Integral relaxation time of single-domain ferromagnetic particles

D. A. Garanin*
I. Institut für Theoretische Physik, Universität Hamburg, Jungiusstrasse 9, D-20355 Hamburg, Germany
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The integral relaxation time $\tau_{\rm int}$ of thermoactivating noninteracting single-domain ferromagnetic particles is calculated analytically in the geometry with a magnetic field H applied parallel to the easy axis. It is shown that the drastic deviation of $\tau_{\rm int}^{-1}$ from the lowest eigenvalue of the Fokker-Planck equation Λ_1 at low temperatures, starting from some critical value of H, is the consequence of the depletion of the upper potential well. In these conditions the integral relaxation time consists of two competing contributions corresponding to the overbarrier and intrawell relaxation processes. [S1063-651X(96)09409-3]

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I. INTRODUCTION

At present, a single-domain ferromagnetic particle with uniaxial anisotropy attracts the attention of researchers, in particular as one of the models of information storage. The hysteretic rotation of the magnetization of such a particle over the potential barrier under the influence of an arbitrary directed magnetic field **H** was studied by Stoner and Wohlfart [1]. At nonzero temperatures the magnetization vector of the particle can surmount the barrier due to the thermal agitation, as argued by Neél [2]; this effect becomes especially pronounced for small particles having lower values of the potential barrier ΔU. Such a ''superparamagnetic'' behavior was observed in many experiments on magnetic liquids, on polymers with magnetic inclusions, as well as on very thin magnetic layers forming ''islands.''

An initial accurate calculation of the thermoactivation rate of a uniaxial ferromagnetic particle is due to Brown [3], who derived the Fokker-Planck equation for an assembly of particles and solved it in the presence of a longitudinal magnetic field $\mathbf{H}=H\mathbf{e}_z$, perturbatively in the low-barrier case $\Delta U \ll T$ and with the use of the Kramers transition-state method [4] in the high-barrier limit $T \ll \Delta U$ (the Boltzmann constant k_B is set to unity). In both limiting cases considered by Brown the time dependence of the average magnetization $\langle M_z \rangle$ is a single exponential and the relaxation rate of ferromagnetic particles is given by the lowest eigenvalue Λ_1 of the Sturm-Liouville equation associated with the Fokker-Planck equation. Subsequently, Λ_1 was calculated numerically by Aharoni for arbitrary values of $\Delta U/T$ without a magnetic field [5] and with a longitudinal magnetic field [6]. The correction terms for the high-barrier result for Λ_1 were given by Brown [7]. Later the analytical expression for Λ_1 in the high-barrier case was rederived in Ref. [8] with a more rigorous method. In Refs. [9–11] various approximate analytical formulas for Λ_1 for the arbitrary $\Delta U/T$ were proposed. Recently the thermoactivation rate of single-domain magnetic particles, as described by Λ_1, was calculated numerically by Coffey *et al.* [12] for the arbitrarily directed magnetic field **H**, i.e., in the geometry considered by Stoner and Wohlfart [1].

Apart from limiting cases, the Fokker-Planck equation for an assembly of single-domain ferromagnetic particles cannot be solved analytically. The magnetization relaxation curve consists of an infinite number of exponentials and the overall deviation of the linear dynamic susceptibility from the Debye form can be as large as about 7% for *isotropic* particles in a static magnetic field, as shown in Ref. [13]. In this case it is convenient to introduce the so-called integral relaxation time $\tau_{\rm int}$, determined as the area under the relaxation curve after a sudden infinitesimal change of the magnetic field. The quantity $\tau_{\rm int}$ depends on all eigenvalues Λ_k, $k=1,2,\ldots$, and is therefore more informative than Λ_1; also it can be directly measured. Moreover, it turned out that, unlike Λ_1, the integral relaxation time $\tau_{\rm int}$ can be calculated *analytically* for uniaxial particles in the longitudinal magnetic field for the arbitrary values of parameters [13] and $\tau_{\rm int}^{-1}$ recovers the analytical results of Brown for Λ_1 in the asymptotic regions.

The integral relaxation time was also the subject of a recent series of papers [14–16], where it was called the ''correlation time.'' In Ref. [14] $\tau_{\rm int}$ for uniaxial ferromagnetic particles was calculated *analytically* with an alternative method for zero magnetic field, the resulting expression being, however, much more complicated than the original formula for $\tau_{\rm int}$ of Ref. [13]. In Ref. [16] a *numerical* calculation of $\tau_{\rm int}$ in the case with nonzero longitudinal magnetic field was presented. In Ref. [15] the congeneric model of rotating dipoles describing the dielectric relaxation was considered. The results of Ref. [14] show that in zero magnetic field $\tau_{\rm int}^{-1}$ is very close to Λ_1 in the whole region of $\Delta U/T$. On the contrary, numerical calculations of Ref. [16] reveal a striking behavior $\tau_{\rm int}^{-1} \gg \Lambda_1$ for relatively small longitudinal fields in the region $T \ll \Delta U$. This region of parameters was not analyzed in Ref. [13], whereas in Ref. [16] the effect was not physically interpreted.

The aim of this paper is thus to consider in more detail the integral relaxation time of uniaxial ferromagnetic particles in the longitudinal magnetic field with the help of the method of Ref. [13]. As we shall see, the effect found in Ref. [16] can be explained by the depletion of the upper biased potential well, which leads to the dominance of the fast relaxation inside the lower well in the integral relaxation time.

The remainder of the paper is organized as follows. In Sec. II the derivation of the Fokker-Planck equation for an

* Electronic address: garanin@physnet.uni-hamburg.de

assembly of single-domain ferromagnetic particles from the stochastic Landau-Lifshitz equation is outlined. In Sec. III the known results for the thermoactivation rate of uniaxial ferromagnetic particles are briefly reviewed. Then the integral relaxation time $\tau_{\rm int}$ is introduced and analyzed, and it is shown that the effect discovered in Ref. [16] can be explained without an explicit calculation of $\tau_{\rm int}$. In Sec. IV the derivation of a general formula for $\tau_{\rm int}$ of uniaxial particles in a longitudinal magnetic field is presented and its behavior is studied analytically and numerically for the whole region of parameters. Some concluding remarks are given in Sec. V.

II. THE FOKKER-PLANCK EQUATION

The magnetization of a single-domain ferromagnetic particle $\mathbf{M}$ can be considered not too close to the Curie point T_c as a vector of fixed length $|\mathbf{M}|=M_s(T)$, whose direction can fluctuate due to the thermal agitation. This fluctuative motion of $\mathbf{M}$ can be described semiphenomenologically with the help of the stochastic equation

$$\dot{\mathbf{M}}=\gamma[\mathbf{M}\times(\mathbf{H}_{\rm eff}+\boldsymbol{\zeta})]-\mathbf{R}(\mathbf{M}), \tag{2.1}$$

where γ is the gyromagnetic ratio,

$$\mathbf{H}_{\rm eff}=-\frac{\partial W}{\partial \mathbf{M}}, \quad W=-\mathbf{H}\cdot\mathbf{M}-KM_z^2 \tag{2.2}$$

are the effective field and the energy density, $\mathbf{H}$ is the external magnetic field, and K is the uniaxial anisotropy constant. The energy of a particle is given by

$$\mathcal{H}=VW, \tag{2.3}$$

where V is the particle volume. The correlators of different components of the white-noise field $\boldsymbol{\zeta}(t)$ can be conveniently written as

$$\langle\zeta_i(t)\zeta_j(t')\rangle=\frac{2\lambda T}{\gamma V}\delta_{ij}\delta(t-t'). \tag{2.4}$$

The relaxation term $\mathbf{R}$ in (2.1) describes, like $\boldsymbol{\zeta}$, the influence of the heat bath on the particle and, as we shall see immediately, it has the Landau-Lifshitz form [17]

$$\mathbf{R}=\gamma\lambda[\mathbf{M}\times[\mathbf{M}\times\mathbf{H}_{\rm eff}]]. \tag{2.5}$$

The Fokker-Planck equation corresponding to (2.1) is formulated for the distribution function $f(\mathbf{N},t)=\langle\delta(\mathbf{N}-\mathbf{M}(t))\rangle$ on the sphere $|\mathbf{N}|=M_s$, where the average is taken over the realizations of $\boldsymbol{\zeta}$. Differentiating f over t with the use of (2.1) and calculating the right-hand side of the resulting equation analogously to the derivations given, e.g., in Refs. [18,19], one comes to the Fokker-Planck equation

$$\frac{\partial f}{\partial t}=-\frac{\partial}{\partial \mathbf{N}}\left\{\gamma[\mathbf{N}\times\mathbf{H}_{\rm eff}]-\mathbf{R}(\mathbf{N})+\frac{\gamma\lambda T}{V}\left[\mathbf{N}\times\left[\mathbf{N}\times\frac{\partial}{\partial\mathbf{N}}\right]\right]\right\}f. \tag{2.6}$$

One can easily see that the equilibrium distribution function

$$f_0(\mathbf{N})\propto\exp[-\mathcal{H}(\mathbf{N})/T], \tag{2.7}$$

is the solution of (2.6) if and only if $\mathbf{R}$ has the double-vector product form (2.5), which reflects the way of how magnetization is coupled to the heat-bath fluctuations in (2.1). If, e.g., the correlators of $\boldsymbol{\zeta}$ components in (2.4) are anisotropic, the expression (2.5) also changes [13].

Brown used in his derivation of the Fokker-Planck equation [3] the stochastic equation of motion (2.1) with the Gilbert relaxation term $\mathbf{R}=\gamma\eta[\mathbf{M}\times\dot{\mathbf{M}}]$ [20]. Redefining $\gamma\Rightarrow\gamma_G$ in Brown's equation, one can transform the latter to the form (2.1) and (2.5) with $\gamma=\gamma_G/(1+\eta^2\gamma_G^2M_s^2)$ and $\lambda=\eta\gamma_G$, where the Langevin field $\boldsymbol{\zeta}$ enters also the expression for $\mathbf{R}$ (2.5) as being added to $\mathbf{H}_{\rm eff}$. This means using a stochastic model somewhat different from the one described above. Both models coinside, however, in the actual small-damping case $\eta\gamma M_s\ll 1$.

The equation of motion for the magnetization $\langle\mathbf{M}\rangle=\int d^3N\mathbf{N}f(\mathbf{N},t)$ of an assembly of particles can be easily derived from (2.6) and has the form

$$\frac{d}{dt}\langle\mathbf{M}\rangle=\gamma\langle[\mathbf{M}\times\mathbf{H}_{\rm eff}]\rangle-\gamma\lambda\langle[\mathbf{M}\times[\mathbf{M}\times\mathbf{H}_{\rm eff}]]\rangle-\Lambda_N\langle\mathbf{M}\rangle \tag{2.8}$$

[cf. (2.1)], where the characteristic diffusional relaxation rate Λ_N is given by

$$\Lambda_N\equiv\tau_N^{-1}\equiv 2\gamma\lambda T/V. \tag{2.9}$$

One can see that even in the case without anisotropy $\mathbf{H}_{\rm eff}=\mathbf{H}$, this equation is not closed since it is connected to the second-order correlation functions $\langle M_iM_j\rangle$ in the Landau-Lifshitz term of (2.8). Therefore, the resonance and relaxational behavior of the Fokker-Planck equation (2.6) is in general *not* described by Lorentz and Debye curves, and the deviations from the latter can be about 7% [13]. Neglecting these features, one can obtain the *best isolated* equation of motion for the magnetization of an assembly of particles in the isotropic case $K=0$, choosing the distribution function in the form $f(\mathbf{N},t)\propto\exp[V\mathbf{A}(t)\mathbf{N}/T]$ [cf. (2.7)], where the temporal evolution of the vector $\mathbf{A}(t)$ is governed by (2.8). Such a generalized Landau-Lifshitz-Bloch equation [13] contains both transverse and longitudinal relaxation terms. In the high-temperature limit (in the isotropic case $K=0$ this requires $T\gg VHM_s$) Eq. (2.8) becomes closed and takes on the form of the Bloch equation with the relaxation rate Λ_N.

III. THE LOWEST EIGENVALUE Λ_1 AND THE INTEGRAL RELAXATION TIME $\tau_{\rm int}$

To parametrize effects of thermal agitation on ferromagnetic particles, it is convenient to introduce the dimensionless energy $u\equiv\mathcal{H}/T=VW/T$, which in the case with a longitudinal magnetic field has the form

$$u=-\xi x-\alpha x^2, \quad x\equiv\cos\theta=M_z/M_s, \tag{3.1}$$

with

$$\xi\equiv\frac{VHM_s}{T}, \quad \alpha\equiv\frac{VKM_s^2}{T}. \tag{3.2}$$

The top of the barrier corresponds to

$$x=x_m=-h, \quad h\equiv\frac{\xi}{2\alpha}=\frac{H}{2KM_s}. \tag{3.3}$$

The barrier height $\Delta u\equiv u(x_m)-u(-1)$ is given by

$$\Delta u=\alpha-\xi+\xi^2/(4\alpha)=\alpha(1-h)^2. \tag{3.4}$$

In the case $\alpha,\xi\sim 1$ a general solution of the Fokker-Planck equation (2.6) cannot be found analytically and the relaxation of any initial state is described by a sum of exponentials of the type $A_i\exp(-\Lambda_i t)$, where Λ_i are the eigenvalues of the Sturm-Liouville equation associated with the Fokker-Planck equation (for the longitudinal relaxation all Λ_i are real). In the low-barrier case $\alpha,\xi\ll 1$, the problem can be solved perturbatively [3] and the longitudinal relaxation is governed with a good accuracy by the single exponential corresponding to the lowest eigenvalue Λ_1, which is given by [3]

$$\Lambda_1\cong\Lambda_N\left(1-\frac{2}{5}\alpha+\frac{48}{875}\alpha^2+\frac{1}{10}\xi^2+\cdots\right), \tag{3.5}$$

with Λ_N determined by (2.9).

In the high-barrier case $\alpha\gg 1$, the relaxation is dominated again by Λ_1 describing now the slow overbarrier thermoactivation, whereas all other eigenvalues Λ_i correspond to the fast intrawell processes with small amplitudes. Brown's result for the high-barrier case, which was derived with the help of the transition-state method of Kramers [4], can be written in the form [6]

$$\Lambda_1\cong\Lambda_N\pi^{-1/2}\alpha^{3/2}(1-h^2)\{(1+h)\exp[-\alpha(1+h)^2] +(1-h)\exp[-\alpha(1-h)^2]\}, \tag{3.6}$$

where h is given by (3.3). The factor $(1+h)$ before the exponential function in (3.6) is irrelevant since the first term of (3.6) is only essential for $\xi\lesssim 1$ which for $\alpha\gg 1$ implies $h\ll 1$. We will, however, keep this factor here and in analogous expressions below for the sake of symmetry.

In the intermediate region $\alpha,\xi\sim 1$, it is convenient to introduce the integral relaxation time determined as the area under the magnetization relaxation curve after a sudden infinitesimal change of the applied field H by ΔH at $t=0$:

$$\tau_{\rm int}\equiv\int_0^\infty dt\frac{\langle M_z(\infty)\rangle-\langle M_z(t)\rangle}{\langle M_z(\infty)\rangle-\langle M_z(0)\rangle}. \tag{3.7}$$

Unlike Λ_1, the integral relaxation time $\tau_{\rm int}$ can be found analytically from the Fokker-Planck equation (2.6) in the whole range of parameters in the geometry with a longitudinal magnetic field [13], as will be described in detail in Sec. IV. Here we discuss the results of recent calculations of $\tau_{\rm int}$ by Coffey *et al.* [14,16]. At first note that the relaxation curve can be represented in the form

$$\langle M_z(\infty)\rangle-\langle M_z(t)\rangle=\Delta H\chi_z\sum_i A_i e^{-\Lambda_i t}, \tag{3.8}$$

where $\chi_z=\partial\langle M_z\rangle/\partial H$ is the static longitudinal susceptibility. This form of writing the response function is more convenient than that of Refs. [14,16] since here the amplitudes A_i obey the sum rule $\Sigma_i A_i=1$. Now $\tau_{\rm int}$ of (3.7) can be rewritten as (cf. [14])

$$\tau_{\rm int}=\sum_i A_i\Lambda_i^{-1}. \tag{3.9}$$

In Refs. [14,16] the integral relaxation time $\tau_{\rm int}$ is called the correlation time since according to the fluctuation-dissipation theorem $\tau_{\rm int}$ can be also considered as the area under the autocorrelation function. The term ''correlation time,'' however, seems to be rather artificial because the autocorrelation function does not appear in the actual calculation of $\tau_{\rm int}$ with the help of (3.7) or (3.9), as well as in Sec. IV below, and really considering autocorrelations would imply going unnecessarily beyond the Fokker-Planck equation.

According to the numerical results of Coffey *et al.* [14] in a zero magnetic field the amplitudes A_i satisfy $A_i\ll A_1$, $i=2,3,\ldots$, for all values of α and the difference between Λ_1 and $\tau_{\rm int}^{-1}$ is small everywhere reaching only 1.2% at $\alpha=5$. On the contrary, the subsequent calculations for $H\neq 0$ [16] revealed a striking behavior $\tau_{\rm int}^{-1}\gg\Lambda_1$ at sufficiently low temperatures. The formal reason for this is that A_1 becomes small in this region and the terms with $k=4,5$ dominate in (3.9), as shown in Ref. [16]. But the effect can also be interpreted on a physical level as the consequence of the depletion of the upper potential well and quantitatively described without a general calculation of $\tau_{\rm int}$, as will be demonstrated below.

The reduced equilibrium magnetization of an ensemble of noninteracting ferromagnetic particles $m_z\equiv\langle M_z\rangle/M_s$ is given by the generalized Langevin function $B(\xi,\alpha)$:

$$m_z=\int_{-1}^1 x f_0(x)dx=\frac{\partial}{\partial\xi}\ln Z=B(\xi,\alpha), \tag{3.10}$$

where, according to (2.7) and (3.1),

$$f_0=\frac{1}{Z}\exp(-\mathcal{H}/T)=\frac{e^{-u}}{Z}, \quad Z=\int_{-1}^1 e^{-u}dx. \tag{3.11}$$

In the high-barrier case $\alpha\gg 1$, the partition function Z is a sum of two contributions corresponding to the two potential wells $Z=Z_+ + Z_-$,

$$Z_\pm\cong\frac{e^{\alpha\pm\xi}}{2\alpha\pm\xi}\left[1+\frac{2\alpha}{(2\alpha\pm\xi)^2}+\cdots\right], \tag{3.12}$$

where the correction terms account for the curvature of the potential-energy function u of (3.1). Neglecting these small terms, one can represent $B(\xi,\alpha)$ of Eq. (3.10) by two mutually complementing expressions

$$B(\xi,\alpha)\cong\tanh\xi-\frac{1}{2\alpha}\left(\frac{\xi}{\cosh^2\xi}+\tanh\xi\right)+\frac{\xi}{(2\alpha)^2} \tag{3.13}$$

for $1\sim\xi\ll\alpha$ and

$$B(\xi,\alpha)\cong 1-2e^{-2\xi}\frac{2\alpha+\xi}{2\alpha-\xi}-\frac{1}{2\alpha+\xi} \tag{3.14}$$

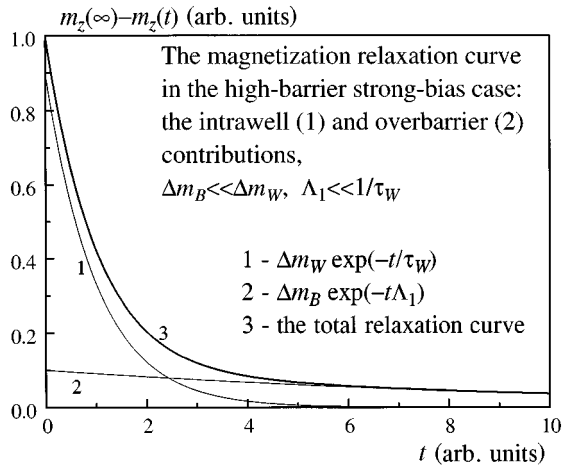

FIG. 1. Schematic look of the two-exponential relaxation curve of single-domain ferromagnetic particles in the strong-bias high-barrier case $\alpha,\xi \gg 1$.

for $1 \ll \xi \sim \alpha$. Here, in the first limiting expression the second term is small and irrelevant; the third term is kept since it yields a contribution to the derivative $B' = \partial B/\partial \xi$ that is not exponentially small for $\xi \gg 1$. In the second limiting expression (the strong-bias case) the deviation of B from unity separates into two parts: The second exponentially small term is due to the population of the upper well ($x \sim -1$), whereas the third one accounts for the thermal agitation in the lower well ($x \sim 1$). The response of magnetization to an infinitesimal change of the magnetic field ΔH is related to the derivative of the generalized Langevin function B': $\Delta m = B' \Delta H/T$. The latter can be determined from (3.13) and (3.14) and put in the whole region $2\alpha - \xi \gg 1$ into the unique expression

$$B' \cong B'_B + B'_W \cong \frac{1}{\cosh^2 \xi} \frac{2\alpha+\xi}{2\alpha-\xi} + \frac{1}{(2\alpha+\xi)^2} \cong \frac{1-h^2}{c^2(\xi,h)} + \frac{1}{(2\alpha+\xi)^2}, \tag{3.15}$$

where

$$c(\xi,h) \equiv \frac{1}{2}[(1+h)e^{-\xi} + (1-h)e^{\xi}] \tag{3.16}$$

and B'_B accounts for the redistribution of particles between the two wells across the potential barrier and B'_W that inside the lower well. Henceforth we will use the second of the equivalent forms of B' in (3.15) for the sake of symmetry [cf. the comments after Eq. (3.6)].

Now, in the low-temperature strong-bias case $\alpha,\xi \gg 1$, the relaxation curve (3.8) consists of only two exponentials (see Fig. 1)

$$m_z(\infty) - m_z(t) = \Delta m_B \exp(-t\Lambda_1) + \Delta m_W \exp(-t\Lambda_W), \tag{3.17}$$

where Λ_1 is given by (3.6) and

$$\Lambda_W \cong 2\gamma\lambda H_{\text{eff}} = \Lambda_N(2\alpha+\xi) \tag{3.18}$$

is the temperature-independent relaxation rate in the lower well, which can be obtained from the deterministic Landau-Lifshitz equation (2.1) and (2.5) without $\boldsymbol{\zeta}$. The integral relaxation time τ_{int} calculated according to the definition (3.7) can be written as

$$\tau_{\text{int}} \cong \tau_{\text{int},B} + \tau_{\text{int},W}, \tag{3.19}$$

where

$$\tau_{\text{int},B} = \frac{B'_B}{B'} \Lambda_1^{-1}, \quad \tau_{\text{int},W} = \frac{B'_W}{B'} \Lambda_W^{-1}. \tag{3.20}$$

One can see that in the low-temperature strong-bias case $\alpha,\xi \gg 1$, the barrier contribution $\tau_{\text{int},B}$ into the integral relaxation time τ_{int} can be substantially reduced due to the depletion of the upper potential well manifesting itself in the exponential smallness of the magnetization change due to overbarrier transitions $\Delta m_B \propto B'_B$ [see (3.15)]. In this case the overbarrier and intrawell terms in (3.19) can compete with each other since Λ_1 is exponentially small and $B'_W/B' \cong 1$. On the contrary, for small or zero bias one has $B'_B/B' \cong 1$ and $B'_W/B' \ll 1$, so that the intrawell process can be completely ignored.

The expressions for $\tau_{\text{int},B}$ and $\tau_{\text{int},W}$ in (3.20) are valid in the whole high-barrier region $2\alpha - \xi \gg 1$ and will be obtained independently in the framework of a general method in Sec. IV. In the strong-bias case $\xi \gg 1$, the barrier contribution $\tau_{\text{int},B}$ in (3.20) can be represented with the use of (3.6), (3.15), and (3.3) as

$$\tau_{\text{int},B} = 16\Lambda_N^{-1}(\pi\alpha)^{1/2} \frac{(1+h)^2}{(1-h)^3} \exp[\alpha(1-6h+h^2)]. \tag{3.21}$$

It changes its behavior as a function of α at the critical value of the applied field

$$h = h_c = 3 - 2\sqrt{2} \approx 0.17, \tag{3.22}$$

which is substantially smaller than the field of the barrier disappearance $h=1$ [see (3.3) and (3.4)]. For h in the vicinity of h_c the exponential factor in (3.21) can be written as $\exp[-4\sqrt{2}\alpha(h-h_c)]$. It can be seen that for $h < h_c$ the quantity $\tau_{\text{int},B}$ exponentially increases with lowering temperature (i.e., with increasing of α) and brings the dominant contribution into τ_{int} of Eq. (3.20). On the contrary, for $h > h_c$ the quantity $\tau_{\text{int},B}$ exponentially decreases at large α, so that τ_{int} tends to the temperature-independent value $\tau_{\text{int},W}$ of (3.18). One can also see that for h only slightly higher than h_c the quantity $\tau_{\text{int},B}$ increases as $\alpha^{1/2}$ at smaller α; then the decreasing exponential becomes to dominate. Thus, in this case $\tau_{\text{int},B}$, and hence τ_{int} of (3.20), has a maximum at some $\alpha \gg 1$ and τ_{int}^{-1} has the corresponding minimum, as was obtained numerically in Ref. [16]. It should be noted, however, that the actual position of this minimum can be described only taking into account in (3.20) the general form of B' given by (3.15).

The results above completely describe the observations made in Ref. [16] in the low-temperature strong-bias region.

In the next section we present the analytical calculation of $\tau_{\rm int}$ in the whole range of parameters.

IV. CALCULATION OF THE INTEGRAL RELAXATION TIME $\tau_{\rm int}$

All the information about the relaxation curve (3.8) is contained in the longitudinal linear dynamic susceptibility $\chi(\omega)$. In the presence of a small alternating field $\Delta H_z(t)=\Delta H_{z0}\exp(-i\omega t)$ the deviation of the distribution function f from the equilibrium function (3.11) can be represented as

$$\delta f = f_0(x) q(x) V M_s \Delta H_z(t)/T, \tag{4.1}$$

where the function $q(x)$ satisfies an equation following from (2.6):

$$\left(\frac{d}{dx}+2\alpha x+\xi\right)(1-x^2)\frac{dq}{dx}+2i\omega\Lambda_N^{-1}q = (1-x^2)(2\alpha x+\xi)-2x. \tag{4.2}$$

The dynamic susceptibility of the particle's assembly is then determined by

$$\chi_z(\omega)=\frac{VM_s^2}{T}\int_{-1}^{1} dx f_0(x) q(x). \tag{4.3}$$

Using the linear-response theory one can easily show that $\chi_z(\omega)$ can be represented in the form

$$\chi_z(\omega)=\chi_z\sum_i \frac{A_i}{1-i\omega\Lambda_i^{-1}}, \tag{4.4}$$

where χ_z, A_i, and Λ_i are the parameters of the magnetization relaxation curve (3.8). Calculating this sum of Debye terms requires knowing all eigenvalues Λ_i and amplitudes A_i associated with the Fokker-Planck equation and cannot be done analytically in the general case. Accordingly, Eq. (4.2) has no general analytical solution and its behavior is to be studied analytically in the limiting cases of high and low temperatures and high and low frequencies, as was done in Ref. [13]. In particular, generating high-frequency expansions of $\chi(\omega)$ does not require solving differrential equations and can be carried out up to high orders. The corresponding results, however, are not very interesting here since they describe only fast intrawell processes. The information about the slow process of thermoactivation is contained in the low-frequency expansion of $\chi(\omega)$, which can be written in the form

$$\chi_z(\omega)\cong\chi_z(1+i\omega\tau_{\rm int}+\cdots). \tag{4.5}$$

Comparing (4.5) with (4.4), one can show that the quantity $\tau_{\rm int}$ in (4.5) is exactly the integral relaxation time given by the formula (3.9).

The perturbative solution of (4.2) for small ω can be done analytically since for $\omega=0$ there are only terms of the type $q'(x)$ and $q''(x)$ in the equation. Hence one can introduce a new variable $g(x)\equiv q'(x)$ and solve successively the first-order differential equations for $g(x)$ and $q(x)$. After calculation of the susceptibility (4.3) one gets the analytic expression the for integral relaxation time $\tau_{\rm int}$ [13]:

$$\tau_{\rm int}=\frac{2}{\Lambda_N B'}\int_{-1}^{1}\frac{dx}{1-x^2}\Phi^2(x) f_0^{-1}(x), \tag{4.6}$$

where f_0 is given by (3.11), $B'=\partial B/\partial\xi$, and

$$\Phi(x)=\int_{-1}^{x}(B-x')f_0(x')dx'. \tag{4.7}$$

Recalling the general formula for $B(\xi,\alpha)$ Eq. (3.10), one can conclude that $\Phi(\pm1)=0$, i.e., the integrand of (4.6) goes to zero at $x=\pm1$. The function $\Phi(x)$ can be easily calculated analytically in two particular cases. In the unbiased case $\xi=0$ one gets

$$\Phi(x)=\frac{1}{2\alpha}[f_0(1)-f_0(x)],\quad f_0(x)=\frac{\exp(\alpha x^2)}{Z(\alpha)}, \tag{4.8}$$

whereas in the isotropic case $\alpha=0$

$$\Phi(x)=\frac{f_0(x)}{\xi}\left[\coth\xi-x-\frac{\exp(-\xi x)}{\sinh\xi}\right] \tag{4.9}$$

with $f_0(x)=\xi\exp(\xi x)/(2\sinh\xi)$. An alternative analytical expression for $\tau_{\rm int}$ in the particular case $\xi=0$ was derived recently by Coffey *et al.* [14] with the help of the development of the solution of the Fokker-Planck equation in Legendre polynomials. Their expression contains Kummer functions and is essentially more complicated than (4.6) with (4.8).

Now we proceed with the analysis of the general expression (4.6) in different limiting cases. In the high-temperature limit $\alpha,\xi\ll1$, the calculation can be done perturbatively with respect to α and ξ. In particular, at very large temperatures one has $f_0(x)\cong1/2$, $B\cong0$, $B'\cong1/3$, and $\Phi\cong(1-x^2)/4$. Thus the whole phase space of the ferromagnetic particle, $-1\leqslant x\leqslant1$, contributes to (4.6) and one gets $\tau_{\rm int}^{-1}\cong\Lambda_N$. A more accurate calculation yields

$$\tau_{\rm int}^{-1}\cong\Lambda_N\left(1-\frac{2}{5}\alpha+\frac{2}{35}\alpha^2+\frac{1}{9}\xi^2+\cdots\right), \tag{4.10}$$

which is very close to Brown's expression for Λ_1 given by (3.5).

In the unbiased low-temperature case $\xi=0$, $\alpha\gg1$, the function Φ given by (4.8) is constant in the main part of the x interval, except for near the borders. Thus the main contribution to the integral (4.6) comes from the barrier region $x\sim\alpha^{-1/2}\ll1$ cut by the function $f_0^{-1}\propto\exp(-\alpha x^2)$. With the use of (3.12) with $\xi=0$ one gets, in the leading order,

$$\tau_{\rm int}^{-1}\cong2\Lambda_N\pi^{-1/2}\alpha^{3/2}e^{-\alpha}, \tag{4.11}$$

which coincides with the expression for Λ_1 in (3.6) in the unbiased case $h=0$. It is not difficult to calculate also the correction terms for the formula (4.11), which coincide with those given by Brown [7]. In the isotropic strong-field limit $\alpha=0$, $\xi\gg1$, the function Φ given by (4.9), as well as the whole integrand of (4.6), is peaked in the vicinity of the potential minimum $x=1$, where the exponentially small term

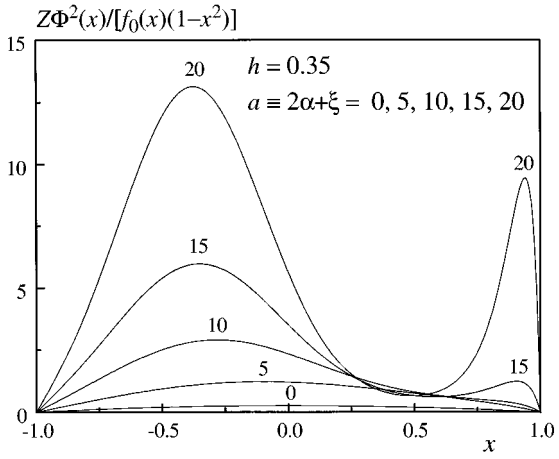

FIG. 2. Integrand of the formula (4.6) for the integral relaxation time τ for different values of parameters.

with $\exp(-\xi x)$ can be neglected. In the leading order one gets for $\tau_{\rm int}^{-1}$ the temperature-independent expression (3.18) with $\alpha=0$.

Now we consider, as a corollary, the biased low-temperature case $\xi\neq 0$, $\alpha\gg 1$. As argued in Sec. III, in this case $\tau_{\rm int}$ can be the sum of intrawell and overbarrier contributions, $\tau_{{\rm int},W}$ and $\tau_{{\rm int},B}$ [see (3.19)]. Accordingly, the integrand of (4.6) can consist, for $\alpha,\xi\gg 1$, of two peaks (see Fig. 2) corresponding to the barrier top $x\sim x_m=-h$ in (3.3) and the lower well $x\sim 1$. The function $\Phi(x)$ of (4.7) is determined by two well-separated potential wells and is therefore practically independent of x for x not too close to the boundaries. In the the lower-well region $1-x\ll 1$, the calculation yields $\Phi(x)$ as a sum of two contributions $\Phi=\Phi_B+\Phi_W$, where to the leading order

$$\Phi_B(x)\cong\frac{1-h^2}{2c^2(\xi,h)}\{1-\exp[-(2\alpha+\xi)(1-x)]\},\qquad(4.12)$$

$c(\xi,h)$ is given by (3.16), and

$$\Phi_W(x)\cong(1-x)\exp[-(2\alpha+\xi)(1-x)].\qquad(4.13)$$

The term Φ_B of (4.12) goes over to the constant mentioned above in the region not too close to the border $[1-x\gg 1/(2\alpha+\xi)\ll 1]$. For $\xi\gg 1$ it acquires the small factor $e^{-2\xi}$ accounting for the depletion of the upper potential well [cf. (3.15) and (3.20)]. On the contrary, such a factor is not present in Φ_W in (4.13), but the corresponding contribution into $\tau_{\rm int}$ given by (4.6) is reduced due to $f_0^{-1}(x)$. Now calculating the integral (4.6) one gets Eq. (3.19), where

$$\tau_{{\rm int},B}\cong\frac{B_B'}{B'}\frac{\pi^{1/2}}{2\Lambda_N\alpha^{3/2}}\frac{\exp[\alpha+\xi^2/(4\alpha)]}{(1-h^2)c(\xi,h)}\qquad(4.14)$$

coincides with the expression given by (3.20) and in the strong-bias case

$$\tau_{{\rm int},W}^{-1}\cong\Lambda_W\cong\Lambda_N\left(2\alpha+\xi-\frac{10\alpha+\xi}{2\alpha+\xi}\right).\qquad(4.15)$$

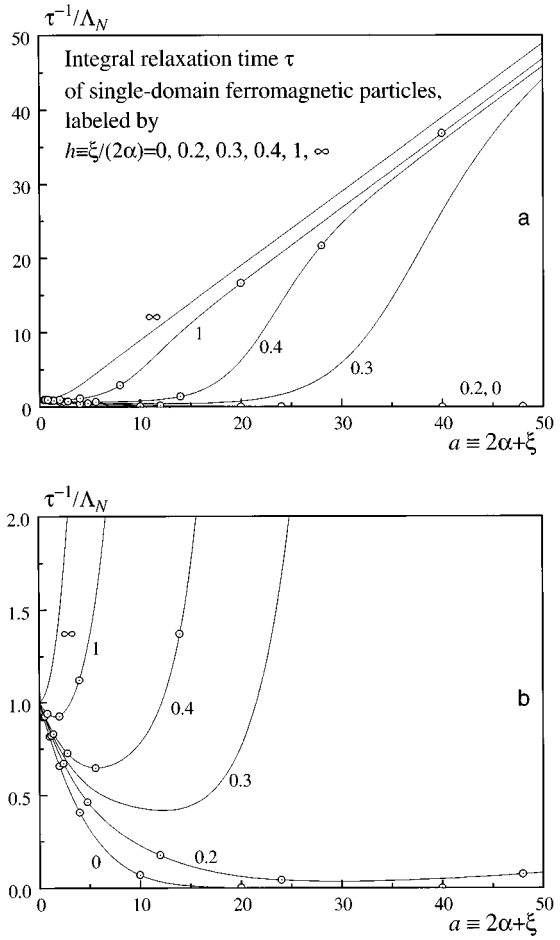

FIG. 3. Inverse of the integral relaxation time τ as a function of the parameter a of (4.16). Circles denote data of Coffey *et al.* [16] taken from their Table II.

In (4.15) [cf. (3.18)] the correction terms, in particular of the type present in (3.12), have been taken into account.

Numerical calculation of the integral relaxation time $\tau_{\rm int}$ given by (4.6) in the whole range of parameters α and ξ poses no difficulties. For the representation of the results for the arbitrary relation between α and ξ, including the case $\alpha=0$, it is more convenient to use the variable

$$a\equiv 2\alpha+\xi=\frac{VM_s}{\gamma T}\omega_R,\qquad(4.16)$$

where $\omega_R=\gamma(2KM_s+H)$ is the ferromagnetic resonance frequency in the lower potential well. One can see that $2\alpha=a/(1+h)$ and $\xi=ah/(1+h)$. In terms of the variables a and h the asymptotic formula (4.15) can be rewritten as $\tau_{{\rm int},W}^{-1}\cong\Lambda_N[a-(5+h)/(1+h)]$, which shows that all curves $\tau_{{\rm int},W}^{-1}(a)$ for different h are parallel to each other. The results of the numerical integration in the formula (4.6) are represented in Fig. 3. These results confirm all the considerations made above, as well as the numerical findings of Coffey *et al.* [16].

V. DISCUSSION

The formula for the integral relaxation time $\tau_{\rm int}$ of an assembly of single-domain ferromagnetic particles (4.6) is

the all-temperature solution of the problem of the thermoactivation escape rate of the particles over a potential barrier. In the unbiased case $\xi=h=0$, the quantity $\tau_{\rm int}$ is very close in the whole temperature range to the inverse of the lowest eigenvalue of the Fokker-Planck equation Λ_1. Besides, in comparison to the latter, $\tau_{\rm int}$ has more physical significance and is given by a simple quadrature (4.6) with (4.8). In the high-barrier strong-bias case $\alpha,\xi\gg 1$, as a result of the depletion of the upper potential well the relaxation is described by two exponentials with uncomparable characteristic times, which correspond to the intrawell and overbarrier processes. In this case it would be naturally an oversimplification to describe the relaxation with a common integral relaxation time, but it is possible to separate analytically both contributions in the general formula (4.6) [see (4.14) and (4.15)] and thus to give a complete description of the relaxation process.

Going beyond the integral relaxation time, one can conceive the calculation of the whole dynamic susceptibility $\chi_z(\omega)$ given by the formula (4.4). In this case the numerical approach of Ref. [16], consisting of the calculation of all A_i and Λ_i, is really useful. However, for the model with a high potential barrier such a calculation is probably not very interesting for the reasons mentioned above: there are, to a good accuracy, only one (in the unbiased case) or two (in the strong-bias case) terms in (4.4). More appealing would be to produce calculations for an isotropic model in a field ($\alpha=0$, $\xi\neq 0$) where the eigenvalues Λ_i are not so well separated. In this case deviations from the Debye form of the longitudinal dynamic susceptibility were studied analytically in Ref. [13], where it was shown that these deviations can be about 7% at $\xi\approx 3$. In Ref. [13] an effective two-relaxator formula for $\chi_z(\omega)$ was proposed, which was argued to describe the main part of deviations from the simple Debye form. It would be interesting to check and improve these results by a direct numerical calculation.

One more unsolved problem is the calculation of the *transverse* integral relaxation time of superparamagnetic particles. Up to now it seems to be considered only for the model of rotating dipoles in Ref. [15].

Although many experimental investigations are currently done on systems showing superparamagnetism, these investigations are practically confined to the certification of a superparamagnetic behavior and to rough estimation of relaxation times. It would be worth making more purposeful measurements aimed at a comparison with existing theories. For this purpose it would be important to eliminate the distribution of particle volumes and orientations of the anisotropy axes.

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