Theoretical relaxation times of large superparamagnetic particles with cubic ansiotropy*

A. Aharoni and I. Eisenstein
Department of Electronics, The Weizmann Institute of Science, Rehovoth, Israel
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The superparamagnetic relaxation time is calculated for spherical particles with cubic magnetocrystalline anisotropy in zero applied field. Computation is extended to larger particle size, namely larger energy barriers, than could be evaluated in the previous work. Still, no asymptotic formula could be found to describe the behavior in this size region. It is found that the increase of the relaxation time with increasing particle size is much faster than can be obtained from a simple exponential, from the asymptotic formula for uniaxial anisotropy, or from a formula obtained by an analogous procedure to the uniaxial case. Unless these asymptotic formulas take over only for much larger barriers than studied here, it seems that the complications imposed by the cubic anisotropy potential cannot be ignored.

I. INTRODUCTION

When a ferromagnetic particle is smaller than a certain critical size, its magnetization $\vec{M}$ is uniform so that it constitutes a single-domain particle.[1,2] In the absence of an applied field, $\vec{M}$ is along some easy direction of magnetization. Under the influence of thermal fluctuations, $\vec{M}$ may change its direction, overcoming an anisotropy energy barrier. The parameter that counts is the ratio of this barrier energy to the thermal energy, which is proportional to the absolute value of a quantity α defined by

$$\alpha = KV/kT, \tag{1}$$

where K is the anisotropy constant, V is the particle volume, k is Boltzmann's constant, and T is the absolute temperature. An ensemble of such particles approaches a thermal equilibrium, for the statistical distribution of magnetization orientations, with a characteristic relaxation time τ. When τ is smaller than the experimental measurement time, the particles are called "superparamagnetic"[3] with respect to that experiment. Under the assumption of uniform rotation of the magnetization, Brown[4] found $1/\tau$ to be the smallest nonvanishing eigenvalue of a Fokker-Planck-type differential equation. He[4] has also deduced the following expression for the asymptotic behavior of $1/\tau$ for large α in the case of uniaxial anisotropy:

$$\frac{1}{\tau} = \frac{2|K|\gamma_0}{M_s}\left(\frac{\alpha}{\pi}\right)^{1/2} e^{-E_B/kT}, \tag{2}$$

where γ_0 is the gyromagnetic ratio e/mc, M_s is the saturation magnetization, and E_B is the anisotropy barrier energy. Solving the relevant equation numerically, formula (2) was shown by Aharoni[5] to be already satisfactory for values of α of the order of 1.

A numerical solution in a limited range of α ($1 \le |\alpha| \le 10$) was given by Aharoni[6] also for the case of cubic anisotropy. Limitations set by the computer used made it practically impossible to extend this range to larger values of $|\alpha|$. Such an extension (up to $|\alpha| \le 24.5$ for $K<0$ and $\alpha \le 24$ for $K>0$) is reported here together with an attempt to match the results with some simple and meaningful asymptotic formulas.

II. THEORY AND COMPUTATIONS

The basic theory appears in Ref. 6 and will be described here in brief. The anisotropy energy density is

$$F = \tfrac{1}{4}K(\sin^2 2\theta + \sin^4\theta \sin^2 2\phi) \tag{3}$$

(with $K>0$ for [100] as an easy direction and $K<0$ for [111] as the easy axis), where θ and ϕ are the polar and azimuthal angles, respectively, of $\vec{M}$ with respect to the system of crystallographic axes. F is substituted into Brown's differential equation[4] and the eigenfunction Φ is expanded according to

$$\Phi(\theta,\phi) = \sum_{l,m} a_{l,m} P_l^m(\cos\theta)\, e^{im\phi}, \quad 0 \le m \le l \tag{4}$$

where P_l^m are the associated Legendre functions of the first kind. After some manipulations one obtains the following infinite set of algebraic equations:

$$\sum_{k=-4}^{4} [A_k(l,m)a_{l+k,m+4} + B_k(l,m)a_{l+k,m} + C_k(l,m)a_{l+k,m-4}] = 0, \tag{5}$$

where the 27 complex coefficients A_k, B_k, C_k (for given l and m) are given in the Appendix of Ref. 6. The smallest nonvanishing eigenvalue λ of the equation appears in B_0 only and is related to the relaxation time τ by

$$\lambda = \left(\frac{2VM_s}{\gamma_0 kT}\right)\frac{1}{\tau} \quad . \tag{6}$$

Each pair of indices (l, m) is replaced by a single index

$$n = \tfrac{1}{2}l(l+1) + m + 1 \ , \tag{7}$$

and the problem is essentially that of diagonalizing the infinite matrix of the complex coefficients A_k, B_k, C_k.

In diagonalizing an infinite matrix the standard procedure is to diagonalize some finite-order matrix, then increase the order and repeat the computation until a further increase in the matrix order causes a negligible change in the smallest nonvanishing eigenvalue (which is the only eigenvalue that counts for the present problem). However, it turns out that the final order needed for the matrix of the coefficients of (5) increases very rapidly with increasing $|\alpha|$. This caused the previous computations[6] to stop for $|\alpha| = 10$, when the limit of matrix size, imposed by the computer memory space, was reached. In order to extend the computations to larger values of $|\alpha|$, the original matrix was first transformed into a real one, which saves half the required computer memory for storing such a matrix. The transformation used included two steps. In the first place the $a_{l+k,m}$ coefficients were replaced by $a_{l+k,m}/i^{l+k}$, and the $A(l, m)$ matrix elements were replaced by $i^{l+k}A_k(l, m)$. [The $C_k(l, m)$'s and $B_k(l, m)$'s were transformed similarly.] This is legitimate since the $a_{l+k,m}$ coefficients depend on the sum $l+k$ rather than on each index separately. In the second place, each equation in (5) was divided by i^l and the matrix elements $i^{l+k}A_k$ were replaced by $i^k A_k$. The complete transformation is then

$$\begin{aligned} &a_{l+k,m} \to a'_{l+k} = a_{l+k,m}/i^{l+k} \ , \\ &A_k(l,m) \to A'_k(l,m) = i^k A_k(l,m) \end{aligned} \tag{8}$$

(and similarly for C_k, B_k). It does not change the contents of the basic equations (5) but on the other hand, it is easily seen that the matrix elements A'_k, B'_k, C'_k are all real.

This transformation to a real matrix allowed pushing further the values of $|\alpha|$ for which standard computer subroutines could be used for the diagonalization. For still larger values of $|\alpha|$, the limit of the computer memory capacity was reached and a different method had to be used. In this method, for a given α, we allow λ to assume arbitrary values—say x. The determinant of coefficients A'_k, B'_k, C'_k should then vanish whenever x is equal to one of the eigenvalues of Brown's equation,[4] and it therefore changes sign in passing through such an eigenvalue. From the curve of λ vs α for preceding values of α, we estimate λ for the present α. Taking far enough values of x on both sides of this λ (and taking care not to pass nearby roots) we find the determinant to be positive for one value of x and negative for the other. By successive interpolations we narrow the difference between these two x values, and the desired λ that is between them can be obtained with any desired accuracy available by the computer. The order of the determinant is then increased, and similar computations follow until there is a negligible change in λ upon further increase of order. In this way only one eigenvalue is computed, but others can also be found by the same method if desired. The main advantage is that unlike the first method in which the subroutines available from the computer library required the whole matrix to be introduced into the computer memory, this second method used a routine which required only a part of the matrix at a time. Determinants up to the order of 650 were used for $\alpha \leq 24$, and it seems that increasing the range of α would require very large determinants and excessive computer time ($\alpha = 25$ is estimated to require a matrix order of about 900).

As a check on the computer program, numerical results were compared with the previous work[6] in the region of overlap, and a complete agreement was found. (The routines used in the two cases were written independently, not to mention the passage from a complex to a real matrix.) The second method, used for high values of $|\alpha|$, was checked against the first one for some lower values and was found to yield the same results. All computations were done in double precision (on the IBM model 370 computer), and some of the results were checked by using quadruple precision, which yielded the same results.

Results for the three lowest roots are shown in Fig. 1 and Table I for $K > 0$ and in Fig. 2 and Table II for $K < 0$. They are expressed in terms of

$$\mu = \frac{\lambda}{|\alpha|} = \frac{2M_s}{\gamma_0 |K| \tau} \ . \tag{9}$$

III. ASYMPTOTIC BEHAVIOR

Following Brown,[4] we look at the magnetization vectors of the particles in the ensemble as describing points on the unit sphere. These points move under the influence of the anisotropy potential and thermal fluctuations and describe a Brownian motion whose statistics is governed by Brown's Fokker-Planck-type differential equation.[4]

In the asymptotic region $|\alpha| \to \infty$, i.e., for $|K|V \gg kT$, it is expected that almost all points are concentrated at the energy minima of the anisotropy potential where they are under conditions of thermal equilibrium governed by Boltzmann's statistics. Only a small fraction of the points are outside the energy minima, allowing a small cur-

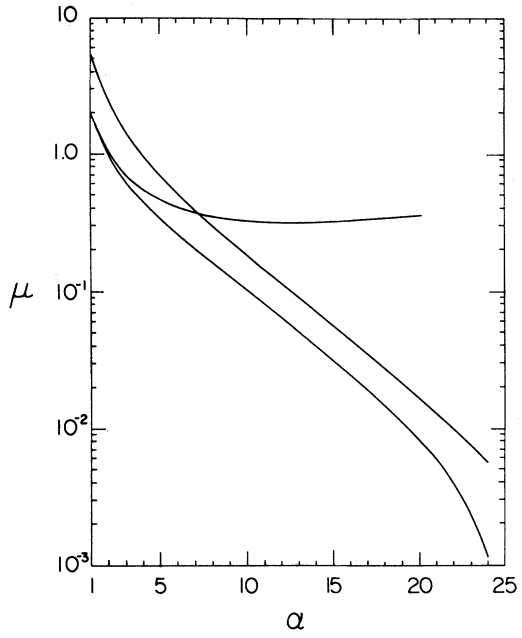

FIG. 1. Parameter μ, which is inversely proportional to the theoretical superparamagnetic relaxation time τ, according to Eq. (9), for cubic anisotropy with [100] the easy axis, i.e., Eq. (3) with $K>0$. The abscissa α is defined in Eq. (1) and is proportional to the energy barrier.

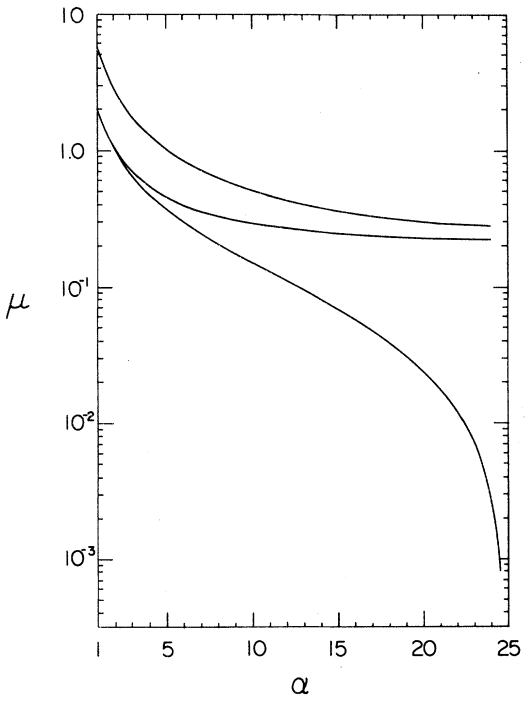

FIG. 2. Same as Fig. 1 for the case of a cubic anisotropy with the easy axis along [111], i.e., Eq. (3) with $K<0$.

TABLE I. Numerical values for Fig. 1.

α	First-lowest parameter μ	Second-lowest parameter μ	Third-lowest parameter μ
1	1.991	2.012	5.445
2	0.981	1.025	2.460
3	0.636	0.704	1.476
4	0.457	0.551	0.991
5	0.345	0.464	0.707
6	0.268	0.411	0.523
7	0.210	0.377	0.395
8	0.166	0.304	0.355
9	0.132	0.236	0.342
10	0.105	0.185	0.334
11	0.828×10^{-1}	0.145	0.330
12	0.655	0.115	0.329
13	0.515	0.908×10^{-1}	0.330
14	0.411	0.719	0.332
15	0.324	0.569	0.336
16	0.255	0.450	0.341
17	0.202	0.355	0.346
18	0.150	0.282	0.351
19	0.114	0.215	0.356
20	0.819×10^{-2}	0.166	0.362
21	0.597	0.128	
22	0.406	0.983×10^{-2}	
23	0.247	0.749	
24	0.115	0.569	

rent between them that decays with time.

Let us discuss the case $K>0$. There are six minima; two of them are at $\theta=0$ and $\theta=\pi$, and will be denoted by I and III, respectively. The other four minima are at $\theta=\frac{1}{2}\pi$ for $\phi=0, \frac{1}{2}\pi, \pi, \frac{3}{2}\pi$, and all four of them taken together will be denoted by II. As has been mentioned before, each particle in the assembly is represented by a point on the unit sphere. Let the number of such points at the minima I, II, and III be denoted by n_1, n_2, and n_3, respectively. Let ν be the probability per point and per unit time for a point to pass from one minimum to any one of the four surrounding minima. (Because of symmetry it is the same for all minima.) And let $\bar{\nu}$ be the probability per point and per unit time for a point to pass directly from I to III or vice versa. (The two probabilities are the same because of symmetry.)

If n is the total number of these points, the initial conditions at $t=0$ are

$$n_1=n,\quad n_2=0,\quad n_3=0\ . \tag{10}$$

Since particles are not created or annihilated, it is required that at any time

$$n_1+n_2+n_3=n\ . \tag{11}$$

Using symmetry of the minima, and in particular the symmetry of minima II with respect to the initial conditions (which means that there is no net transfer of points among themselves), we write down the "evolution" equations:

TABLE II. Numerical values for Fig. 2.

$\|\alpha\|$	First-lowest parameter μ	Second-lowest parameter μ	Third-lowest parameter μ
1	1.993	2.012	5.675
2	0.986	1.024	2.722
3	0.648	0.702	1.761
4	0.476	0.546	1.294
5	0.372	0.456	1.022
6	0.301	0.399	0.845
7	0.249	0.360	0.723
8	0.210	0.332	0.633
9	0.178	0.311	0.565
10	0.152	0.295	0.512
11	0.130	0.282	0.469
12	0.111	0.272	0.435
13	0.950×10^{-1}	0.263	0.407
14	0.808	0.257	0.384
15	0.684	0.251	0.364
16	0.574	0.246	0.348
17	0.476	0.242	0.335
18	0.389	0.238	0.324
19	0.311	0.235	0.314
20	0.241	0.233	0.307
21	0.179	0.231	0.300
22	0.123	0.229	0.295
23	0.732×10^{-2}	0.227	0.290
24	0.285	0.226	0.287
24.5	0.799×10^{-3}		

$$\dot{n}_1 = \tfrac{1}{4}\nu n_2 - \nu n_1 + \bar{\nu} n_3 - \bar{\nu} n_1 , \tag{12a}$$

$$\dot{n}_3 = \tfrac{1}{4}\nu n_2 - \nu n_3 + \bar{\nu} n_1 - \bar{\nu} n_3 . \tag{12b}$$

The equation for n_2 is then determined by differentiating (11) with respect to t.

The general solution of Brown's equation for the distribution of points on the unit sphere is

$$W(\theta, \phi, t) = \sum_{n=0}^{\infty} A_n(\theta, \phi)\, e^{-p_n t} . \tag{13}$$

In our case we find a solution of the form (13) with two decay constants (aside from $p_0 = 0$),

$$p_1 = \tfrac{3}{2}\nu, \quad p_2 = \nu + 2\bar{\nu} , \tag{14}$$

this solution being

$$n_1 = \tfrac{1}{6} n\,(1 + 2e^{-p_1 t} + 3e^{-p_2 t}) , \tag{15a}$$

$$n_2 = \tfrac{4}{6} n\,(1 - e^{-p_1 t}) , \tag{15b}$$

$$n_3 = \tfrac{1}{6} n\,(1 + 2e^{-p_1 t} - 3e^{-p_2 t}) . \tag{15c}$$

(Similar calculation holds for $K < 0$ with the only difference that three decay constants are needed for the solution.) Let us denote the three lowest nonvanishing roots, obtained by the numerical computations, by μ_1, μ_2, and μ_3 in increasing order. Then, they are proportional to p_1, p_2, and p_3. From Fig. 1 we see that, indeed, only two roots μ_1 and μ_2 may be significant at large values of α. (On the other hand, the numerical computations for $K < 0$ indicate that only one root may be significant for large $|\alpha|$.)

Let us go further and assume that for large enough α there is no direct passage of points from minimum I to III, or vice versa. That means that a point arriving at III from I has passed through one of the minima II and participated in the thermal equilibrium in this minimum before jumping to III and that during an infinitesimal interval of time dt no point that begins at I ends at III. Mathematically it means that $\bar{\nu} = 0$ so that the two decay constants p_1 and p_2 are related by

$$p_1 = \tfrac{3}{2} p_2 . \tag{16}$$

However, the numerical results indicate that for large α the ratio μ_2/μ_1 tends to grow with α (exceeding $\frac{3}{2}$) so that, at least in the region used, this assumption of $\bar{\nu} = 0$ is not correct. Since, according to (14), $p_1/p_2 \leq \frac{3}{2}$ and the ratio μ_2/μ_1 increases with α and exceeds $\frac{3}{2}$, we must assume that, if we are really in the asymptotic region, μ_1 corresponds to p_1 and μ_2 correcponds to p_2 and that the ratio $\bar{\nu}/\nu$ increases with α. It is interesting to note that, using a method similar to that used by Brown for the uniaxial case (the Kramers method[7]), one can obtain, in the case $\bar{\nu} = 0$, an expression for p_1 that goes like $e^{-\alpha/4}$ (and consequencly for p_2, too) whereas the numerical computations indicate a faster decrease of μ_1 with α for large α.

Assuming that we are in the asymptotic region, p_1, which is identified with the smallest nonvanishing decay constant, is essentially the decay constant relating a transfer between two adjacent minima. In view of this, an attempt was made to deduce an expression for it by averaging over the asymptotic expression in the uniaxial case.

A point going from the minimum at $\theta = 0$ to the minimum at $\theta = \frac{1}{2}\pi$, $\phi = 0$ has to overcome an anisotropy potential barrier in the shape of a saddle stretching between $\theta = \cos^{-1}(1/\sqrt{3})$, $\phi = -\frac{1}{4}\pi$ and $\theta = \cos^{-1}(1/\sqrt{3})$, $\phi = \frac{1}{4}\pi$. Points that go astray to other minima at $\theta = \frac{1}{2}\pi$ are compensated for by similar points related to these minima. On the saddle, to be denoted by S, we have the relation

$$\frac{\partial}{\partial \theta} F(\theta, \phi) = 0 , \tag{17}$$

where F is the anisotropy potential density defined in (3). Equation (17) gives θ in terms of ϕ on S. For any given ϕ there is a potential barrier whose height $E_B(\phi)$ equals the height of the saddle S at that ϕ. We assume that the differential decay constant $\xi(\phi)\,dl$ for passing the saddle through a length element dl at ϕ is proportional to the asymptotic expression for the decay constant in the uniaxial case, namely

$$\xi(\phi)\,dl = C\left(\frac{E_B(\phi)}{kT}\right)^{1/2} \exp[-E_B(\phi)/kT]\,dl , \tag{18}$$

where C is a constant and dl is given by

$$dl = \left[\sin^2\theta + \left(\frac{d\theta}{d\phi}\right)^2\right]_S^{1/2} d\phi \ . \tag{19}$$

The subscript S in (19) indicates that the quantities in the square brackets should be evaluated on the saddle S. The average decay constant is then

$$\xi = \frac{1}{l}\int_S \xi(\phi)\, dl \ , \tag{20}$$

which, for large-enough α, should be proportional to the numerically calculated decay constant μ_1. After some manipulations one arrives at the integral

$$\xi = A\alpha^{1/2} e^{-\alpha/4} \int_0^2 \left(\frac{x+6-x^2}{x(2-x)(6-x)}\right)^{1/2} e^{-(\alpha/24)x} dx \ , \tag{21}$$

where A is a constant

In the range of integration, the factor $(x+6-x^2)/(6-x)$ in the integrand changes between 1 and 2 whereas the rest of the integrand is singular at $x=0$ and at $x=2$ and for large α the exponential factor changes rather fast. We therefore replace this slowly varying factor by a constant and obtain

$$\xi = A'\alpha^{1/2} e^{-\alpha/4} \int_0^2 \frac{e^{-(\alpha/24)x}}{[x(2-x)]^{1/2}}\, dx$$
$$= A''\alpha^{1/2} e^{-(7\alpha/24)} I_0(\alpha/24) \ , \tag{22}$$

where A'' is a constant and I_0 is the modified Bessel function of the first kind.

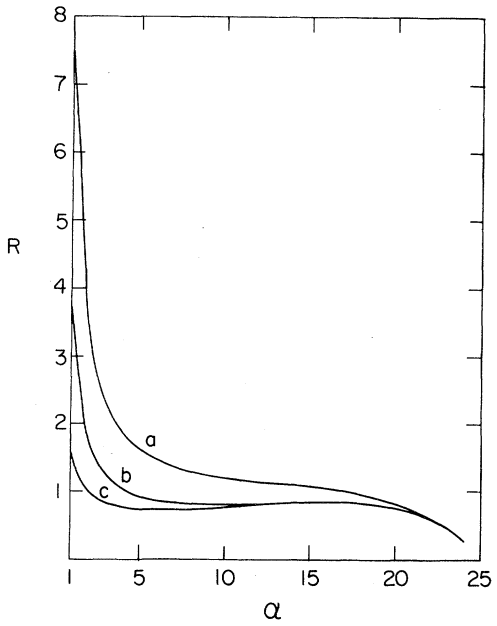

FIG. 3. Ratio of the lowest curve of μ vs α of Fig. 1 to the following possible asymptotic behaviors: (a) $\alpha^{1/2}e^{-\alpha/4}$; (b) $\alpha^{1/2}I_0(\alpha/24)e^{-(7\alpha/24)}$; (c) $e^{-\alpha/4}$. I_0 is the modified Bessel function of the first kind. All three curves are normalized in such a way that they have the same value at $\alpha = 24$.

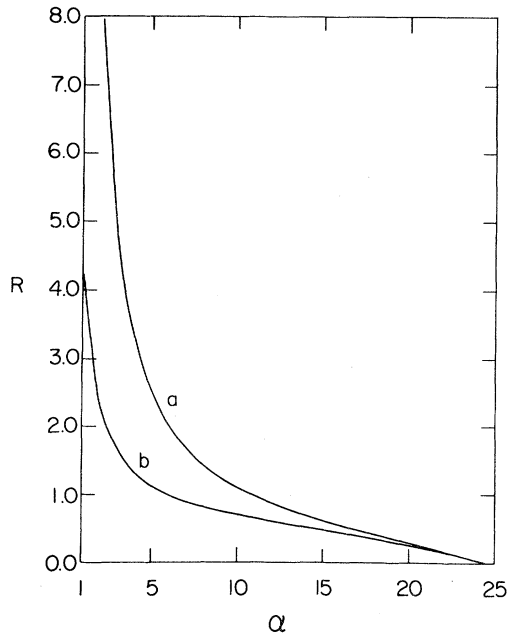

FIG. 4. Ratio of the lowest curve of μ vs α of Fig. 2 to the following asymptotic behaviors: (a) $|\alpha|^{1/2}e^{-|\alpha|/12}$; (b) $e^{-|\alpha|/12}$. The two curves are normalized in such a way that they have the same value at $|\alpha| = 24.5$.

Bessel function of the first kind.

In order that the asymptotic formula (22) be valid, the ratio of the numerically calculated decay constant μ_1 to ξ should tend asymptotically towards being a constant for large values of α. This ratio μ_1/ξ is plotted in Fig. 3 (curve b). Also plotted are the ratio of μ_1 to the uniaxial asymptotic formula, $\mu_1/\alpha^{1/2}e^{-\alpha/4}$ (curve a), and the ratio of μ_1 to a simple exponential, $\mu_1/e^{-(\alpha/4)}$ (curve c). All three ratios were normalized in such a way that they are the same for $\alpha = 24$. Since none of them shows an asymptotic tendency towards being a constant, it is concluded that if we are in the asymptotic region, the behavior of μ_1 cannot be represented by a simple exponential or by the uniaxial asymptotic formula, or by formula (22) since μ_1 falls down faster than any one of them.

Similar conclusions apply to the case $K<0$. Here a calculation analogous to that resulting in (22) was not tried, and in Fig. 3 we show only the ratio of μ_1 to a simple exponential, $\mu_1/e^{-|\alpha|/12}$ (curve b), and to the uniaxial asymptotic behavior, $\mu_1/|\alpha|^{1/2}e^{-|\alpha|/12}$ (curve a). Here μ_1 is that corresponding to the case $K<0$ and the factor $\frac{1}{12}$ appears in the exponential since in the case $K<0$ the anisotropy barrier energy is $\frac{1}{12}|K|V$.

IV. DISCUSSION

Concerning the results obtained, the first question to be asked is whether at about $\alpha = 24$ we have

really reached the asymptotic region in the sense that some asymptotic formula for the decay constant, based on the assumption that the energy barrier for flipping the magnetization is much higher than the thermal energy of fluctuations, is valid there. In the uniaxial case where the anisotropy barrier energy is KV, the asymptotic formula is already valid at about $\alpha = 3$. We may therefore expect that for a cubic anisotropy the asymptotic region is reached at about $\alpha = 12$ for $K > 0$ and only at about $|\alpha| = 36$ for $K < 0$, since the lowest barrier energies in these two cases are $\frac{1}{4}KV$ and $\frac{1}{12}|K|V$, respectively. On the other hand, a different criterion for the asymptotic region may be the magnitude of the (numerically calculated) decay constant μ_1 since this is the physically observed quantity directly related to the relaxation time τ. If we adopt this criterion we are certainly in the asymptotic region (by comparison with the uniaxial case). The rather fast decrease of μ_1 with $|\alpha|$ in both cased ($K \lesseqgtr 0$) may then be attributed to the greater number of possibilities of the magnetization to rotate. In the uniaxial case there is only the possibility of a 180° rotation, and the asymptotic formula (2) applies. In the cubic case, for $K > 0$, there is a competition between falling into the minima at $\theta = \frac{1}{2}\pi$ (a 90° rotation) and falling into the minimum at $\theta = \pi$ (a 180° rotation). For $K < 0$ there is a competition between three such processes: falling into the nearest minima [a rotation by $\theta = 2\sin^{-1} \times (1/\sqrt{3})$], into the next ones [a rotation by $\theta = 2\sin^{-1} \times (\sqrt{2}/\sqrt{3})$], and into the minimum at $\theta = \pi$ (a 180° rotation). In such circumstances we may not expect formula (2) to hold. The failure of (22) for the case $K > 0$ may be attributed to the role played by the shape of the potential density. This latter argument (as well as that concerning the number of minima) is favored by the observation that going from $K > 0$ to $K < 0$, namely, to a more complicated potential density (from the point of view of the minima), the decrease of μ_1 with $|\alpha|$ is more pronounced. It looks as if the complicated shape of the potential brings about an increase in the effective height of the energy barrier. Of course, if we have not reached the asymptotic region in our calculations, one of the formulas (2) or (22) may still describe the true asymptotic behavior which may be reached at much higher values of $|\alpha|$.

Experiments that are directly related to our computations and that include all relevant data are rather rare. Krop and Williams[8] have measured two values of $|\alpha|$ and the corresponding λ in β-Co particles that are of the fcc structure with $K < 0$. Their results, $\lambda = 0.0109$ for $|\alpha| = 1.67$ and $\lambda = 0.0105$ for $|\alpha| = 1.84$, are not consistent with the numerical results quoted here (and not with the uniaxial results, either) for which we find $\lambda = 2.04$ for $|\alpha| = 1.67$ and $\lambda = 1.97$ for $|\alpha| = 1.84$. As suggested by the authors themselves, the measurements may not have been accurate enough. A much better agreement is obtained to later, more detailed measurements of Krop.[9]

Weil[10] has performed granulometry of Ni particles that have cubic structure with $K = -4.5 \times 10^4$ ergs/cm^3. He estimated his measuring time as 10 sec and analyzed his results in terms of Néel's formula.[11] Since $M_s = 500$ G we find from Eq. (9) $\mu \simeq 10^{-10}$. From the slope of the curve of μ vs $|\alpha|$ at $|\alpha| = 24.5$ we find that, if there is no critical change in the following region, this value of μ corresponds to $|\alpha|$ that is between 25 and 30. Using the T values of Weil's experiment we find that this value of $|\alpha|$ corresponds to particles that are about 1.5 times bigger than his estimation (whose average was checked independently by direct methods) which is of the same order of magnitude.

Jordan[12] applied the uniaxial formula (2) for τ in order to compute the susceptibility of cubic Co particles that have a distribution of magnitudes in a time-varying field, and obtained poor agreement with experiment. An attempt that was made to follow his[12] calculation using the present results brought no improvement; however, this may be due to using a range of α that was not large enough.

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