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First-Passage-Time Approach to Overbarrier Relaxation of Magnetization

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We consider the irreversible dynamics of the magnetization vector **M** in a single-domain particle. The dynamics is given by a stochastic phenomenological equation due to Gilbert. It contains a damping field proportional to $\dot{\mathbf{M}}$ and a corresponding white noise field component. The probability distribution function satisfies a Fokker–Planck equation derived by Brown. We give the overbarrier decay rate κ out of a metastable minimum. First we rederive the well-known expression for κ for an axially symmetric model. We argue that this result is unphysical. For systems of general point symmetry of the magnetic anisotropy energy we give κ in both the low-damping and intermediate- to high-damping limits.

KEY WORDS: First passage times; magnetization relaxation; superparamagnets.

1. INTRODUCTION

The problem of thermally activated escape of a particle trapped in a metastable state of a potential field has been exhaustively studied ever since the pioneering work of Kramers[1] and has found applications in a wide range of physical systems described by formally equivalent Langevin equations. A number of methods for obtaining the decay rate has been developed, the most notable recently being the uniform expansion of the first passage time of Matkowsky *et al.*,[2] which results in an iterated expression valid for all dissipation regimes.

By contrast, the thermal fluctuations of the magnetic moment of a single-domain ferromagnetic particle and its decay toward thermal equilibrium have received scant attention. The problem was introduced by Néel[3] and further developed by Brown.[4] The latter author considered the

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stochastic dynamics of the magnetization vector of a small ferromagnetic single-domain (superparamagnetic) particle, subject to a damping field and interacting with a random magnetic field. The magnetization is taken to be uniform throughout the particle at all times. Brown's starting point was a Langevin equation in Gilbert's form where the effective magnetic field has a damping and a stochastic external field component:

$$\frac{d\mathbf{M}}{dt} = \gamma_0 \mathbf{M} \mathbf{x} \left[-\frac{\partial V}{\partial \mathbf{M}} - \eta \frac{d\mathbf{M}}{dt} + \mathbf{h}(t) \right] \tag{1.1}$$

Here $\mathbf{M}(t)$ is the total magnetic moment of the ferromagnetic particle, V is its Gibbs free energy, γ_0 is the ratio of magnetic moment to angular momentum, η is a dissipation constant, and $\mathbf{h}(t)$ is a random isotropic magnetic field such that at temperature T

$$\langle h_i(t)\, h_j(t+t') \rangle = 2\eta T \delta_{ij} \delta(t') \tag{1.2}$$

It is easy to see from Eq. (1.1) that $\mathbf{M}^2(t) = M_s^2$ is conserved at all times, with M_s the saturation magnetization. From Eqs. (1.1) and (1.2), Brown derived a Fokker–Planck equation for the probability distribution of the magnetic moment vector $\mathbf{M}(t)$ and gave the decay rate when the magnetic anisotropy energy is axially symmetric. The corresponding Fokker–Planck equation is in this case $1+1$ dimensional; it was studied subsequently by Aharoni[5] and Agarwal *et al.*[6] In Section 2 we give another derivation of the decay rate in strictly axially symmetric models. This derivation is then the basis for our argument that this model fails to reflect the properties of even the simplest realistic model as far as the prefactor is concerned. It is the purpose of this paper to give the correct prefactor of the thermal decay rate in a system of arbitrary magnetic symmetry and to show where it differs from the predictions based on axial symmetry and why. We use the mean first passage time formalism of Matkowsky *et al.*[2]

Our derivation is based on Brown's theory, which, however, makes two assumptions not in strict accordance with reality. Both of these assumptions are accepted in this work; nonetheless, they merit some discussion. First, it is clear that magnetization density cannot be uniform throughout the volume of the particle, since this assumption neglects all effects on the edge of the domain, i.e., on the particle surface. A damped phenomenological equation of motion allowing for spatial dispersion of magnetization and of the relaxation terms was derived by Bar'yakhtar[7]; the boundary conditions are discussed by Aharoni.[8] The problem thus formulated presents formidable mathematical difficulties. Also, the surface properties appear to depend drastically, and in a way that is not well understood, on the manner of sample preparation.[9] Thus, a realistic

model, for a given particle volume and shape, is essentially unattainable. For this reason we neglect surface effects and assume uniform rotation of magnetization within the particle. Brown's approach was also criticized by Walton,[10] who pointed out that the interaction of a magnetic moment with a heat bath is constrained by quantum selection rules, so that only a fraction of the bath bosons will couple to the magnetization at any given time. Under such circumstances the response and collision times may be of similar order of magnitude and the white noise model fails. The specific form of the colored noise correlation function and of the corresponding memory kernel depend of course on the specific damping model. A theoretical study of the damping mechanism in superparamagnets was carried out by Garg and Kim,[11] who considered magnetoelastic coupling and energy dissipation by sound waves in the elastic environment. These authors arrive at the spectral density $J(\omega) \sim \omega^3$ and conclude that the damping is weak. Competing models shall indubitably be published in the future. In the present work we restrict ourselves to the memoryless case in both the weak and the intermediate-to-high (IHD) limits. Our results on decay in systems with memory are reported in a separate work,[12] where we derive the corresponding phenomenological equations of motion and also introduce a generalized Euclidean action on phase space for path integral applications.

After these preliminaries, let us specify our model. Let there be a single-domain ferromagnetic particle whose Gibbs free energy is a function of the magnetic moment **M** and an external magnetic field **B** only. We neglect all changes in the internal structure occurring during the irreversible decay process. Under these conditions the Gibbs free energy is effectively the Hamiltonian H. Orientation in space is given by spherical coordinates. We introduce the notation $p = \cos\theta$ (so that $M_3 = M_s p$) and define for magnetization the conjugate pair $P = (M_s/\gamma_0)\, p$ and φ. To the saturation magnetization M_s there corresponds an angular momentum $P_0 = M_s/\gamma_0$. In the scaled variables p and φ, Gilbert's equation, after some manipulation, becomes

$$\dot{p} = -h'(1-p^2)\, H_p - g' H_\varphi + N_{pi}(p, \varphi)\, h_i \tag{1.3a}$$

$$\dot{\varphi} = g' H_p - h'(1-p^2)^{-1}\, H_\varphi + N_{\varphi i}(p, \varphi)\, h_i \tag{1.3b}$$

Summation over i is assumed. The noise coefficients N_{pi} and $N_{\varphi i}$ are rather complicated; we refer the reader to ref. 4, Eqs. (3.3)–(3.5), for we shall make no use of their explicit form. Further, h' is the effective dissipation constant and g' is the inverse of the effective angular momentum corresponding to the saturation magnetization M. Note (below) that it

is dissipation dependent. Introducing the dimensionless parameters $\alpha = M_s \gamma_0 \eta$ and $\Delta = 1 + \alpha^2$, we write

$$g' = 1/(P_0 \Delta), \qquad h' = g'\alpha \tag{1.4}$$

Subscripts of the Hamiltonian $H(M_i) = H(\varphi, p)$ denote partial derivatives. The Fokker–Planck equation for the probability distribution $W(\varphi, p)$ of the stochastic process (1.1), (1.2) is, according to Brown,[4]

$$\frac{\partial W}{\partial t} = L_T W \tag{1.5a}$$

where the operator L_T is defined on the scaled phase space as

$$L_T = \frac{\partial}{\partial p}\left[-\dot{p}_D(\varphi, p) + h' T(1-p^2)\frac{\partial}{\partial p}\right] + \frac{\partial}{\partial \varphi}\left[-\dot{\varphi}_D(\varphi, p) + h' T(1-p^2)^{-1}\frac{\partial}{\partial \varphi}\right] \tag{1.5b}$$

Here $\dot{p}_D$ and $\dot{\varphi}_D$ are given by the deterministic part of Eqs. (1.3), i.e., with $\mathbf{N}_p$ and $\mathbf{N}_\varphi$ both set identically to zero. It is easy to see that the equilibrium solution of Eq. (1.5) is indeed $W_0 = \exp(-H/T)$. The probability of finding the system in an element $dP\, d\varphi$ of the phase space is $\text{Prob} = W(\varphi, P)\, dP\, d\varphi$.

2. AXIALLY SYMMETRIC MODEL

Let us first turn our attention to the axially symmetric case, where $H(M_3) = H(p)$. A typical Hamiltonian of this kind is

$$H = K'(M_s^2 - M_3^2) - BM_3 = -BM_s \cos\theta + K \sin^2\theta \tag{2.1}$$

where B is an external magnetic field in the 3-direction and $K' = K/M_s^2$ is an anisotropy constant. This Hamiltonian was used by Brown in his original work.[4] The Fokker–Planck equation is $1+1$ dimensional in spherical coordinates and the methods developed for the Smoluchowski equation may be used to advantage. We follow here the asymptotic expansion method of Matkowsky and Schuss[13] to obtain the first nonzero eigenvalue. Let us write the solution of Eq. (1.5) in the form $W(p, t) = \exp(-H/T)\, u(p, t)$, so that

$$\frac{1}{h'}\frac{\partial u}{\partial t} = T\frac{\partial}{\partial p}\left[(1-p^2)\frac{\partial u}{\partial p}\right] - (1-p^2)\, H_p \frac{\partial u}{\partial p} \tag{2.2}$$

and the corresponding eigenvalue problem becomes

$$-h'T\frac{d}{dp}\left[(1-p^2)\frac{dv_n}{dp}\right]+h'(1-p^2)\,H_p\frac{dv_n}{dp}=\lambda_n v_n \tag{2.3}$$

Restricting ourselves to the first nonzero eigenvalue, we see that $u(p, t)$ evolves in time as $v(p)\exp(-\lambda_1 t)$, $\lambda_1>0$, and that the decay rate is $\kappa=\lambda_1$. Let now the Hamiltonian have a single maximum at p_1, $-1<p_1<1$, such that $H\approx E_1+\frac{1}{2}H''(p_1)(p-p_1)^2$ in its vicinity. Let H have further two minima at the edges of the domain $p=\pm 1$. These minima are assumed not to be extrema of H, so that $H\approx E_0+H'(1)(p-1)$ to the right of p_1; obviously, $H'(-1)>0$ and $H'(1)<0$. The calculation is essentially the same as in ref. 13 and only the result will be presented. Let the initial probability distribution be peaked around the energy minimum at $p=1$. Then the decay rate is given by

$$\kappa=h'T(1-p_1^2)\left[\frac{|H''(p_1)|\,[H'(1)]^2}{2\pi T}\right]^{1/2}\exp\left[\frac{-(E_1-E_0)}{T}\right] \tag{2.4}$$

The quantity $h'\,|H'(1)|$ is the drift rate out of the minimum at $p=1$, as can be seen from the equations of motion, and the difference (E_1-E_0) is obviously the barrier height.

This formula was previously given in a slightly different form by Brown,[4] who in his derivation used the assumption that the friction force is strong enough to keep the populations in both wells thermalized at all times. The decay rate (2.4) goes to zero with decreasing dissipation strength, as expected for low dissipation η, since $h'\sim\eta$. Throughout our derivation we never imposed the condition of thermal equilibrium on the well subsystem and thus we conclude that the formula (2.4) holds in the entire range of the dissipation constant η. This circumstance is also quite obvious from the first passage time analysis, since in the axially symmetric case the separatrix coincides with the contour of critical energy $E_C=E_1$ in the phase space, as discussed in the following section. In this context the following observation should be made. The random field $\mathbf{h}(t)$ couples to both the momentum and position, as seen from Eq. (1.1). For this reason it is impossible to claim that one of the conjugate variables becomes rapidly thermalized by strong dissipation. Hence it is impossible to write down a true Smoluchowski equation for the overdamped limit. The $(1+1)$-dimensional equation (2.1) originates from symmetry, not thermodynamic, considerations.

The axially symmetric model exemplified by the Hamiltonian (2.1) has two pathological features which distinguish it from all real systems.[14]

First, there exist no saddle points on the energy surface. This is in contrast to any realistic anisotropy energy expression, in which the minima are always linked by flux paths of least energy leading through a finite number of saddle points. This applies also to realistic uniaxial systems, since the ever-present anisotropy in the basis plane breaks the axial symmetry. The second demerit of the strictly axially symmetric model has to do with the parametrization in spherical coordinates. In these coordinates the 3-direction which is chosen to coincide with the easy axis is singular. The direct consequence of this circumstance is the presence of the singular, cusplike minima which are nothing but cutoffs of the energy at $p = \pm 1$. No well frequency can be defined for such minima, since $H''(\pm 1)$ do not exist. The singular minimum in conjunction with the smooth maximum brings about the $T^{-1/2}$ temperature dependence of the prefactor.[13] Obviously, this dependence is an artifact of the parametrization; the physically equivalent Hamiltonian obtained from (2.1) by a rotation about the 1-axis by $\pi/2$ has in spherical coordinates smooth minima in the 2-direction, but of course no saddles. To sum up: the axially symmetric Hamiltonian lacks saddle points, which are otherwise a common feature of all real systems and the parametrization, in which the easy axis coincides with the singular direction of the spherical coordinates, brings about unphysical effects in the prefactor. For this reason predictions based on the axially symmetric model must be rejected as faulty.

3. SYSTEMS OF GENERAL SYMMETRY, IHD LIMIT

Consider now a system of definite point symmetry. Let the total energy be given as a polynomial in the direction cosines v_i. From this expression one arrives at the Hamiltonian $H(\varphi, p)$ via the obvious substitution

$$\mathbf{v} = \{(1 - p^2)^{1/2} \cos\theta, (1 - p^2)^{1/2} \sin\theta, p\}$$

We recall that the points $p = \pm 1$ are singular points of this parametrization. The Hamiltonian is defined on a phase space which is a closed manifold [the scaled space (φ, p) is the surface of a unit sphere] and thus a local energy minimum is surrounded by two or more saddle points, depending on the symmetry of the problem. The total probability flux out of the metastable minimum equals the sum of fluxes through all the saddle points. In an asymmetric case, e.g., when an external field is applied, some of these fluxes become exponentially small and may safely be neglected. The total flux out of the metastable minimum is in this case dominated by the energetically most favorable path.

For simplicity, now let the metastable minimum and one of the saddle points lie on the line $p=0$, their coordinates being $(\varphi_0, 0)$ and $(\varphi_1, 0)$, respectively. The minimum and the saddle point of interest may always be brought into this position by a suitable rotation of the axes.

When the equation of motion (1.3) is linearized in the vicinity of any point on the equator, the leading terms of the noise coefficients $\mathbf{N}_p$ and $\mathbf{N}_\varphi$ become constant, as can most easily be seen from the linearized form of the Fokker–Planck operator (1.5b) without reference to their explicit form. Thus, we are dealing with additive white noise exhaustively treated in the IHD limit by the formalism of Brinkman,[15] Landauer and Swanson,[16] Langer,[17] and Dygas *et al.*[18] The results of the latter work are readily applicable to the present case. Let us define the well ($i=0$) and saddle ($i=1$) frequencies by the Hessians

$$\omega_i^2 = P_0^{-2} \,|\mathrm{He}[H(\varphi_i, 0)]| = P_0^{-2} \,|H_{pp}^{(i)} H_{\varphi\varphi}^{(i)} - (H_{p\varphi}^{(i)})^2| \tag{3.1}$$

where $H_{pp}^{(i)} = H_{pp}(\varphi_i, 0)$, etc. The Hessian is positive at the minimum and negative at the saddle point. Let further ν_c be the positive eigenvalue of the noiseless dynamics (1.3) linearized about the saddle point. With $\varphi \to \varphi - \varphi_1$, Eqs. (1.3) become

$$\dot{p} = -h'(H_{pp}^{(1)} p + H_{p\varphi}^{(1)} \varphi) - g'(H_{p\varphi}^{(1)} p + H_{\varphi\varphi} \varphi) \tag{3.2a}$$

$$\dot{\varphi} = g'(H_{pp}^{(1)} p + H_{p\varphi}^{(1)} \varphi) - h'(H_{p\varphi}^{(1)} p + H_{\varphi\varphi} \varphi) \tag{3.2b}$$

so that

$$\nu_C = \tfrac{1}{2}[D - h'(H_{pp}^{(1)} + H_{\varphi\varphi}^{(1)})] \tag{3.3}$$

with

$$D = h'^2 (H_{pp}^{(1)} + H_{\varphi\varphi}^{(1)})^2 + 4\omega_1^2/A \tag{3.4}$$

The decay rate in the IHD limit is then given in terms of these quantities as[18]

$$\kappa = \frac{1}{2\pi} \frac{\omega_0}{\omega_1} \nu_C \exp\left[\frac{-(E-E)}{T}\right] \tag{3.5}$$

The transition-state theory result is recovered in the limit $\eta \to 0$, since $\nu_C(\eta=0) = \omega_1$. In the high-dissipation limit $\eta \to \infty$, $\nu_C \sim \eta^{-1}$, so that in this regime the decay rate is inversely proportional to the dissipation constant η.

For illustration, consider now a system of cubic symmetry. The total

energy in terms of the direction cosines is given to the sixth order in the direction cosines by[14]

$$E = K_1[\nu_1^2\nu_2^2 + \nu_2^2\nu_3^2 + \nu_3^2\nu_1^2] + K_2\nu_1^2\nu_2^2\nu_3^2$$

and we assume $K_1 > 0$, as for iron. Usually, K_2 is an order of magnitude smaller than K_1. There are three easy axes and six corresponding energy minima of $E_0 = 0$. Let us choose the one that lies at $(0, 0)$; then $H_{pp}^{(0)} = H_{\varphi\varphi}^{(0)} = 2K_1$. The minimum is surrounded by four equivalent saddle points with energy $E_1 = K_1/4$. One of them is located at $(0, \pi/4)$ and the requisite second derivatives are $H_{pp}^{(1)} = K_1 + K_2/2 > 0$ and $H_{\varphi\varphi}^{(1)} = -2K_1 < 0$. The mixed derivatives vanish along the equator. The decay rate through one saddle follows from (3.5); the total rate is obviously four times as much. The magnetization, originally directed along the $[1, 0, 0]$ axis, decays in the $[0, 1, 0]$ and $[0, 0, 1]$ directions toward thermal equilibrium.

The formulas (3.3)–(3.5) allow us to calculate the prefactor also in the presence of an external magnetic field. The resultant expressions are rather clumsy; we list them in a separate work[14] for magnetic fields parallel to an easy axis of systems of cubic symmetry (both Fe and Ni type) and to the easy axis of uniaxial crystals with tetragonal, triclinic, and hexagonal symmetry in the basis plane.

A small note is due at this point. A Hamiltonian, albeit phenomenological, containing odd powers of the momentum P is aesthetically rather unsettling. Apart from the case of triclinic symmetry, it is always possible,[14] by rotational symmetry, to write down a Hamiltonian that is an even function of the momentum. In this case the mixed derivatives $H_{p\varphi}$ vanish along the equator $p = 0$.

4. SYSTEMS OF GENERAL SYMMETRY, UNDERDAMPED LIMIT

Of some interest also is the low-dissipation limit, in particular in view of the estimate by Garg and Kim[11] that the dissipation strength in superparamagnets is low. The asymptotic expansion method of the first passage time[2] affords a convenient approach to this problem. As expected, the decay rate turns out to be proportional to the energy loss (to first order in the small dissipation constant η) per period of the underdamped motion at $E = E_1$. We prefer to give here a brief sketch of the derivation, since the generalization of published results to our case is not quite obvious.

The low-dissipation limit is distinguished by the fact that the contour of critical energy E_1 lies within the boundary layer near the separatrix Γ.

Thus, we introduce a domain Q such that $E<E_1$ in the interior of Q and $E=E_1$ on the boundary $\partial(Q)$. By assumption, $\partial(Q)$ lies so near to Γ that the passage time from $\partial(Q)$ to Γ is negligible (to leading order in T) in comparison to the mean time $\tau(\varphi, p)$ required to reach $\partial(Q)$ starting from a point $(\varphi, p)\in Q$. The mean first passage time $\tau(\varphi, p)$ is a solution of the equation

$$L_T^+ \tau(\varphi, p) = -1 \tag{4.1}$$

subject to the boundary condition $\tau=0$ on $\partial(Q)$. Here L_T^+ is the adjoint of the Fokker–Planck operator (1.5b).

Following ref. 2, we set

$$\tau(\varphi, p) = \tau(Q)\, u_T(\varphi, p) \tag{4.2}$$

$\tau(Q)$ is an exponentially large quantity independent of the initial point (φ, p), and $\max u_T(\varphi, p)=1$ in Q. Then, to leading order in T, we obtain from (4.1) that $du_T^{(0)}/dt=0$ along any trajectory within Q, so that, by our normalization, $u_T^{(0)}=1$ within Q. In order to satisfy the boundary condition, we introduce now a boundary layer by the stretching transformation $s=(E_1-E)/T$. Equation (4.1) then yields, to leading order in T,

$$u_T = 1-\exp(-s) \tag{4.3}$$

which satisfies the boundary condition $u_T=0$ on $\partial(Q)$ and the matching condition $u_T=1$ within Q, i.e., as $s\to\infty$. Using the fact that $L_T \exp(-H/T)=0$, we now employ Green's theorem to obtain the constant $\tau(Q)$:

$$\tau(Q) = \frac{\int_Q dp\, d\varphi\, e^{-H/T}}{h'T \oint_{\partial(Q)} e^{-H/T}[(1-p^2)(\partial u_T/\partial p)\, d\varphi - (1-p^2)^{-1}(\partial u_T/\partial\varphi)\, dp]} \tag{4.4}$$

The integral in the numerator is easily evaluated in the asymptotic limit and the denominator may be expressed in terms of the Hamiltonian using Eq. (4.3). Thus, we finally arrive at

$$\tau(Q) = (2\pi T/\omega_0 \Delta E)\exp[(E_1-E_0)/T] \tag{4.5}$$

This is the mean first passage time from all points in Q, except for a boundary layer of width $O(T)$, to $\partial(Q)$. We introduced here the energy loss ΔE per period of the underdamped motion at $E=E_1$ by

$$\Delta E = \alpha \oint_{\partial(Q)} [(1-p^2)\, H_p\, d\varphi - (1-p^2)^{-1} H_\varphi\, dp] \tag{4.6}$$

We recall that α is the dimensionless dissipation parameter. In the underdamped limit it is small by assumption and only the leading terms of (4.5)–(4.6) are retained. As argued above, in the underdamped limit the mean first passage time from the interior of Q to $\partial(Q)$ almost coincides with the mean exit time out of the well (i.e., with the mean first passage time to the separatrix Γ) and the decay rate to leading order in T is then given by $\kappa^{-1} = 2\tau(Q)$. We see that the formula (4.6) is formally equivalent to that for the low-dissipation first passage time in the Kramers problem, where the dissipative coupling is such that ΔE is proportional to the action.[1,2]

The transition region of dissipation in which neither the intermediate-to-high limit (3.5) nor the low-dissipation limit (4.5) holds is easily estimated. In this region the ratio of the low-dissipation decay rate to the transition-state-theory result is near to unity, $\kappa/\kappa_{TS} = \Delta E/T \approx 1$. The contour integral in (4.6) has a value $\bar{E}$, which is on the order of magnitude of the barrier height, so that $\Delta E = \alpha\bar{E}$ with $\bar{E}/T \gg 1$ by assumption. In the intermediate region a bridging expression may be found using the formalism of ref. 2 or ref. 20.

5. SUMMARY AND CONCLUSIONS

This paper is concerned with the thermal decay rate of magnetization in small, ferromagnetic, single-domain particles, i.e., in superparamagnets. Our starting point is the Fokker–Planck equation due to Brown[4] for the orientation probability distribution of the magnetization vector, which is assumed to be homogeneous throughout the volume of the particle.

In Section 2 we consider the axially symmetric model studied previously by various authors.[4–6] We write down the corresponding $(1+1)$-dimensional Fokker–Planck equation in spherical coordinates and find its first nonzero eigenvalue. The decay rate thus obtained is formally valid in the entire dissipation range. However, we argue that the prefactor is unphysical. The point is that the easy axis coincides with the singular direction of the spherical coordinates where the minima become cusplike. This is an artefact of the parametrization and it vanishes if the easy axis is placed in a regular direction where the minima become smooth. The cusplike minima lead to the $T^{-1/2}$ dependance of the prefactor (2.4) for any dissipation strength. The axially symmetric model also differs qualitatively from any real system in that it lacks saddle points.

In Section 3 we give the thermal decay rate of magnetization in systems of general point symmetry in the intermediate- to high-dissipation (IHD) limit. We discuss the flux geometry and illustrate our results on the example of Fe-type cubic symmetry. A detailed list of the requisite quan-

tities for various anisotropy energy symmetries encountered in practice is given in ref. 14. In this work we also allow for an external magnetic field to be present along an easy axis. The underdamped limit is considered in Section 4. We show that the decay rate is proportional to the energy loss per period of the underdamped almost periodic motion at the barrier energy. An estimate of the region where neither the IHD nor the underdamped limit holds is given.

The dissipation strength in superparamagnets is unknown. We quote the theoretical prediction of low dissipation strength by Garg and Kim,[11] but no experimental data seem to be available. In principle, it is possible to estimate the dissipation strength (η or α) experimentally, or at least to decide whether the system is underdamped or in the IHD region. Such experiments would involve either direct measurement of the decay rate ($\sim T^{-1}$ in the underdamped, and T independent in the IHD region) or of the mean excess energy as proposed by Naeh *et al.*[19] (T independent in the underdamped, and $\sim T$ in the IHD region). Another possibility is the measurement of the linewidth in the magnetic resonance of superparamagnets.[20] It is not clear how to measure the excess energy, and the evaluation of all measurements (even if carried out with sufficient accuracy) is hampered by the need for extremely uniform particle distribution in the sample. Thus, e.g., in decay rate measurements the observed quantity is the mean rate over the particle ensemble and unless the energy dependence on particle volume and the volume distribution are known *a priori* the data cannot be analyzed. In particular, even the exponential T dependence of the decay rate is affected by the averaging.

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