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Asymptotic superparamagnetic time constants for cubic anisotropy. I. Positive anisotropy

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It is shown that the relaxation rate of superparamagnetic particles with a positive cubic anisotropy is determined by *two* (and *only* two) time constants, in the limit of a high energy-barrier. Their ratio in this limit tends towards 3/2. These findings are confirmed by numerical calculations that correct for an error in previous ones. In addition, theoretical expressions for the time constants are derived, which show an $\exp(E_B/kT)$ behavior when $E_B \gg kT$. Here E_B is the barrier energy, T is the absolute temperature, and k is Boltzmann's constant. The numerical computations give reasonably close results to the theoretical expressions, but an asymptotic tendency towards them is not as yet identified with full confidence, in the limited region $E_B \leq 20kT$, where these computations were carried out.

I. INTRODUCTION

Small ferromagnetic particles have a uniform magnetization $\vec{M}$ that in the absence of an applied field is directed along some easy axis where the magnetic anisotropy energy is at a local minimum. Thermal agitations effect continual fluctuating changes in the orientation of $\vec{M}$ and eventually the flip of $\vec{M}$ from one such minimum to another, overcoming an anisotropy energy barrier with height E_B. Here,

$$E_B = KV \quad \text{for uniaxial anisotropy}, \tag{1a}$$

$$E_B = \tfrac{1}{4}KV \quad \text{for cubic anisotropy, with } K>0, \tag{1b}$$

$$E_B = \tfrac{1}{12}|K|V \quad \text{for cubic anisotropy, with } K<0, \tag{1c}$$

where K is the anisotropy energy constant and V is the particle volume. An ensemble of such particles is characterized by a relaxation time τ with which the distribution of orientations of $\vec{M}$ approaches thermal equilibrium. If τ is smaller than the measuring time of the experiment, the particles are termed "superparamagnetic."[1]

In a spherical coordinate system with θ and ϕ as the polar and azimuthal angles, respectively, let us denote by $W(\theta, \phi, t)\,d\Omega$ the probability that (at time t) $\vec{M}$ is oriented within the element of solid angle $d\Omega = \sin\theta\, d\theta\, d\phi$ centered at (θ, ϕ). Using the theory of Brownian motion, Brown[2] has shown that W is the solution of a certain differential equation and has the form

$$W(\theta, \phi, t) = A_0(\theta, \phi) + \sum_{n=1}^{\infty} A_n(\theta, \phi) e^{-p_n t}, \tag{2}$$

where $A_0(\theta, \phi)$ is the Maxwell-Boltzmann distribution describing thermal equilibrium and where

$$p_1 < p_2 < \cdots p_n < \cdots \tag{3}$$

are the eigenvalues of this differential equation.

For a uniaxial anisotropy the differential equation has been solved numerically,[3] and for $E_B \gg kT$ it has been found that[2,3] the physical system is governed by only one constant

$$1/\tau = p_1 = f_0 e^{-E_B/kT}, \tag{4}$$

since $p_1 \ll p_2$. Here T is the absolute temperature, k is Boltzmann's constant, and

$$f_0 = 2K\gamma_0 (KV/\pi kT)^{1/2}/M_s, \tag{5}$$

where M_s is the saturation magnetization and γ_0 is the gyromagnetic ratio.

The relaxation time associated with large (i.e., $E_B \gg kT$) superparamagnetic particles with cubic anisotropy has for some time been the subject of premises only. In the absence of an asymptotic formula to describe τ as $E_B/kT \to \infty$, most authors used the expression (4) for this case too, with f_0 taken as a constant or as given by (5). This has been done in the face of some indications[4,5] that (4) may not be appropriate for this case.

In this paper we derive an approximate expression for τ for the case of a cubic anisotropy with $K>0$, namely when the easy axis is along [100]. This expression justifies the use of (4) with a constant f_0 when E_B/kT is large enough and seems to be consistent with a numerical solution of Brown's differential equation which corrects for an error that fell in previous calculations.[5,6] It will be shown, however, that one should take into account p_2 as well as p_1, since the assumption $p_2 \gg p_1$ does not hold here, although $p_3 \gg p_2$ still holds.

II. NUMERICAL CALCULATIONS AND RESULTS

In the numerical calculations, the reduced time constants

$$\mu_i = (2M_s/\gamma_0 K)p_i \quad [\mathrm{Re}(\mu_i) < \mathrm{Re}(\mu_j) \quad \text{if } i<j] \tag{6}$$

were computed as functions of the parameter

$$\alpha = KV/kT \, , \tag{7}$$

which is proportional to the barrier energy E_B [Eqs. (1)]. The method of computation has already been described before[5,6] and we shall repeat it only to the extent needed to correct for the error that fell in the previous work. After the anisotropy energy density

$$F(\theta, \phi) = \tfrac{1}{4}K(\sin^2 2\theta + \sin^4\theta \sin^2 2\phi) \tag{8}$$

is substituted into Brown's differential equation,[2] the equation is converted into a time-independent eigenequation whose eigenvalues are

$$\lambda_i = \alpha\,\mu_i \ . \tag{9}$$

These are found by expanding the corresponding eigenfunctions $\Phi_i(\theta, \phi)$ according to

$$\Phi_i(\theta, \phi) = \sum_{lm} a_{ilm} P_l^m(\cos\theta) e^{im\phi} \tag{10}$$

and using matrix algebra. Here, the P_l^m are the associated Legendre functions of the first kind and the a_{ilm} are complex constants. The error in the previous calculations consisted in limiting the values of m to the range

$$0 \le m \le l \, , \tag{11}$$

whereas the range

$$0 \le l, \quad -l \le m \le l \tag{12}$$

should have been taken, as is done here. In Fig. 1 we show the results of computing μ_1, μ_2, and $\mathrm{Re}(\mu_3)$. Whereas μ_1 and μ_2 are real, μ_3 becomes complex above $\alpha = 5$ and $\mathrm{Re}(\mu_3)$ becomes much larger than μ_2 as α grows up. (Note that the matrix diagonalized is real so that its complex eigenvalues occur in complex conjugate pairs). On the other hand, μ_1 and μ_2 seem to obey the same law. The computations here were carried out up to $\alpha = 20$, since for larger values of α an excessive computer time would have been needed. It should be noted that for small values of α the previous results[5,6] do not differ much from the present ones. Thus, at $\alpha = 1$ there is only a 0.25% deviation. But at $\alpha = 10$ and $\alpha = 20$ the deviations are already 10% and 30%, respectively. This fact leaves the conclusions of Krop *et al.*[7] unchanged.

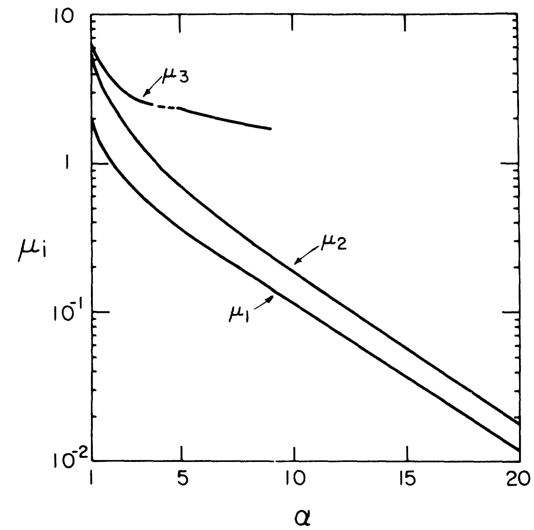

FIG. 1. Reduced numerically computed time constants μ_1, μ_2, and μ_3. [Eqs. (6) and (2)] as functions of the reduced barrier energy α [Eqs. (7) and (1b)] for superparamagnetic particles with cubic anisotropy where the anisotropy constant is $K>0$. For $\alpha \geq 5$, μ_3 becomes complex and $\mathrm{Re}(\mu_3)$ is instead plotted. For $\alpha \gg 1$, only μ_1 and μ_2 determine the relaxation rate.

III. ASYMPTOTIC BEHAVIOR, $\alpha \to \infty$

A. General

Consider an ensemble of uniformly magnetized particles whose magnetization vectors $\vec{M}$ (with M_s constant) can be represented by points on the unit sphere. These points move under the influence of the cubic anisotropy potential $F(\theta, \phi)$ [Eq. (8)] and of thermal agitations, against dissipation forces characterized by a constant η. They describe a Brownian motion whose statistics is governed by Brown's differential equation that can be put in the form of a continuity equation[2]

$$\frac{\partial W}{\partial t} + \vec{\nabla} \cdot \vec{J} = 0 \ . \tag{13}$$

Here $\vec{J}$ is a surface current density of points on the unit sphere whose components are[2]

$$J_\theta = -\left(h' \frac{\partial F}{\partial \theta} - \frac{g'}{\sin\theta}\frac{\partial F}{\partial \phi}\right)W - k'\frac{\partial W}{\partial \theta} \, , \tag{14a}$$

$$J_\phi = -\left(g' \frac{\partial F}{\partial \theta} + \frac{h'}{\sin\theta}\frac{\partial F}{\partial \phi}\right)W - \frac{k'}{\sin\theta}\frac{\partial W}{\partial \phi} \, , \tag{14b}$$

where

$$h' = \frac{\eta}{1/\gamma_0^2 + \eta^2 M_s^2} \, , \quad g' = \frac{h'}{\gamma_0 \eta M_s} \, , \quad k' = \frac{h'}{\beta} \, , \tag{15}$$

with

$$\beta = V/kT \ . \tag{16}$$

In (13) the operator $\vec{\nabla}$ is expressed in spherical coordinates with the radial term omitted. In (14) the terms containing g' constitute a gyromagnetic current density and in the following we shall assume the dissipation constant η to be given by[3]

$$\eta = 1/\gamma_0 M_s \Rightarrow h' = g' = \gamma_0/2M_s \ . \tag{17}$$

In the limit $\alpha \to \infty$ (i.e., $E_B \gg kT$) we assume that a quasistationary state prevails.[8] More specifically, we assume that most of the representative points on the unit sphere are concentrated at the energy minima of the anisotropy potential where they are under conditions of thermal equilibrium and where W coincides with the Maxwell-Boltzmann distribution. Only a small fraction of the points is outside the energy minima allowing a small diffusion current between them that manifests the nonequilibrium conditions. However, we assume that the state is *quasistationary* in that *between* the energy minima

$$\frac{\partial W}{\partial t} = 0 \ . \tag{18}$$

Hence, by the continuity equation (13),

$$\vec{\nabla} \cdot \vec{J} = 0. \tag{19}$$

In the following we shall use (19) to obtain asymptotic (i.e., $\alpha \to \infty$) expressions for the p_i's that are important in the relaxation process. In order to see what these are we shall repeat here, for completeness, a calculation that was misinterpreted in Ref. 5 due to the wrong numerical results.

B. Relations between time constants

Considering the anisotropy potential $F(\theta\phi)$ [Eq. (8)], there are six minima. Two of them are at $\theta = 0$ and $\theta = \pi$, and will be denoted by M_1 and M_3 respectively. The other four minima are at $\theta = \frac{1}{2}\pi$ with $\phi = 0, \ \frac{1}{2}\pi, \ \pi, \ \frac{3}{2}\pi$ and will be denoted by M_2. Let n be the number of points in the ensemble. Since we assume $\alpha \to \infty$, practically all points are concentrated at the energy minima. Their numbers in the minima M_1 and M_3 will be denoted by n_1 and n_3 respectively, and their *total* number in the four minima M_2 by n_2. Since the number of points in the ensemble is constant, we require that at any time t

$$n_1 + n_2 + n_3 = n \ , \tag{20}$$

and we shall further impose the initial conditions

$$n_1(t=0) = n \ , \tag{21a}$$

$$n_2(t=0) = n_3(t=0) = 0 \ . \tag{21b}$$

Let ν be the probability per unit time for a point to pass from one energy minimum to *any* of the four nearest ones. We shall assume that a point arriving at a farther minimum (separated by an angle of π rather than $\frac{1}{2}\pi$, e.g., from M_1 to M_3) must first pass through a nearby minimum, participate in the thermal equilibrium there, and only then may go to the following minimum. This is because α being large and the current density small, the outgoing points are channeled into the nearby minima by the shape of the potential. Taking into account the symmetry of the minima and of the initial conditions (21) (noting, in particular, that the net transfer of points among the minima M_2 themselves vanishes), we obtain the equations

$$\dot{n}_1 = \tfrac{1}{4}\nu n_2 - \nu n_1 \ , \tag{22a}$$

$$\dot{n}_3 = \tfrac{1}{4}\nu n_2 - \nu n_3 \ . \tag{22b}$$

The corresponding equation for n_2 is obtained by differentiating (20) with respect to t and using (22). Equations (20)–(22) have a solution of the form (2) that reads

$$n_1 = \tfrac{1}{6}n(1 + 3e^{-p_1 t} + 2e^{-p_2 t}) \ , \tag{23a}$$

$$n_2 = \tfrac{4}{6}n(1 - e^{-p_2 t}) \ , \tag{23b}$$

$$n_3 = \tfrac{1}{6}n(1 - 3e^{-p_1 t} + 2e^{-p_2 t}) \ , \tag{23c}$$

where

$$p_1 = \nu, \ \ p_2 = \tfrac{3}{2}p_1 \ . \tag{24}$$

In other words, (23) and (24) predict that as $\alpha \to \infty$, *two* (and *only* two) time constants are needed in order to describe the evolution of the system with time, and that the ratio between them is as given by (24). These predictions seem to be fulfilled by the numerical results. In Fig. 2 we show the ratios $\mu_2/\mu_1 (= p_2/p_1)$ and $\mathrm{Re}(\mu_3)/\mu_1 [= \mathrm{Re}(p_3)/p_1]$ as functions of α. Whereas the first ratio seems to converge to the value $\frac{3}{2}$, the second one keeps increasing rapidly. Hence, as $\alpha \to \infty$, the terms

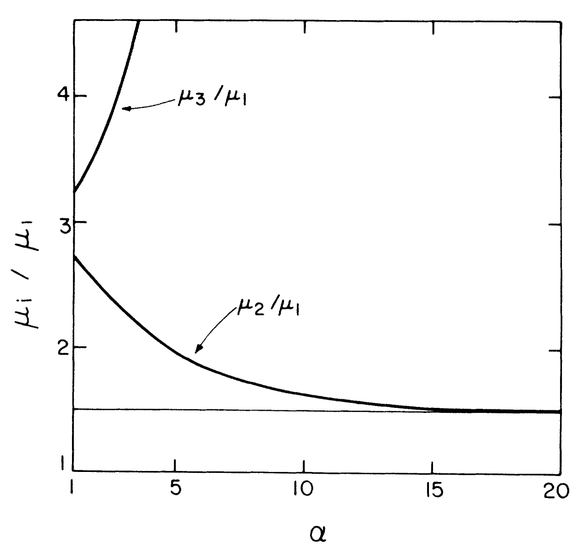

FIG. 2. Numerically computed ratios μ_2/μ_1 and μ_3/μ_1 as functions of α. The μ_i's and α are the same as in Fig. 1.

with $i>2$ in Eq. (2) decay very rapidly to zero and do not influence the relaxation process.

C. Asymptotic expressions $\alpha\to\infty$

Returning to Eq. (19), we write it in the form

$$\frac{1}{\sin\theta}\frac{\partial}{\partial\theta}(\sin\theta J_\theta)+\frac{1}{\sin\theta}\frac{\partial}{\partial\phi}J_\phi=0\ . \tag{25}$$

Integrating over ϕ from $-\frac{1}{4}\pi$ to $\frac{1}{4}\pi$, we get

$$\frac{\partial}{\partial\theta}\int_{-\pi/4}^{\pi/4}J_\theta\sin\theta\,d\phi=-J_\phi\Big|_{\phi=-\pi/4}^{\phi=\pi/4}\ , \tag{26}$$

but since the system and the initial conditions are symmetric under a rotation of $\frac{1}{2}\pi$ in the azimuthal plane, the right-hand side of (26) vanishes, and we have

$$\int_{-\pi/4}^{\pi/4}J_\theta\sin\theta\,d\phi=\tfrac{1}{4}I=\text{const.} \tag{27}$$

Here I is the *total* current of points flowing out of minimum M_1, namely,

$$I=-\dot{n}_1\ . \tag{28}$$

Using (14c), (15), and (16), Eq. (27) becomes

$$\frac{I}{4k'}=-\int_{-\pi/4}^{\pi/4}\left(\beta\frac{\partial F}{\partial\theta}W+\frac{\partial W}{\partial\theta}-\frac{\beta}{\sin\theta}\frac{\partial F}{\partial\phi}W\right)\sin\theta\,d\phi\ . \tag{29}$$

Since $F(\theta,\phi)$ and the initial conditions are symmetric under the transformation $\phi\to-\phi$, we shall *assume* that W is also symmetric under this transformation, namely,

$$W(\theta,\phi)=W(\theta,-\phi)\ . \tag{30}$$

This assumption will be further discussed in Sec. IV.

On the other hand, (8) implies

$$\frac{\partial F}{\partial\phi}(\theta,\phi)=-\frac{\partial F}{\partial\phi}(\theta,-\phi)\ , \tag{31}$$

so that

$$\int_{-\pi/4}^{\pi/4}\frac{\partial F}{\partial\phi}W\,d\phi=0\ . \tag{32}$$

Hence,

$$\frac{I}{4k'}=-\int_{-\pi/4}^{\pi/4}\left(\beta\frac{\partial F}{\partial\theta}W+\frac{\partial W}{\partial\theta}\right)\sin\theta\,d\phi$$
$$=-2\int_0^{\pi/4}e^{-\beta F}\frac{\partial}{\partial\theta}(e^{\beta F}W)\sin\theta\,d\phi\ . \tag{33}$$

Now, W coincides with the Maxwell-Boltzmann distribution inside the potential minima and is small between them. We shall assume that it is a well-behaved function so that $\partial(e^{\beta F}W)/\partial\theta$ is continuous in the region $0\le\phi\le\frac{1}{4}\pi$. Also $e^{-\beta F}>0$ in this region so that we may use the mean value theorem of integral calculus in order to write

$$\frac{I}{4k'}=-2\sin\theta\frac{\partial}{\partial\theta}(e^{\beta F}W)_{\theta,\phi=\zeta}\int_0^{\pi/4}e^{-\beta F}d\phi\ , \tag{34}$$

where ζ is a function of θ and must exist in the region

$$0\le\zeta\le\tfrac{1}{4}\pi\ . \tag{35}$$

Substituting (8) in (34), defining a variable $u\equiv\sin^2 2\phi$ and integrating over u one obtains

$$\frac{I}{4k'}=-\tfrac{1}{2}\pi\sin\theta\frac{\partial}{\partial\theta}(e^{\beta F}W)_{\theta,\zeta}$$
$$\times\exp[-\tfrac{1}{4}\alpha(\sin^2 2\theta+\tfrac{1}{2}\sin^4\theta)]I_0(\tfrac{1}{8}\alpha\sin^4\theta)\ , \tag{36}$$

where I_0 is the modified Bessel function of the first kind and zeroth order, and where a use has been made of (7) and (16). Rearranging terms and integrating over θ we have

$$(e^{\beta F}W)\Big|_{\theta_1,\phi=\zeta(\theta_1)}^{\theta_2,\phi=\zeta(\theta_2)}=-\frac{I}{2\pi k'}\int_{\theta_1}^{\theta_2}G(\theta)\exp(\tfrac{1}{4}\alpha\sin^2 2\theta)d\theta\ , \tag{37}$$

where

$$G(\theta)=\exp(\tfrac{1}{8}\alpha\sin^4\theta)/\sin\theta I_0(\tfrac{1}{8}\alpha\sin^4\theta)\ , \tag{38}$$

and where we look at $0<\theta_1\le\theta\le\theta_2<\frac{1}{2}\pi$. Following Brown,[2] θ_1 and θ_2 are chosen close enough to 0 and $\frac{1}{2}\pi$, respectively, so that a thermal equilibrium exists in $(0,\theta_1)$ and in $(\theta_2,\frac{1}{2}\pi,\phi\lesssim\theta_1)$ (that is, in minimum M_1 and the minima M_2). Still, when $\alpha\to\infty$ θ_1 is assumed (by choice) to obey $e^{-\beta F(0,0)}\gg e^{-\beta F(\theta_1,\phi)}\gg e^{-\beta F_2}$, where F_2 is the value of F at the saddle point $(\theta=\frac{1}{4}\pi,\phi=0)$ separating θ_1 and θ_2. Analogous assumption holds for θ_2. The mean value theorem is again applicable and we have

$$(e^{\beta F}W)\Big|_{\theta_1,\zeta(\theta_1)}^{\theta_2,\zeta(\theta_2)}=-\frac{I}{2\pi k'}G(\xi)\int_{\theta_1}^{\theta_2}\exp(\tfrac{1}{4}\alpha\sin^2 2\theta)d\theta, \tag{39}$$

where

$$\theta_1\le\xi\le\theta_2\ . \tag{40}$$

With due care to the singularity of $1/\sin\theta$ at $\theta=0$, we shall now impose $\theta_1\sim0$ and $\theta_2\sim\frac{1}{2}\pi$. Carrying out the integration

$$(e^{\beta F}W)\Big|_{\theta_1,\zeta_1}^{\theta_2,\zeta_2}=-\frac{I}{4k'}G(\xi)\exp(\tfrac{1}{8}\alpha)I_0(\tfrac{1}{8}\alpha)\ , \tag{41}$$

where

$$\zeta_i\equiv\zeta(\theta_i)\text{ and }0<\xi<\tfrac{1}{2}\pi\ . \tag{42}$$

For a small argument, $I_0(z)\sim e^z\sim1$, and in the limit of a large argument, $I_0(z) \sim e^z/(2\pi z)^{1/2}$, so that when $\alpha \to \infty$, $G(\theta)$ changes much more slowly than $\exp(\frac{1}{4}\alpha \sin^2 2\theta)$ in (θ_1, θ_2). In this limit, most of the contribution to the integral in (37) comes from the region near $\theta = \frac{1}{4}\pi$ [where $\exp(\frac{1}{4}\alpha \sin^2 2\theta)$ has a maximum that corresponds to the saddle of $F(\theta, \phi)$ at $(\theta = \frac{1}{4}\pi, \phi = 0)$] so that we expect ξ to approach $\frac{1}{4}\pi$ as $\alpha \to \infty$. In order to evaluate the left-hand side of (41) we consider times $t \sim 0$ (although this assumption is not essential for the derivation) so that $W(\theta_2, \xi_2) \sim 0$ [Eqs. (21)]. For $0 \le \theta \le \theta_1$, W coincides with the Maxwell-Boltzmann distribution

$$W(\theta, \phi) = C \exp\{-\beta[F(\theta, \phi) - F(0,0)]\}, \quad 0 \le \theta \le \theta_1 , \tag{43}$$

where C is a constant. Hence,

$$(e^{\beta F} W)\Big|_{\theta_1, \xi_1}^{\theta_2, \xi_2} = -C , \tag{44}$$

since $F(0,0) = 0$ [Eq. (8)]. Now, by (43)

$$n_1 = \int_0^{\theta_1} d\theta \int_0^{2\pi} d\phi C \exp\{-\beta[F(\theta, \phi) - F(0,0)]\} \sin\theta , \tag{45}$$

where βF contains α as a factor [Eqs. (7), (8), and (16)]. As $\alpha \to \infty$, the exponential function in (45) decreases rapidly as θ grows above zero. A good approximation is then obtained for the integral if for a given ϕ we expand F in a Taylor's series about $\theta = 0$ up to the θ^2 term, replace $\sin\theta$ by θ, and θ_1 by infinity. The integral obtained does not depend on ϕ and we get

$$n_1 = (\pi/\alpha) C . \tag{46}$$

Using (28), (41), (44), and (46), we have

$$\dot{n}_1 = -[(4k'\alpha/\pi)/G(\xi) \exp(\tfrac{1}{8}\alpha) I_0(\tfrac{1}{8}\alpha)] n_1 . \tag{47}$$

Comparing with (22a) at $t = 0$ [i.e., using (21)], we see that the coefficient of n_1 in (47) equals ν $(= p_1)$. Hence, by (6), (15), and (16),

$$\mu_1 = (4/\pi)/G(\xi) \exp(\tfrac{1}{8}\alpha) I_0(\tfrac{1}{8}\alpha), \quad 0 < \xi < \tfrac{1}{2}\pi , \tag{48}$$

and by (24),

$$\mu_2 = \tfrac{3}{2} \mu_1 .$$

With the same justification discussed after Eq. (42), we first approximate (48) by taking $\xi = \frac{1}{4}\pi$. After substituting from (38),

$$\mu_1 \underset{\alpha \to \infty}{\sim} (4/\pi \times 2^{1/2}) I_0(\alpha/32)/\exp(\tfrac{5}{32}\alpha) I_0(\tfrac{1}{8}\alpha) , \tag{49}$$

or, using $I_0(z) \to e^z/(2\pi z)^{1/2}$ for $z \to \infty$,

$$\mu_1 \underset{\alpha \to \infty}{\sim} (4/\pi) 2^{1/2} e^{-\alpha/4} . \tag{50}$$

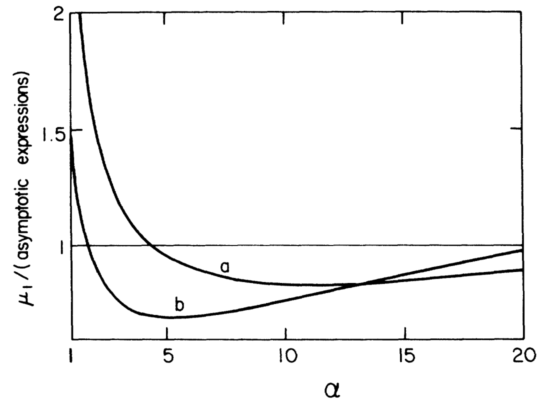

FIG. 3. Ratio of the numerically computed time constant μ_1 [Eqs. (6) and (2)] to the asymptotic expressions (a), $(4/\pi \times 2)^{1/2}) I_0(\frac{1}{32}\alpha)/\exp(\frac{5}{32}\alpha) I_0(\frac{1}{8}\alpha)$ [Eq. (49)], (b), $(4/\pi)2^{1/2}e^{-\alpha/4}$ [Eq. (50)]. The curves are plotted as functions of the reduced barrier energy α [Eqs. (7) and (1b)].

This is different from the asymptotic formula for a uniaxial anisotropy,

$$\mu_1(\text{uniaxial}) \underset{\alpha \to \infty}{\sim} 4(\alpha/\pi)^{1/2} e^{-\alpha} , \tag{51}$$

in the different values of E_B [Eq. (1)] in the exponential function and in the preexponential factor. The ratios of the numerically computed μ_1 to the expressions (49) and (50) are shown in Fig. 3 in curves (a) and (b), respectively. We see that although the ratios are rather close to 1.0 already at $\alpha \sim 5$, there is some overshoot, and they do not quite seem to converge to this value at the vicinity of $\alpha = 20$. But when one is interested in the order of magnitude only, which is very often the case, the approximations (49) and (50) are more or less equivalent and hold down to α as low as 1. If (15) is not assumed, then

$$k' = \eta/\beta(1/\gamma_0^2 + \eta^2 M_s^2) , \tag{52}$$

so that (50) transforms to

$$\mu_1 \underset{\alpha \to \infty}{\sim} [8 \times 2^{1/2}/\pi(\gamma_0 \eta M_s + 1/\gamma_0 \eta M_s)] \exp(-\tfrac{1}{4}\alpha) . \tag{53}$$

IV. DISCUSSION

The underlying assumption that leads to Eqs. (23), (24), and (41) is that α is large enough in order that our ensemble is under quasistationary conditions [with (41) augmented with assumption (30)]. This seems to be justified already at $\alpha = 20$ by the numerical results as mentioned in Sec. III B [Eq. (24) and Fig. 2]. It is consistent with the case of a uniaxial anisotropy since at $\alpha = 20$, $E_B/kT = 5$ [Eq. (1b)] which corresponds to $\alpha = 5$ in the uniaxial case where the above assumption was shown to be well justified.[3] The passage from

(41) to (48) can also be considered as justified by the uniaxial analogy[3] since the same approximations are used successfully in that case.

However, the derivation of (49) and (50) contains another approximation related to the shape of the anisotropy potential. Whether the approximation $\xi = \frac{1}{4}\pi$ is good enough depends on whether the maximum of the term $\exp(\frac{1}{4}\alpha \sin^2 2\theta)$ in (37) is high and sharp enough relative to the other terms in the integrand. Due to the $G(\theta)$ term that comes through the ϕ dependence of F [see Eqs. (33)–(38)], these requirements are not as easily fulfilled as in the uniaxial case. Although by Fig. 3 the approximation is rather good, the curves probably overshoot the value of 1 and only then (or through further oscillations) approach it asymptotically. Another cause for the deviation of (50) from the numerical results is probably assumption (30) that amounts to dropping the gyroscopic contribution to the total polar current. Thus, instead of (30) one can assume $g' \ll h'$ (or $\eta \gg 1/\gamma_0 M_s$) and obtain (53) after minor changes in the calculations. The numerical results, however, were obtained for $g' = h'$ and according to Fig. 3 it is seen that the difference is not large. Moreover, dropping (30) does not change the dependence of μ_1 on α and thus cannot make the slope of curve (b) in Fig. 3 approach the asymptotic convergence. This can be seen in a recent work by Smith and de Rozario that came to our knowledge after completing our calculations. Using a different approach, they obtain the same dependence on α with a different pre-exponential factor that formally tends to our result for a large dissipation. Their results indeed show that generally (30) is not fulfilled.

In Fig. 4 we show the value of ξ for which the relation (48) holds as a function of α. It so happens that for each numerically computed value of $\mu_1(\alpha)$ there exist two values of $\xi(\alpha)$ that fulfill (48). One value decreases rather quickly as α increases (at $\alpha = 20$ it is less than 25°), and the other one is shown in Fig. 4. One cannot tell from the figure whether it is going to converge to $\frac{1}{4}\pi$.

Concerning the passage from (49) to (50), this is certainly not justified *a priori* for $\alpha < 32$. Indeed we see that curves (a) and (b) representing (49) and (50) in Fig. 3 do not seem to converge into one another at $\alpha \leq 20$ as they should do for $\alpha \to \infty$. This fact may show that we should not

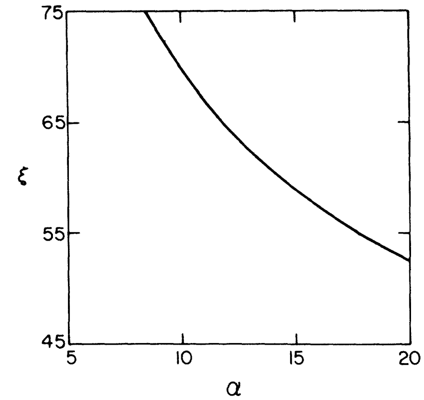

FIG. 4. Value of the angle ξ that satisfies the formula $\mu_1 = (4/\pi)/G(\xi)\exp(\frac{1}{8}\alpha)I_0(\frac{1}{8}\alpha)$ [Eq. (48)] as a function of α. Here μ_1 and α are the same as in Fig. (3), and $G(\xi)$ is defined in (38).

worry too much about their nonconvergence into the value of 1 in the region considered. But at all events the formulas give a reasonably practical approximation in this region.

To conclude, we want to comment on the dependence of the relaxation process on two time constants rather than one [Eqs. (23)]. Consider an assembly of small noninteracting ferromagnetic particles with cubic anisotropy that is represented by the ensemble discussed in the foregoing. Suppose that all particles are aligned with their [001] axes in the same direction and that they are magnetized in this direction by means of a large magnetic field. Suppose further that at time $t = 0$ the magnetic field is switched off and the remanent magnetization M_r is measured as a function of time. According to the discussion in Sec. III B,

$$M_r = M_s(n_1 - n_3)n \,, \tag{54}$$

and according to (23)

$$M_r = M_s e^{-p_1 t} \,, \tag{55}$$

which is independent of p_2.

The second time constant p_2 can be measured if an experiment is contrived which determines some other combination of the n_i's.

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