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THE RESERVOIR - INDUCED BEHAVIOR OF MAGNETIZATION IN A SINGLE-DOMAIN MAGNETIC NANOPARTICLE

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Abstract: *It is proposed a theoretical approach for magnons at finite temperature in a single-domain nanoparticle. Due to the spin wave quantization, the magnons have a finite spectrum, depending on the type, size and shape of the nanoparticle. The thermal fluctuations of the environment - considered as a thermal reservoir – is considered to be the cause of the nanoparticle nonequilibrium behavior, especially of the magnetization.*
Keywords*: Fine particle systems, spin waves, magnetic anisotropy, Brownian motion*

1. Introduction

The single-domain magnetic nanoparticles were intensively studied in the last decades, mostly as components of a system (e.g. ferrofluid or magnetic composite). An interesting physical property which appears in such a system at nonzero temperature is the superparamagnetism. The first interpretation of this phenomenon was given by Néel, based on the behavior of the magnetization vector **M** of a single-domain nanoparticle, whose uniaxial anisotropy determines an internal magnetic potential, consisting of two minima separated by a barrier. Néel affirmed that, due to thermal agitation, **M** could surmount the potential barrier and move from one minimum to another; this happens even in the absence of an external magnetic field favorable to lower the barrier. Developing this idea, most of the theoretical approaches treated the single-domain nanoparticle as if it has a single magnetic moment and **M** is considered to obey a phenomenological equation of damped precession of Landau-Lifshitz or Gilbert type. The first model, based on the solving of the Fokker-Planck equation for **M** of an assembly of nanoparticles, in order to find the damping constant, is due to Brown. It was followed by many others which use similar methods.

Here is presented a new approach for the magnetization behavior: a model based on the nonequilibrium theory for the magnons in a single-domain magnetic nanoparticle with

uniaxial anisotropy. The environment is considered as a thermal reservoir which, due its fluctuations, determines nonequilibrium states in the magnon system. Using the evolution equation for the statistical operator, it is deduced an expression for the time-dependent magnon number. A simplified model, similar to the Einstein model for phonons, is then introduced.

2. The Theoretical Framework

This paper starts from the observation that a single-domain nanoparticle, containing a relatively small number of spins ($\sim 10^4$), is a so-called mesoscopic system, too small to have equilibrium thermodynamic properties. On the other hand, the well-known quantum theory of a bulk (macroscopic) magnetic insulator (having enormous number of spins) provides successfully that its equilibrium properties at finite temperatures, especially the magnetization, are determined by the quantized spin waves or magnons, which are thermally generated by the contact of the system with a thermal reservoir and due to the specific interactions among lattice spins (exchange, dipole-dipole etc.). Therefore, this nanoparticle is much more sensitive to the fluctuations of the thermal reservoir, but its magnetic behavior may be somewhat similar to the equilibrium case of the macroscopic system. Consequently, in this work it is proposed a nonequilibrium magnon theory for the magnetization evolution in a single-domain insulator nanoparticle at finite temperatures. This approach has two main ideas, partly similar with those of the macroscopic theory. Firstly, the magnons do not a priori exist in an insulator magnetic system; they are quantas of spin waves thermally generated in it by the contact with the environment (considered as a thermal reservoir) due to all the spin-spin quantum interactions. But in the same time, the thermal contact allows the reservoir fluctuations to influence the system; if the system is macroscopic, this influence is negligible, but in a nanoparticle it may be of crucial importance, determining a nonequilibrium behavior. Secondly, all the magnetic properties of the system, especially the magnetization, are determined only by the magnons; for a macroscopic system, these properties are typical of equilibrium and are constant in time, but for a nanoparticle, they characterize nonequilibrium and depend of time. The Hamiltonian of the magnon system A within the nanoparticle, denoded by H_A, describing the spin-spin interactions in the absence of the external magnetic field, is considered to be

$$\begin{aligned}\hat{\mathcal{H}}_A &= \hat{\mathcal{H}}_{ex} + \hat{\mathcal{H}}_{an} \\ \hat{\mathcal{H}}_{ex} &= -\frac{1}{2} J \sum_{<i,j>} \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j \\ \hat{\mathcal{H}}_{an} &= -\mathcal{K} \sum_i \hat{S}_{iz}^2.\end{aligned} \tag{1}$$

where the angular brackets denote neighboring spins. In order to avoid confusions, all the operators are written with hats. H_{ex} is the exchange Hamiltonian and H_{an} is an effective Hamiltonian representing both the magneto-crystalline and dipole-dipole anisotropic interactions The resulting anisotropy is considered uniaxial, defining the *oz* axis; the nanoparticle shape is considered to posses a cylindrical symmetry around the magneto-crystalline anisotropy axis. Using the well-known Holstein-Primakoff formalism - which is entirely linear, neglecting the magnon-magnon interaction - this Hamiltonian may be finally written in the second quantization

$$\hat{\mathcal{H}}_A = \sum_{\mathbf{q}} \hbar\omega_{\mathbf{q}} \hat{a}_{\mathbf{q}}^{+} \hat{a}_{\mathbf{q}} \tag{2}$$

where a^{+}_{q} and a_q are respectively the creation and annihilation operators for the quantized spin waves or magnons with **q** wavevector. The general energy spectrum of
these magnons is given by the dispersion relation

$$\hbar\omega_{\mathbf{q}} = \hbar\omega_{\mathbf{q}}^{0} + \hbar\omega_{an}. \tag{3}$$

The kinetic energy of magnons, in the case of a simple cubic lattice it is given by

$$\hbar\omega_{\mathbf{q}}^{0} = \begin{cases} \mathcal{D}q^2 \\ \mathcal{C}q \end{cases} \tag{4}$$

where the first expression corresponds to the ferro- and ferrimagnetic-, and respectively the second to the antiferromagnetic insulators. The potential energy of magnons determined by the uniaxial anisotropy, also in the linear approximation, is

$$\hbar\omega_{an} = 2NS\mathcal{K} = 2KV \tag{5}$$

where K is the effective anisotropy constant and N is the number of spins in the nanoparticle. On the other hand, it is very important to remember that the quantization of spin waves must be made in the small nanoparticle. Considering periodic boundary conditions, this implies, as is well known from quantum mechanics, only standing spin waves. The smallest wavelength is assumed to be equal to the lattice constant a. Similarly to Debye model for phonons, magnon wavelengths shorter than a are considered meaningless; in this way, the magnon energy spectrum becomes superiorly limited. It is also assumed that the spin waves propagates

only on the crystallographic axes (chosen to be *ox,oy,oz* for the cubic lattice), due to the relatively small number of spins on each direction. The assumptions above represent selection rules for magnon wavevectors and energies, depending obviously on the chemical structure, size and shape of the nanoparticle. Consequently, the magnon energy spectrum is discrete and finite. It is very restricted compared to the continuum spectrum in the bulk case, where quantization is made on a huge number of spins; this fact marks a very important difference. Also, the magnon wavevectors, considered with directions on the crystallographic axis, have discrete magnitudes given by

$$q = |\mathbf{q}| \in \{2\pi/L, ..., 2\pi/a\}\,. \tag{6}$$

L is the greatest dimension of the nanoparticle, obviously on the *oz* axis, and *a* is the lattice constant; usually L~10 a. The operators for magnon numbers, defined by the relations

$$\hat{N}^A = \sum_{\mathbf{q}} \hat{N}^A_{\mathbf{q}} = \sum_{\mathbf{q}} \hat{a}^+_{\mathbf{q}} \hat{a}_{\mathbf{q}} \tag{7}$$

do not depend on the representation type and are constants of the time evolution. In order to calculate every average, it is necessary to know the statistical operator of the magnon system *A*. Its time dependence is the key of the nonequilibrium behavior of the magnon system, and is caused, as it was emphasized in the introduction, by the thermal reservoir fluctuations. Consequently, the average magnon numbers are time-dependent

$$\begin{aligned} N^A_{\mathbf{q}}(t) &= <\hat{N}^A_{\mathbf{q}}>_T (t) = Tr_A \left\{\hat{\sigma}(t)\hat{a}^+_{\mathbf{q}}\hat{a}_{\mathbf{q}}\right\} \\ N^A(t) &= <\hat{N}^A>_T (t) = \sum_{\mathbf{q}} N^A_{\mathbf{q}}(t). \end{aligned} \tag{8}$$

Like in the magnon theory for macroscopic systems, the magnetization of the nanoparticle **M** has a component on the *oz* axis defined by the relation

$$M_z = M - \frac{\gamma}{V} N^A(t) = M_z(t) \tag{9}$$

and is obviously time-dependent because of the magnon number. *M* is the spontaneous magnetization at *T=0* (where the magnons do not exist), *V* is the volume of the nanoparticle and the constant is $\gamma = g\mu_B$, with *g* the gyromagnetic factor and μ_B the Bohr magneton.

Using nonequilibrium statistical methods, one may find the time-dependent expressions of the total number of magnons, the magnetization and the energy

$$\mathcal{N}^A(t) = \sum_{\mathbf{q},\mathbf{k}} \left\{ \left[\mathcal{N}^A_{\mathbf{q}(t_0)} - \mathcal{N}^B_{\mathbf{k}} \right] e^{-\alpha_{\mathbf{q}} \frac{4\sin^2\left[\frac{1}{2}(\Omega_{\mathbf{k}}-\omega_{\mathbf{q}})(t-t_0)\right]}{(\Omega_{\mathbf{k}}-\omega_{\mathbf{q}})^2}} + \mathcal{N}^B_{\mathbf{k}} \right\} \tag{10}$$

$$\mathcal{M}^A_z(t) = \mathcal{M}^A_0 - \gamma \sum_{\mathbf{k},\mathbf{q}} \mathcal{N}^B_{\mathbf{k}} \left\{ 1 - e^{-\frac{4g^2_{\mathbf{q}}\sin^2\left[\frac{1}{2}(\Omega_{\mathbf{k}}-\omega_{\mathbf{q}})(t-t_0)\right]}{\hbar^2(\Omega_{\mathbf{k}}-\omega_{\mathbf{q}})^2}} \right\} \tag{11}$$

$$\mathcal{E}^A(t) = \sum_{\mathbf{k},\mathbf{q}} \hbar\omega_{\mathbf{q}} \left\{ \left[\mathcal{N}^A_{\mathbf{q}(t_0)} - \mathcal{N}^B_{\mathbf{k}} \right] e^{-\frac{4g^2_{\mathbf{q}}\sin^2\left[\frac{1}{2}(\Omega_{\mathbf{k}}-\omega_{\mathbf{q}})(t-t_0)\right]}{\hbar^2(\Omega_{\mathbf{k}}-\omega_{\mathbf{q}})^2}} + \mathcal{N}^B_{\mathbf{k}} \right\}. \tag{12}$$

Here t_0 is the initial time, $N^A_{q(t_0)}$ is the initial number of magnons with **q** wavevector and ω_q, N^B_k is the number of reservoir quantas with **k** wavevector and frequency Ω_k.

3. Conclusions.

The behavior of the nanoparticle, especially its magnetization, may be explained with the nonequlibrium theory of magnons. They are thermally generated in the nanoparticle due the contact with the environment, considered to be a thermal reservoir. Using the nonequilibrium statistical methods, one may calculate the expression of total magnon number, magnetization and magnon energy; they all have a periodic time-dependence. This may suggest that the superparamagnetism is caused by this periodic motion of the magnetization.

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