# Computer-simulation study of magnetic relaxation in anisotropic magnetic systems

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We present a computer-simulation study of the effect of the distribution of energy barriers in an anisotropic magnetic system on the relaxation behavior of the magnetization. While the relaxation law for the magnetization can be approximated in all cases by a time logarithmic decay, the law for the dependence of the magnetic viscosity with temperature is found to be quite sensitive to the shape of the distribution of barriers. The low-temperature region for the magnetic viscosity never extrapolates to a positive no-null value. Moreover our computer simulation results agree reasonably well with some recent relaxation experiments on highly anisotropic single-domain particles.

## I. INTRODUCTION

Relaxation phenomena from an arbitrary initial state to an equilibrium or stationary one are of great importance in physics. This is both because they are present in many situations of interest in science and technology and because they represent particular situations of physics out of equilibrium, which is an actively developing part of physics. In this paper we will address the particular case of magnetic relaxation processes of systems of small noninteracting particles with two spin states available to each particle and a distribution of barriers of potential energy between the two states. This situation has become one of actual interest because experimental observations of such relaxation phenomena provide relevant information on important problems in magnetism and superconductivity.

For the simplest theoretical case of systems of particles with just a single size for the barriers, the problem of the relaxation has an easy solution. If a magnetic field that aligns all magnetic moments in one direction is removed, the total magnetic moment of that system will decay according to the law $M(t)=M(0)\exp(-\Gamma t)$, $\Gamma$ being the rate of the thermal transitions determined by the Boltzman factor $\Gamma \propto \exp(-U/k_B T)$, where $U$ is the size of the barrier.

The problem of magnetic relaxation in the realistic situation in which there is some distribution of barrier heights is actually one of great interest. In fact, it has been noticed long ago[1] that a broad distribution of energy barriers may provide a time relaxation law of the type

$$M(t)=M(t_0)-S(H,T)\ln(t/t_0)\ ,$$

valid for times greater than $t_0$, where $S(H,T)$ is the so-called magnetic viscosity that will depend on both the magnetic field $H$ and the temperature $T$.[2] This logarithmic law in time has been observed in a variety of systems.[3–5] For the case of superconductors, this type of magnetic relaxation can also be explained within the Anderson-Kim model.[6,7] Moreover, very recently, Lottis, White, and Dan Dahlberg[8] have published a model consisting of a spin system with the spins interacting via the dipole-dipole interaction; the dynamics of such system is also described by a quasilogarithmic or stretched-exponential time dependence. The common fact of these different models and the key to understanding the time logarithmic relaxation law is that as the observation time is running, the system arrives to greater and greater barriers which are more difficult to overcome.

The quantity of interest here is the magnetic viscosity whose dependence on temperature $T$ and magnetic field $H$ will characterize the relaxation behavior of the system. It deserves to be noted that the magnetic viscosity dependence on the applied field $H$ is due to the fact that the field modifies the energy-barrier height. Therefore it is convenient to express the viscosity as $S\equiv f(H)g(T)$, where $f(H)$ depends on the value and direction of the external magnetic field and $g(T)$ is a function of temperature. In this paper we will concentrate on the dependence of the magnetic viscosity on temperature, $S(T)$. Despite the interest of the problem, only few computer-simulation studies have been performed on this issue.[9,10] Those performed until now are for systems of identical interacting particles such that the barriers are equal for all the particles and the distribution of energy barriers is entirely due to the interactions. Then, in the models used, the shape of the distribution of energy barriers is not a variable that can be controlled easily. We will present in this paper a computer-simulation study of a model in which the shape of the distribution of barriers between the states is introduced explicitly, so that it can be used to get some insight into the effect of the shape of the distribution of barriers on the relaxation phenomena. We will limit ourselves here to systems of noninteracting particles (i.e., the ideal gas version of the model), which seem to be a good approximation in many experimental situations.[2–5]

The paper is arranged as follows. In Sec. II we give a detailed description of the theoretical model which is the basis of our calculations. Section III is devoted to present the results of the computer simulations. In Sec. IV a comparison is made between our computer-simulation results and some recent experiments on single-domain particles. Finally, in Sec. V we discuss and summarize the main results of this work.

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## II. THEORETICAL MODEL

To study the relaxation behavior of systems of the type described above by means of computer simulations, we have devised a theoretical model whose basic features are as follows. Our model is a system of $N$ noninteracting particles. The relevant property of a particle is its volume $V_i$, $i \in \{1,2,\ldots,N\}$. Associated to a particle of volume $V_i$, there are two additional interesting properties: (i) a magnetic moment $\mathbf{M}_i$ with $M_i = \mu V_i$, $\mu$ being a constant, and such that $\sigma_i \equiv (\mathbf{M}_i \hat{\mathbf{e}})/M_i \in \{+1,-1\}$, $\hat{\mathbf{e}}$ being a convenient unitary vector, so that the particle can be in one of two states $\{+M_i, -M_i\}$, and (ii) a barrier of energy between these two states, $U_i = \nu V_i$, with $\nu$ a constant. A state of the system is given by the enumeration of the individual states of its particles, $\tilde{\sigma} = \{\sigma_1, \sigma_2, \ldots, \sigma_N\}$. Then the macroscopic quantity that characterizes the state of the system will be the total magnetization

$$M = \sum_{i=i}^{N} \sigma_i M_i \ .$$

The particles being independent, the system will evolve as a succession of thermally induced single-particle events of the type $\sigma_i \rightarrow -\sigma_i$. The probability of one of those events to happen is just the probability of the particle to jump its own barrier, which is given by

$$P(U_i) \propto \exp\left[-\frac{U_i}{k_B T}\right] ,$$

$k_B$ being the Boltzmann constant and $T$ the temperature.

A system of $N$ particles will be characterized by some distribution function of volumes, $f(V)$, such that the number of particles in the system with volume $V$ between the $V+dV$ is $dN = Nf(V)dV$. A variety of reasonable choices are available for the function $f(V)$. We have considered here three that are very different from each other and whose shapes are representative of reasonable physical situations, so that our study will be representative of situations commonly found in experiment. On these bases we have studied the distribution functions that follow, whose parameters are a normalization constant $A$ and a characteristic volume $V_0$: (i) the uniform distribution

$$f(V) = \begin{cases} A & \text{if } V \le V_0 \ , \\ 0 & \text{if } V > V_0 \ , \end{cases}$$

which is representative of the situation where the distribution of particles is flat so that all volumes up to a maximum are practically equal represented in the system, (ii) the exponential distribution

$$f(V) = A \exp\left[-\frac{V}{V_0}\right] ,$$

which represents the situation where the number of particles with a given volume decreases monotonously with the value of the volume, and (iii) the Maxwellian distribution

$$f(V) = A \left[\frac{V}{V_0}\right]^2 \exp\left[-\frac{V}{V_0}\right]^2 ,$$

which corresponds to the case in which there is a finite non-null volume preferred by the particles.

To make our study as general as possible, the calculations have been performed in a system of reduced unities so that $V_0$ is the unit of volume, $\nu V_0$ is the unit of energy, and $\nu V_0/k_B$ is the unit of temperature. With regard to the magnetization, the constant $\mu$ in the relation $M_i = \mu V_i$ has been taken to be equal to 1.

Our computer simulations have been performed under the following dynamics. We first set a table in which volumes, distributed along one of the above functions $f(V)$, are assigned to $N \sim 10^4$ particles. We will call this our computer sample. The initial state of the system is chosen with all the particles in the same state $\sigma_i = +1$ so that $M_0 = \mu \Sigma V_i$ is the initial value for the magnetization. Then a value of the reduced temperature $T^*$ is selected and the state of the computer sample is evolved in a computer-simulated time $t^*$ to get the law for the relaxation of the magnetization $M(t^*) = \mu \Sigma \sigma_i(t^*) M_i$ at this temperature. To do this the following process is repeatedly iterated: A particle $i$ is randomly chosen, and its state is changed if a random number between 0 and 1 is smaller than $P = \exp(-U_i/T^*)$. We define a time step as a bunch of $N$ of this single-particle process. Then our unit of time, $\tau^*$, is the length of one of those time steps; the computer-simulated time is given by $t^* = n\tau^*$, $n$ being an integer. The evolution of the system is followed until a computer-simulated time $t_N^* \sim N\tau^*$ has elapsed. This algorithm is in fact a particular case of one of the algorithms to simulate time-dependent processes reviewed by Binder[11] (algorithm 3). Performing runs such as these for any one of the above distributions of volumes and a wide range of temperatures for each $f(V)$, we obtain a discretized version of the time evolution of the magnetization $M^*(t^*) = M(t^*)/\mu$ and from it, by means of a least-squares fit performed in the time interval where $M^*(t^*)$ versus $\ln(t^*)$ is linear, we obtain the magnetic viscosity $S^*(T^*) = dM^*/d[\ln(t^*)]$.

It deserves to be noted that the dynamical process implied in this computational algorithm includes two important features of the real systems: (i) thermal fluctuations given by the temperature-dependent stochastic nature of the process and (ii) the possibility of the particles to jump back and forth between the two single-particle states available because a particle suffers one process each time unit in the average. In consequence, jumps back into the original configuration of the system are allowed by the dynamics. However, from the statistical point of view they are very improbable because we begin in a state of the system with all spins aligned, which is far from equilibrium, so that it evolves from it toward a state of more disorder.

## III. COMPUTER-SIMULATION RESULTS

The simulations reported here have been performed for a system of $N = 3.6 \times 10^4$ particles, evolved up to $t_N^* = 3.6 \times 10^4$ time steps for all temperatures. Then a

particle suffers an average of $3.6\times10^4$ attempts of flipping from its actual state in a run, being the actual number of flips dependent on temperature. The temperature range studied in the above reduced units was chosen as follows. The highest temperature considered was such that the magnetization completely relaxes to zero in the fixed value of $t_N^*$ considered. The lowest temperature was such that the magnetization relaxes to a magnitude of about 1% of its initial value in the same time. In Fig. 1 we show a set of time relaxations of the magnetization for each of the above distributions and for ten equally spaced temperatures chosen in a wide interval of reduced temperatures.

When using a logarithmic scale in time, we found that a time logarithmic decay is a good approximation in a time interval that roughly begins between $t^*\approx2.5\times10^2$ steps and $t^*\approx10^3$ steps, and ends between $t^*\approx10^3$ steps and $t^*\approx3.6\times10^4$ steps, depending on temperature and distribution. The results for the three distributions considered appear in Fig. 2. The qualitative resemblance of the results in this figure and some previously published experimental results for this phenomenon in different systems deserves to be noted.[2–5]

The magnetic viscosities obtained by means of a fit to a straight line of these relaxations performed in the ap-

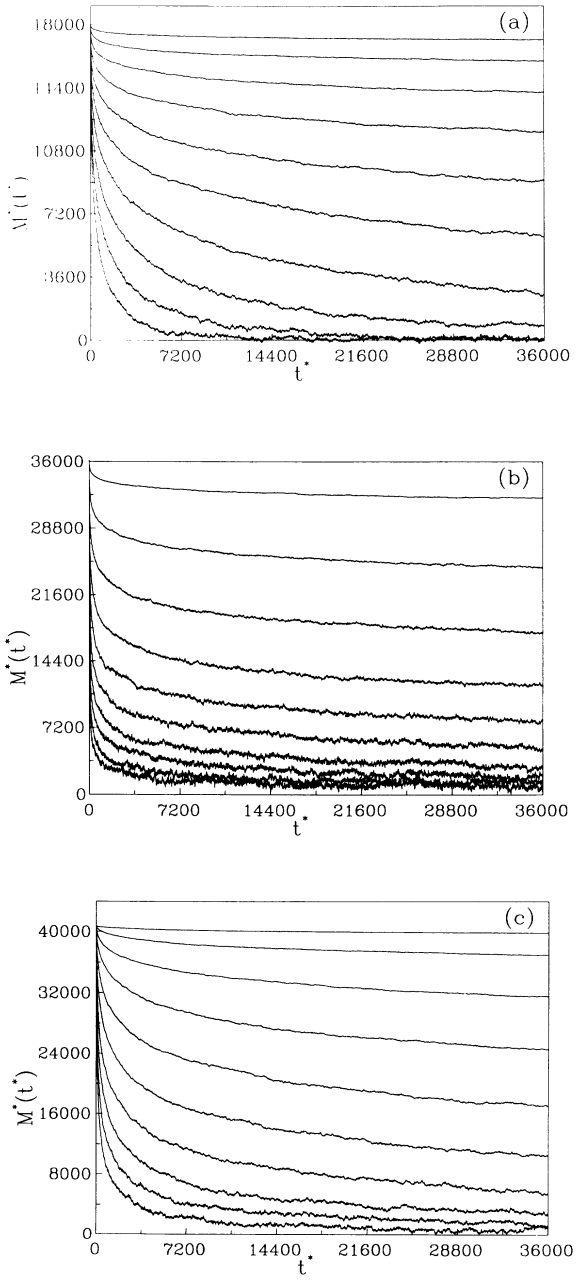


FIG. 1. Examples of the time relaxation of magnetization for ten different temperatures of the reduced temperature $T^*$: (a) Uniform distribution with $T^*\in[0.02,0.11]$ at intervals of $\Delta T^*=0.01$, (b) exponential distribution with $T^*\in[0.05,0.50]$ at intervals of $\Delta T^*=0.05$, and (c) Maxwellian distribution with $T^*\in[0.04,0.22]$ at intervals of $\Delta T^*=0.02$.

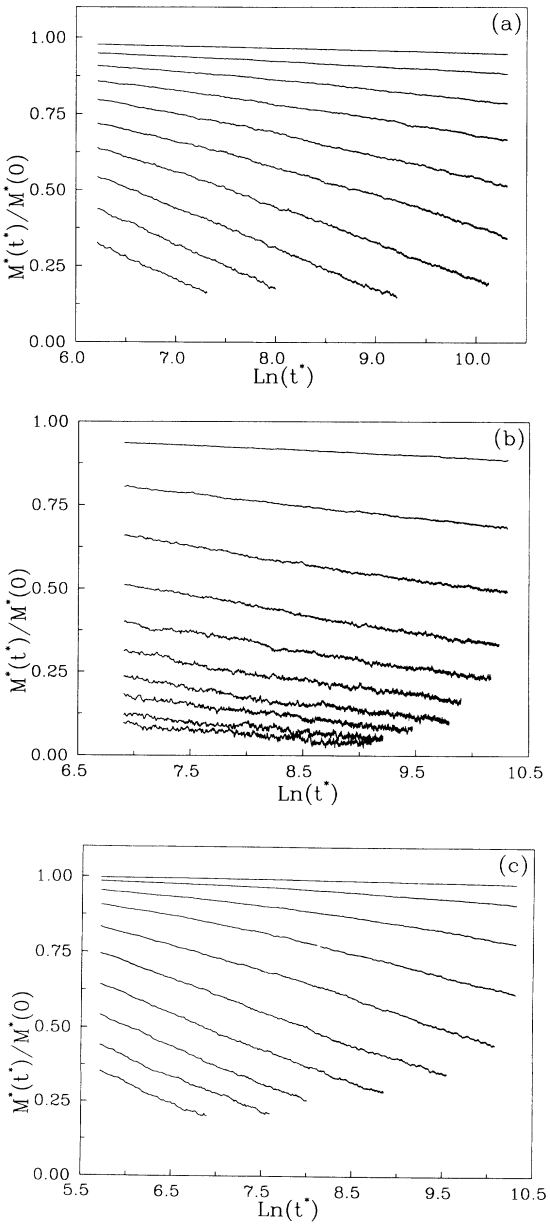


FIG. 2. Magnetization vs logarithmic time for the relaxations shown in Fig. 1.

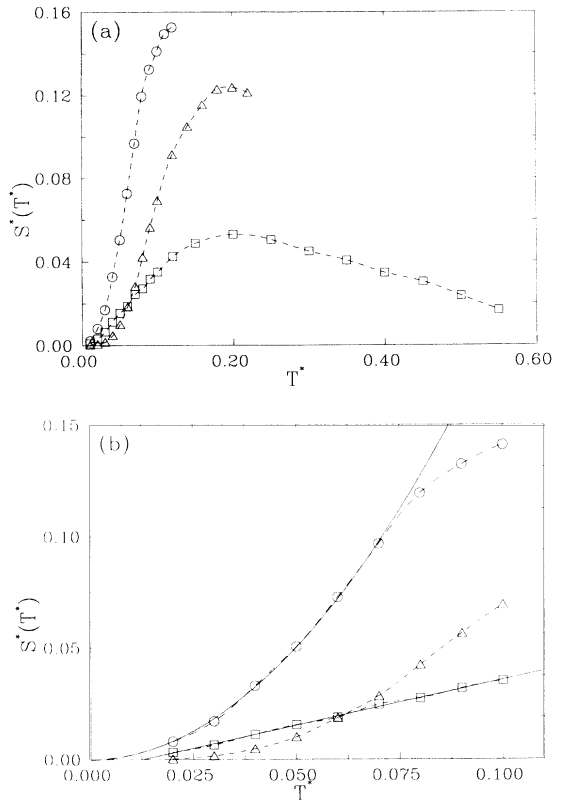


FIG. 3. (a) Magnetic viscosity as a function of temperature for the uniform distribution (circles), the exponential distribution (squares), and the Maxwell distribution (triangles). (b) View of the low-temperature region of (a); solid lines have been added to show fits to a straight line for the exponential distribution and to a quadratic law for the uniform distribution.

propriate interval are displayed in Fig. 3(a). The functions obtained increase with temperature toward what to seems to be a maximum or a plateau. For the case of the exponential distribution, a pronounced decay of $S^*$ with $T^*$ is clearly observable after the maximum. For temperatures above the highest considered here for every distribution, the time logarithmic relaxation law is lost in the sense that the region where linear approximation is feasible becomes narrower and noisy with increasing temperature until it becomes practically inaccessible to the computer simulation. In Fig. 3(b) we show in more detail the low-temperature behavior of $S^*(T^*)$. From this figure it is clear that the extrapolation to zero temperature intersects the $S^*$ axis at $S(0) \leq 0$ for all distributions. Moreover, the shape of $S^*(T^*)$ is also affected by the shape of the distribution. This seems to be linear for the exponential distribution, quadratic for the uniform distribution, and something not adjustable by a simple function for the Maxwell distribution.

## IV. COMPARISON WITH EXPERIMENT

An experimental situation where this model is easily applicable is the case of single-domain particles with uniaxial anisotropy recently studied by Tejada *et al.*[12] In this case the metastability is due to the existence of two states with minimal energy which correspond to the magnetization vector pointing along the easy-axis distribution. The switching of the magnetization from one stable direction to another involves overcoming an energy barrier usually via a process of thermal activation. The height of the barriers when no external field is applied is proportional to the volume $V$ of the particles. It is determined by the energy of magnetocrystalline anisotropy combined with the shape anisotropy. This is given by

$$U_0=(\kappa+2\pi M_s^2)V=K_{\text{eff}}V \ ,$$

where $\kappa$ is the first-order anisotropy constant, which is given by the anisotropy field $H_K$ and the magnetic moment density (or magnetic saturation) $M_S$ of the switching unit through $\kappa=\frac{1}{2} H_K M_S$. The particles studied in Ref. 12 where highly anisotropic, with characteristic anisotropy fields of $H_K \sim 10^4$ Oe. As the typical orders of magnitude for saturation magnetization density and size of those particles were $M_s \sim 5\times 10^2$ emu/cm$^3$ and $V \sim 5\times 10^{-19}$ cm$^3$, the typical size of those barriers is $U_0 \sim 10^{-12}$ erg.

A sample of single-domain particles being a many-particle system, one must consider the effect of the dipolar interactions between the particles to properly describe the system. The effect of these interactions is shown by the presence of a magnetic field $H_D$ on a particle, created by the remainder of the system. This field varies with the magnetization of the sample. Moreover, we must consider the possibility of an external field $H$, applied to the sample. These two fields are seen by a particle combined as a field acting on it, $H=H_D+H_e$, whose effect is to modify the height of its barrier in a quantity given by the expression for the activation energy proposed by Néel,[13]

$$U=K_{\text{eff}}\left[1-\frac{H}{H_K}\right]^2 V \ .$$

Moreover, as $H$ varies with the magnetization of the sample through $H_D$, this introduces a change of the effective height of the barriers during the process of relaxation of magnetization.

In the materials studied in Ref. 12, the particles in the sample were diluted, and in practice only 1% of the total relax during a typical experiment. The particles that do not relax can be seen as a set of fixed magnetic moments randomly directed. They create an average field in the sample that does not vary with time. So one can properly define the system that we study as the set of particles that relax and treat the remainder as part of the environment. Then, in the above description of the field external to a particle (that relaxes) as a field composed of the field external to the sample and the dipolar fields of the surrounding particles, one must consider this last decomposed into two contributions $H_D=H_{DR}+H_{DN}$, where $H_{DR}$ stands for the dipolar field due to the particles that relax and $H_{DN}$ for the particles that do not relax. This last can properly be seen as a constant field, external to the system of interest. The dependence on magnetization of $H_D$ is entirely due to $H_{DR}$. How important this effect

is will be determined by the comparison of the above size of the barriers $U_0$ and the magnitude of the dipolar interaction, which is given by $(M_s V)^2/R^3$, $R$ being the average distance between the particles.

Let us first note that in the relaxation experiments in Ref. 12 an external field of $H_e = 10^2$ Oe was applied. This is to be added to the constant average field created by the particles that do not relax, $H_{DN}$, which is estimated to be smaller than $10^2$ Oe. With regard to $H_{DR}$, one must note that the energy of the dipole interaction between two particles that are in close contact with each other is of the order of $10^{-13}$ erg. However, as only 1% of the particles in the sample relax, and this is very diluted, the average distance between them will be greater than $10^{2/3}$ of the closest distance so that the corresponding energy will be smaller than $10^{-15}$ erg, which is $10^{-3}$ of $U_0$. So we can neglect the effect of the interactions between the relaxing particles in the relaxation process. As the variation with magnetization is entirely due to the field created by these particles, this effect must be negligible. This implies that, although $H_{DR}$ varies with $M$, its bounds are such that the magnitude of $H_{DR}(M)$ is at most of the same order of magnitude as $H_{DN}$. As the total external field on a particle is the one that results from the combination of these three fields, it happens that (even considering the variation with $M$) its order of magnitude is about $10^{-2}$ of the anisotropy field. Then the factor $(1-H/H_K)^2$ in the Néel expression is practically equal to 1. So, despite the fact that this equation holds for the dilute systems, it can be effectively written in the present highly anisotropic case as $U = K_{\text{eff}} V + O(H/H_K)$, where $O(H/H_K)$ stands for higher-order terms in $H/H_K$, which in this case are about 1% of $K_{\text{eff}} V$, so that they can be neglected, and in practice it is $U = K_{\text{eff}} V$ in very good approximation.

Similarly, $H/H_K$ being very small, the activation enthalpy for jumps back to the original single-particle configuration

$$U = K_{\text{eff}} \left[ 1 + \frac{H}{H_K} \right]^2 V$$

can be approximated by $U = K_{\text{eff}} V$, neglecting terms that are two orders of magnitude smaller. This means that in the present highly anisotropic case the barriers are symmetric in the first approximation, so that back and forth processes must overcome practically the same barrier height. Then the relaxation processes studied here are dominated by the shape of the distribution of energy barriers, so that this shape is the convenient control parameter to study them. Moreover, neglecting effects that in this particular case are very small, the quantity $\nu$ in our computational model can be properly identified as $\nu = K_{\text{eff}} = \kappa + 2\pi M_S^2$, and it stands for both back and forth processes.

In this experimental situation, the distribution functions selected above can be justified having in mind that the actual distributions of energies is determined by the fluctuations in the anisotropy constant $\kappa$ and volume $V$, the distribution of easy axes with respect to the orientation of the applied field plus the distribution of $\kappa$, $V$, and $\widehat{MH}$ angle values. Then a flat distribution of energy barriers with a maximum barrier height may be a good representation for the energy barriers corresponding to a magnetic system with high metastability. It may be a system of interacting single-domain particles or, alternatively, a system of identical particles with a broad distribution of $\kappa$, $V$, and $\widehat{MH}$ angle values. An exponential distribution of energy barriers may correspond to a system of highly defected single-domain particles; the smallest barriers may be associated with the coercitivity barriers which clearly depend on the nature of the pinning centers. A Maxwell distribution of barrier heights may be invoked to represent the barrier height distribution of a system of single-domain particles with very high anisotropy constants $\kappa$. In this case the distribution function could only be associated with the dispersion of the $\kappa V$ values, if we admit that the barrier heights go to zero.

Tejada *et al.*[12] reported experimental results on the magnetic viscosity as a function of temperature for several ferrofluid systems constituted by single-domain particles. In particular, their results for $Fe_3O_4$, and FeC, and $CoFe_2O_4$ can be used for meaningful comparison with our computer-simulation results. Given the simplicity of the particular selections of $f(V)$, we are just looking for an overall qualitative agreement between them and the numerical values that we compute.

The experimental results for $Fe_3O_4$ show a broad maximum at high temperature just as our simulations for the exponential distribution. A reasonable good scaling between experiment and computer-simulation results is shown in Fig. 4(a). This has been achieved by means of the following scale transformation of computer-simulation results: $T = 33T^*$ and $S = 115S^*$. In a similar manner, the experimental results for FeC can be reasonably scaled with the computer-simulation results for the uniform distribution when the transformation $T = 102T^*$ and $S = 127S^*$ is used as shown in Fig. 4(b). For the case of $CoFe_2O_4$, the computer-simulation results that gave a best fit were found to be those for the Maxwellian distribution as shown in Fig. 4(c), where the transformation used was $T = 245T^*$ and $S = 831S^*$.

From the expressions and the comments on the reduced unit system in Sec. II and the transformations used to scale the functions, one obtains the following estimations for the characteristic barrier heights: $U_0(Fe_3O_4) = 33.0Kk_B$, $U_0(FeC) = 102Kk_B$, and $U_0(CoFe_2O_4) = 245Kk_B$. From Tejada *et al.*,[12] one can obtain only estimations of the average size of the barriers. These are $U_0(Fe_3O_4) = 15Kk_B$, $U_0(FeC) = 30Kk_B$, and $U_0(CoFe_2O_4) = 75Kk_B$. As expected, the results from computer experiments agree with those from real experiments in order of magnitude.

It deserves to be noted that the distribution of volumes measured in the above samples was found to be of the log-normal type. As in the simulation in this paper the distribution of barriers and the distribution of volumes are identical, it follows that in real systems the distribution of barriers is not necessarily equal to that of volumes as discussed in this section.

## V. COMMENTS AND CONCLUSIONS

We have investigated the magnetic relaxation behavior of highly anisotropic systems by means of the simulation of a computational model of systems of noninteracting particles with two available states separated by a barrier of energy. Thermal fluctuations and the possibility of jumps back to the original configuration have been included in the computer simulation by means of a convenient stochastic dynamics. Our investigation has been concerned with the effect of having a distribution of energy barriers instead of a single-barrier height. We have neglected other possible effects on relaxation such as external or internal fields, which is a good approximation in some real experimental situations. Moreover, this model is interesting from a theoretical point of view because the effect of the distribution of barriers appears isolated from other contributions to the relaxation process, which is relevant from an analytical point of view.

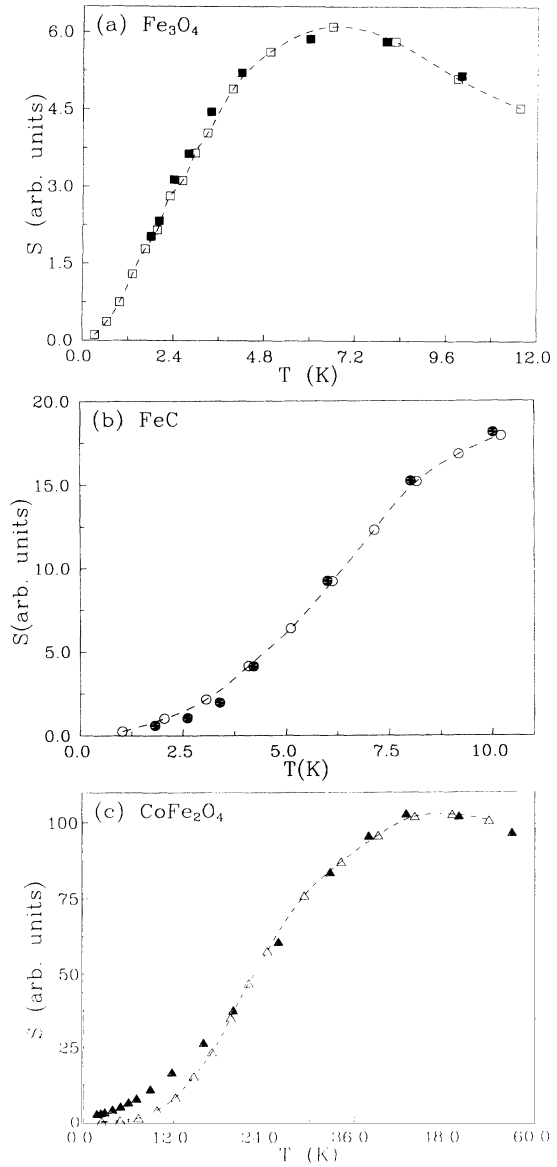


FIG. 4. (a) Computer-simulation results for the exponential distribution (open squares) scaled to experimental measurements for $Fe_3O_4$ (solid squares), (b) uniform distribution (open circles) scaled to the experimental measurements for FeC (solid circles), and (c) Maxwellian distribution (open triangles) scaled to the experimental measurements for $CoFe_2O_4$ (solid triangles).

The variety of shapes for the distribution law of barriers considered allows us to conclude that, except for very singular distributions, we will be able to obtain a time interval for which the time logarithmic law decay is a good approximation. So it follows that slow relaxation can also be obtained, neglecting interactions and considering only a distribution of barriers plus thermal agitation. Moreover, magnetic viscosity appears as a convenient quantity to characterize this phenomenon.

Another major result of this paper is the study of the magnetic viscosity. The dependence of the magnetic viscosity with temperature has been found to be sensitive to the shape of the distribution of barriers both at high and low temperatures. Particularly interesting is the case of the exponential distribution of barriers where we have been able to observe very clearly a $S(T)$ function that increases toward a maximum at some value of $T$ and then falls down toward zero. This type of behavior has been reported in various experiments in small particles,[5,12] superconductors,[14–16] and spin glasses.[17] We will note that we have observed different low-temperature behaviors, but none of them was convergent to a positive value of the magnetic viscosity as the temperature goes to zero. This last observation is relevant in relation to the interpretation of plateaus in the low-$T$ region of $S(T)$ as evidence of quantum tunneling of magnetization.[12,18,19]

When compared with experiment, our $S(T)$ functions look similar to the observed in experiments. Moreover, we obtained reasonable estimations of the size of the barriers from a scaling of our functions with the experimental ones, despite the fact that no attempt of fitting multiple parameters has been done. This is reliable evidence that the computational model proposed here is a good approach to understand the phenomena of slow relaxation that appear in magnetism and superconductivity. To get better agreement in comparisons of this nature, two improvements are needed. First, in the experiments, the distribution functions of the energies (or volumes of the particles) must be determined more precisely and the relation between volume and energy barriers must be well estimated so that average values are properly defined and computed. Second, extensive computer simulations are needed to get a more precise idea of the relation between the shape of the distribution of barriers and the function $S(T)$.

Finally, we will note that as our calculations have been done for an ideal system, we have obtained evidence that for, the experimental systems considered here, weak interactions between the particles is a fair approach. In fact, the effect of the interactions between the particles on the relaxation of the system, in the type of systems considered here and in other related systems, both in magnetism and superconductivity, is an interesting issue that will be addressed in future investigations by means of natural generalizations of the model presented here.

## ACKNOWLEDGMENT

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