy.

For ferromagnetic nanoparticles the situation becomes more complicated since thermal fluctuations not only lead to a reduction of the magnetization of the nanoparticles but also to random fluctuations of its direction around its easy axis which affects physical quantities like the magnetic susceptibility. Thus, depending on the size of the particle one has to distinguish two different temperature effects: thermal fluctuations of the moments of the atoms within the particle resulting in a temperature-dependent reduced net magnetization of the particle and thermal fluctuations of this net magnetization around its easy axis. The first of these effects depends on the ratio T/J where J denotes the exchange interaction between atoms within the particle and T the temperature while the second one primarily depends on the size of the particle. In the experiments reported in Ref. 8 on single-domain particles the ratio T/J is such that the magnetization is practically saturated but the particles are so small that the thermal fluctuations of the net magnetization are important. In this situation the nanoparticle again can be described by a "macrospin" coupled to a heat bath. This is the situation we want to study in the present paper. We assume that the dynamics of the particles is governed by the stochastic Landau-Lifshitz-Gilbert (LLG) equation which takes the temperature fluctuations fully into account.[9] It is the aim of the present contribution to study their influence on the FMR signal in detail.

The dynamics of independent magnetic moments with LLG dynamics has been studied before. The paper closest to ours in spirit is that of Ref. 10 in which the dynamical susceptibility was calculated numerically starting from the LLG equations for various sets of system parameters. Unfortunately, from these results a rather complete picture of the dependence of the resonance field and the width of the resonance line on temperature cannot be obtained since fields (or external frequencies) are not varied systematically. Other approaches concentrate primarily on the derivation of temperature-averaged quantities—i.e., the distribution function for the moments[7]—or they treat temperature effects on a phenomenological basis.[11,12] Thus, to the best of our knowledge, a systematic study of the ferromagnetic resonance of nanoparticles taking temperature fluctuations fully into account is not available.

In the experiments mentioned above the anisotropy axis of the nanoparticles most likely are randomly distributed and additionally there is a size distribution. To entangle the effects coming from this structural disorder from the temperature effects is rather difficult. Therefore in a first step to understand temperature effects on linewidths and line shifts of the FMR signal we consider for simplicity in the present paper nanoparticles all having the same magnetic moment μ_s and the same uniaxial anisotropy energy, leaving the more complicated problem of temperature effects in a random assembly of nanoparticles to a future publication.[13] Note that when thinking about applications a common uniaxial anisotropy axis for all particles is essential. Experimentally this challenging goal has not yet been reached.

The main result of the present paper is a detailed numerical calculation of the shift of the FMR resonance field as

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function of temperature taking thermal fluctuations fully into account. It is shown that this shift decreases with increasing temperature proportional to m_0^2 where m_0 denotes the equilibrium magnetization of the particles at resonance. The decrease with temperature is in qualitative agreement with experiments.[8] Additionally we show that the FMR linewidth strongly depends on temperature, making it difficult to determine the Gilbert damping parameter from FMR experiments.

II. MODEL

The Hamiltonian of a nanoparticle is given by

$$\mathcal{H} = -\mu_s \mathbf{S}\cdot\mathbf{B} - \mu_s \mathbf{S}\cdot\mathbf{H}(t) - DS_z^2, \tag{1}$$

where $\mathbf{S}=\boldsymbol{\mu}_s/\mu_s$ denotes a unit vector parallel to its magnetic moment, $\mathbf{B}$ the external static magnetic field, $\mathbf{H}(t)$ the high frequency field, and D the anisotropy energy. The static field is applied parallel to the z direction while the high-frequency (HF) field is given by

$$\mathbf{H}(t) = H_{hf}\cos(\omega t)\mathbf{e}_x, \tag{2}$$

where $\mathbf{e}_x$ denotes a unit vector perpendicular to the anisotropy axis.

If the nanoparticles are well separated, their interaction which is of dipole type is small and will be neglected in the following.

The spin dynamics is governed by LLG equation

$$\frac{\partial \mathbf{S}}{\partial t} = -\frac{\gamma}{(1+\alpha^2)}\mathbf{S}\times\{\mathbf{B}_e(t) + \alpha[\mathbf{S}\times\mathbf{B}_e(t)]\}, \tag{3}$$

with the effective field $\mathbf{B}_e(t) = -\frac{1}{\mu_s}\frac{\partial \mathcal{H}}{\partial \mathbf{S}} + \zeta(t)$ which contains the external fields and a thermal noise $\zeta(t)$ which is Gaussian distributed with zero mean and correlator:

$$\langle \zeta_i(0)\zeta_j(t)\rangle = \delta_{ij}\delta(t) 2\alpha k_B T/\mu_s\gamma. \tag{4}$$

γ denotes the gyromagnetic ratio, α denotes a Gilbert damping parameter, i,j are Cartesian components, and the other symbols have their usual meaning. The power absorbed in a FMR experiment is given by

$$P = -\frac{1}{t_0}\int \mathbf{S}(t)\cdot\frac{\partial \mathbf{H}(t)}{\partial t}\mathrm{d}t, \tag{5}$$

where t_0 denotes the measuring time and the integral runs over a time interval of length t_0 in the stationary state.

In experiments very often the frequency ω is kept fixed while the strength of the applied static field is varied. For an easier comparison with experiments we follow this line in the present paper. The fixed frequency ω is expressed in terms of the resonance field $B_L=\omega/\gamma$ at $D=0$, $T=0$. Introducing then the reduced units $b=B/B_L$, $h=H_{hf}/B_L$, and $\tau=\omega t$ the LLG equation can be rewritten as

$$\frac{\partial \mathbf{S}}{\partial \tau} = -\frac{1}{(1+\alpha^2)}\mathbf{S}\times\{\mathbf{h}_e(\tau) + \alpha[\mathbf{S}\times\mathbf{h}_e(\tau)]\}, \tag{6}$$

where the effective field is now given explicitly by

$$\mathbf{h}_e(\tau) = b\mathbf{e}_z + h\cos(\tau)\mathbf{e}_x + 2dS_z\mathbf{e}_z + \Theta(\tau), \tag{7}$$

with

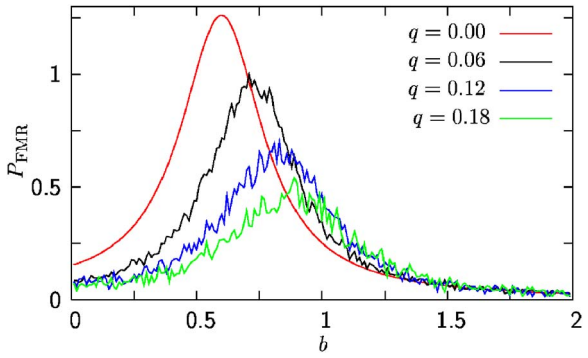

FIG. 1. (Color online) Absorbed FMR power (arbitrary units) vs reduced magnetic field b for different values of reduced temperature q.

$$\langle \Theta_i(0)\Theta_j(\tau)\rangle = \delta_{ij}\delta(\tau)q. \tag{8}$$

Thus the physics only depends on the reduced anisotropy energy d, $d=D/\mu_s B_L$, and the reduced temperature q, $q=2\alpha k_B T/\mu_s B_L$.

The experiments reported in Ref. 8 were done on $Fe_{0.7}Pt_{0.3}$ nanoparticles with a mean diameter of 2.3 nm. The magnetic moment was estimated to 8.45×10^{-2} eV/Tesla from which values for the reduced anisotropy energy d of about $d=0,\ldots,0.2$ can be obtained assuming reasonable values for D.

III. RESULTS

The stochastic differential equation (3) is solved numerically using the Heun method which converges in a quadratic mean to a solution of the LLG equation when interpreted in the sense of Stratonovich.[10,14] This ensures that the stationary solution of the corresponding Fokker-Planck equation is given by the Boltzmann distribution (for details see Ref. 7 and 10).

FMR resonance curves are calculated numerically for different temperatures starting from the ground state in a small external static field $b=0.01$ (in reduced units); i.e., initially all spins are pointing into the direction of the external field. Note that experimentally this state can be reached after slow cooling in an external field. The absorbed power is obtained from Eq. (5) using the solution of Eq. (3). Note that an average over a large number of field cycles after a stationary state is reached is necessary to reduce the thermal fluctuations. For the damping parameter $\alpha=0.2$ used in this paper stationarity is obtained after about 100 field cycles. For obtaining the complete resonance curve the external field is increased in small steps of $db=0.01$ each time until a field is reached which is well above the resonance field ($b=2$).

Figure 1 shows results for $d=0.2$ and for different values of reduced temperature q. The $q=0$ curve agrees with the exact solution available in this limit. At this temperature the position of the maximum is at $b_{max,T=0}=1-2d$ (in the reduced units used). With increasing temperature two features are observed clearly: a shift of the maximum of the resonance curve to higher fields mainly due to a strong decrease of the power absorbed for small b and a decrease of the

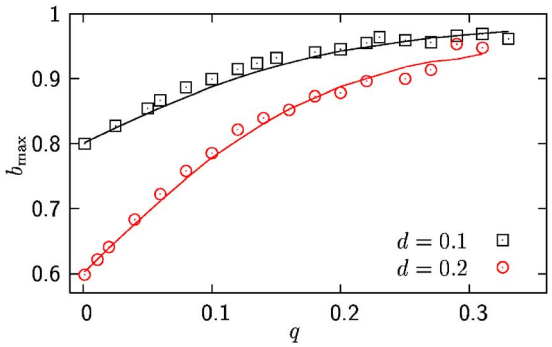

FIG. 2. (Color online) Position of the maximum of the resonance curve vs reduced temperature for two values of the reduced anisotropy.

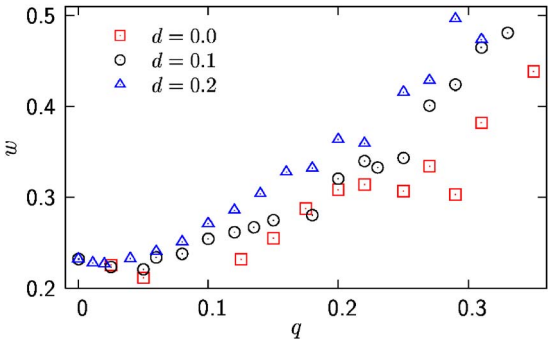

FIG. 3. (Color online) Width of the resonance curves vs temperature.

maximum of the absorbed power. Additionally, we observe in Fig. 1 an increase in the fluctuations which is due to the fact that we use for all temperatures deliberately the same amount of averaging over the thermal disorder: the FMR signal for a given value of the applied static field is averaged over 400 000 cycles of the HF field. Note that this time is still very small compared to the measuring time in a typical FMR experiment in which of the order of 10^3 as many field cycles are considered.[8] However, a corresponding increase in the simulation time is difficult since the CPU time needed for one resonance curve is a couple of days on a power PC.

From the numerically obtained resonance curves we calculate the position of its maximum, b_{max}, as a function of temperature. It is obtained by first fitting the resonance curves to the sum of two Lorentzians from which a rather accurate position of the maximum can be obtained. Two Lorentzians are used for this fitting since this is the form of the exact result known in the zero-temperature limit. Note that for elevated temperatures a fit with a single Lorentzian gives practically the same results. In Fig. 2 results are shown for $d=0.1$ and $d=0.2$, respectively. With increasing temperature b_{max} goes to 1 in qualitative agreement with the results found in Ref. 8 in which the temperature-dependent shift of the resonance signal defined as $1-b_{max}$ is interpreted as the temperature dependence of the magnetic anisotropy. Such an interpretation which is suggested by the fact that this shift is directly given by $2d$ at $T=0$ has been applied with success to thin ferromagnetic films where the observed reduction of the shift of the FMR signal with increasing temperature can be understood as a reduction of the effective anisotropy energy with temperature.[5] For thin films the observed reduction follows very closely a m_0^3 law which is due to temperature fluctuations within the film reducing its magnetization.[15] Here, m_0 denotes the equilibrium magnetization of the film.

However, in the present case we found that for nanoparticles this power law definitely does not fit the data. Instead the temperature shift $1-b_{max}$ is fitted very well by a term proportional to the square of the equilibrium magnetization at the resonance field, $1-b_{max}=2dm_0^2$, shown as solid lines in Fig. 2. The magnetization m_0 is calculated from

$$\mathbf{m}_0 = \langle \mathbf{S} \rangle = \frac{\mathrm{Tr}\, \mathbf{S} e^{-\beta \mathcal{H}_0}}{\mathrm{Tr} e^{-\beta \mathcal{H}_0}} \tag{9}$$

at the resonance field where $\mathcal{H}_0$ denotes the Hamiltonian given in Eq. (1) but without the high-frequency field.

The interpretation of this finding is that for nanoparticles it is the direction of the magnetization which fluctuates thermally and not the spin fluctuations within the particles.

Another quantity of interest is the width w of the resonance curve. A measure of this width often used in experiments is the difference between the zeros of the second derivative of the resonance signal. We follow this convention in the follwing. At $T=0$ this width is given by 2α for small damping constant α. With the value of α used in this paper this width is a few percent larger than 2α.

At finite temperature the width of the resonance curve as explained above is obtained by first fitting the numerical data to the sum of two Lorentzians and then calculating the zeros of the second derivatives of this function. w is shown in Fig. 3 for three values of the anisotropy energy, $d=0$, 0.1, and 0.2.

Despite extensive averaging there is still a strong scatter in the data. The important point, however, is that the width is increasing as a function of temperature by a factor of 2 to almost 3. This means that an effective damping parameter deduced from the measured width of the resonance curve is strongly temperature dependent. For a precise determination of the damping parameter used in the LLG equation an extrapolation of the data to $T=0$ is therefore necessary.

IV. SUMMARY

In this work we have studied in detail the shift of the resonance curves and the increase of its width for nanoparticles as a function of temperature. The particles all have the same uniaxial anisotropy axis with a static applied field parallel to this axis and a FMR field perpendicular to it. This is a special situation which, however, is of particular interest when thinking of assemblies of nanoparticles as storage media. We have found a strong decrease of the anisotropy-induced shift of the resonance line with increasing temperatures and have shown that this shift can very well be fitted by the square of the equilibrium magnetization at the resonance field. Additionally we observed a strong increase of the linewidth with temperature. Both these findings should be helpful in determining the microscopic parameters D and α from experiments.

At present, however, only assemblies of particles with random anisotropy axis have been studied experimentally.

Since the resonance field depends strongly on the easy axis of the particle relative to the applied fields, the experimentally observed FMR lines are superpositions of a large number of different lines, resulting in a large broadening of the resulting FMR signal. In this case the width of the measured resonance line at zero temperature is determined primarily by the strength of the anisotropy energy and eventually its distribution and not so much by the damping parameter α entering the LLG equation. Finite temperatures again lead to line shifts and an additional broadening of the resonance line. A study of these effects is under way.[13]

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