Interpolation formula between very low and intermediate-to-high damping Kramers escape rates for single-domain ferromagnetic particles

P. M. Déjardin,[1,*] D. S. F. Crothers,[1] W. T. Coffey,[2] and D. J. McCarthy[3]
[1]*Department of Applied Mathematics and Theoretical Physics, David Bates Building, The Queen's University of Belfast, Belfast BT7 1NN, Northern Ireland, United Kingdom*
[2]*Department of Electronics and Electrical Engineering, Trinity College, Dublin 2, Ireland*
[3]*School of Mathematics, Statistics and Computer Science, Dublin Institute of Technology, Kevin Street, Dublin 8, Ireland*
(Received 15 August 2000; published 18 January 2001)

It is shown that the Mel'nikov-Meshkov formalism for bridging the very low damping (VLD) and intermediate-to-high damping (IHD) Kramers escape rates as a function of the dissipation parameter for mechanical particles may be extended to the rotational Brownian motion of magnetic dipole moments of single-domain ferromagnetic particles in nonaxially symmetric potentials of the magnetocrystalline anisotropy so that both regimes of damping occur. The procedure is illustrated by considering the particular nonaxially symmetric problem of superparamagnetic particles possessing uniaxial anisotropy subject to an external uniform field applied at an angle to the easy axis of magnetization. Here the Mel'nikov-Meshkov treatment is found to be in good agreement with an exact calculation of the smallest eigenvalue of Brown's Fokker-Planck equation, provided the external field is large enough to ensure significant departure from axial symmetry, so that the VLD and IHD formulas for escape rates of magnetic dipoles for nonaxially symmetric potentials are valid.

DOI: 10.1103/PhysRevE.63.021102 PACS number(s): 05.40.−a, 75.50.Tt

I. INTRODUCTION

Renewed interest in the Kramers theory of escape rates [1] has been stimulated by the recent success of Wernsdorfer *et al.* [2] in isolating single-domain ferromagnetic particles and in measuring the time of reversal (for barriers significantly greater than the thermal energy, the Kramers escape rate) of the magnetization of these particles as a function of the damping parameter predicted by the Néel-Brown [3,4] theory of superparamagnetic relaxation.

In effect, the latter theory is an adaptation of the Kramers theory of chemical reaction rates to longitudinal relaxation of the magnetization of single-domain ferromagnetic particles. The validation of that theory by experiment [2,5] furthermore confirms the Kramers conception of a thermal relaxation process over a potential barrier. The Kramers theory of chemical reaction rates was initially adapted to longitudinal relaxation of spins (i.e., reversal of the direction of precession of the magnetic moment over the internal anisotropy potential barrier) by Brown [4]. In his first calculations of escape rates, however, he confined himself to axially symmetric potentials of the magnetocrystalline anisotropy [4]. Hence no coupling between the longitudinal and the transverse modes of motion exists thus the longitudinal mode are governed by a single state variable, namely the colatitude ϑ, the polar angle of the magnetization vector.

As a result of this consideration Brown was able to demonstrate that the Kramers escape rate theory for particles may be easily adapted to yield an expression for the escape rate for spins which is valid for all values of the damping parameter in his Langevin equation and for any axially symmetric potential of the magnetocrystalline anisotropy.

An important comment should be made concerning the fact that the axially asymptotic equation for the escape rate is valid for all values of the damping. First we remark that the Fokker-Planck equation for the longitudinal relaxation of spins (the second state variable, namely, the azimuthal angle φ of the magnetization vector manifests itself merely as a steady precession of that vector) is effectively a one-dimensional Fokker-Planck equation (FPE), since the inertia of the particle plays no role, so that escape rates calculated from it are valid for all values of the damping parameter. In the Kramers problem for mechanical particles, on the other hand, the underlying FPE in position and momentum as state variables always has a two-dimensional state space and a reduction to a one-dimensional FPE (in this case the Smoluchowski equation) can be achieved only through the strong damping of the momentum. The particle problem may also be reduced to a one-dimensional problem if the damping is very small by writing the FPE in angle-action variables, averaging over the fast angle variable and considering the slow diffusion of the (total) energy. Thus in the mechanical Kramers problem, the following three regimes of damping appear.

(a) Intermediate-to-high damping (IHD): the general picture in this case [6] being that inside the well the distribution function is almost the Maxwell-Boltzmann distribution obtaining in the depth of the well. However, near the barrier the distribution function deviates from the equilibrium distribution due to the slow draining of particles across the barrier. The barrier region is assumed to be so small that one may approximate the potential in this region by an inverted parabola.

(b) Very low damping (VLD): here the damping is so small that the assumption in (a), namely that the particles approaching the barrier region from the depth of the well, have the Maxwell-Boltzmann distribution completely breaks

*Corresponding author.

down. Thus the region where deviations from that distribution occur extends far beyond the interval where the potential shape may be approximated by an inverted parabola. Thus we may now, by transforming the FPE into an equation in the energy and phase variables, and by supposing that the motion of a particle attempting to cross the barrier is almost conservative, and is the librational motion in the well of a particle with energy equal to the barrier energy, derive an equation of diffusion in energy. We remark that the assumption of almost conservative behavior meaning that the energy loss per cycle is almost negligible and is equal to the friction times the action of the undamped motion at the barrier energy ensures that the Liouville term in the FPE vanishes (unlike in IHD where there is strong coupling between the diffusive and Liouville term) so that one is left with only the diffusion term in the energy variable. The dependence on the phase having been eliminated by averaging the distribution function in energy-phase variables along a closed trajectory of the energy since we assume a librational motion in the well.

(c) An intermediate (crossover) region where neither IHD nor VLD formulas apply: in this region neither of the approaches to the problem may be used. In contrast to the VLD case the Liouville term in the FPE does not vanish meaning that one cannot average out the phase dependence of the distribution function which is ultimately taken account of by constructing from the FPE an equation for the distribution function with the energy and action as independent variables. This procedure allows one to express the energy distribution function at a given action, which in this case may be manipulated so as to pose the problem in terms of the energy loss per cycle at the barrier energy as a Wiener-Hopf equation yielding an integral formula the product of which with the IHD escape rate yields an expression for the escape rate which is valid for all values of the damping, so allowing the complete solution of Kramers's problem. The integral formula derived from the Wiener-Hopf equation effectively allowing for the coupling between the Liouville and dissipative term in the Kramers equation when written in terms of energy-phase variables which is ignored in the VLD limit.

However, as mentioned above the analogous spin problem is fundamentally different in that the one-dimensional FPE in the single state variable ϑ does not arise from *damping of the momentum* but rather from *axial symmetry*. Thus in order to construct Kramers formulas—equivalent to that for mechanical particles—for spins, one has to consider in Brown's Fokker-Planck equation nonaxially symmetric potentials of the magnetocrystalline anisotropy, i.e., magnetic systems with coupling between the two degrees of freedom.

We have mentioned that Kramers obtained two formulas for the escape rate, one valid in the so-called intermediate-to-high damping regime, and the other in the low damping regime, where it is assumed in both cases that the energy barrier is much greater than the thermal energy so that the concept of an escape rate applies. He mentioned in his paper, however, that he could not find a general method of attack for the purpose of obtaining a formula which would be valid for any damping regime [see (c) above] which was effectively solved by Mel'nikov and Meshkov [7].

As far as single-domain ferromagnetic particles are concerned, the equivalent of the Kramers IHD formula for spins was derived by Brown in 1979 [8], while the corresponding LD formula was established in 1990 by Klik and Gunther [9]. Furthermore, the latter authors emphasised that Brown's 1979 IHD calculation was in effect a special case of Langer's treatment of the decay of metastable states [10]. In addition, the LD formula of Klik and Gunther holds for escape from a single well, while in magnetic relaxation of single-domain ferromagnetic particles, the Gibbs free energy has in general a bistable structure due to the anisotropy term. Hence their formula does not take account of the possibility of recrossings of the anisotropy barrier by the magnetic moments in the LD case.

For the sake of clarity, we recall that the original Kramers problem, referring to the undamped motion, is characterized by the state variables, namely, q the position coordinate and p the momentum of a particle of mass m moving in a potential $V(q)$. Thus the Hamiltonian is

$$E=\frac{p^2}{2m}+V(q) \tag{1.1}$$

and the canonical variables (q, p) satisfy Hamilton's equations, namely,

$$\dot{q}=\frac{\partial E}{\partial p},\quad \dot{p}=-\frac{\partial E}{\partial q}. \tag{1.2}$$

In the treatment of the escape rate given by Mel'nikov and Meshkov, which yields a formula valid for all values of the friction for systems governed by the Hamiltonian (1.1), it is assumed that one may write the escape rate as

$$\kappa=A\kappa_{\mathrm{TST}}, \tag{1.3}$$

where κ_{TST} is the escape rate predicted by the transition state theory (TST) which for a double-well potential reads as

$$\kappa_{\mathrm{TST}}=\frac{\omega_1}{2\pi}\exp(-E_1/kT)+\frac{\omega_2}{2\pi}\exp(-E_2/kT). \tag{1.4}$$

Here, E_i is the energy barrier a given particle has to overcome when it is in well i and ω_i is an attempt frequency in well i (which is the frequency of oscillation in that well). In Eq. (1.3) A is a quantity termed the prefactor and contains corrections to the TST rate, namely, the effects of the surroundings of the particle.

Mel'nikov and Meshkov proceeded from the energy-action diffusion equation mentioned above by deriving a functional form for A in the LD limit, which bridges the two Kramers formulas. They then obtained the escape rate in the whole damping range by multiplying their functional form with the Kramers IHD prefactor.

The bridging between the two friction regimes has however never been effected for magnetic relaxation of single-domain ferromagnetic particles. Such a bridging formula is important here because in the low-damping limit one often encounters difficulties in implementing numerical simulations of the dynamics of the magnetization and longest lived

relaxation mode for the purpose of reproducing the exact solution for the escape rate [11]. It follows that a formula for the escape rate valid for all values of the friction would be useful (a) in view of its relative ease of computation and (b) as a check on the accuracy of numerical simulations as this formula, in principle, is able to qualitatively reproduce the frictional behavior of escape rates.

The purpose of this work is to demonstrate how to bridge LD and IHD magnetic Kramers formulas by suitably adapting the Mel'nikov-Meshkov procedure. Such adaptations are necessary because, unlike mechanical particles, the undamped equation of motion of the magnetization of a single-domain ferromagnetic particle is the gyromagnetic equation. Thus the Hamiltonian of the system is the Gibbs free energy which is in general not separable in terms the canonical variables of the problem. Furthermore, the magnetic system has two degrees of freedom, namely, the polar and azimuthal angles. This is in contrast with the original Kramers problem characterized by one degree of freedom and a two-dimensional state space. Again, unlike that problem the inertia of the particle plays no role with the result that even though the magnetic system has inherently two degrees of freedom, it still has a two-dimensional state space as in the Kramers problem. Having taken account of these considerations, the results obtained for single-domain ferromagnetic particles with uniaxial anisotropy when a uniform magnetic field is applied at an angle to the easy axis are compared to an exact solution in terms of matrix continued fractions. The range of applicability of this formula as the axially symmetric limit is approached will also be discussed.

II. PROBABILITY DENSITY DIFFUSION EQUATION FOR SPINS IN TERMS OF ENERGY-ACTION VARIABLES

The starting point of our investigation is the gyromagnetic equation

$$\frac{d\mathbf{M}}{dt}=\gamma(\mathbf{M}\times\mathbf{H}), \tag{2.1}$$

where γ is the gyromagnetic ratio, $\mathbf{M}$ is the magnetization vector of a single-domain ferromagnetic particle, and the field $\mathbf{H}$ which may comprise the field due to the magnetocrystalline anisotropy and external applied fields is

$$\mathbf{H}=-\frac{\partial V}{\partial \mathbf{M}}. \tag{2.2}$$

Here V is the Gibbs free energy density.

The equations of motion of the magnetization vector namely (2.1) may be written as an equation for the rate of change of the angular momentum $\mathbf{P}$ of a spin namely

$$\dot{\mathbf{P}}=\dot{\mathbf{u}}\frac{M_s}{\gamma}=\mathbf{u}\times\mathbf{h}, \tag{2.3}$$

where

$$\mathbf{u}=\frac{\mathbf{M}}{M_s},\mathbf{h}=-\frac{\partial V}{\partial \mathbf{u}}, \tag{2.4}$$

and M_s is the magnetic moment magnitude of a nonrelaxing particle. Equation (2.3), by introducing the orientations (ϑ,φ) of the magnetization vector $\mathbf{M}$ may in turn be written as

$$\frac{M_s}{\gamma}\dot{\vartheta}=\frac{1}{\sin\vartheta}\frac{\partial V}{\partial\varphi}(\vartheta,\varphi), \tag{2.5}$$

$$\dot{\varphi}\sin\vartheta=-\frac{\gamma}{M_s}\frac{\partial V}{\partial\vartheta}(\vartheta,\varphi). \tag{2.6}$$

If we now introduce as variable $x=\cos\vartheta$, Eqs. (2.5) and (2.6) assume the form of Hamilton's canonical equations *viz.*

$$\dot{x}=-\frac{\gamma}{M_s}\frac{\partial V}{\partial\varphi}(x,\varphi)=-\frac{\partial E}{\partial\varphi}(x,\varphi), \tag{2.7}$$

$$\dot{\varphi}=\frac{\gamma}{M_s}\frac{\partial V}{\partial x}(x,\varphi)=\frac{\partial E}{\partial x}(x,\varphi), \tag{2.8}$$

where $E(x,\varphi)=(\gamma/M_s)V(x,\varphi)$ is the Hamiltonian of the system. The set $\{\varphi,x\}\equiv\{q,p\}$ constitutes the canonical variables of the magnetic problem and consequently generates a two-dimensional state space as in Kramers's problem (1.1)–(1.2) (again referring to the undamped motion) with the difference that unlike mechanical particles the Hamiltonian $E(q,p)$ is no longer separable. Equations (2.7) and (2.8) describe the undamped motion of the system in the absence of thermal agitation and may be used to sketch the phase space trajectories. We also note in passing that the distribution W in phase space obeys Liouville's theorem of conservation of density in phase, namely (d/dt denoting a hydrodynamical derivative):

$$\frac{dW}{dt}=\frac{\partial W}{\partial t}+\dot{x}\frac{\partial W}{\partial x}+\dot{\varphi}\frac{\partial W}{\partial\varphi}=0, \tag{2.9}$$

which, by virtue of Eqs. (2.7)–(2.8) reads as

$$\frac{dW}{dt}=\frac{\partial W}{\partial t}+\frac{\gamma}{M_s}\left[\frac{\partial V}{\partial x}\frac{\partial W}{\partial\varphi}-\frac{\partial V}{\partial\varphi}\frac{\partial W}{\partial x}\right]=0. \tag{2.10}$$

If we include thermal agitation by regarding the Landau-Lifshitz equation as the Langevin equation of the system Eq. (2.1) becomes [12]

$$\frac{d\mathbf{M}}{dt}=\gamma(\mathbf{M}\times\mathbf{H})+\frac{\alpha\gamma}{M_s}(\mathbf{M}\times\mathbf{H})\times\mathbf{M}, \tag{2.11}$$

where this time

$$\mathbf{h}=-\frac{\partial V}{\partial\mathbf{u}}+\mathbf{h}_r(t), \tag{2.12}$$

with $\mathbf{h}_r(t)$ being the normalized δ-correlated white-noise force due to thermal fluctuations, and α is a dimensionless damping constant. Now, Eq. (2.11) accounts for orientation changes of the magnetization only, so that the FPE obtained from Eqs. (2.11)–(2.12) will govern the distribution function of magnetization orientations on the unit sphere. Hence, we have Brown's FPE [12,13]

$$2\tau_N\frac{\partial W}{\partial t}=\Lambda^2 W+\frac{\beta}{\sin\vartheta}\frac{\partial}{\partial\vartheta}\left[\sin\vartheta\frac{\partial V}{\partial\vartheta}W-\frac{1}{\alpha}\frac{\partial V}{\partial\varphi}W\right]+\frac{\beta}{\sin\vartheta}\frac{\partial}{\partial\varphi}\left[\frac{1}{\sin\vartheta}\frac{\partial V}{\partial\varphi}W+\frac{1}{\alpha}\frac{\partial V}{\partial\vartheta}W\right],\quad(2.13)$$

where in Eq. (2.13),

$$\Lambda^2=\frac{1}{\sin\vartheta}\frac{\partial}{\partial\vartheta}\left(\sin\vartheta\frac{\partial}{\partial\vartheta}\right)+\frac{1}{\sin^2\vartheta}\frac{\partial^2}{\partial\varphi^2}\quad(2.14)$$

is the angular part of the Laplacian and

$$\tau_N=\frac{\beta M_s}{2\alpha\gamma}\quad(2.15)$$

is the diffusion relaxation time, $\beta=\nu/kT$, and ν is the volume of a single-domain ferromagnetic particle. In the presence of thermal agitation, Liouville's theorem of conservation of density in phase no longer holds, so that one has

$$\frac{dW}{dt}=\frac{\partial W}{\partial t}+\frac{\gamma}{M_s}\left[\frac{\partial V}{\partial x}\frac{\partial W}{\partial\varphi}-\frac{\partial V}{\partial\varphi}\frac{\partial W}{\partial x}\right]=\mathbf{D}W,\quad(2.16)$$

where **D** denotes a dissipation operator. We remark that if we set $x=\cos\vartheta$ and make the transformation $\vartheta\rightarrow x$ in Eq. (2.13), then the term $\mathbf{D}W$ assumes the form

$$\mathbf{D}W=\frac{\alpha\gamma}{\beta M_s}\left\{\frac{\partial}{\partial x}\left[(1-x^2)\left(\frac{\partial W}{\partial x}+\beta W\frac{\partial V}{\partial x}\right)\right]+\frac{1}{1-x^2}\frac{\partial}{\partial\varphi}\left[\left(\frac{\partial W}{\partial\varphi}+\beta W\frac{\partial V}{\partial\varphi}\right)\right]\right\},\quad(2.17)$$

which is the form of **D** corresponding to the Langevin Eq. (2.11). We remark that we have split Brown's FPE into the two equations namely

$$\frac{dW}{dt}=\frac{\partial W}{\partial t}+\frac{\gamma}{M_s}\left[\frac{\partial V}{\partial x}\frac{\partial W}{\partial\varphi}-\frac{\partial V}{\partial\varphi}\frac{\partial W}{\partial x}\right]\quad(2.18)$$

which describes the undamped precessional motion and is entirely equivalent to Eq. (2.1) or (2.5)–(2.8), and

$$\frac{dW}{dt}=\frac{\alpha\gamma}{\beta M_s}\left\{\frac{\partial}{\partial x}\left[(1-x^2)\left(\frac{\partial W}{\partial x}+\beta W\frac{\partial V}{\partial x}\right)\right]+\frac{1}{1-x^2}\frac{\partial}{\partial\varphi}\left[\left(\frac{\partial W}{\partial\varphi}+\beta W\frac{\partial V}{\partial\varphi}\right)\right]\right\}.\quad(2.19)$$

Equation (2.19) expresses the irreversible total rate of change of the distribution function, i.e., irreversible evolution of the system from one phase point to an other.

In order to adapt the Mel'nikov-Meshkov approach for mechanical particles to the problem at hand, we must consider the quasistationary regime where

$$\frac{\partial W}{\partial t}\approx 0.\quad(2.20)$$

Equation (2.18) then becomes

$$\frac{dW}{dt}=\frac{\gamma}{M_s}\left[\frac{\partial V}{\partial x}\frac{\partial W}{\partial\varphi}-\frac{\partial V}{\partial\varphi}\frac{\partial W}{\partial x}\right].\quad(2.21)$$

We proceed by introducing the reduced energy variable

$$\varepsilon=\beta V(x,\varphi)\quad(2.22)$$

and transform Eqs. (2.19) and (2.21) into energy-azimuthal angle variables (ε,φ) by using the chain rule. We have

$$\left(\frac{\partial W}{\partial x}\right)_\varphi=\left(\frac{\partial\varepsilon}{\partial x}\right)_\varphi\left(\frac{\partial W}{\partial\varepsilon}\right)_\varphi,\quad(2.23)$$

$$\left(\frac{\partial W}{\partial\varphi}\right)_x=\left(\frac{\partial W}{\partial\varphi}\right)_\varepsilon+\left(\frac{\partial\varepsilon}{\partial\varphi}\right)_x\left(\frac{\partial W}{\partial\varepsilon}\right)_\varphi,\quad(2.24)$$

(where for simplicity we do not use a new symbol for the transformed distribution function) so that according to Eqs. (2.23) and (2.24) the dissipative term Eq. (2.19) becomes

$$\frac{dW}{dt}=\frac{\alpha\gamma}{\beta M_s}\left(\frac{\partial\varepsilon}{\partial x}\right)_\varphi\frac{\partial}{\partial\varepsilon}\left\{(1-x^2)\left(\frac{\partial\varepsilon}{\partial x}\right)_\varphi\left[W+\left(\frac{\partial W}{\partial\varepsilon}\right)_\varphi\right]\right\}_\varphi+\frac{\alpha\gamma}{\beta M_s}\frac{1}{1-x^2}\left\{\frac{\partial}{\partial\varphi}\left[\left(\frac{\partial W}{\partial\varphi}\right)_\varphi+W\left(\frac{\partial\varepsilon}{\partial\varphi}\right)_x\right]\right\}_\varepsilon$$
$$+\frac{\alpha\gamma}{\beta M_s}\frac{1}{1-x^2}\left\{\frac{\partial}{\partial\varphi}\left[\left(\frac{\partial W}{\partial\varepsilon}\right)_\varphi\left(\frac{\partial\varepsilon}{\partial\varphi}\right)_x\right]\right\}_\varepsilon+\frac{\alpha\gamma}{\beta M_s}\frac{1}{1-x^2}\left(\frac{\partial\varepsilon}{\partial\varphi}\right)_x\left\{\frac{\partial}{\partial\varepsilon}\left[\left(\frac{\partial W}{\partial\varphi}\right)_\varepsilon\right]\right\}_\varphi$$
$$+\frac{\alpha\gamma}{\beta M_s}\frac{1}{1-x^2}\left(\frac{\partial\varepsilon}{\partial\varphi}\right)_x\frac{\partial}{\partial\varepsilon}\left\{\left(\frac{\partial\varepsilon}{\partial\varphi}\right)_x\left[W+\left(\frac{\partial W}{\partial\varepsilon}\right)_\varphi\right]\right\}_\varphi,\quad(2.25)$$

and the Liouville term Eq. (2.21) reads as

$$\frac{dW}{dt}=\frac{\gamma}{\beta M_s}\left(\frac{\partial\varepsilon}{\partial x}\right)_{\varphi}\left(\frac{\partial W}{\partial\varphi}\right)_{\varepsilon}. \tag{2.26}$$

Now we have seen that x (effectively the magnetic momentum) and φ (the azimuthal angle) are canonical conjugate in the zero damping limit [13,14]. In this limit, the energy $V(x,\varphi)$ is a constant of the motion which is completely described by Eqs. (2.7) and (2.8). Regarding the damped motion, the energy is no longer conserved. However, in the crossover region the dissipation process may be perceived as consisting of small changes in the energy of the system, i.e., in the crossover dissipation process, ε is a slow variable (at given φ) while φ (keeping the energy fixed) is a fast variable. This means that in the first approximation we may neglect the effect of the operators $(\partial/\partial\varphi)_{\varepsilon}$ in the dissipative term Eq. (2.25). However, the angle φ is maintained in Eq. (2.26) because unlike the case of very low damping one needs to retain in first approximation the effect of the coupling between the Liouville and dissipation terms which must be taken into account if we wish to describe accurately the crossover region. Hence the dissipation operator contains only derivatives with respect to ε and Eq. (2.25) becomes

$$\frac{dW}{dt}=\frac{\alpha\gamma}{\beta M_s}\left(\frac{\partial\varepsilon}{\partial x}\right)_{\varphi}\frac{\partial}{\partial\varepsilon}\left\{(1-x^2)\left(\frac{\partial\varepsilon}{\partial x}\right)_{\varphi}\left[W+\left(\frac{\partial W}{\partial\varepsilon}\right)_{\varphi}\right]\right\}_{\varphi}+\frac{\alpha\gamma}{\beta M_s}\frac{1}{1-x^2}\left(\frac{\partial\varepsilon}{\partial\varphi}\right)_{x}\frac{\partial}{\partial\varepsilon}\left\{\left(\frac{\partial\varepsilon}{\partial\varphi}\right)_{x}\left[W+\left(\frac{\partial W}{\partial\varepsilon}\right)_{\varphi}\right]\right\}_{\varphi}. \tag{2.27}$$

In order to derive the energy-action diffusion equation one has to show that x depends on φ only. This is readily demonstrated by noting that the energy is supposed quasiconstant, so that we have

$$d\varepsilon(x,\varphi)=\left(\frac{\partial\varepsilon}{\partial x}\right)_{\varphi}(x,\varphi)dx+\left(\frac{\partial\varepsilon}{\partial\varphi}\right)_{x}(x,\varphi)d\varphi\approx 0, \tag{2.28}$$

leading to the differential equation of the phase space trajectories

$$\frac{dx}{d\varphi}\approx-\frac{\left(\frac{\partial\varepsilon}{\partial\varphi}\right)_{x}}{\left(\frac{\partial\varepsilon}{\partial x}\right)_{\varphi}}. \tag{2.29}$$

This equation, by imposing a suitable boundary condition (the value of x at the saddle point in general) and solving for x guarantees that x depends *explicitly* on φ only, and not on ε (see Appendix). This means that we can rewrite Eq. (2.27) as

$$\frac{dW}{dt}=\frac{\alpha\gamma}{\beta M_s}\left[(1-x^2)\left(\frac{\partial\varepsilon}{\partial x}\right)_{\varphi}^{2}+\frac{1}{1-x^2}\left(\frac{\partial\varepsilon}{\partial\varphi}\right)_{x}^{2}\right]\times\left\{\frac{\partial}{\partial\varepsilon}\left[W+\left(\frac{\partial W}{\partial\varepsilon}\right)_{\varphi}\right]\right\}_{\varphi}. \tag{2.30}$$

On equating the right-hand sides of Eqs. (2.25) and (2.30) we obtain finally

$$\left(\frac{\partial W}{\partial\varphi}\right)_{\varepsilon}\approx\alpha\left[(1-x^2)\left(\frac{\partial\varepsilon}{\partial x}\right)_{\varphi}-\frac{1}{1-x^2}\left(\frac{\partial\varepsilon}{\partial\varphi}\right)_{x}\frac{dx}{d\varphi}\right]\times\left\{\frac{\partial}{\partial\varepsilon}\left[W+\left(\frac{\partial W}{\partial\varepsilon}\right)_{\varphi}\right]\right\}_{\varphi}, \tag{2.31}$$

where we have used Eq. (2.29). By introducing the dimensionless action s via the differential equation

$$\frac{ds}{d\varphi}=(1-x^2)\left(\frac{\partial\varepsilon}{\partial x}\right)_{\varphi}-\frac{1}{1-x^2}\left(\frac{\partial\varepsilon}{\partial\varphi}\right)\frac{dx}{d\varphi}, \tag{2.32}$$

and using the chain rule again, we arrive at the energy-action diffusion equation

$$\frac{\partial W}{\partial s}=\alpha\frac{\partial}{\partial\varepsilon}\left(W+\frac{\partial W}{\partial\varepsilon}\right), \tag{2.33}$$

where we have suppressed the subscripts of the derivatives as their meaning is now obvious. The action variable is

$$s=\int_{\varepsilon=\text{constant}}(1-x^2)\left(\frac{\partial\varepsilon}{\partial x}\right)d\varphi-\frac{1}{1-x^2}\frac{\partial\varepsilon}{\partial\varphi}dx. \tag{2.34}$$

Here, we are interested in the critical energy trajectories which are given by

$$\varepsilon=\varepsilon_c, \tag{2.35}$$

where ε_c defines the energy contour through the saddle point of the energy from which the moments may overcome the barrier. When the energy of the system attains this value ε_c, the magnetization may leave the initial well, i.e., can reverse. In addition, we remark that Eq. (2.35) constitutes a boundary condition for Eq. (2.29) whence it follows that the action variable (2.34) is to be evaluated on the critical energy contour. We emphasise that the solution W of Eq. (2.33) will effectively assume the Maxwell-Boltzmann distributions deep in the wells and will change in a relatively narrow region about the top of the barrier, the behavior being analogous to that in the VLD case with the difference that the azimuthal dependence of W which in this case is described by the left-hand side of Eq. (2.33) may not be neglected near the top of the barrier. We further remark that Eq. (2.34) defines an action variable analogous to that introduced by Mel'nikov and Meshkov for mechanical particles. It is more complicated, however, because we reiterate that unlike mechanical systems, magnetic ones are not completely separable (the Hamiltonian is not additive in x and φ), so that

with the exception of time, one cannot separate the variables in the correponding (deterministic) Hamilton-Jacobi equation. As Eq. (2.33) is of the same form as the energy action-diffusion equation of Ref. [7] (details of the derivation of the Mel'nikov-Meshkov theory is given in Ref. [17]), it follows that the magnetic prefactor has a form identical to that for mechanical particles. For a double-well potential, this prefactor in the intermediate-to-low damping (ILD) limit (crossover region) where neither IHD nor VLD treatments apply is [7]

$$A(\alpha s_1, \alpha s_2) = \frac{A(\alpha s_1) A(\alpha s_2)}{A(\alpha s_1 + \alpha s_2)}, \tag{2.36}$$

where

$$A(\zeta) = \exp\left\{ \frac{1}{\pi} \int_0^\infty \ln(1 - e^{-\zeta(z^2+1/4)}) \frac{dz}{z^2+1/4} \right\} \tag{2.37}$$

is the prefactor for a single escape path, $\zeta = \alpha s_i$ is the energy loss per cycle in well i [justified following Eq. (3.4) below] and

$$s_i = \int_{\varepsilon = \varepsilon_{c_i}} (1-x^2) \frac{\partial \varepsilon}{\partial x} d\varphi - \frac{1}{1-x^2} \frac{\partial \varepsilon}{\partial \varphi} dx, \quad i = 1,2 \tag{2.38}$$

is the magnetic action in the well i. The line integral is taken along the direction of precession in well i.

Equation (2.36) is an accurate formula for the magnetic prefactor in the crossover region. In order to write down a formula for the magnetic prefactor which is valid for all values of the damping, we recall that the IHD prefactor derived by Brown has essentially the same form as that for mechanical particles and is [8,13,15] (the details of derivation of this prefactor which are rather lengthy are given in Geoghegan *et al.* [15] and also derived in detail using Langer's method [10] in Ref. [17]):

$$\frac{1}{2\omega_C \tau_N} \left[\left(\omega_C^2 + \frac{\alpha^2 (c_2^C - c_1^C)^2}{4} \right)^{1/2} - \frac{(c_2^C + c_1^C)\alpha}{2} \right], \tag{2.39}$$

where c_i^C is the coefficient of the second term in the Taylor expansion of the potential energy about the saddle point in terms of the magnetization direction cosines, and [8,13]

$$\omega_C \propto \sqrt{-c_1^C c_2^C} \tag{2.40}$$

so that the product of Eqs. (2.23) and (2.26) yields the magnetic prefactor in the whole damping range. It follows that a formula for the escape rate of magnetic moments across the anisotropy potential barrier which is valid for all values of the damping is

$$\kappa = \frac{1}{2\omega_C \tau_N} \left[\left(\omega_C^2 + \frac{\alpha^2 (c_2^C - c_1^C)^2}{4} \right)^{1/2} - \frac{(c_2^C + c_1^C)\alpha}{2} \right] \frac{A(\alpha s_1) A(\alpha s_2)}{A(\alpha s_1 + \alpha s_2)} \kappa_{\mathrm{TST}}. \tag{2.41}$$

We note that the frequencies ω_1 and ω_2 in each well are here respectively given by

$$\omega_i = \sqrt{c_1^i c_2^i}, \quad i = 1,2. \tag{2.42}$$

III. APPLICATION TO THE PROBLEM OF RELAXATION OF A SYSTEM HAVING UNIAXIAL ANISOTROPY WITH A UNIFORM FIELD AT AN ANGLE TO THE ANISOTROPY AXIS

We now apply the above considerations to the calculation of the greatest relaxation time of single-domain ferromagnetic particles having uniaxial anisotropy when an external uniform magnetic field is applied at an angle to the easy axis [i.e., in the (x, z) plane]. The dimensionless free Gibbs energy is given by

$$\varepsilon(x, \varphi) = -\sigma x^2 - \xi x \cos\psi - \xi \sqrt{1-x^2} \sin\psi \cos\varphi, \tag{3.1}$$

where ψ is the angle the direction of the magnetic field makes with the easy axis of magnetization. This potential has a saddle point in the Greenwich meridian $\varphi = 0$ (zero longitude) and an absolute maximum at $\varphi = \pi$. Furthermore, in Eq. (3.1) we have introduced the notations (reduced barrier height and reduced field parameter):

$$\sigma = \frac{K\nu}{kT}, \quad \xi = \frac{M_s H}{kT}. \tag{3.2}$$

Here, K is the anisotropy constant, M_s the saturation magnetization, and **H** is the external uniform magnetic field. Before proceeding, we note that it is customary to introduce the reduced field parameter h defined as [16]

$$h = \frac{\xi}{2\sigma}. \tag{3.3}$$

In order to evaluate the escape rate (2.41) for this problem, one must first numerically evaluate the action integrals (2.38) as well as Eq. (2.24). The quantity $A(\zeta)$ is best calculated by using its rerpresentation in terms of the complementary error function *viz.* [7]

$$A(\zeta) = \exp\left\{ -\sum_{p=1}^{\infty} \frac{\mathrm{erfc}(\sqrt{p\zeta}/2)}{p} \right\}.$$

We evaluate the action integrals (2.38) by reiterating that in the VLD limit, the free energy will be almost a constant of the motion. Hence, on using Eq. (2.29) *viz.* the fact that the energy is almost conserved and the Hamiltonian Eq. (3.1), we have the differential equation

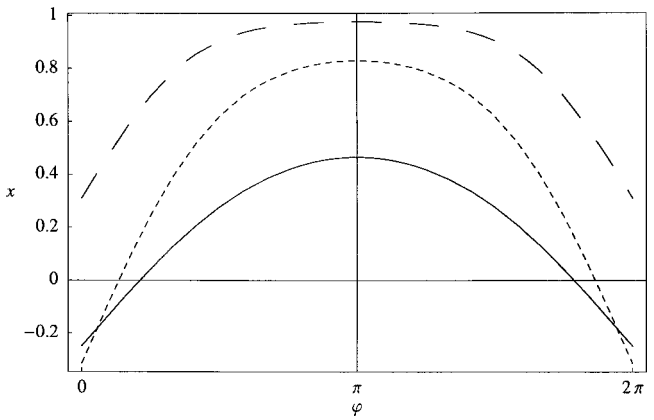

FIG. 1. The phase space (x,φ) for $\sigma=10$ and $\psi=\pi/6$, and $h=0.25$ (solid line); $\psi=\pi/4$, and $h=0.33$ (small dashed line); $\psi=\pi/3$, and $h=0.4$ (large dashed line).

$$\frac{dx}{d\varphi}=\frac{h(1-x^2)\sin\psi\sin\varphi}{x\sqrt{1-x^2}+h\cos\psi\sqrt{1-x^2}-hx\sin\psi\cos\varphi}. \tag{3.4}$$

This differential equation must be integrated subjected to the boundary condition

$$x(0)=x_C, \tag{3.5}$$

since the saddle point of the free energy surface is at $\varphi=0$. This boundary condition is needed since we are required to evaluate the action integral on the critical energy trajectory. We remark that the solution of Eq. (3.4) with the boundary condition (3.5) is unique on the interval $[0,2\pi]$. This procedure avoids solving the usual quartic equation involved [17] in the determination of x as a function of φ, together with all its disadvantages (selection of the correct root in particular). Having solved this equation numerically for $x(\varphi)$, one may calculate the actions in each well. Here, these integrals are equal because the regions contributing to the action variables in each well are symmetric. The contours of integration are delimitated by noting that the solution of Eq. (3.4) is periodic with period 2π and exhibits a maximum at $\varphi=\pi$ for any value of h and the angle ψ (see Fig. 1). This fact may be used to define the regions of integration, namely, $0\leqslant\varphi\leqslant\pi$ for the action variable in the first well, and $\pi\leqslant\varphi\leqslant2\pi$ in the second. To put this in another way, the same arc length is involved in the calculation of the two action variables corresponding to the two wells, so that the energy loss per cycle in each well is the same. The actual calculation of the line integrals (transformed into ordinary integrals) is usually performed numerically. The sole exception to this is the calculation of the action variables when a small field is applied perpendicular to the easy axis and is illustrated in section 4 of the present paper.

Having evaluated the escape rate rendered by Eq. (2.41) as described above, we compare the result with that obtained by calculating the escape rate exactly. In order to accomplish this, one expands the time-dependent probability density on the basis of the spherical harmonics, namely,

$$W(x,\varphi,t)=\sum_{l=0}^{\infty}\sum_{m=-l}^{l}a_{l,m}(t)N_{l,m}P_l^m(x)e^{im\varphi}, \tag{3.6}$$

where

$$N_{l,m}=(-1)^m\sqrt{\frac{(2l+1)(l-m)!}{4\pi(l+m)!}},\quad m>0, \tag{3.7}$$

and the $P_l^m(x)$ are the associated Legendre functions [18]. We also have [19]

$$P_l^{-m}(x)=(-1)^m\frac{(l-m)!}{(l+m)!}P_l^m(x).$$

The time-dependent Fokker-Planck equation may be reduced to the set of differential recurrence relations

$$\dot{a}_{l,m}(t)=\sum_{r=-2}^{2}\sum_{s=-r}^{r}d_{l,m,l+r,m+s}a_{l+r,m+s}(t), \tag{3.8}$$

where the $d_{l,m,l',m'}$ are the elements of Brown's Fokker-Planck operator and have been given in detail elsewhere [20]. The thirteen-term differential recurrence relation (3.8) now may be cast into the tridiagonal form [20]:

$$\dot{\mathbf{C}}_l(t)=\mathbf{Q}_l^-\mathbf{C}_{l-1}(t)+\mathbf{Q}_l\mathbf{C}_l(t)+\mathbf{Q}_l^+\mathbf{C}_{l+1}(t), \tag{3.9}$$

where $\mathbf{C}_l(t)$ is the column vector given by

$$\mathbf{C}_l(t)=\begin{pmatrix}a_{2l,-2l}(t)\\ a_{2l,-2l+1}(t)\\ \vdots\\ a_{2l,2l}(t)\\ a_{2l-1,-2l+1}(t)\\ \vdots\\ a_{2l-1,2l-1}(t)\end{pmatrix}, \tag{3.10}$$

and the elements of the matrices $\mathbf{Q}_l^{\pm}$ and $\mathbf{Q}_l$ are expressed in terms of the $d_{l,m,l',m'}$ [21]. The smallest eigenvalue of the Fokker-Planck operator may be calculated by calculating the smallest eigenvalue of the (8×8) matrix [20]

$$\mathbf{S}=-\frac{[\mathbf{Q}_1-\mathbf{Q}_1^+\Delta_2\mathbf{Q}_2^-]}{\left[\mathbf{I}+\sum_{n=2}^{\infty}\left(\prod_{m=1}^{n-1}\mathbf{Q}_m^+\right)\left(\prod_{k=1}^{n-1}\Delta_{n-k+1}^2\mathbf{Q}_{n-k+1}^-\right)\right]}, \tag{3.11}$$

where $\mathbf{I}$ is the (8×8) identity matrix and Δ_n is the infinite matrix continued fraction [20]

$$\Delta_n=\cfrac{\mathbf{I}}{-\mathbf{Q}_n-\mathbf{Q}_n^+\cfrac{\mathbf{I}}{-\mathbf{Q}_{n+1}-\mathbf{Q}_{n+1}^+\cfrac{\mathbf{I}}{-\mathbf{Q}_{n+2}-\cdots}\mathbf{Q}_{n+2}^-}\mathbf{Q}_{n+1}^-}. \tag{3.12}$$

This procedure allows us to compute the escape rate of the magnetization from a stable state of orientations for a wide range of anisotropy parameters σ and α and avoids the solu-

tion of a high order polynomial, provided that the smallest eigenvalue of Brown's Fokker-Planck operator is significantly lower than all the other eigenvalues.

We note that in order to be able to compare the rate given by Eq. (2.41) and those obtained from the diagonalization of Eq. (3.12), all approximate formulas given in this paper should be divided by α. In fact, on considering Eq. (2.13) and converting it to an eigenvalue problem we have

$$2\lambda\tau_N W+\frac{1}{\alpha}\left(\frac{\partial\varepsilon}{\partial x}\frac{\partial W}{\partial\varphi}-\frac{\partial\varepsilon}{\partial\varphi}\frac{\partial W}{\partial x}\right)=\frac{\partial}{\partial x}\left[(1-x^2)\left(\frac{\partial W}{\partial x}+\beta W\frac{\partial V}{\partial x}\right)\right]+\frac{1}{1-x^2}\frac{\partial}{\partial\varphi}\left[\left(\frac{\partial W}{\partial\varphi}+\beta W\frac{\partial V}{\partial\varphi}\right)\right], \quad (3.14)$$

where we have again set $x=\cos\vartheta$ in Eq. (2.13). This eigenvalue problem is solved in terms of the matrix continued fraction (3.12). However, in deriving Eq. (2.33) we have multiplied Eq. (3.14) by α. Thus we have implicitly considered the following eigenvalue problem

$$2\lambda'\tau_N W+\left(\frac{\partial\varepsilon}{\partial x}\frac{\partial W}{\partial\varphi}-\frac{\partial\varepsilon}{\partial\varphi}\frac{\partial W}{\partial x}\right)=\alpha\frac{\partial}{\partial x}\left[(1-x^2)\left(\frac{\partial W}{\partial x}+\beta W\frac{\partial V}{\partial x}\right)\right]+\frac{\alpha}{1-x^2}\frac{\partial}{\partial\varphi}\left[\left(\frac{\partial W}{\partial\varphi}+\beta W\frac{\partial V}{\partial\varphi}\right)\right], \quad (3.15)$$

with $\lambda'=\alpha\lambda$ (we note in passing that since $\alpha>0,\lambda_1'=\alpha\lambda_1$ will still be the smallest nonvanishing eigenvalue). This means that in order to compare Kalmykov's solution with, in particular, our formula (2.41) we have to divide it by α, including the generalization of the Klik and Gunther LD formula for a double-well potential, namely,

$$\kappa_{\mathrm{LD}}\tau_N=\frac{\alpha s_1}{4}\kappa_{\mathrm{TST}}, \quad (3.16)$$

where we have taken into account that the actions in each well are equal, $s_1=s_2$.

In addition, in the context of numerical calculations, we emphasize that care must also be taken in solving the differential equation (3.4). Namely, this equation is valid in so far as the concept of an escape rate has a meaning. In particular, according to the Stoner-Wohlfarth calculation of the critical reduced field h_c [12,22], one must be aware that the escape rate problem is meaningful only if

$$h\leqslant\frac{1}{(\cos^{2/3}\psi+\sin^{2/3}\psi)^{3/2}}, \quad (3.17)$$

which guarantees that the Hamiltonian (3.1) retains its bistable structure, so that the concept of an escape rate can be applied. Indeed, if this condition is violated, the only process involved is fast relaxation inside the only remaining well and not barrier crossing.

IV. LIMITING CASES

In order to check our formula (2.41), we study below known limiting cases which are (a) the explicit evaluation of the escape rate in the VLD regime for escape from a single well already considered by Klik and Gunther and (b) the calculation of the energy loss per cycle for a transverse applied field as already obtained by Garanin *et al.* [23].

First, we show how to obtain the Klik and Gunther formula from the single-well version of Eq. (2.41). For a single well, we have

$$\kappa=\frac{1}{2\omega_C\tau_N}\left[\left(\omega_C^2+\frac{\alpha^2(c_2^C-c_1^C)^2}{4}\right)^{1/2}-\frac{(c_2^C+c_1^C)\alpha}{2}\right]A(\alpha s)\kappa_{\mathrm{TST}}, \quad (4.1)$$

where only one term is retained in the expression (1.4) for κ_{TST} since there is only one well. In the limit of small α, Eq. (4.1) becomes

$$\kappa=\frac{1}{2\tau_N}A(\alpha s)\kappa_{\mathrm{TST}}. \quad (4.2)$$

Now, the behavior of $A(\zeta)$ for small ζ will be demonstrated. In that limit, the exponential in Eq. (2.37) may be expanded to first order in ζ to yield

$$A(\zeta)\approx\exp\left\{\frac{1}{\pi}\left[\ln\zeta\int_0^\infty\frac{dz}{z^2+1/4}+\int_0^\infty\frac{\ln(z^2+1/4)}{z^2+1/4}dz\right]\right\}. \quad (4.3)$$

Now [24]

$$\int_0^\infty\frac{\ln(z^2+1/4)}{z^2+1/4}dz=0, \quad (4.4)$$

so that Eq. (4.3) becomes

$$A(\zeta)\approx\exp\left[\frac{1}{\pi}(\pi\ln\zeta)\right]\approx\zeta, \quad (4.5)$$

and Eq. (4.2) finally reads

$$\kappa=\frac{\alpha s}{2\tau_N}\kappa_{\mathrm{TST}}, \quad (4.6)$$

which is, in our notation, the formula of Klik and Gunther.

Here we evaluate the action integral in Eq. (2.38) for a small uniform transverse applied field, yet large enough to ensure departure from axial symmetry. This is the only case in which the calculation may be carried out explicitly without encountering severe algebraic difficulties. It will also

serve to illustrate the difference between the action integral used by Garanin *et al.* which is evaluated by expressing x directly as a function of φ and the action integral used in the present problem which is calculated by using the differential Eq. (3.4) of the trajectories in the (x,φ) space. We remark that Garanin *et al.* [23], by noting that the potential due to a transverse field is a symmetric bistable potential, showed how the action integral may be calculated by considering the result for a single potential well only. Here, we will determine the action integrals in each well separately and we shall show how the action integral may be reduced to that of Garanin *et al.* We first note that Eq. (3.4) with $\psi=\pi/2$ is

$$\frac{dx}{d\varphi}=h\frac{(1-x^2)\sin\varphi}{x(\sqrt{1-x^2}-h\cos\varphi)}. \tag{4.7}$$

This equation is singular at the equator $x=0$. If $\psi=\pi/2$ the boundary condition for Eq. (4.7) is $x_c=0$ (because the saddle point is at the equator $\vartheta=\pi/2$ for a transverse field), thus we make the change of variable $u=x^2$ and transform Eq. (4.7) into an equation for u

$$\frac{du}{d\varphi}=2h\frac{(1-u)\sin\varphi}{(\sqrt{1-u}-h\cos\varphi)}, \tag{4.8}$$

with the boundary condition

$$u(0)=0. \tag{4.9}$$

Now, the field is relatively small so that, following Garanin *et al.* [23], when $h\to 0$, we may ignore terms of the order $h^{3/2}$. We may also write in the vicinity of the saddle point

$$1-u\approx 1. \tag{4.10}$$

Hence Eq. (4.8) becomes

$$\frac{du}{d\varphi}(\varphi)=2h\sin\varphi, \tag{4.11}$$

which may be readily integrated between 0 and φ to yield

$$u(\varphi)=2h(1-\cos\varphi), \tag{4.12}$$

leading to the two possible solutions for $x(\varphi)$

$$x(\varphi)=\pm 2\sqrt{h}\sin\frac{\varphi}{2}. \tag{4.13}$$

The two phase trajectories are equally possible. In physical terms, the solution with the plus sign corresponds to spins rotating clockwise, the minus sign to spins rotating anticlockwise. We note that for a transverse uniform applied field, this is always so, as the solution of Eq. (4.7) cannot be carried out without Eq. (4.9) which involves the square of the magnetic momentum.

Having expressed x in terms of φ one may evaluate the line integrals as follows. As the actions in each well are equal, it suffices to calculate the action variable in the first well only. As the reduced field parameter h is small and as on the critical energy curve we still have $1-u\approx 1$, we may rewrite Eq. (2.38) as

$$s_1\approx\int_{\varepsilon=\varepsilon_C}\frac{\partial\varepsilon}{\partial x}d\varphi-\frac{\partial\varepsilon}{\partial\varphi}dx. \tag{4.14}$$

Following Garanin *et al.* [23] and Coffey *et al.* [27], one can ignore the second term in this integral as it is of the order $h^{3/2}$ so that, accounting for the two phase paths given by Eq. (4.13) we have

$$s_1\approx\int_{\varepsilon=\varepsilon_C^+}\frac{\partial\varepsilon}{\partial x}d\varphi+\int_{\varepsilon=\varepsilon_C^-}\frac{\partial\varepsilon}{\partial x}d\varphi, \tag{4.15}$$

where ε_C^+ and ε_C^- denote the critical energy values associated with the two phase paths (4.13). These two values are equal. We have simply introduced the notation ε_C^+, ε_C^- in order to emphasise the difference between the respective contributions of the two possible integration paths.

Again, following Garanin *et al.* [23] we utilize the fact that

$$\frac{\partial\varepsilon}{\partial x}\approx -2\sigma x, \tag{4.16}$$

so that Eq. (4.15) can be finally evaluated to yield

$$\begin{aligned}s_1&=-4\sigma\sqrt{h}\left[\int_0^{\pi}-\sin\frac{\varphi}{2}d\varphi+\int_{\pi}^{0}\sin\frac{\varphi}{2}d\varphi\right]\\&\approx 16\sigma\sqrt{h}.\end{aligned} \tag{4.17}$$

Since $s_1=s_2$ and that the prefactor $A(\zeta)$ Eq. (2.37) behaves as ζ in the limit of small energy losses per cycle, we find, by evaluating the prefactor for the double-well potential Eq. (2.36),

$$A(\alpha s_1,\alpha s_2)\approx\frac{\alpha^2 s_1^2}{2\alpha s_1}=\alpha\frac{s_1}{2}=8\alpha\sigma\sqrt{h}, \tag{4.18}$$

in agreement with the calculation of Garanin *et al.* [23]. We reiterate that the splitting of the action integral in Eq. (4.15) arises because the differential equation (4.7) admits of two solutions corresponding to the two possible phase trajectories. Such a separation in the precession directions is, in general, not possible for arbitrary $\psi(\neq 0)$ because the solution of Eq. (3.4) with boundary condition (3.5) is then unique.

V. RESULTS AND CONCLUSIONS

We have indicated above how a formula (2.41) for the escape rate for single-domain ferromagnetic particles which is valid in the whole damping range may be obtained. This is accomplished by neglecting the azimuthal dependence of the distribution function in the dissipation term.

When one tries to apply the formalism described earlier to a particular problem in magnetism, we remark that the integration paths necessary to calculate the actions in each well are obtained by solving Eq. (3.4) with boundary condition

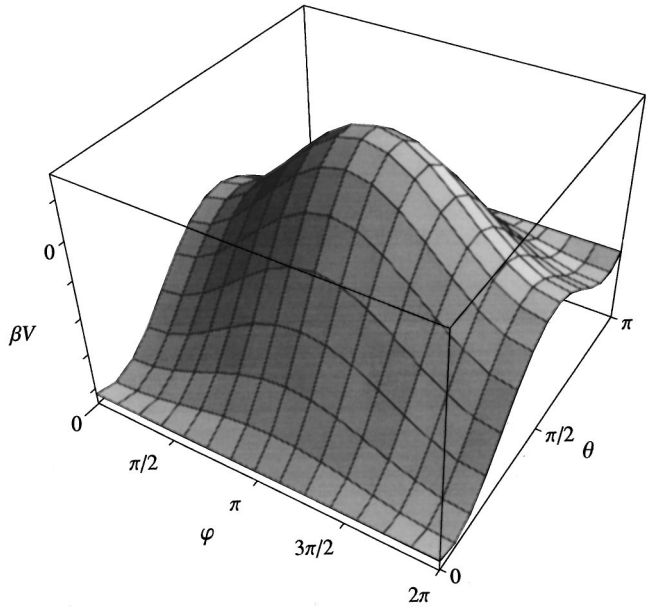

FIG. 2. 3D plot of the Hamiltonian as a function of ϑ and φ for $\psi=\pi/6$, and $h=0.2$. The value $\sigma=20$ has been arbitrarily chosen.

(3.5). We note from Fig. 1 that the phase trajectories are open (unlike those for mechanical particles) as one may expect for a motion of rotational type [25]. These phase trajectories are unique for any angle ψ except for $\psi=\pi/2$ for which two open trajectories are possible. These open trajectories are symmetric with respect to the straight line $x=0$. One may also remark that in order to calculate action integrals such as (2.38) transformed into ordinary integrals, one integrates over φ from 0 to π in the first well and from π to 2π in the second. This is because as is apparent from the open nature of the trajectories and from Fig. 2, both the Hamiltonian and the phase trajectories attain a maximum at $\varphi=\pi$.

In Fig. 3 the logarithm of the exact, LD and IHD, and the Mel'nikov-Meshkov magnetic formula (2.41) are plotted as a function of the logarithm of the friction parameter α. All the

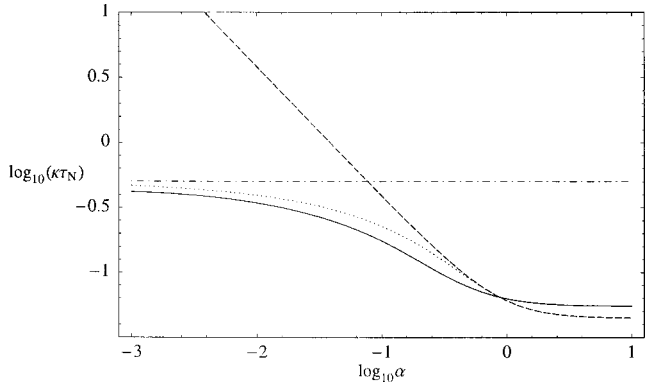

FIG. 3. $\log_{10}\kappa\tau_N$ vs $\log_{10}\alpha$ for $\sigma=10$, $\psi=\pi/6$, and $h=0.25$. The solid line is the exact rate; the dotted line is Eq. (2.41). The dashed line is Brown's nonaxially symmetric IHD formula and the dot-dashed line is the Klik-Gunther LD formula for the double-well potential (3.16).

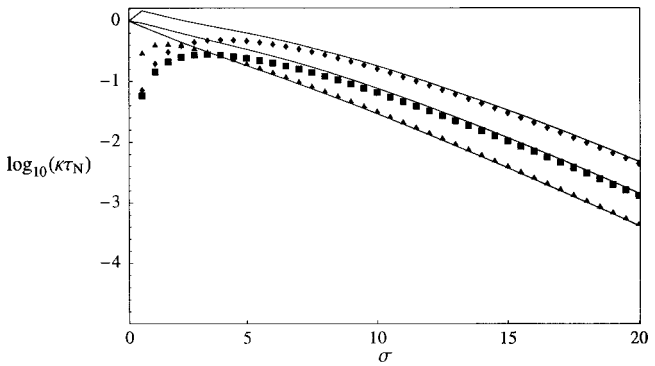

FIG. 4. $\log_{10}\kappa\tau_N$ as a function of σ. The solid line is the exact solution. Diamonds denote formula (2.41) for $\alpha=0.001$ (LD); squares denote formula (2.41) for $\alpha=0.1$ (crossover); and triangles denote formula (2.41) for $\alpha=10$ (IHD).

rates are normalized with respect to the characteristic diffusion time τ_N so that the problem of the choice of the starting stochastic differential equation namely Landau-Lifshitz or Gilbert is irrelevant. We remark that the qualitative behavior of the exact escape rate is well reproduced over the whole damping range, and especially in the crossover region where neither the LD, nor the IHD formulas are valid.

We have plotted on Fig. 4 $\log_{10}(\kappa\tau_N)$ as a function of σ for 3 typical values of the damping parameter α. It is apparent from this figure that the agreement between Eq. (2.41) and the exact solution is good for large σ as expected. The qualitative agreement in frictional behavior may be explained as follows. The behavior of the escape rate as a function of σ for large σ is approximately Arrheniuslike [8] and this behavior arises from an equilibrium property of the system (namely the Maxwell-Bolttzmann distribution at the bottom of the well). On the other hand, the frictional dependence of the escape rate is due to nonequilibrium (dynamical) properties of the system and so is contained in the prefactor A only, the detailed nature of which depends on the precise form of the asymptotic expression used to obtain it. In other words, not only does one have to postulate a high barrier (hence a Maxwell-Boltzmann distribution at the bottom of the well), one must also postulate the behavior of the distribution function at the barrier top. We remark that as emphasised by Kramers, it is hardly ever of any practical importance to improve on the accuracy of the IHD or LD formulas themselves because in experimental situations where relaxation is studied, one is left with estimates of the prefactor within a certain degree of accuracy which is difficult to evaluate. For example information on α is scanty. On the other hand it is important to predict the behavior of the relaxation times as a function of friction using analytical methods such as the one described in this paper because of the detailed information such methods yield concerning the mechanisms underlying the relaxation process.

Referring again to Eq. (2.41) we emphasise that this formula can be used when the angle ψ the field makes with the easy axis is large enough to ensure significant departures from axial symmetry [26] (see explanation below). Nevertheless, as one can see from Fig. 5 the calculation fails because the assumptions made in the derivation of the IHD and

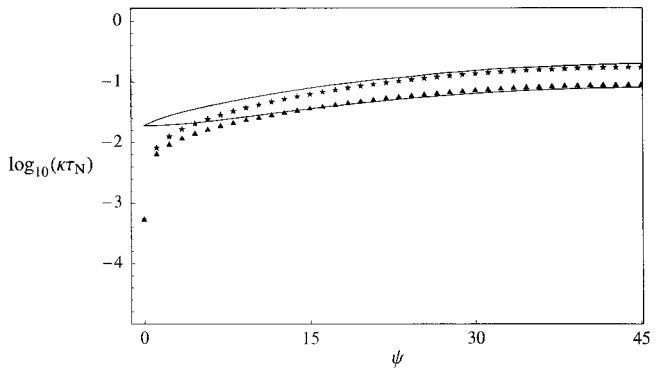

FIG. 5. $\log_{10}\kappa\tau_N$ as a function of the angle ψ (degrees) for $\sigma=10$, $h=0.2$. (1) Stars denote formula (2.41) for $\alpha=0.001$ (LD); (2) triangles denote formula (2.41) for $\alpha=0.1$ (crossover). The calculation fails for small angles.

VLD asymptotic formulas are invalid for small departures from axial symmetry [26,27]. Thus our formula Eq. (2.41) may not be used in the neighborhood of uniaxial crossovers because such action integrals as Eq. (2.38) are zero for any angle ψ (for small fields) or small departures of the latter angle from zero in any field.

In conclusion, it is apparent that Eq. (2.41) provides a good qualitative account of the behavior of the exact escape rate for magnetic particles having uniaxial anisotropy when an external magnetic field is applied at an angle to the easy axis. We finally remark that although our formula (2.41) provides a good approximation to the exact escape rate when the potential is truly non-axially symmetric, it has more limitations than its mechanical equivalent as it cannot be used for small angles ψ and low fields since the action as rendered by Eq. (2.38) then vanishes. Thus, if one desires a formula which embraces both uniaxial and frictional crossovers, one must suitably combine the techniques developed in Reference [23] with those of the present paper.

ACKNOWLEDGMENTS

We are grateful to Professor Yu. P. Kalmykov for communication of his results prior to publication. One of us (P.M.D.) thanks the United Kingdom EPSRC (Grant No. GR/L06225) for the support of this work. W. T. C. acknowledges the support of Enterprise Ireland Research (Grant No. SC/97/701), and D. J. McC. thanks the Dublin Institute of Technology for financial support.

APPENDIX: DETAILS OF DERIVATION OF EQ. (2.33)

In this appendix we give some details concerning the derivation of the energy-action diffusion Eq. (2.33) for magnetic dipole moments. In particular, we explain why we may derive Eq. (2.30). We have inferred in the text that x and the derivatives of ε with respect to x and φ are not explicit functions of ε, so that these may be taken out of the outer derivative sign with respect to ε. This may be justified as follows. We recall that in the LD limit the motion is quasideterministic. Thus the total energy is approximately conserved. Hence we have the equations of the trajectories in phase space

$$\varepsilon = f(x,\varphi) \approx \text{constant}. \tag{A1}$$

On taking the total differential of this equation we have

$$d\varepsilon = df(x,\varphi) \approx 0, \tag{A2}$$

Now, if the energy is constant, we will have, solving Eq. (A2)

$$x = g(\varphi). \tag{A3}$$

Furthermore, if the energy is *quasiconstant* as in the present problem we will then have

$$x \approx g(\varphi). \tag{A4}$$

Thus

$$\left(\frac{\partial \epsilon}{\partial x}\right)_{\varphi} = g_1[x(\varphi),\varphi], \tag{A5}$$

say, if the energy constant, and if it is quasiconstant, then

$$\left(\frac{\partial \varepsilon}{\partial x}\right)_{\varphi} \approx g_1[x(\varphi),\varphi], \tag{A6}$$

which is not an explicit function of ε and so may be taken outside the $(\partial/\partial\varepsilon)_{\varphi}$ operator in Eq. (2.27). Similar considerations hold for the derivative of the energy with respect to the angle φ. *With these approximations*, the dissipation term finally becomes Eq. (2.30).

[1] H. A. Kramers, Physica (Utrecht) **7**, 284 (1940).
[2] W. Wernsdorfer, E. Bonet Orozco, K. Hasselbach, A. Benoit, B. Barbara, N. Demoncy, A. Loiseau, H. Pascard, and D. Mailly, Phys. Rev. Lett. **78**, 1791 (1997).
[3] L. Néel, Ann. Geophys. (C.N.R.S.) **5**, 99 (1949).
[4] W. F. Brown, Phys. Rev. **130**, 1677 (1963).
[5] W. T. Coffey, D. S. F. Crothers, J. L. Dormann, Yu. P. Kalmykov, E. C. Kennedy, and W. Wernsdorfer, Phys. Rev. Lett. **80**, 5655 (1998).
[6] N. G. van Kampen, *Stochastic Processes in Physics and Chemistry*, 2nd ed. (North-Holland, Amsterdam, 1992).
[7] V. I. Mel'nikov and S. V. Meshkov, J. Chem. Phys. **85**, 1018 (1986).
[8] W. F. Brown, IEEE Trans. Magn. **15**, 1197 (1979).
[9] I. Klik and L. Gunther, J. Stat. Phys. **60**, 473 (1990).
[10] J. S. Langer, Ann. Phys. (N.Y.) **54**, 258 (1969).
[11] U. Nowak, R. W. Chantrell, and E. C. Kennedy, Phys. Rev. Lett. **84**, 163 (1999).
[12] W. T. Coffey, Yu. P. Kalmykov, and J. T. Waldron, *The Langevin Equation* (World Scientific, Singapore, 1996).
[13] W. T. Coffey, Adv. Chem. Phys. **103**, 259 (1998).
[14] W. T. Coffey, J. Mol. Struct. **479**, 261 (1999).

[15] L. J. Geoghegan, W. T. Coffey, and B. Mulligan, Adv. Chem. Phys. **100**, 475 (1997).
[16] W. T. Coffey, Yu. P. Kalmykov, and J. T. Waldron, Ref. [12].
[17] W. T. Coffey, D. A. Garanin, and D. J. McCarthy, Adv. Chem. Phys. (to be published).
[18] *Handbook of Mathematical Functions*, edited by M. Abramowitz and I. Stegun (Dover, New York, 1964).
[19] A. R. Edmonds, *Angular Momentum in Quantum Mechanics* (Princeton University Press, Princeton, NJ, 1957).
[20] Yu. P. Kalmykov, Phys. Rev. E **62**, 227 (2000).
[21] Yu. P. Kalmykov and S. V. Titov, Phys. Rev. Lett. **82**, 2967 (1999).
[22] E. C. Stoner and E. P. Wolfarth, Philos. Trans. R. Soc. London, Ser. A **240**, 599 (1948).
[23] D. A. Garanin, E. C. Kennedy, D. S. F. Crothers, and W. T. Coffey, Phys. Rev. E **60**, 6499 (1999).
[24] I. S. Gradshteyn and I. M. Ryzhik, *Table of Integrals, Series and Products* (Academic, New York, 1980).
[25] H. Goldstein, *Classical Mechanics*, 2nd ed. (Addison Wesley, Reading, MA, 1980).
[26] W. T. Coffey, D. S. F. Crothers, J. L. Dormann, L. J. Geoghegan, E. C. Kennedy, and W. Wernsdorfer, J. Phys.: Condens. Matter **10**, 9093 (1998).
[27] W. T. Coffey, D. A. Garanin, H. Kachkachi, and D. J. McCarthy, J. Magn. Magn. Mater. (to be published).