Fluctuation theory of magnetic relaxation for two-dimensional ensembles of dipolar interacting nanoparticles

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(Received 12 March 2001; published 23 October 2001)

The fluctuation theory of magnetic relaxation has been developed for the two-dimensional ensembles of ferromagnetic nanoparticles. The particles have random locations on a square lattice, interact via dipolar interaction, and their easy axes of magnetization are perpendicular to the lattice plane. The derivation of the equation that describes the time evolution of the magnetization has been based on the Fokker-Planck equation for the distribution function of a nanoparticle magnetic moment. The influence of the mean value and the fluctuations of the dipolar magnetic field on the process of magnetic relaxation is studied in detail. It has been shown that, in contrast to the case of noninteracting nanoparticles which is characterized by the single relaxation time, magnetic relaxation in those ensembles is characterized by two different relaxation times, and that the rate of relaxation decreases with time.

DOI: 10.1103/PhysRevB.64.184433 PACS number(s): 76.20.+q, 75.50.Tt, 05.10.Gg

I. INTRODUCTION

The study of ferromagnetic nanoparticles, i.e., nanometer-sized single-domain ferromagnetic particles, has both theoretical and experimental interest stimulated by their technological applications in data storage, composite materials, ferrofluids, etc.[1,2] The magnetic properties of ensembles of noninteracting nanoparticles are fully defined by their individual characteristics such as magnetic moments and magnetic anisotropy. A nanoparticle with uniaxial anisotropy is characterized by an internal potential that has two stable stationary points which correspond to the antiparallel orientation of the magnetic moment along the easy axis of magnetization and which are separated by a potential barrier. At nonzero temperatures the nanoparticle magnetic moment overcomes the barrier due to thermal agitation, as described by Néel.[3] A mathematically correct description of this phenomenon, known for the low-barrier case as superparamagnetism, first was given by Brown.[4] He derived and in specific cases solved the Fokker-Planck equation for the distribution function of the magnetic moment based on the stochastic Landau-Lifshitz equation. Now this approach is widely used for the study of the magnetic properties of ensembles of noninteracting nanoparticles and, specifically, of the magnetic relaxation.[5–9]

The approach of noninteracting nanoparticles is valid when the mean dipolar field acting on a nanoparticle is much less than the anisotropy field. If this condition is violated then, as the analytical results[10–19] and Monte Carlo simulations[20–25] have demonstrated, the dipolar interaction essentially changes the magnetic properties of the three-dimensional nanoparticle ensembles. From the mathematical point of view, the solution of the problem of magnetic relaxation requires the solution of the Fokker-Planck equation for the joint distribution function of the magnetic moments. Since this equation does not have exact solutions the study of magnetic relaxation for ensembles of dipolar interacting nanoparticles is always carried out by approximate methods. Within those methods different versions of the mean-field approximation are usually used and, as a rule, the final phase of magnetic relaxation is studied. The mean-field approximation has two main drawbacks. Firstly, as a result of the dipolar interaction the directions of the nanoparticle magnetic moments are correlated, i.e., a dynamical correlation exists between the magnetic moments. This means that different groups of nanoparticles can be under the influence of different local dipolar fields and, consequently, in nanoparticle ensembles the long-range spatial and temporal fluctuations in the local field exist. But within the mean-field approximation this fact is ignored. Secondly, since under thermal agitation magnetic moments perform random rotations, the dipolar field contains a high-frequency fluctuating part. The fluctuations of the dipolar field act on the magnetic moments in a way similar to the thermal agitation, but within the mean-field approximation they are not taken into account. One expects that for ensembles of close-packed nanoparticles (when the average distance between the nanoparticles is comparable with the nanoparticle size) both the long- and short-range fluctuations of the local dipolar field can strongly influence the magnetic relaxation.

During the last years much attention is paid to the two-dimensional (2D) nanoparticle ensembles that has been motivated by the significant developments in the fabrication techniques of such ensembles including ordered ones,[26–29] and by their interesting physical properties arising, in particular, from the dipolar interaction between nanoparticles.[30–34] Among the 2D ensembles there is a class in which the easy axes of magnetization are perpendicular to the plane where the nanoparticles are distributed. For such ensembles the influence of the dipolar interactions at low temperatures had been already investigated through the magnetization curves of thin films including the CoCr and CoCrPt nanoparticles.[35–37] Thermally activated magnetic relaxation from the initial state, when all magnetic moments

are oriented along a certain direction of the easy axis, to the demagnetized ground state was considered within a simplified version of the mean-field approximation.[38] The authors numerically showed that magnetic relaxation, in the restricted time interval, occurs slower than a simple Debye relaxation model predicts and they approximated the relaxation law by the stretched-exponential time dependence. But this stretched-exponential form does not hold for all times, therefore the behavior of magnetic relaxation for those ensembles remains an open question even within the mean-field theory.

In this paper we develop the fluctuation theory of magnetic relaxation for the 2D ensembles of uniaxial nanoparticles with easy axes of magnetization which are perpendicular to the lattice plane, which takes into account the influence of the high-frequency fluctuations of the dipolar field. In our approach, for the description of the dynamics of the nanoparticle magnetic moment we use the stochastic Landau-Lifshitz equation that takes into account the thermal agitation. We consider the dipolar interaction by means of the dipolar field, which we write as the sum of the mean and the fluctuating fields. We approximate the components of the fluctuating dipolar field by effective white noises and we derive the Fokker-Planck equation for the distribution function of the magnetic moment. In the general case this equation is not closed with respect to that distribution function since it contains the intensities of the components of the fluctuating dipolar field, which are defined by the joint distribution function. We perform the closing of this equation, i.e., the representation of these intensities via the distribution function, in the case when the potential barrier between the equilibrium directions of the magnetic moment essentially exceeds the thermal energy. In this approximation we solve the Fokker-Planck equation by the Kramers method[39] and we derive the law of magnetic relaxation.

The paper is organized as follows. In Sec. II we introduce the basic equations. We derive the equation for the distribution function of the magnetic moment in Sec. III, where we also express the intensities of the components of the fluctuating dipolar field through the correlation functions of the magnetic moment, and for the high-barrier case we calculate these intensities. In Sec. IV we derive the law of magnetic relaxation. Concluding remarks are contained in Sec. V, and some technical details are given in the Appendixes.

II. MEAN MAGNETIC MOMENT

We consider the 2D ensemble of spherical nanoparticles with a radius r randomly distributed on the sites of a square lattice with a lattice constant d (see Fig. 1). The easy axes of the nanoparticles magnetization are perpendicular to the xy plane. A lattice site is occupied by a nanoparticle with probability p. We describe the dynamics of the magnetic moments $\mathbf{m}_i=\mathbf{m}_i(t)$ (the index i labels the nanoparticles) by the system of stochastic Landau-Lifshitz equations

$$\dot{\mathbf{m}}_i=-\gamma\mathbf{m}_i\times[\mathbf{H}_i+\mathbf{n}_i(t)]-(\lambda\gamma/m)\mathbf{m}_i\times\mathbf{m}_i\times\mathbf{H}_i, \quad (1)$$

where $\gamma(>0)$ is the gyromagnetic ratio, $\lambda(\ll 1)$ is the damping parameter, $m=|\mathbf{m}_i|$, $\mathbf{H}_i=-\partial W/\partial\mathbf{m}_i$ is an effective magnetic field acting on $\mathbf{m}_i$, W is the magnetic energy of a nanoparticle ensemble, and $\mathbf{n}_i(t)$ is the thermal magnetic field which models the action of the heat bath. The thermal magnetic field is defined by its zero mean values $\overline{\mathbf{n}_i(t)}=0$ (the overbar denotes averaging with respect to the thermal field) and by the correlation functions

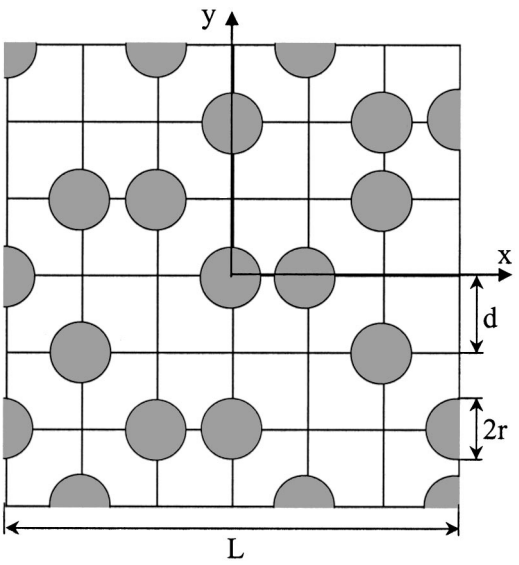

FIG. 1. Schematic representation of our model.

$$\overline{n_{i\alpha}(t)n_{j\beta}(t+\tau)}=2\Delta\delta_{ij}\delta_{\alpha\beta}\delta(\tau). \quad (2)$$

Here $n_{i\alpha}(t)$ $(\alpha=x,y,z)$ are the components of $\mathbf{n}_i(t)$, Δ is the intensity of the thermal field, δ_{ij} is the Kronecker symbol, and $\delta(\tau)$ is the δfunction. Within this model we write the magnetic energy W in the form

$$W=-\frac{\beta}{2V}\sum_i(\mathbf{m}_i\mathbf{e}_z)^2+\frac{1}{2}\sum_{i\neq j}\frac{(\mathbf{m}_i\mathbf{m}_j)r_{ij}^2-3(\mathbf{m}_i\mathbf{r}_{ij})(\mathbf{m}_j\mathbf{r}_{ij})}{r_{ij}^5}, \quad (3)$$

$\beta(>0)$ is the uniaxial anisotropy constant, V is the nanoparticle volume, $\hat{\mathbf{k}}$ is the unit vector along the z axis, $r_{ij}=|\mathbf{r}_{ij}|$, $\mathbf{r}_{ij}=\mathbf{r}_i-\mathbf{r}_j$, and $\mathbf{r}_i$ $(\mathbf{r}_i\hat{\mathbf{k}}=0)$ is the position vector of the lattice site where the ith nanoparticle is sitting.

Since $|\mathbf{m}_i|$ are conserved values, we can describe the dynamics of the magnetic moments by a system of stochastic equations for polar $\psi_{i1}=\psi_{i1}(t)$ and azimuthal $\psi_{i2}=\psi_{i2}(t)$ angles of $\mathbf{m}_i$

$$\dot{\psi}_{ik}=f_k\{\psi\}+g_{k\alpha}(\psi_{i1},\psi_{i2})n_{i\alpha}(t) \quad (k=1,2). \quad (4)$$

Here, and further on, twice repeated indices, which are not nanoparticle labels, imply summation, and $0\leqslant\psi_{i1}\leqslant\pi$, $0\leqslant\psi_{i2}<2\pi$. The functions $g_{k\alpha}(\psi_{i1},\psi_{i2})$ and $f_k\{\psi\}$ (ψ in angular brackets denotes the polar and azimuthal angles of all magnetic moments in the nanoparticle ensemble), which we will write as $g_{k\alpha}$ and f_k, respectively, are defined as follows:

$$g_{1\alpha}=\frac{\gamma}{m\sin\psi_{i1}}\frac{\partial m_{i\alpha}}{\partial\psi_{i2}}, \quad g_{2\alpha}=-\frac{\gamma}{m\sin\psi_{i1}}\frac{\partial m_{i\alpha}}{\partial\psi_{i1}}, \quad (5)$$

$$f_1 = -\frac{\gamma}{m \sin\psi_{i1}}\left[\lambda \sin\psi_{i1}\frac{\partial}{\partial\psi_{i1}} + \frac{\partial}{\partial\psi_{i2}}\right]W,$$

$$f_2 = \frac{\gamma}{m \sin^2\psi_{i1}}\left[\sin\psi_{i1}\frac{\partial}{\partial\psi_{i1}} - \lambda\frac{\partial}{\partial\psi_{i2}}\right]W. \qquad (6)$$

Since the thermal magnetic field approximates the real one having finite correlation time, the stochastic equation (4) should be interpreted according to Stratonovich.[40] In this case using the known from Ref. 39 techniques we find the Fokker-Planck equation

$$\frac{\partial P}{\partial t} = \sum_i \frac{\partial}{\partial\psi_{ik}}\left[-\left(f_k + \Delta g_{h\alpha}\frac{\partial g_{k\alpha}}{\partial\psi_{ih}}\right)P + \Delta\frac{\partial}{\partial\psi_{ih}}(g_{h\alpha}g_{k\alpha}P)\right] \qquad (7)$$

for the joint distribution function $P = P(\{\psi\},t)$ of the angles ψ_{ik}. The solution of this equation must satisfy the normalization condition

$$\int P(\{\psi\},t)\prod_i d\psi_{i1}d\psi_{i2} = 1 \qquad (8)$$

and the initial condition which for $\mathbf{m}_i(0) = m\hat{\mathbf{k}}$ has the form

$$P(\{\psi\},0) = \prod_i \frac{1}{2\pi}\delta(\psi_{i1}). \qquad (9)$$

Using Eq. (5) we find

$$g_{h\alpha}\frac{\partial g_{k\alpha}}{\partial\psi_{ih}} = \delta_{k1}\gamma^2\cot\psi_{i1},$$

$$g_{h\alpha}g_{k\alpha} = \gamma^2(\delta_{h1}\delta_{k1} + \delta_{h2}\delta_{k2}\sin^{-2}\psi_{i1}). \qquad (10)$$

Now it is not difficult to show that the stationary solution $P_s(\{\psi\})$ of Eq. (7) is given by

$$P_s(\{\psi\}) = Z^{-1}\exp(-\lambda W/\Delta\gamma m)\prod_i \sin\psi_{i1} \qquad (11)$$

(Z is the normalization constant). From Eq. (11), identifying $P_s(\{\psi\})$ with the Boltzmann distribution, we find the intensity of the thermal field[4]

$$\Delta = \lambda kT/\gamma m, \qquad (12)$$

where k is the Boltzmann constant, and T is the absolute temperature.

With the help of the joint distribution function P the mean value $\overline{m_z(t)} = \langle\overline{m_{jz}(t)}\rangle$ of the z component of the magnetic moment of a nanoparticle is written in the form

$$\overline{m_z(t)} = m\int \cos\psi_{j1}\langle P(\{\psi\},t)\rangle\prod_i d\psi_{i1}d\psi_{i2}. \qquad (13)$$

Here the angular brackets denote an average over the possible locations of nanoparticles, which are supposed to be equiprobable. In Eq. (13) the choice of the particle with the number j is arbitrary, so for convenience we consider the particle at the origin and we give it the number $j=0$. Then, having designated $\psi_{01} = \psi_1$ and $\psi_{02} = \psi_2$, from Eq. (13) we obtain

$$\overline{m_z(t)} = m\int_0^{\pi}\int_0^{2\pi}\cos\psi_1 P_0(\psi_1,\psi_2,t)d\psi_1 d\psi_2, \qquad (14)$$

where

$$P_0(\psi_1,\psi_2,t) = \int\langle P(\{\psi\},t)\rangle\prod_{i\neq 0} d\psi_{i1}d\psi_{i2} \qquad (15)$$

is the distribution function of the selected nanoparticle.

III. EQUATION FOR THE DISTRIBUTION FUNCTION

Since the calculation of the distribution function $P_0 = P_0(\psi_1,\psi_2,t)$, using the exact expression (15), is not possible, we obtain first the equation for P_0 using the approximation of the Gaussian fluctuations of the dipolar field. Within this approximation, the dipolar magnetic field

$$\mathbf{h}(t) = \sum_{i\neq 0}\frac{3\mathbf{r}_i(\mathbf{m}_i\mathbf{r}_i) - r_i^2\mathbf{m}_i}{r_i^3} \qquad (16)$$

acting on the magnetic moment $\mathbf{m} = \mathbf{m}_0(t)$ located at the origin is represented by the sum of the mean field $\mathbf{H}(t) = \langle\overline{\mathbf{h}(t)}\rangle$ and the Gaussian field $\tilde{\mathbf{n}}_0(t)$ with zero mean value. Notice that, since the dipolar interaction is long ranged, the Gaussian approximation for $\mathbf{h}(t)$ is quite justified. In addition, if the condition $\omega_m/\omega_T \ll 1$ [ω_m is the characteristic frequency of the macroscopic evolution of $\mathbf{m}$, and ω_T is the characteristic frequency of $\tilde{\mathbf{n}}_0(t)$] holds, then we can approximate the total thermal field $\mathbf{q}(t) = \mathbf{n}_0(t) + \tilde{\mathbf{n}}_0(t)$ by the δ-correlated one with the following correlation functions:

$$\overline{q_\alpha(t)q_\beta(t+\tau)} = 2\Delta\Delta_{\alpha\beta}(t)\delta(\tau), \qquad (17)$$

where $\Delta_{\alpha\beta}(t)$ are the functions to be calculated. As $\omega_T \sim kT/\hbar$. ($\hbar$ is the Planck constant), Eq. (17) is valid for $\omega_m\hbar/kT \ll 1$. Specifically, if $\beta \gg 1$ then $\omega_m \sim \gamma H_a$ ($H_a = m\beta/V$ is the anisotropy field) and the latter condition yields $T \gg \gamma\hbar H_a/k \sim 10^{-4}H_a$ ([H_a] = Oe).

Within the considered approximation we can write the system of stochastic equations for polar $\psi_1 = \psi_1(t)$ and azimuthal $\psi_2 = \psi_2(t)$ angles of $\mathbf{m}$ in the form

$$\dot{\psi}_k = f_k(\psi_1,\psi_2,t) + g_{k\alpha}(\psi_1,\psi_2)q_\alpha(t) \quad (k=1,2). \qquad (18)$$

Here the functions $g_{k\alpha} = g_{k\alpha}(\psi_1,\psi_2)$ and $f_k = f_k(\psi_1,\psi_2,t)$ are given by Eqs. (5) and (6) in which the index i should be dropped and W should be replaced by $W_0 = -(H_a/2m)m_z^2 - \mathbf{m}\cdot\mathbf{H}(t)$ (W_0 is the magnetic energy of the nanoparticle in the mean-field approximation). The equation for the distribution function P_0 corresponding to Eq. (18) can be obtained by transforming Eq. (18) to the form of Eq. (4). For this reason we use the representation $q_\alpha(t) = \lambda_{\alpha\eta}(t)n_\eta(t)$ [$\mathbf{n}(t) \equiv \mathbf{n}_0(t)$], and from Eq. (17) we find that the functions $\lambda_{\alpha\eta}(t)$ are defined by

$$\lambda_{\alpha\eta}(t)\lambda_{\beta\eta}(t) = \Delta_{\alpha\beta}(t). \qquad (19)$$

Denoting $g_{k\beta}\lambda_{\beta\alpha}(t)$ as $\tilde{g}_{k\alpha}$ by analogy with Eq. (7) we obtain

$$\frac{\partial P_0}{\partial t}=\frac{\partial}{\partial \psi_k}\left[-\left(f_k+\Delta\tilde{g}_{h\alpha}\frac{\partial\tilde{g}_{k\alpha}}{\partial\psi_h}\right)P_0+\Delta\frac{\partial}{\partial\psi_h}(\tilde{g}_{h\alpha}\tilde{g}_{k\alpha}P_0)\right]. \tag{20}$$

Now we express the functions $\mathbf{H}(t)$ and $\Delta_{\alpha\beta}(t)$, which are contained in Eq. (20), through the statistical characteristics of **m**. From the form of the initial condition (9) and the equiprobability of the nanoparticle distributions it follows that $\mathbf{H}(t)=H(t)\hat{\mathbf{k}}$ and

$$H(t)=-\left\langle\sum_{i\neq 0}\frac{\overline{m_{iz}(t)}}{r_i^3}\right\rangle. \tag{21}$$

In order to perform the average in Eq. (21), we consider the lattice region of size $L\times L$ containing S sites. The number of permutations of $N-1$ indistinguishable particles over $S-1$ sites (the site $\mathbf{s}=0$ is occupied by a particle) is equal to $R=(S-1)!/(N-1)!(S-N)!$. The number of permutations in which two sites $\mathbf{s}=0$ and $\mathbf{s}=d(n_1\hat{\mathbf{i}}+n_2\hat{\mathbf{j}})$ ($\hat{\mathbf{i}}$ and $\hat{\mathbf{j}}$ are the unit vectors along the x and y axis, respectively, n_1 and n_2 are integers, not equal to zero simultaneously) are occupied by nanoparticles, equals to $R_1=R(N-1)/(S-1)$. Let us number the last permutations by the index l and denote the z component of the magnetic moment in the site **s** as $m_{sz}^{(l)}(t)$. Then, since the probability of each permutation is equal to $1/R$, we can rewrite Eq. (21) in the form

$$H(t)=-\lim_{L\to\infty}\frac{R_1}{R}\sum_s\frac{1}{|\mathbf{s}|^3}\left(\frac{1}{R_1}\sum_{l=1}^{R_1}\overline{m_{sz}^{(l)}(t)}\right). \tag{22}$$

As $L\to\infty$ the ratio R_1/R tends to p, and the expression in the brackets tends to $\overline{m_z(t)}$. Therefore, by introducing

$$S_1=\frac{1}{8}\sum_{n_1,n_2}\frac{1}{(n_1^2+n_2^2)^{3/2}}\approx 1.1291, \tag{23}$$

from Eq. (22) we obtain

$$H(t)=-8pS_1\overline{m_z(t)}/d^3. \tag{24}$$

In order to find the connection between $\Delta_{\alpha\beta}(t)$ and the statistical characteristics of **m** we first replace in Eq. (17) τ by $-\tau$ and t by $t+\tau$. This yields $\Delta_{\alpha\beta}(t)=\Delta_{\beta\alpha}(t)$, and so we can define the functions $\Delta_{\alpha\beta}(t)$ as

$$\Delta_{\alpha\beta}(t)=\frac{1}{2\Delta}\int_0^\infty[K_{\alpha\beta}(t,t+\tau)+K_{\beta\alpha}(t,t+\tau)]d\tau, \tag{25}$$

with

$$\begin{aligned}K_{\alpha\beta}(t,t+\tau)&=\overline{\langle(n_\alpha(t)+h_\alpha(t)-\overline{h_\alpha(t)})(n_\beta(t+\tau)+h_\beta(t+\tau)-\overline{h_\beta(t+\tau)})\rangle}\\&=2\Delta\delta_{\alpha\beta}\delta(\tau)+\langle\overline{n_\alpha(t)h_\beta(t+\tau)}\rangle+\langle\overline{n_\beta(t+\tau)h_\alpha(t)}\rangle+\langle\overline{h_\alpha(t)h_\beta(t+\tau)}-\overline{h_\alpha(t)}\cdot\overline{h_\beta(t+\tau)}\rangle.\end{aligned} \tag{26}$$

For the calculation of the mean values in Eq. (26) we use the Chandrasekhar approximation[42] according to which the magnetic moments of nanoparticles are considered noncorrelated. Within this approximation, Eq. (25) yields (see Appendix A)

$$\Delta_{ll}(t)=1+\frac{20S_2p}{\Delta d^6}\int_0^\infty R_{ll}(t,t+\tau)d\tau, \tag{27}$$

$$\Delta_{zz}(t)=1+\frac{8S_2p}{\Delta d^6}\int_0^\infty R_{zz}(t,t+\tau)d\tau, \tag{28}$$

$$\Delta_{xy}(t)=-\frac{8p}{\Delta d^6}(S_2-9S_3)\int_0^\infty[R_{xy}(t,t+\tau)+R_{yx}(t,t+\tau)]d\tau, \tag{29}$$

$$\Delta_{lz}(t)=-\frac{2S_2p}{\Delta d^6}\int_0^\infty[R_{lz}(t,t+\tau)+R_{zl}(t,t+\tau)]d\tau, \tag{30}$$

where

$$R_{\alpha\beta}(t,t+\tau)=\overline{m_\alpha(t)m_\beta(t+\tau)}-\overline{m_\alpha(t)}\cdot\overline{m_\beta(t+\tau)} \tag{31}$$

are the correlation functions of the components of **m**, and $l=x$ or y. Equations (27)–(30) show that, in contrast to the mean field $H(t)$, the functions $\Delta_{\alpha\beta}(t)$ are not defined by P_0. This means that the Fokker-Planck equation (20) is not closed with respect to P_0.

We shall carry out the closing of this equation, which is necessary for its solution, in the case that the height δU of the potential barrier between the equilibrium directions of the magnetic moment essentially exceeds the thermal energy kT_{eff}, where T_{eff} is the effective temperature of the thermal field $\mathbf{q}(t)$ (see Sec. IV). For $\varepsilon=\delta U/kT_{\text{eff}}\gg 1$ the magnetic moments perform random fluctuations inside small solid angles along the positive and negative directions of the z axis. In this case the average time, which the magnetic moments spend in these directions, essentially exceeds the average time of their reorientation [for noninteracting particles the order of these times is[4] $(\lambda\gamma H_a)^{-1}a^{-1/2}\exp a(a=H_am/2kT)$ and $(\lambda\gamma H_a)^{-1}$, respectively]. Therefore we can write $\mathbf{m}=\mathbf{m}^\sigma(t)$, with $\mathbf{m}^\sigma(t)=m_x^\sigma(t)\hat{\mathbf{i}}+m_y^\sigma(t)\hat{\mathbf{j}}+\sigma m\hat{\mathbf{k}}$, $|\mathbf{m}^\sigma(t)-\sigma m\hat{\mathbf{k}}|\ll m$, and $\sigma=+$ or $-$ indicating fluctuations of the vector **m** along the positive (+) or negative (−) direction of the z axis. This behavior of **m** allows us to write

the functions $R_{\alpha\beta}(t,t+\tau)$ in the form of the weighted average values

$$R_{\alpha\beta}(t,t+\tau)=p_+(t)R^+_{\alpha\beta}(t,\tau)+p_-(t)R^-_{\alpha\beta}(t,\tau). \quad (32)$$

Here

$$p_+(t)=\int_0^{\theta_0(t)}\int_0^{2\pi}P_0(\psi_1,\psi_2,t)d\psi_1 d\psi_2,$$

$$p_-(t)=1-p_+(t) \quad (33)$$

are the probabilities that **m** fluctuates in the positive and negative direction of the z axis, respectively,

$$\theta_0(t)=\arccos\left(-\frac{H(t)}{H_\alpha}\right) \quad (|H(0)|<H_a) \quad (34)$$

is the angle where the energy $W_0=-(1/2)H_a m\cos^2\psi_1 - H(t)m\cos\psi_1$ has the maximum value, and

$$R^\sigma_{\alpha\beta}(t,\tau)=\overline{m^\sigma_\alpha(t)m^\sigma_\beta(t+\tau)}-\delta_{z\alpha}\delta_{z\beta}m^2. \quad (35)$$

Since $m^\sigma_z(t)=\sigma m$ and $\overline{m^\sigma_\alpha(t)}=\sigma m\delta_{z\alpha}$, Eqs. (32) and (35) give $R_{\alpha z}=R_{z\alpha}=0$ and from Eqs. (B1) and (B3) we obtain $\Delta_{\alpha z}(t)=\delta_{\alpha z}$. The solution of the linearized Landau-Lifshitz equation shows (see Appendix B) that the matrix $[\Delta_{\alpha\beta}(t)]$ has the diagonal form and $\Delta_{xx}(t)=\Delta_{yy}(t)\equiv\Delta_1(t)$. The function $\Delta_1(t)$ is defined by Eq. (27). Using Eqs. (35), (B5), and (B8) we obtain

$$R^\sigma_{ll}(t,\tau)=\Delta\gamma^2m^2\int_0^t[e^{\lambda^\sigma_1(t)t'+\lambda^\sigma_2(l+\tau)(t'+\tau)} + e^{\lambda^\sigma_2(t)t'+\lambda^\sigma_1(t+\tau)(t'+\tau)}]\Delta_1(t-t')dt'. \quad (36)$$

On the time intervals of the order $[\lambda\gamma H_+(t)]^{-1}$ the function $H(t)$ and, consequently, the functions $\Delta_1(t)$ and $\lambda^\sigma_{1,2}(t)$ practically do not change. This means that for the calculation of the integrals in Eqs. (27) and (36) we can replace the functions $\lambda^\sigma_{1,2}(t+\tau)$ by $\lambda^\sigma_{1,2}(t)$, and the function $\Delta_1(t-t')$ by $\Delta_1(t)$. In this approximation

$$\int_0^\infty R^\sigma_{xx}(t,\tau)d\tau=\Delta\Delta_1(t)m^2\frac{1-e^{-2\lambda\gamma H_\sigma(t)t}}{H^2_\sigma(t)(1+\lambda^2)}, \quad (37)$$

and Eq. (27) yields

$$\Delta_1(t)=\left\{1-\frac{20pS_2m^2}{(1+\lambda^2)d^6}\left(p_+(t)\frac{1-e^{-2\lambda\gamma H_+(t)t}}{H^2_+(t)} + p_-(t)\frac{1-e^{-2\lambda\gamma H_-(t)t}}{H^2_-(t)}\right)\right\}^{-1}. \quad (38)$$

As $\Delta_1(t)\geqslant 1$ (the equality holds for non-interacting nanoparticles), the dipolar interaction increases the intensity of those components of the total thermal field which lie on the lattice plane.

IV. LAW OF MAGNETIC RELAXATION

According to the results of the previous section, for $\varepsilon \gg 1$ the components of the matrix $[\Delta_{\alpha\beta}(t)]$ are expressed through the function $\Delta_1(t)$, which is fully defined by the distribution function P_0. This means that the Fokker-Planck equation (20) now is closed with respect to P_0. Let us find its solution in this case. Using the expressions

$$\tilde g_{h\alpha}\frac{\partial\tilde g_{k\alpha}}{\partial\psi_h}=\delta_{k1}\gamma^2\Delta_1(t)\cot\psi_1,$$

$$\tilde g_{h\alpha}\tilde g_{k\alpha}=\gamma^2\{\delta_{h1}\delta_{k1}\Delta_1(t)+\delta_{h2}\delta_{k2}[1+\Delta_1(t)\cot^2\psi_1]\} \quad (39)$$

which follow from Eqs. (5) and (B7), we can rewrite Eq. (20) in the form

$$\frac{\partial P_0}{\partial t}=\frac{\partial}{\partial\psi_1}\left(\frac{\lambda}{m}\frac{\gamma dW_0}{d\psi_1}-\gamma^2\Delta\Delta_1(t)\cot\psi_1\right)P_0 + \gamma^2\Delta\Delta_1(t)\frac{\partial^2P_0}{\partial\psi_1^2}-\frac{\gamma}{m\sin\psi_1}\frac{dW_0}{d\psi_1}\frac{\partial P_0}{\partial\psi_2} + \gamma^2\Delta[1+\Delta_1(t)\cot^2\psi_1]\frac{\partial^2P_0}{\partial\psi_2^2}. \quad (40)$$

Since the magnetic energy W_0 and the initial distribution $P_0(\psi_1,\psi_2,0)$ do not depend on ψ_2, we define $P_0=Q(\psi_1,t)/2\pi$, where the distribution function $Q(\theta,t)$ obeys the initial condition $Q(\theta,0)=\delta(\theta)$ and satisfies the equation

$$\frac{\partial Q(\theta,t)}{\partial t}=\frac{1}{t_r a(t)}\frac{\partial}{\partial\theta}\left\{\{a(t)[\sin 2\theta+2b(t)\sin\theta]-\cot\theta\} \times Q(\theta,t)+\frac{\partial Q(\theta,t)}{\partial\theta}\right\}. \quad (41)$$

Here $a(t)=a/\Delta_1(t)$, $b(t)=H(t)/H_a$, and $t_r=2/\lambda\gamma H_a$.

We define also the distribution function

$$Q_{\rm qe}(\theta,t)=C(t)\sin\theta e^{\alpha(t)[\cos^2\theta+2b(t)\cos\theta]} \quad (42)$$

[$C(t)$ is the normalization factor], which satisfies the equation

$$\frac{\partial Q_{\rm qe}(\theta,t)}{\partial\theta}+\{a(t)[\sin 2\theta+2b(t)\sin\theta]-\cot\theta\}Q_{\rm qe}(\theta,t)=0 \quad (43)$$

and at $t>t_{\rm qe}\sim t_r a(t)$ describes the quasiequilibrium (for $t=\infty-$ equilibrium) distribution of **m**. Expressing the function $Q_{\rm qe}(\theta,t)$ in the form of the Boltzmann distribution, i.e., $Q_{\rm qe}(\theta,t)=C(t)\sin\theta\exp(-W_0/kT_{\rm eff})$, and using the relation $W_0=-(1/2)H_a m[\cos^2\theta+2b(t)\cos\theta]$ we find the connection between the effective and absolute temperatures: $T_{\rm eff}=T\Delta_1(t)$.

Using Eq. (43) we rewrite Eq. (41) as

$$\frac{\partial Q(\theta,t)}{\partial t}=\frac{1}{t_r a(t)}\frac{\partial}{\partial\theta}\widetilde{Q}_{\rm qe}(\theta,t)\frac{\partial}{\partial\theta}\frac{Q(\theta,t)}{\widetilde{Q}_{\rm qe}(\theta,t)} \tag{44}$$

and with the help of the Kramers method[39] for the value $\rho(t)=2p_+(t)-1$ we obtain the following equation (see Appendix C):

$$\dot{\rho}(t)=-\rho(t)\left(\frac{1}{t_s^+(t)}+\frac{1}{t_s^-(t)}\right)-\frac{1}{t_s^+(t)}+\frac{1}{t_s^-(t)}. \tag{45}$$

Here

$$t_s^\sigma(t)=\frac{t_r\sqrt{\pi/a(t)}}{2[1-b^2(t)][1+\sigma b(t)]}\exp\{a(t)[1+\sigma b(t)]^2\} \tag{46}$$

are the times that the magnetic moment **m** spends on the average in the positive ($\sigma=+$) and negative ($\sigma=-$) directions of the z axis. According to Eq. (C5) $m_z(t)=m\rho(t)$, consequently Eq. (45) describes the process of the magnetic relaxation.

Equation (45) defines the function $\rho(t)$ at $t>t_{\rm qe}$ when the quasiequilibrium distribution of **m** has already been formed. The characteristic time $t_{\rm qe}$ satisfies the conditions $t_{\rm qe}/t_s^+(t)\ll 1$ and $\lambda\gamma H_\sigma(t)t_{\rm qe}\gg 1$. The former shows that for $t\sim t_{\rm qe}$ the probability for transition of **m** from the state $\sigma=+$ to the state $\sigma=-$ is negligible. Hence, having transferred the origin of time to an arbitrary point $t\sim t_{\rm qe}$, we can write the initial condition for Eq. (45) as $\rho(0)=1$. From the latter condition it follows that, for $t>t_{\rm qe}$, the exponential functions in Eq. (38) can be neglected, so the right-hand part of Eq. (45) depends on t only by means of $\rho(t)$. Denoting this part by $-F[\rho(t)]$ and using the condition $\rho(0)=1$, we can transform Eq. (45) to the form

$$\int_{\rho(t)}^{1}\frac{dx}{F(x)}=t. \tag{47}$$

With the help of Eq. (46) it is easy to verify that $F(x)>0$ if $0<x\leqslant 1$, and $F(0)=0$. This means that the solution of Eq. (47) is a decreasing function of time: $\rho(t_1)>\rho(t_2)(t_1<t_2)$, $\rho(\infty)=0$. Using this fact and rewriting Eq. (38) as

$$\Delta_1(t)=\left\{1-\chi^2\xi^2\frac{1+2\xi\rho^2(t)+\xi^2\rho^2(t)}{[1-\xi^2\rho^2(t)]^2}\right\}^{-1} \tag{48}$$

[$\chi^2=5S_2/16(1+\lambda^2)S_1^2 p$, and $\xi=-b(0)=8pS_1 m/H_a d^3$ is the parameter characterizing the intensity of the dipolar interaction] it is not difficult to show that $\Delta_1(t_1)>\Delta_1(t_2)$. Thus, the effective temperature $T_{\rm eff}=T\Delta_1(t)$ decreases with time while the height of the potential barrier $\delta U=(H_a m/2)[1-\xi\rho(t)]^2$ increases, therefore the process of the magnetic relaxation delays.

Notice that, within the developed approach the degree of influence of dipolar field fluctuations on the process of magnetic relaxation is determined by that how strongly the effective temperature at $t=0$ differs from the absolute one. We will characterize the difference $T_{\rm eff}|_{t=0}$ from T by the parameter $u=T_{\rm eff}|_{t=0}/T-1$, which, as it follows from Eq. (48), is given by

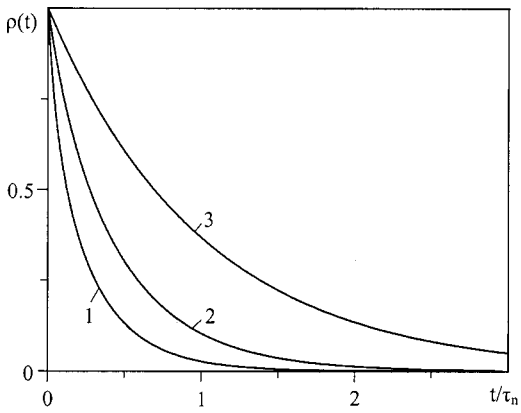

FIG. 2. Time dependence of $\rho(t)$ for ensembles of Co nanoparticles with the parameters (Ref. 41) $H_a=6400$ Oe, $m/V=1400$ G, $T=300$ K, $\lambda=0.2$, $r=40$ Å, $d=6r$ ($a\approx 28.995$, $\xi\approx 0.038p$, $\chi^2 p\approx 0.137$, $\tau_n\approx 28.6$ s) and $p=1$ (curve 1), $p=0.5$ (curve 2), $p=0$ (curve 3).

$$u=\frac{\chi^2\xi^2}{(1-\xi)^2-\chi^2\xi^2}. \tag{49}$$

If we use the parameter $\kappa=(1-\xi)^2-\chi^2\xi^2$ ($0<\kappa\leqslant 1$), then $\xi=(1-\chi^2)^{-1}[1-\sqrt{1-(1-\chi^2)(1-\kappa)}]$ [since $H_+(0)>0$ (see Appendix B) we chose the solution of equation $\kappa=(1-\xi)^2-\chi^2\xi^2$ which satisfies the condition $0\leqslant\xi<1$] and Eq. (49) yields

$$u=\frac{1}{\kappa}\frac{\chi^2}{(1-\chi^2)^2}[1-\sqrt{1-(1-\chi^2)(1-\kappa)}]^2. \tag{50}$$

According to Eq. (50) u is a decreasing function of κ and $u\sim(1-\kappa)^2\chi^2/4\to 0$ for $\kappa\to 1$, and $u\sim\kappa^{-1}\chi^2(1+\chi)^{-2}\to\infty$ for $\kappa\to 0$. But the parameter κ cannot take very small values. Indeed, the condition $\varepsilon=a(t)[1+b(t)]^2\gg 1$ is equivalent to $a(0)(1-\xi)^2\gg 1$ which in turn leads to the condition $a\kappa\gg 1$. Usually $a\sim 10-10^2$, therefore min $\kappa\sim 0.1$ and the parameter u, as a rule, does not exceed unity. As $a(0)=a/(u+1)$ and $a\gg 1$, from Eq. (46) it follows that even in this case the fluctuations of dipolar field essentially enhance magnetic relaxation in comparison with that which predicts the mean-field approximation (when $u=0$ or $T_{\rm eff}=T$). In particular, if $u\sim 1$ then, designating $t_s^\sigma(0)$ in the mean-field approximation as $[t_s^\sigma(0)]_{mf}$, we obtain $[t_s^\sigma(0)]_{mf}/t_s^\sigma(0)\sim\exp[a(1-\sigma\xi)^2/2]\gg 1$. If $u\ll 1$ then the fluctuations of the dipolar field are small and we can describe the magnetic relaxation within the mean-field approximation.

The dependence of $\rho(t)$ on t for different ensembles of Co nanoparticles is shown in Fig. 2. As can be seen the decrease of p, leading to the decrease of the intensity of the dipolar interaction, gives rise to reduction of magnetic relaxation.

Equation (47) can be solved analytically for $p=0$, $t\ll t_s^\sigma(0)$, and $t\gg t_s^\sigma(\infty)$. In the first case $F(x)=(1/\tau_n)x$, and

Eq. (47) yields $\rho(t)=\exp(-t/\tau_n)$, where $\tau_n = t_r\sqrt{\pi/16a}\exp a$ is the relaxation time in the case of noninteracting nanoparticles. For $t \ll t_s^{\sigma}(0)$ the condition $\rho(t) \approx 1$ holds. Therefore, replacing $F(x)$ by $F(1)=1/\tau_0$ ($\tau_0 = t_s^{+}(0)/2$) in Eq. (47), we obtain $\rho(t)=1-t/\tau_0$, where

$$\tau_0 = t_r \sqrt{\frac{\pi}{a(0)}}\,\frac{e^{a(0)(1-\xi)^2}}{4(1-\xi^2)(1-\xi)}. \tag{51}$$

Finally if $t \gg t_s^{\sigma}(\infty)$ then $\rho(t)\approx 0$, consequently, the main contribution into the left part of Eq. (47) comes from the lower limit region of the integration. Since $F(0)=0$, in this region we can approximate the function $F(x)$ by the function $xF'(0)$ [$F'(0)=dF(x)/dx|_{x=0}$]. For this case Eq. (47) gives $\rho(t)\sim\exp(-t/\tau_\infty)$, where

$$\tau_\infty = \frac{1}{F'(0)} = t_r \sqrt{\frac{\pi}{a(\infty)}}\,\frac{e^{a(\infty)}}{4[1+(2a(\infty)-1)\xi]}. \tag{52}$$

The analysis of expressions (51) and (52) shows that for $a(0)(1-\xi)^2 \gg 1$ and $\xi \neq 0$ the inequalities $\tau_0 < \tau_\infty < \tau_n$ hold ($\tau_0=\tau_\infty=\tau_n$ for $\xi=0$), and $\tau_0 \ll \tau_\infty$ for not very small ξ.

The asymptotic expressions for $\rho(t)$ at $t\to 0$ and $t\to\infty$ allow us to derive the approximate expression for $\rho(t)$ which is valid for all the times

$$\rho(t) = \frac{\tau_0/\tau_\infty}{e^{t/\tau_\infty}+\tau_0/\tau_\infty-1} \tag{53}$$

[the exact solution of Eq. (47) slightly exceeds it]. Equation (53) shows that, in contrast to the case of noninteracting nanoparticles, which is characterized by the single relaxation time τ_n, magnetic relaxation in ensembles of dipolar interacting nanoparticles can be approximately characterized by two different relaxation times, viz., the initial relaxation time τ_0 and the final one τ_∞. In the meanfield approximation Eq. (53) is also valid but the values $a(0)$ and $a(\infty)$ must be replaced by a. Notice that from a mathematical point of view deceleration of magnetic relaxation occurs due to a transition from one asymptotic behavior of $\rho(t)$ characterized by the relaxation time τ_0, to another asymptotic behavior characterized by the larger relaxation time τ_∞.

V. CONCLUSIONS

We have developed an analytical theory of the magnetic relaxation for the 2D ensembles of nanoparticles with uniaxial anisotropy that takes into account the fluctuations of the dipolar magnetic field. Our approach is based on the consideration that each nanoparticle is an isolated one and its magnetic moment **m** interacts with the mean dipolar field of the other nanoparticles and with an effective heat bath having the effective temperature $T_{\rm eff}$. The difference of $T_{\rm eff}$ from the absolute temperature T results from the contribution of the dipolar field fluctuations into the total fluctuating magnetic field acting on **m**. We have approximated the components of the total fluctuating field by the white noises, and we have described the dynamics of **m** by the stochastic Landau-Lifshitz equation. In the case when the height of the potential barrier between two equilibrium directions of **m** essentially exceeds the thermal energy we expressed $T_{\rm eff}$ and the noise intensities through the distribution function of **m**. Solving the Fokker-Planck equation for that distribution function we derived the equation for magnetization and we found its solution in the limiting cases. We showed that the effective temperature decreases with time and the height of the potential barrier increases. Consequently, the rate of magnetic relaxation in such ensembles decreases with time. Also, we have derived a simple approximate expression for the law of magnetic relaxation which is valid for all times.

ACKNOWLEDGMENTS

One of the authors (S.I.D.) would like to thank V.L. Safonov for helpful discussion and comments. This work was supported in part by NATO Grant No. PST.CLG.978108.

APPENDIX A

From Eq. (16), using the Chandrasekhar approximation[42] we can write

$$\begin{aligned}\langle \overline{h_\alpha(t)h_\beta(t+\tau)} - \overline{h_\alpha(t)}\cdot\overline{h_\beta(t+\tau)}\rangle &= \Big\langle \sum_{i\neq 0}\frac{1}{r_i^{10}}(r_i^4\delta_{\alpha\varepsilon}\delta_{\beta\eta} - 3r_i^2 r_{i\alpha}r_{i\varepsilon}\delta_{\beta\eta} \\ &\quad - 3r_i^2 r_{i\beta}r_{i\eta}\delta_{\alpha\varepsilon} + 9r_{i\alpha}r_{i\beta}r_{i\varepsilon}r_{i\eta}) \\ &\quad \times[\overline{m_{i\varepsilon}(t)m_{i\eta}(t+\tau)} - \overline{m_{i\varepsilon}(t)}\cdot\overline{m_{i\eta}(t+\tau)}]\Big\rangle\end{aligned} \tag{A1}$$

and, because $\overline{n_\alpha(t)m_{i\beta}(t')}=0$ ($i\neq=0$), we obtain

$$\langle \overline{n_\alpha(t)h_\beta(t+\tau)}\rangle = \langle \overline{n_\beta(t+\tau)h_\alpha(t)}\rangle = 0. \tag{A2}$$

Using the same approximations as for the calculation of $H(t)$, we transform Eq. (A1) to the form

$$\begin{aligned}\langle \overline{h_\alpha(t)h_\beta(t+\tau)} - \overline{h_\alpha(t)}\cdot\overline{h_\beta(t+\tau)}\rangle &= P\sum_s \frac{1}{|\mathbf{s}|^{10}}(|\mathbf{s}|^4\delta_{\alpha\varepsilon}\delta_{\beta\eta} - 3|\mathbf{s}|^2 s_\alpha s_\varepsilon\delta_{\beta\eta} \\ &\quad - 3|\mathbf{s}|^2 s_\beta s_\eta\delta_{\alpha\varepsilon} + 9s_\alpha s_\beta s_\varepsilon s_\eta)R_{\varepsilon\eta}(t,t+\tau).\end{aligned} \tag{A3}$$

From this, having defined the lattice sums as

$$\begin{aligned}S_2 &= \frac{1}{8}\sum_{n_1,n_2}\frac{1}{(n_1^2+n_2^2)^3} \approx 0.5824, \\ S_3 &= \frac{1}{8}\sum_{n_1,n_2}\frac{n_1^2 n_2^2}{(n_1^2+n_2^2)^5} \sim 0.0174\end{aligned} \tag{A4}$$

and using the relations

$$\sum_s \frac{s_\alpha s_\varepsilon}{|\mathbf{s}|^8} = \frac{4S_2}{d^6}(\delta_{x\alpha}\delta_{x\varepsilon} + \delta_{y\alpha}\delta_{y\varepsilon}),$$

$$\sum_s \frac{s_\alpha s_\beta s_\varepsilon s_\eta}{|\mathbf{s}|^{10}} = \frac{4}{d^6}(S_2-2S_3)(\delta_{x\alpha}\delta_{x\beta}\delta_{x\varepsilon}\delta_{x\eta} + \delta_{y\alpha}\delta_{y\beta}\delta_{y\varepsilon}\delta_{\gamma\eta}) + \frac{8S_3}{d^6}(\delta_{x\alpha}\delta_{x\beta}\delta_{y\varepsilon}\delta_{y\eta} + \delta_{x\alpha}\delta_{y\beta}\delta_{x\varepsilon}\delta_{y\eta} + \delta_{x\alpha}\delta_{y\beta}\delta_{y\varepsilon}\delta_{x\eta} + \delta_{y\alpha}\delta_{y\beta}\delta_{x\varepsilon}\delta_{x\eta} + \delta_{y\alpha}\delta_{y\beta}\delta_{x\varepsilon}\delta_{x\eta} + \delta_{y\alpha}\delta_{x\beta}\delta_{x\varepsilon}\delta_{y\eta}) \tag{A5}$$

[at their proof we took into account that $\Sigma_s s_x^4/|\mathbf{s}|^{10} = \Sigma_s s_y^4/|\mathbf{s}|^{10} = 4(S_2-2S_3)/d^6$], we find

$$\langle \overline{h_\alpha(t)h_\beta(t+\tau)} - \overline{h_\alpha(t)}\cdot\overline{h_\beta(t+\tau)}\rangle = \frac{4pS_2}{d^6}[2R_{\alpha\beta} - 3\delta_{x\alpha}R_{x\beta} - 3\delta_{y\alpha}R_{y\beta} - 3\delta_{x\beta}R_{\alpha x} - 3\delta_{y\beta}R_{\alpha y} + 9\delta_{xa}\delta_{x\beta}R_{xx} + 9\delta_{y\alpha}\delta_{y\beta}R_{yy}] + \frac{72pS_3}{d^6}[\delta_{x\alpha}\beta_{x\beta}(R_{yy}-R_{xx}) + \delta_{y\alpha}\delta_{y\beta}(R_{xx}-R_{yy}) + (\delta_{x\alpha}\delta_{y\beta}+\delta_{y\alpha}\delta_{x\beta})(R_{xy}+R_{yz})]. \tag{A6}$$

Finally, using Eqs. (26), (31), and (A6) we obtain Eqs. (27)–(30).

APPENDIX B

Neglecting the terms $\sim m_\alpha^\sigma(t)q_\beta(t)$, the linearized Landau-Lifshitz equation takes the form

$$\dot{\boldsymbol{\mu}}(t) = A\boldsymbol{\mu}(t) + B\mathbf{w}(t) \quad [\boldsymbol{\mu}(0)=\mathbf{0}]. \tag{B1}$$

Here $\mu(t)$ is a vector with components $m_x^\sigma(t)$ and $m_y^\sigma(t)$, $\mathbf{w}(t)$ is a vector with components $q_x(t)$ and $q_y(t)$,

$$A = -\gamma H_\sigma(t)\begin{pmatrix} \lambda & \sigma 1 \\ -\sigma 1 & \lambda \end{pmatrix}, \quad B = \sigma\gamma m\begin{pmatrix} 0 & 1 \\ -1 & 0 \end{pmatrix}, \tag{B2}$$

and $H_\sigma(t) = H_a + \sigma H(t)$. The matrix A has the eigenvalues $\lambda_{1,2}^\sigma(t) = -\gamma H_\sigma(t)(\lambda \mp i)$ [since both directions of the z axis must correspond to the minimum of W_0 we suppose that $\mathrm{Re}\,\lambda_{1,2}^\sigma(t) < 0$, i.e., $H_+(0) > 0$] to which the characteristic times $[\lambda\gamma H_\sigma(t)]^{-1}$ and $[\gamma H_\sigma(t)]^{-1}$ of the macroscopic evolution of $\mathbf{m}^\sigma(t)$ correspond. The maximum characteristic time $[\lambda\gamma H_+(t)]^{-1}$, is much less than the characteristic time of the change of $H(t)$ (see Sec. IV) so in Eq. (B1) we can consider the matrix A as a constant one. In this case the solution of Eq. (B1) is given by the expression[43]

$$\boldsymbol{\mu}(t) = \int_0^t \exp(At')B\mathbf{w}(t-t')dt'. \tag{B3}$$

Using the relation

$$e^{At'} = e^{\lambda_1^\sigma(t)t'}\frac{A-\lambda_2^\sigma(t)I}{\lambda_1^\sigma(t)-\lambda_2^\sigma(t)} - e^{\lambda_2^\sigma(t)t'}\frac{A-\lambda_1^\sigma(t)I}{\lambda_1^\sigma(t)-\lambda_2^\sigma(t)} \tag{B4}$$

(I is the unit matrix) which follows from the Sylvester theorem,[44] we obtain from Eq. (B3)

$$m_x^\sigma(t) = \frac{\gamma m}{2i}\int_0^t [(e^{\lambda_1^\sigma(t)t'} - e^{\lambda_2^\sigma(t)t'})q_x(t-t') + \sigma i(e^{\lambda_1^\sigma(t)t'} + e^{\lambda_2^\sigma(t)t'})q_y(t-t')]dt',$$

$$m_y^\sigma(t) = \frac{\gamma m}{2i}\int_0^t [(e^{\lambda_1^\sigma(t)t'} - e^{\lambda_2^\sigma(t)t'})q_y(t-t') - \sigma i(e^{\lambda_1^\sigma(t)t'} + e^{\lambda_2^\sigma(t)t'})q_x(t-t')]dt'. \tag{B5}$$

Let us make the assumption that the matrix $[\Delta_{\alpha\beta}(t)]$ has the diagonal form

$$[\Delta_{\alpha\beta}(t)] = \mathrm{diag}[\Delta_1(t), \Delta_1(t), 1]. \tag{B6}$$

Then according to Eq. (19) we have

$$[\lambda_{\alpha\beta}(t)] = \mathrm{diag}[\sqrt{\Delta_1(t)}, \sqrt{\Delta_1(t)}, 1] \tag{B7}$$

and, consequently,

$$q_x(t) = \sqrt{\Delta_1(t)}n_x(t), \quad q_y(t) = \sqrt{\Delta_1(t)}n_y(t), \quad q_z(t) = n_z(t). \tag{B8}$$

Using Eqs. (B5), (B8), and (2) it is not difficult to show that $R_{xy}^\sigma(t,\tau) = -R_{yx}^\sigma(t,\tau)$, therefore, $\Delta_{xy}(t) = 0$. Hence, taking into account the condition $\Delta_{\alpha z}(t) = \delta_{\alpha z}$ derived earlier, we confirm that the matrix $[\Delta_{\alpha\beta}(t)]$ has the diagonal form.

APPENDIX C

In accordance to the Kramers method[39] we find the solution of Eq. (44) for $t > t_{\mathrm{qe}}$ in the form

$$Q(\theta,t) = \tilde{Q}_{\mathrm{qe}}(\theta,t)\begin{cases} p_+(t)/n_+(t), & 0 \leqslant \theta \leqslant \theta_0(t), \\ p_-(t)/n_-(t), & \theta_0(t) < \theta \leqslant \pi, \end{cases} \tag{C1}$$

where $\tilde{Q}_{\mathrm{qe}}(\theta,t) = Q_{\mathrm{qe}}(\theta,t)/C(t)$, and

$$n_+(t) = \int_0^{\theta_0(t)} \tilde{Q}_{\mathrm{qe}}(\theta,t)d\theta, \quad n_-(t) = \int_{\theta_0(t)}^{\pi} \tilde{Q}_{\mathrm{qe}}(\theta,t)d\theta. \tag{C2}$$

According to Eqs. (24),(38) the functions $a(t)$ and $b(t)$ are expressed through the probabilities $p_\pm(t)$ and the mean magnetic moment $\overline{m_z(t)}$ which satisfies the equation

$$\overline{m_z(t)} = m\frac{p_+(t)}{n_+(t)}\int_{-b(t)}^1 xe^{a(t)[x^2+2b(t)x]}dx + m\frac{p_-(t)}{n_-(t)}\int_{-1}^{-b(t)} xe^{\alpha(t)[x^2+2b(t)x]}dx. \tag{C3}$$

Since $\varepsilon = a(t)[1+b(t)]^2$ $(\delta U = W_0|_{\theta=\theta_{0(t)}} - W_0|_{\theta=0} = (H_a m/2)[1+b(t)]^2)$ and for $\varepsilon \gg 1$ the asymptotic expressions

$$\int_{-b(t)}^{1} x e^{a(t)[x^2+2b(t)x]} dx = \frac{e^{a(t)[1+2b(t)]}}{2a(t)[1+b(t)]} = n_+(t),$$

$$\int_{-1}^{-b(t)} x e^{a(t)[x^2+2b(t)x]} dx = -\frac{e^{a(t)[1-2b(t)]}}{2a(t)[1-b(t)]} = -n_-(t) \tag{C4}$$

hold, Eq. (C3) is reduced to

$$\overline{m_z(t)} = m[p_+(t) - p_-(t)]. \tag{C5}$$

As $p_-(t) = 1 - p_+(t)$, the single unknown function in Eq. (C1) is the probability $p_+(t)$. Using the equation

$$\int_0^{\theta_0(t)} \frac{1}{\tilde{Q}_{\rm qe}(\theta,t)} \frac{\partial}{\partial t}\left(\int_0^{\theta} Q(\vartheta,t) d\vartheta \right) d\theta$$

$$= \frac{1}{t_r a(t)} \int_0^{\theta_0(t)} \frac{\partial}{\partial \theta}\left(\frac{Q(\theta,t)}{\tilde{Q}_{\rm qe}(\theta,t)} \right) d\theta, \tag{C6}$$

which follows from Eqs. (44) and (C1), the asymptotic expression

$$\int_0^{\theta_0(t)} \frac{1}{\tilde{Q}_{\rm qe}(\theta,t)} \frac{\partial}{\partial t}\left(\int_0^{\theta} Q(\vartheta,t) d\vartheta \right) d\theta$$

$$= \dot{p}_+(t) \frac{1}{2} \sqrt{\frac{\pi}{a(t)}} \frac{e^{a(t)b^2(t)}}{1-b^2(t)} \quad (\varepsilon \gg 1) \tag{C7}$$

and the relation

$$\frac{\partial}{\partial \theta}\left(\frac{Q(\theta,t)}{\tilde{Q}_{\rm qe}(\theta,t)} \right) = \left(\frac{p_-(t)}{n_-(t)} - \frac{p_+(t)}{n_+(t)} \right) \delta[\theta - \theta_0(t)], \tag{C8}$$

we obtain Eq. (45).

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