Micromagnetics at a finite temperature using the ridge optimization method

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The thermally activated transitions of the magnetization are studied for a chain of three identical interacting ferromagnetic particles. The saddle points are located by implementation of the ridge method with an initial direction of search chosen at random using the statistical properties of small collective thermal excitations. The solution is refined using a technique that combines aspects of the conjugate gradient and a Newton-type optimization. The methodology used identifies the transitions of relatively high probability of occurrence and deduces the energy barrier of least height. From the study of the topology of the energy surface, it is shown that the irreversible response of the system to an external field is modified at finite temperature. The model predicts a reduction in the symmetry of the "fanning" mode of magnetic reversal and a finite probability for transitions to states other than the state of negative saturation that is attributed to the local uniaxial anisotropy of the particles. The field dependence of the energy barrier exhibits a discontinuity when the final state following the activation process is no longer stable.

I. INTRODUCTION

In most theoretical micromagnetic studies of hysteresis in ferromagnetic media, the assumption is usually made that magnetic reversal occurs in the absence of any thermal perturbations. The free energy of the system, which comprises the anisotropy energy, the magnetostatic self-energy, the exchange energy, and the energy of interaction with an applied field, is first minimized, subject to the boundary conditions for the magnetic potential, to evaluate the equilibrium state of the magnetization. The magnetic reversal then occurs for some critical value of the applied field, the switching field,[1] when the local energy minimum is transformed to a point of inflection, whereupon an irreversible transition occurs into a new stable state.

The magnetization reversal, however, is intrinsically driven by thermal activation over finite energy barriers which arise for example from the intrinsic anisotropy in granular materials or the pinning mechanisms in materials whose magnetic behavior is predominantly governed by domain-wall motion. The thermally activated magnetic reversal results in phenomena of theoretical and practical interest, such as superparamagnetism[2] and the viscous decay of a recorded magnetic signal.[3] A framework for studying thermally activated processes is therefore needed.

The rate of escape over an energy barrier E_b in a bistable or metastable system can be evaluated using a variety of methods[4] and is given, under the condition of weak thermal noise $E_b/kT \gg 1$, by the Arrhenius-Néel law[5,6]

$$r = \nu e^{-E_b/kT} . \tag{1}$$

For a single-domain ferromagnetic particle, analytic expressions for the rate constant r were derived by Brown[7] using the Fokker-Planck differential equation that describes the time evolution of the probability-density distribution function of moment orientations. Numerical solutions were given by Aharoni[8,9] for a single particle and Rodé, Bertram, and Fredkin[10] for a pair of magnetic dipoles. The Fokker-Planck method, however, is difficult to apply in extended ferromagnetic systems with many degrees of freedom, since it requires the detailed knowledge of the energy surface of the system[11] which is not in general available.

An alternative approach is to simulate the thermal activation by generating stochastic moment trajectories from the metastable state by integrating the Langevin equations of motion.[12] The computational effort, however, increases exponentially with the size of the energy barrier E_b, so that the usefulness of this approach is limited to the simulation of near-spontaneous activation processes in systems with few degrees of freedom.

Although the prefactor ν is dependent in general on the temperature and the morphology of the energy surface,[7,13] in the limit of weak thermal noise, it can be regarded to a good approximation as a constant of the order 10^9-10^{10} sec^{-1},[6,7] The rate of escape r then depends to first order on the barrier energy E_b and the path of minimum energy through the saddle point of the transition becomes a good approximation of the magnetization reversal mechanism.[12] It is useful therefore to employ an optimization algorithm to evaluate the transition state of the system. Transition states are saddle points with Morse index 1. This means that the Hessian matrix H (i.e., the matrix of the second derivatives of the energy) at those points has a single negative eigenvalue. The calculation of the transition states is required to determine the prefactor ν,[14] as well as the precise heights of the energy barriers that are essential in any treatment of magnetic viscosity.

Analytic expressions for the energy barrier of a pair of magnetic dipoles have been given[15] and numerical estimates for more complex systems have also been report-

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ed.[16–18] In principle, the numerical methods used involve some intuitive choice of an internal variable that is assumed to vary monotonically during the activation process, while the other variables are optimized at fixed intervals. The resultant pathway, however, is not guaranteed to pass through a genuine saddle point and different choices of the variable that is assumed to vary monotonically may produce different results.[19,20] An additional difficulty is that the equilibrium magnetization configuration following the activation process is not always known in advance and may depend, as will be demonstrated later, not simply on the geometry of the system but also on intrinsic magnetic properties such as the anisotropy and the saturation magnetization.

There are considerable difficulties in applying existing saddle-point methods to multidimensional systems. The number of saddle points to be found is not known in general. The topology of the energy surface may be complex. Any method used should also be efficient and use for the most part only first-order derivative information of the energy surface.

There is no one optimization method for finding saddle points that is rigorous. Most of the progress has been motivated for application to the isomerization reactions of organic molecules. For instance, a stable algorithm has been constructed by minimization along conjugate directions.[21] Two directions p,q, are conjugate with respect to the Hessian H when $p^THq=0$. If an initial search direction p of negative curvature is found ($p^THp<0$), minimization in the subspace conjugate to p should converge to the saddle point.[21] The minimization part of the algorithm can be carried out using the quasi-Newton method that is quadratically convergent.[22] This results in an hybrid method that combines the stability of the conjugate gradient with the efficiency of quasi-Newton minimization. A good initial guess of the solution, however, is still required so that the energy can be approximated as a quadratic function. The method is therefore best suited to refining a solution and was adopted here as described in Sec. II.

For more difficult nonquadratic energy surfaces, a good initial guess of the solution is not available. Convergence in this case can be achieved iteratively by making use of only local information on the energy surface. An efficient method, introduced by Ionova and Carter,[23] finds transition states by minimization of the energy along the ridge, i.e., along the hypersurface that forms the boundary between the domains of attraction of two stable states. The ridge method uses only gradient information to achieve convergence and explores a smaller area of the coordinate space, so it can more readily be applied to systems with a large number of degrees of freedom.

The ridge method converges to a transition state starting from any arbitrary starting point on the ridge. This represents an important advantage over previous methods that allows a systematic investigation of the possible transitions from a metastable state. This search is needed, for example, to evaluate the transition state of lowest barrier height, since no saddle-point method can guarantee convergence to a globally optimal state. In our implementation of the ridge method, the search for transition states is carried out using a technique that considers the normal modes of thermal excitation of the system. This is more similar to "importance sampling" than the search using a random initial direction. We note that the computational method is thus intimately linked with the physics of the problem rather than being developed as a general optimization method. A description of the implementation is given in Sec. II. In Sec. III, the thermoactivated transitions are studied for a model three-particle micromagnetic system that is simple enough to be physically transparent while demonstrating interesting transition phenomena due to its relatively complex energy surface.

II. DESCRIPTION OF THE METHOD

In the present study, we address the problem of finding multiple transition states in a micromagnetic system that are associated with a given metastable state. The transition states were located by implementation of the ridge method of Ionova and Carter.[23]

The ridge method[23] relies on the fact that any genuine saddle point, i.e., a saddle point with one unstable direction, must satisfy the mountain pass theorem.[24] The method, shown schematically in Fig. 1, can briefly be described as follows. Suppose x_0, x_1 are two points in a n-dimensional coordinate space that represent stable configurations (local energy minima) that belong to neighbor domains of attraction so that they are separated by an hypersurface (ridge) of greater functional value. A linear search is carried out along the direction of negative curvature $[x_0,x_1]$ for a maximum of the potential energy on the ridge at some point x_m. A small interval $[x_0',x_1']$ around the point x_m is then defined by setting $x_0'=x_m-p$, $x_1'=x_m+p$, where p is a side step vector parallel to $[x_0,x_1]$. Two new points x_0'', x_1'' are then defined from $x_0''=x_0'+\alpha p_0, x_1''=x_1'+\alpha p_1$ where $\alpha>0$ and p_0,p_1 are directions downhill in the energy surface. A line search along $[x_0'',x_1'']$ is then carried out to obtain

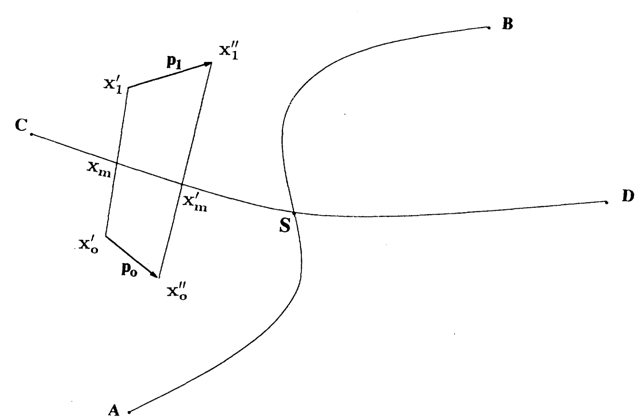

FIG. 1. A schematic illustration of the ridge method. The saddle point, shown by the symbol S is connected through valleys to the minima, A, B and through the ridge to the maxima C, D.

a maximum x'_m on the ridge. This procedure is repeated and a sequence of points $x_m, x'_m \ldots$ on the ridge is constructed that should gradually converge to a saddle point x_s.

In our numerical tests, the ridge method was found not to be always stable so that convergence to a stationary point is not guaranteed. For instance, if the ridge curves severely, it is possible that the points x''_0, x''_1 that are generated belong to the same domain of attraction. A maximum point x'_m may still be located that does not belong to the ridge but rather to some local undulation in the energy surface on the same domain of attraction that decays eventually on a "mountain slope." A linear search in the direction $[x''_0, x''_1]$ will not necessarily reveal the existence of a second maximum so that no error can be perceived by the algorithm until at some stage no maximum point x'_m can be found. It is difficult to envisage a test that avoids failure in this situation, although some ideas contained in the detour algorithm of Ionova and Carter[23] may be useful. The fundamental problem appears to be that the algorithm uses only local information of the energy surface and it is not possible to know *a priori* that the sequence of iterations does not lead to a saddle point.

The rate of convergence of the ridge method is linear. The position of the saddle point was therefore refined in our study by implementation of the quadratically convergent method of Bell, Crighton, and Fletcher,[22] when the gradient on the ridge becomes sufficiently small $|g(x'_m)| < \epsilon_1$. The basic requirement of the method, that the energy surface is of nearly quadratic form, is satisfied in most cases when $\epsilon_1 = 0.001$. The method of Bell, Crigton, and Fletcher[22] involves an energy minimization in the subspace Z that is conjugate to the direction of negative curvature (that is provided here by the ridge method). For the minimization part of the algorithm, we have chosen a Newton-type rather than quasi-Newton optimization, since it was found to be more robust in the case when the Hessian matrix in conjugate space $Z^T H(x_m) Z$ is ill conditioned, for example close to some critical points, when the energy curvature at the saddle point x_s about some stable direction becomes rather flat. The algorithm was terminated when the condition $|g(x'_m)| < \epsilon_2 = 2 \times 10^{-6}$ was satisfied.

The main difficulty in the implementation of the ridge method in a micromagnetic system is that in general one has knowledge only of the initial metastable state x_0 but the position of the second stable state x_1 is not known. If a direction of negative curvature is found and the ridge method[23] can be initiated, then a method has been suggested by Ionova and Carter[23] that has the potential of finding all the transition states that may be present in the system. In complex micromagnetic systems, however, the total number of transition states may be excessively large and our objective is to restrict the search for those that involve a given metastable state x_0.

In principle, it is possible to search for transition states by generating the initial direction of negative curvature $p \equiv [x_0, x_1]$, without knowledge of x_1, completely at random. However, when studying processes of thermoactivated magnetic reversal, one is normally interested in those transitions only that have relatively low activation energies and are therefore more likely to occur. In this case, it may be of advantage to initiate the ridge method with a direction of negative curvature $[x_0, x_1]$ generated using the normal modes of excitation of the system[25] about the metastable state x_0.

If $[u_i, \lambda_i, i = 1, n]$ are the orthonormal eigenvectors and respective eigenvalues of the Hessian matrix $H(x_0)$, the direction of search p for a maximum on the ridge is generated using

$$p = \sum_{i=1}^{n} y_i u_i \ , \tag{2}$$

where the amplitudes y_i of the normal modes are obtained numerically at random from the Gaussian distribution defined by

$$\langle y_i \rangle = 0 \ , \quad \langle y_i^2 \rangle = kT/\lambda_i \ . \tag{3}$$

Equation (3) is consistent with the requirement of equipartition of energy $kT/2$ on average for each mode, in the classic limit. The directions p thus generated are representative of the spontaneous response of the system in the low-temperature thermodynamic limit.

The implicit assumption in using Eq. (2) is that the saddle point x_s of the thermally activated transition in a micromagnetic system can be located in a direction that involves primarily the modes of lower frequency. The limitations involved in the assumption can be determined by considering in more detail the variation in the topology of the energy surface.

The topology of the energy surface is determined by the number and the Morse index i $(0 \le i \le n)$ of the stationary points. If the energy surface is dependent on k external (control) parameters $c_\alpha, \alpha = 1, \ldots, k$, then the response of a stationary point x to small changes in c_α is given by[26]

$$\frac{\partial x_i}{\partial c_\alpha} = -\sum_j H_{ij}^{-1} H_{j\alpha} \ , \tag{4}$$

where H_{ij}, $H_{j\alpha}$ refer to the matrices of the second-order energy derivatives of dimension $n \times n$ and $n \times k$, respectively.

If the Hessian matrix H_{ij} is nonsingular, the response of the stationary point x to small variations in the control parameters c_α is linear. A discontinuous response occurs at critical points x^0 in the energy surface, when both the gradient and the determinant of the Hessian vanish $(|g| = 0, |H| = 0)$. At such critical points, the Morse index of a stationary point changes and the topology of the energy surface is modified. The locus of all critical points c^0 in the space of the control parameters is a $(k-1)$-dimensional hypersurface, that is defined as the separatrix of the system in the framework of catastrophe theory.[26]

The switching field H_{sw} of a micromagnetic system as defined by Schabes and Bertram[1] is a generalization of the concept of a nucleation field[27] that is applicable for systems with nonuniform magnetization states at equilibrium. The switching field is an example of a critical point c^0 such that the local minimum in the energy surface is transformed to a saddle point. In the absence of any degeneracies, the eigenvalue of the lowest frequency then vanishes, ($\lambda_1=0$) and an irreversible magnetic reversal occurs initially along the direction of the associated eigenmode u_1. If the applied field is smaller but allowed to approach the switching field there will be a small but finite energy barrier. During the process, the principal axes of the saddle point and the energy minimum become gradually parallel and the eigenvalues associated with at least one direction tend towards zero.[26] It is evident that the saddle point must be approaching from the direction of the principal axis of vanishing eigenvalue, i.e., the "soft" mode of the system. The soft mode of the system is therefore a good guess of the position of the saddle point of the transition in the limit $H_a \to H_{sw}$.

Transition states other than the one traced through the soft mode may also exist, for a given value of the applied field. These involve, in general, energy barriers of different height. The magnetization reversal will be determined, however, only by transition states of low activation energies that are expected to involve mainly the low-frequency normal modes of excitation. For this reason, the generation of the initial direction of search using Eq. (2) should be of some practical advantage.

There are some important limitations in using Eq. (2). The method can be computationally expensive, since searches from different starting points on the ridge may converge to the same solution. A large number of searches may also be necessary since one has no advance knowledge whether all the transition states of interest have been located. It is more efficient to employ the original method[23] when the position of the stable states x_1 following the activation process are known in advance. Finally, the method adopted is not applicable to any arbitrary system but limited, as discussed above, to physical systems close to a first-order phase transition, when the energy of activation is relatively small.

The contribution of different normal modes to the activation process can be determined by expressing the deviation $x_s - x_0$ required for the magnetization to attain the transition state, as a sum over the complete set of orthonormal eigenvectors u_k of the Hessian matrix at the metastable state $H(x_0)$.

$$x_s - x_0 = \sum_{k=1}^{n} a_k u_k \ . \tag{5}$$

The amplitude a_k then represent the relative contribution of the kth normal mode to the activation process in the limit of weak thermal noise $E_b/kT \gg 1$.

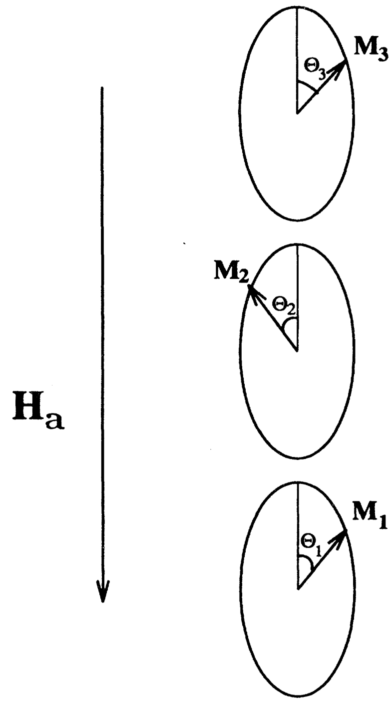

FIG. 2. A system of three anisotropic single-domain particles.

III. THERMAL ACTIVATION IN A SIMPLE MICROMAGNETIC SYSTEM

We consider a system of three identical interacting fine ferromagnetic particles as shown in Fig. 2. Following a description of the implementation of the method for this system in Sec. III A, the thermally activated magnetic reversal is studied in detail in Sec. III B and a discussion of the importance of the local particle anisotropy is given in Sec. III C.

A. Implementation of the ridge method

The fine particles in Fig. 2 may be allowed to have a point contact and are assumed to possess uniaxial (shape or magnetocrystalline) anisotropy in the direction of alignment. An external field H_a is applied along the common anisotropy axis tending to cause magnetic reversal. The direction of the magnetization of the single-domain particles is defined in general by polar and azimuthal angles θ_i, ϕ_i $(-\pi \le \theta_i \le \pi, 0 \le \phi_i \le \pi,\ \ i=1,2,3)$ that represent the internal variables of the system. Using a dipole approximation for the demagnetizing field, the free energy of the system can be expressed as

$$E = \frac{1}{2} m H_k \sum_i \sin^2\theta_i + m^2 \sum_i \sum_{j>i} \frac{\sin\theta_i \sin\theta_j \cos(\phi_i - \phi_j) - 2\cos\theta_i \cos\theta_j}{r_{ij}^3} + m H_a \sum_i \cos\theta_i \ , \tag{6}$$

where m is the magnetic moment of a particle, H_k is the local anisotropy field of a particle, and r_{ij} is the particle separation.

The azimuthal dependence of the energy was ignored in the calculations. This simplification is possible in systems of uniaxial symmetry[15,28,29] since the path of minimum energy through the low-lying transition states in the energy surface involves the collective rotation of polar angles only and the deviation of any moment in the azimuthal direction results in an expense in dipole interaction energy. Transition states such that the magnetic moments are not confined on the same plane are ignored here since they involve relatively high-energy barriers.

The configuration of perfect magnetic alignment $(\theta_1,\theta_2,\theta_3)=(000)$ is a local energy minimum, provided that the reverse field H_a is smaller than the nucleation field H_n $(H_a<H_n)$. The nucleation field H_n (Ref. 27) is the zero-temperature switching field $[H_n=H_{\rm sw}(T=0)$ (Ref. 1)] that causes a spontaneous transition by making the energy barrier vanish $(E_b=0)$. Here it is distinguished from the switching field $H_{\rm sw}(T)$ that can "nucleate" a magnetic reversal at some point in the system by thermal activation $(E_b>0)$, as will be demonstrated later. We stress that the concept of a switching field as introduced by Schabes and Bertram[1] is here generalized to allow it to be temperature dependent.

The nucleation field H_n and the normal modes of excitation of the system are obtained by diagonalization of the Hessian H at the energy minimum $(\theta_1,\theta_2,\theta_3)=(000)$, given by

$$H_{ij}=mH_k\left[\frac{\kappa}{|i-j|^3}(1-\delta_{ij})+\left[\sum_{l\neq i}\frac{2\kappa}{|i-l|^3}+1-h\right]\delta_{ij}\right], \qquad (7)$$

where δ_{ij} is the Kronecker delta function, $\kappa=m/(H_k r^3)$ is a dimensionless parameter that is a measure of the strength of the dipole interaction between neighbor particles at a distance r and $h=H_a/H_k$ is the applied field in reduced form. The nucleation field H_n is then given as the value of the applied field H_a that will make the lowest positive eigenvalue of the Hessian vanish.

The implementation of the ridge method starts by defining the point x_0 to be the metastable state (000). The nucleation field H_n is determined and the applied field is chosen to be close but smaller than that value. The direction $[x_0,x_1]$ is then defined as the direction of the soft mode (i.e., the eigenvector of lowest eigenvalue) and a linear search is carried out to obtain the maximum point x_m on the ridge. The saddle point x_s is then located using the ridge/Newton-type optimization procedure. The value of the applied field is then changed by a small amount $H_a\to H_a-\delta H_a$ and two different methods can be employed to define the initial direction of search $p=[x_0,x_1]$ for the maximum on the ridge.

In the first method, the initial direction of negative curvature p is chosen from the previous solution by setting $x_1=x_s$, prior to the implementation of the ridge method. The evolution of the saddle point in coordinate space as the field H_a is varied can then be traced. At critical values of the field, when the Morse index of the saddle point changes, convergence to a nongenuine saddle point is likely. Since the method is deterministic, no further progress can be achieved, so it is then necessary to modify the initial direction of negative curvature by a small random perturbation and initiate a new search.

In the second method, the initial direction of negative curvature p is generated using Eq. (2). This allows saddle points that can not be traced by the former method to be located.

It is possible that a direction is generated such that a maximum on the ridge is not found before one of the variables θ_i reaches its boundary value $\pm\pi$. The imposition of linear inequality constraints would increase substantially the complexity of the algorithm and it is preferable to treat the problem as one of unconstrained optimization. The periodic boundary condition $\theta_i=\theta_i+2m\pi$ (where m is an integer) is imposed, if necessary, when the solution x_s is found. This procedure is of advantage, for example, when the trajectory of a saddle point in coordinate space is traced close to the boundary surface.

The initial value of the side step size $|p|$ was chosen so that at the points x_0,x_1 the maximum declination of any moment from the orientation at the ridge did not exceed 10°, i.e., $p_i^{\max}=10°$. The downhill step constant α was initialized using $\alpha_{\rm in}|g(x_m)|/|p|=r_{\max}=0.5$ and in subsequent reductions of the step sizes[23] the minimum ratio was set to $r_{\min}=0.25$. For each update of the applied field, convergence to a saddle point was achieved in typically less than eight iterations using the first method and less than 30 iterations using the second method. When searching for multiple transition states, six independent searches were conducted using the second method for each field update.

B. Results for a system of three coupled particles

There are two main considerations in a study of thermally activated processes. First, it is necessary to evaluate the transition state of minimum barrier height. The path of minimum energy is associated with the mode of activation that is most likely to occur. Secondly, it is also useful to consider whether transitions to some different states are possible. Evaluation of the barrier heights would then provide an estimate of their relative probability.

The energy barrier of lowest height, for some constant value for κ, was evaluated as follows. The external field H_a was set to a value close to the nucleation field H_n. The saddle point x_s associated with the soft mode was first located and the associated energy barrier was evaluated. Then an attempt to sample other transition states was carried out by stochastic generation of the direction of negative curvature using Eq. (2). The associated energy barriers were compared with the energy barrier for the soft mode. The minimum energy barrier was found at the saddle point associated with the soft mode. The applied field H_a was subsequently reduced in magnitude in small discrete steps. The same procedure was repeated for each step. The minimum energy barrier was found in all cases to be associated with the saddle point traced through the soft mode (using the first method described in the previous subsection).

Clearly, the position of the saddle point $x_s=(\theta_1,\theta_2,\theta_3)$ traced through the soft mode provides important information on the mode of thermally activated magnetic reversal that is most likely to occur. To identify the different types of thermally activated magnetic reversal, we also considered the dependence on applied field. In Fig. 3, the polar orientations of the three moments at the saddle point x_s of lowest barrier height are plotted as a function of field for different strengths of interaction ($\kappa=0.3$, 0.5, 0.9). In general, two distinct types of magnetic reversal have been observed.

In the case when the coupling strength is strong $\kappa>0.89$ or the field is sufficiently close to the nucleation field $|H_a|\le|H_n|$, the nucleation process occurs by asymmetric "fanning"[30] with the inclination of the two particles at the end of the chain being precisely identical $\theta_1=\theta_3$, $|\theta_2|<|\theta_1|$. The symmetric fanning mode $(\theta_1=-\theta_2=\theta_3)$ is, however, a good approximation as is illustrated in Fig. 3(a). Two saddle points can be located that arise from the invariance in the transformation $(x_s\rightarrow -x_s)$. In recent work on the same system, it was reported that $|\theta_2|>|\theta_1|$.[17] The discrepancy may arise from a small error in the convergence of the optimization method used (simulated annealing) and indicates the importance of refining the solution to a good accuracy.

In the case when the coupling strength $\kappa<0.89$, a reduction in the symmetry of the fanning mode $(\theta_1\neq\theta_3)$ occurs at a critical value of the applied field $h_{\rm cr}$ [Figs. 3(b) and 3(c)]. The transition occurs when the lowest positive eigenvalue of the Hessian matrix at the saddle point vanishes. The energy surface along the associated eigendirection becomes flat and the saddle point becomes a degenerate non-Morse critical point. The topology of the energy surface changes and a discontinuous variation of the position of the saddle point with applied field is observed [Figs. 3(b) and 3(c)]. The number of saddle points increases from 2 to 4. The fourfold symmetry results from the additional invariance in the transformation $(\theta_1,\theta_2,\theta_3)\rightarrow(\theta_3,\theta_2,\theta_1)$. A similar behavior has been observed for a system of two interacting particles.[15] In the case of moderately strong coupling $(0.45<\kappa<0.89)$, a second transition to symmetric fanning occurs at $h=-h_{\rm cr}$, i.e., when the applied field is in the direction resisting magnetic reversal. The field dependence of the polar components θ_1,θ_3 at the saddle point therefore forms a loop, as shown in Fig. 3(b).

When the coupling strength $\kappa>0.45$, the transition is always to the state with the moments reversed $(\pi\pi\pi)$. A different behavior is observed when $\kappa<0.45$ as shown in Fig. 3(c). At some value of the field $h_0<h_{\rm cr}$ [not shown in Fig. 3(c)], the transition is no longer to the state $(\pi\pi\pi)$ but to the state with one of the moments at the edges reversed, i.e., $(\pi 00)$ or (00π). The saddle point then gradually converges to the state $(\pi 00)$ as the field is varied. Eventually the state $(\pi 00)$ becomes a saddle and a transition to that state is no longer possible. Minimization of the energy in either direction of steepest descent retrieves

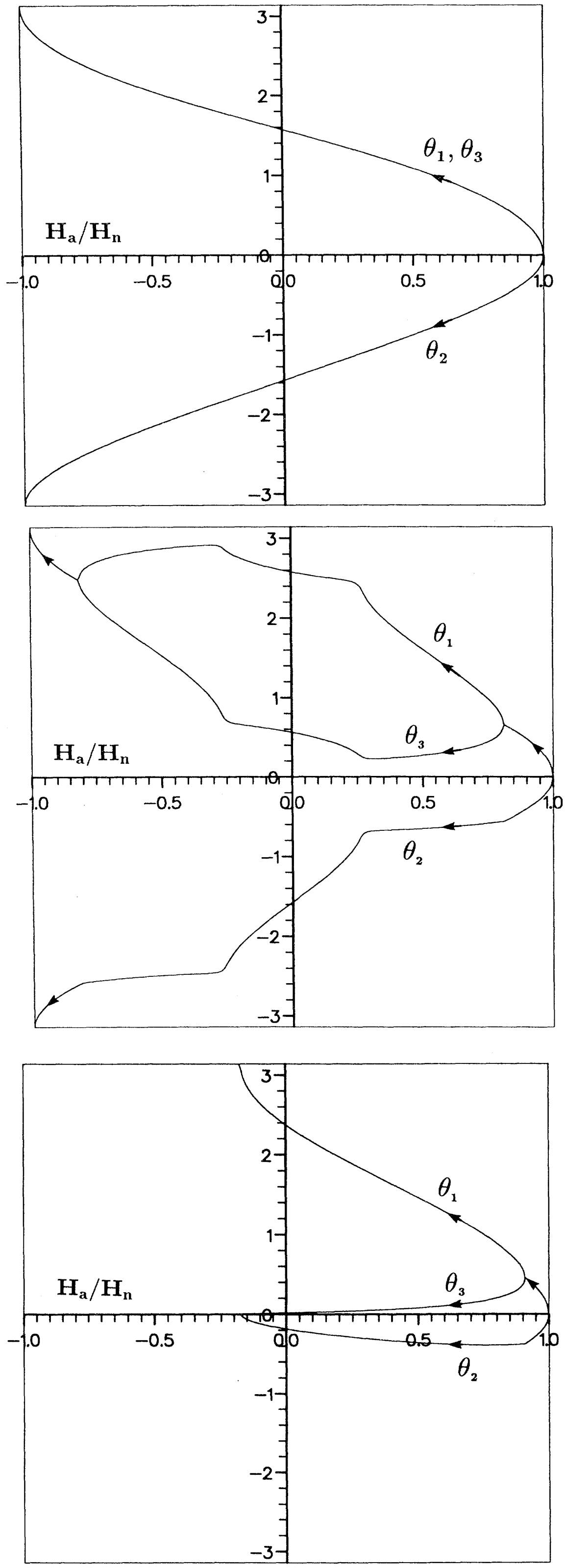

FIG. 3. The polar angles $\theta_1,\theta_2,\theta_3$ of the moments at the saddle point as a function of the reduced applied field H_a/H_n. (a)–(c) were obtained with the constant of magnetostatic coupling given by $\kappa=0.9$, 0.5, 0.3, respectively.

the original state (000), so the saddle point (π00) is not associated with any particular transition. A more detailed discussion of the field h_0 is given later. A similar behavior has been observed for a system of two particles,[12] where activation to a state with the two moments being antiparallel is possible when the dipole interaction is sufficiently weak.

Next, we test the validity of the assumption made regarding the importance of the low-frequency normal modes in the activation process. We consider the case when x_s is associated with the minimum activation energy and express $x_s - x_0$ as a sum over the eigenvectors u_k of the Hessian matrix at the metastable state $x_0 = (000)$ as in Eq. (5), i.e., $x_s - x_0 = \sum_{k=1}^{3} y_k u_k$. The coefficients y_1, y_2, y_3 that provide a measure of the contribution of the normal modes to the thermal activation are shown in Fig. 4 as a function of the applied field in the case when the constant of magnetostatic coupling $\kappa = 0.3$. When the applied field is sufficiently close to the value at nucleation $H_a \rightarrow H_n \approx 1.43 H_k$, the thermal activation process can be described, in principle, by a single mode of the lowest frequency, i.e., the soft mode ($y_1 \neq 0, y_2, y_3 = 0$). This observation is consistent with the discussion given in Sec. II. The amplitudes y_2, y_3 vanish probably as a result of the uniaxial symmetry of the system. At the critical field h_{cr}, the reduction in the symmetry of the fanning mode of magnetic reversal ($\theta_1 \neq \theta_3$), is found to be associated with the onset of a finite amplitude for the contribution of the high-frequency modes of excitation y_2, y_3 to the position of the transition state. The change in y_2, y_3 at this critical point is small but discontinuous. A further reduction in the magnitude of the applied field is associated with an increase in activation energy E_b. The contribution of the high-frequency modes of excitation to the activation process as shown in Fig. 4 then becomes more significant, as has already been discussed in Sec. II.

Next we consider the situation when, for a given choice of the control parameters (κ, h), more than a single transition is possible. The current implementation of the ridge method provides the framework for a systematic analysis by evaluating the contributions to the separatrix of the system from the different local energy minima to which the system may relax, following the activation process. These energy minima may be metastable and transitions into new states (including the original energy minimum) are possible. The separatrix (see Sec. II) here consists of curves that divide the two-dimensional phase space of the control parameters k, h into a finite number of regions that represent structurally stable energy surfaces of different topology.

The separatrix was evaluated as follows. The ridge method was implemented a number of times using Eq. (2). The stable states associated with the saddle points were noted and the procedure repeated for different values of the parameters κ, h. It was therefore possible to verify that transitions to stable states x_1 with any number of moments reversed are possible in general, e.g., (π00), ($\pi\pi$0), (0π0), etc. The energy gradient at these states is zero. The set of values of the control parameters κ, h

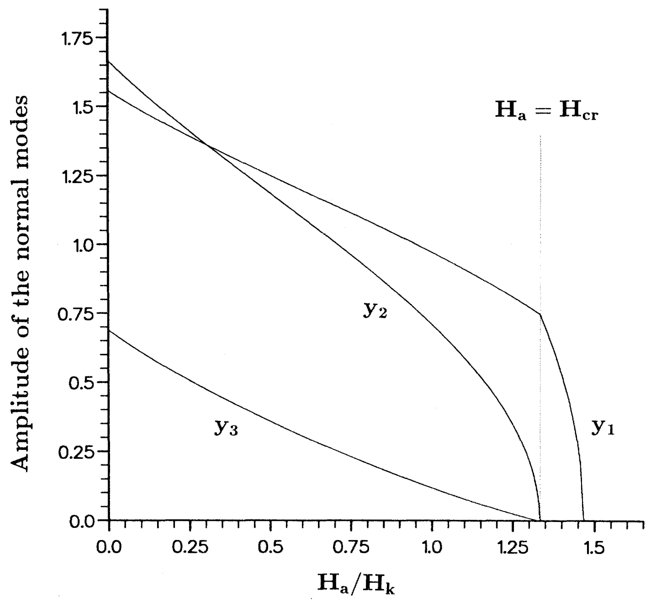

FIG. 4. The amplitudes of the normal modes of excitation about the metastable state (000), at the saddle point of the transition (y_1, y_2, y_3), are shown as a function of the reduced applied field $h = H_a / H_k$. The constant of magnetostatic coupling is $\kappa = 0.3$.

when the Hessian matrix $H(x_1)$ becomes positive definite was evaluated. These are the critical points in parameter space when the states x_1 become local energy minima. The contribution to the separatrix from the states (π00), ($\pi\pi$0), (0π0), (π0π) in the two-dimensional parameter space κ, h are the curves c, d, e, f shown in Fig. 5. The area within each curve represents the locus of all points in parameter space for which that state is stable and a transition possible. The curves for (π00) and ($\pi\pi$0) exhibit reflection symmetry with respect to the κ axis, and it can be easily verified that they represent the same physical process as the applied field changes sign $H_a \rightarrow -H_a$. The same argument also applies for the contributions from (0π0) and (π0π). In the same figure, the critical points associated with the nucleation field H_n are represented by curves a. The increase of H_n with coupling strength indicates that the magnetostatic coupling tends to resist the magnetic reversal (a phenomenon that is also associated with superferromagnetism[31]). Curve b represents the critical field h_{cr} associated with the reduction in the symmetry of the fanning mode. For any point within the area enclosed by curve b, the trajectories of the two particles at the end of the chain will be different in the thermoactivated process ($\theta_1 \neq \theta_3$).

Figure 5 provides, in general, information on the mode of magnetic reversal that is most likely (curve b) and the stable states where a transition is possible (curves c, d, e, f). The existence of a stable state, however, does not guarantee that a direct transition to that state is possible, i.e., that the two domains of attraction share a common boundary. A search for direct transitions was carried out as follows. The applied field was reduced in small discrete steps from the value at nucleation H_n. At each step the ridge method was implemented with stochastic initial directions of negative curvature using Eq. (2) (method 2 in Sec. III A). The first instance of a transition to a new stable state was recorded. The trajectory of the associated saddle point in coordinate space was then traced as the magnitude of the applied field was varied (method 1 in Sec. III A). Thus the range of values of the applied field for which a direct transition to that state is possible was determined.

The activation energy E_b for all direct transitions from the state (000) is shown in Figs. 6(a)–6(c) as a function of field for different values of coupling strength ($\kappa=0.1,0.3,0.4$). The dependence of the energy barrier of minimum height on the applied field can easily be deduced, as shown in Fig. 7. It is important to note that the additional transition states that are located using Eq. (2) cannot be found by an approach based on the soft mode alone. Thus the approach adopted allows a systematic study of the magnetization reversal mechanisms that are possible at a finite temperature.

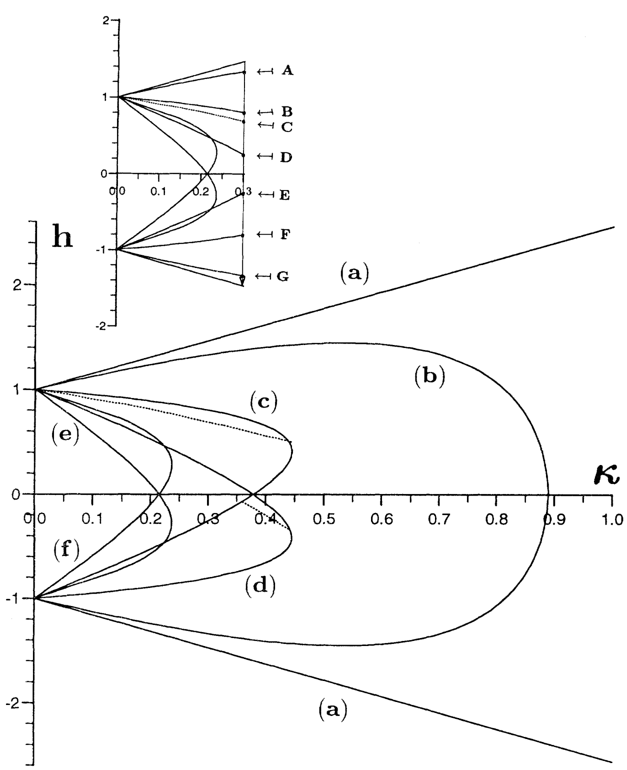

FIG. 5. Contributions to the separatrix in the space of the two control parameters κ, h. Curves a represent the intrinsic nucleation field H_n, curve b the reduction in the symmetry of the fanning mode of reversal, curves c,d,e,f the critical points when the states (π00), ($\pi\pi$0), (0π0), and (π0π) become metastable. The dashed curves do not belong to the separatrix and are associated with a change of the stable state following the activation process. The points $A, B, \ldots G$ associated with qualitative changes in the activation process are shown for clarity separately in the inset.

A direct transition to a particular state, as is evident from Figs. 5 and 6, appears to be possible only in a region of control parameter space that is a subset of the region where the state is stable. In Fig. 6 these regions are represented by "bands" that are, in general, disconnected. A transition to a new stable state is also possible for values of the applied field that do not belong to these bands, however, the transition in this case must be indirect through the domain of attraction of an intermediate state.

The set of metastable states where a direct transition is possible changes as the control parameters are varied. In the first type of change, a state that is directly accessible is replaced by another, the former being thereafter accessible only indirectly. For example, when the field is gradually reduced from the value at nucleation (point A in Fig. 6), the state ($\pi\pi\pi$) ceases to be accessible by direct transition at some point B, when $h=h_0$. The saddle point then becomes associated with a transition to the (π00) state. The noncritical point B is not part of the separatrix of the system and there is no change in the topology of the energy surface. Other examples include a change of the transition from the state ($\pi\pi\pi$) to (0π0) [point E in Fig. 6(a)] and from the state ($\pi\pi\pi$) to ($\pi\pi$0) [point E in Fig. 6(c)]. The locus of such noncritical points in control parameter space is represented by the dashed curves in Fig. 5.

A second type of change of the set of states where a transition is possible is illustrated by point C in Fig. 6(a). The state (π00) ceases to be a local minimum and the most likely transition is to the ($\pi\pi$0) state. A similar change from the state ($\pi\pi$0) to the state ($\pi\pi\pi$) occurs at the point H. A small change in the control parameters at such critical points results to activation through some different path of minimum energy to a new state. The energy barrier of lowest height therefore exhibits a discontinuous variation as is illustrated in Fig. 7.

A more complete account of the variations in the path of minimum energy occurring when the field is gradually reduced from the maximum value H_n can now be given. We consider the case $\kappa=0.3$, represented by an arrow in the inset of Fig. 5. The nucleation of magnetic reversal occurs initially by near-symmetric fanning to the state ($\pi\pi\pi$). The symmetry of the fanning mode is reduced at the critical point A. The state (π00) then becomes metastable (point B) and replaces the state ($\pi\pi\pi$) as the state where a direction transition is possible (point C). At this stage transitions to the state ($\pi\pi\pi$) are possible only indirectly, through the domain of attraction of the state (π00). Similarly the state ($\pi\pi$0) becomes metastable (D) but is not yet accessible directly [Fig. 6(b)]. The state (π00) ceases to be a minimum (E) and the only direct transition is to the state ($\pi\pi$0). The latter is similarly replaced by ($\pi\pi\pi$) (F).

The behavior for other values of the coupling contrast $\kappa<0.45$ is similar. The only difference is that for $0.36<\kappa<0.45$, it is the state ($\pi\pi\pi$) that replaces (π00) when the latter ceases to be a local minimum.

It is interesting that transitions to states such as (0π0) and $(\pi 0 \pi)$ whose contribution to the separatrix are curves enveloped by those of other transitions have relatively larger activation energies. The implication is that in extended systems with a large number of metastable states, the thermal activation may adequately be described by considering a relatively small number of transitions.

The temperature and time dependence of the switching field $H_{sw}(T)$ that causes the first irreversible response on average over a time interval t can be evaluated from Fig. 7, by setting $E_b = kT\ln(\nu t)$ [Eq. (1)] and assuming that the temperature remains low so that transitions other than the most probable can be ignored. The result is shown in Fig. 8 for different values of the coupling constant k. The curves can be fitted to a good approximation by an analytic expression of the form

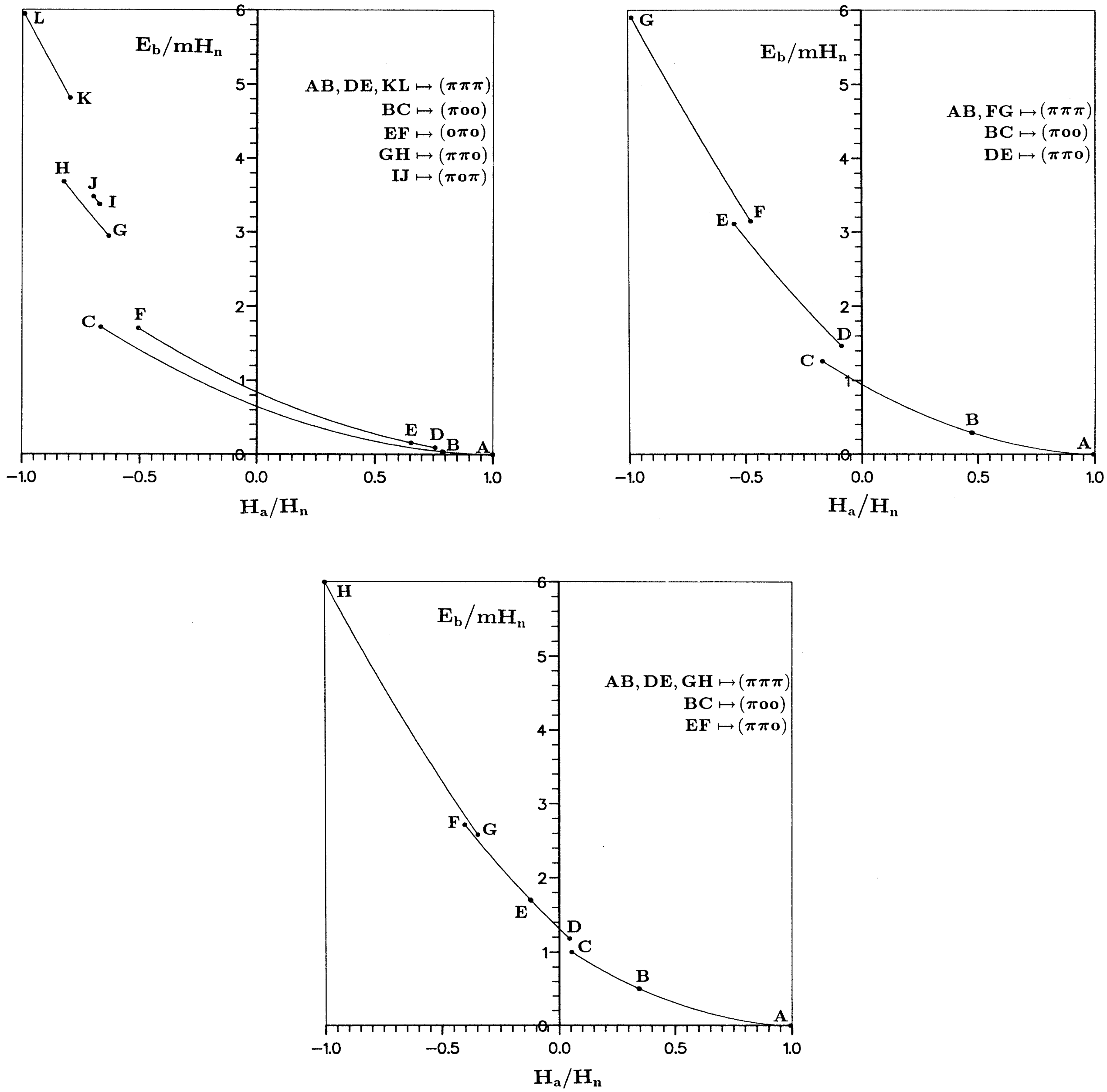

FIG. 6. The reduced energy barrier E_b/mH_n of all transitions from the state of perfect magnetic alignment (000), as a function of reduced applied field H_a/H_n. (a)–(c) were obtained with a coupling constant $\kappa = 0.1, 0.3, 0.4$, respectively. The states to which transitions occur are shown in each figure separately.

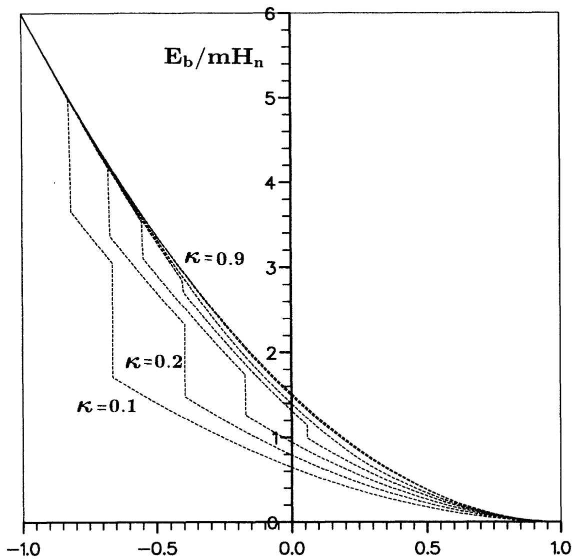

FIG. 7. The reduced energy barrier of lowest height E_b/mH_n for transitions from the state (000) as a function of the reduced applied field H_a/H_n. Results are shown for a range of values of the coupling constants $\kappa=0.1$, 0.2, 0.3, 0.4, 0.5, 0.7, 0.9.

$$kT\ln(\nu t)=C(k)\Delta H^2 , \tag{8}$$

where C is independent of the field and $\Delta H=H_{sw}(0)-H_{sw}(T)$. A model by Victora[32] predicts a $\Delta H^{3/2}$ dependence. The basic assumption of that model, however, that the third derivative of the energy with respect to the normal modes does not vanish at the critical point when $H_a=H_{sw}(0)=H_n$ is not satisfied here because of the uniaxial symmetry of the system.

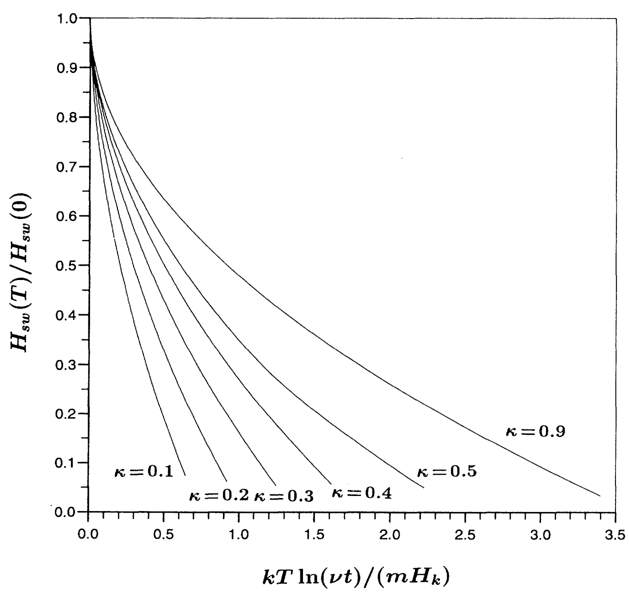

FIG. 8. The temperature and time dependence of the switching field H_{sw} resulting in the first irreversible magnetic change. Results are presented for different values of the coupling constant k.

C. The effect of particle anisotropy on magnetic reversal

The system studied in the present treatment (Fig. 2) is similar to the "chain-of-spheres" model of Jacobs and Bean.[30] In a chain of spheres, however, the particles are assumed to have no anisotropy ($H_k=0$), so the nucleation field of the chain is determined from the magnetostatic interaction. The behavior of a chain of spheres is obtained in our model in the limit of strong coupling $\kappa=m/(H_k r^3)\rightarrow\infty$. A comparison between the two models should therefore provide information on the effect of particle anisotropy.

Jacobs and Bean[30] only considered the spontaneous magnetic reversal when the energy barrier vanishes $E_b=0$. For chains of n spheres, where $n\geq 3$, the moments were found to reverse by asymmetric fanning. In the case $n=3$, however, symmetric fanning ($\theta_1=-\theta_2=\theta_3$) turned out to be a good approximation. Assuming reversal by symmetric fanning, analytic expressions can be obtained for the nucleation field and energy barrier that include the effect of particle anisotropy:

$$H_n=H_k+\frac{m}{r^3}[6K_n-4L_n] , \tag{9}$$

$$E_b=\frac{nmH_n}{2}\left[1-\frac{H}{H_n}\right]^2 , \tag{10}$$

where

$$K_n=\sum_{j=1}^{n}\frac{n-j}{nj^3} , \quad L_n=\sum_{j=1}^{(n-1)/2<j<(n+1)/2}\frac{n-(2j-1)}{n(2j-1)^3} . \tag{11}$$

For a chain of three particles ($n=3$), Eq. (9) reduces to

$$\frac{H_n}{H_k}=1+\frac{19}{12}\kappa . \tag{12}$$

The result of a precise calculation, shown by curve a in Fig. 5, indicates that the symmetric fanning approximation [Eq. (12)] is satisfactory for all values of κ. The particle anisotropy does not modify the magnetic reversal mode (by near-symmetric fanning) in this case ($E_b=0$).

We consider next the case when magnetic reversal is by thermal activation ($E_b>0$). For a chain of spheres ($\kappa\rightarrow\infty$), the superposition of the curves in Fig. 7 is in good agreement with Eq. (9) and indicates that the mode of activation is well approximated by symmetric fanning. Comparison with data obtained for low values of κ, in Figs. 3(b), 3(c), 5, and 7 indicates that the particle anisotropy can modify the mode of activation by reducing its symmetry and by allowing activation to new stable configurations.

IV. DISCUSSION

The thermally assisted magnetic reversal in a micromagnetic system of three interacting ferromagnetic particles in the low-temperature limit was studied by application of the ridge optimization method. The magnetic reversal occurs in general by asymmetric fanning of the moments. The remaining symmetry of the fanning mechanism was shown to be broken at critical values of the field and coupling strength (the control parameters) by the presence of the particle anisotropy which also allows transitions to new stable configurations. A direct or indirect transition is only possible in the space of control parameters defined by the separatrix of the system. The final state following the activation process may change at a noncritical point that constitutes the boundary of a direct and indirect transition. When the applied field is sufficiently close to the value at nucleation, the activation process involves primarily the soft normal mode of excitation of the system and has the lowest activation energy. If the applied field is reduced in magnitude, a discontinuous change in the value of the minimum activation energy and the associated mode of activation was observed at some critical point, when the final state, following the thermal activation, ceases to be metastable.

The current implementation of the ridge method uses the normal modes of excitation about the metastable point to restrict effectively the search for transition states to a relatively small section of the phase space of all possible directions. This technique may be of advantage in the study of more complex micromagnetic systems when the external applied field is sufficiently close to the switching field. For instance, it is useful to apply the method to evaluate exact values of the barrier heights in the magnetic reversal of fine particles of irregular morphology. The detailed geometry of the particles determines to a large extent their viscous properties[17] that are of importance in applications such as magnetic recording.

The derivation of the energy barrier dependence on applied field using the ridge method is useful for the analysis of experimental data, for example, the dependence derived by measurement of the waiting time for a magnetization jump to occur[33] and of the statistical distribution of the field that must be applied before a jump is observed.[33] Exact values for the activation volume V_{act} of the particles can also be evaluated numerically using the expression derived by Gaunt[35] [$V_{\mathrm{act}}=(1/M_s)dE_b/dH_a$]. These can be compared with the experimental values[37] derived from measurement of the magnetic viscosity S and the irreversible susceptibility χ_{irr} using the relation $V_{\mathrm{act}}=kT\chi_{\mathrm{irr}}/(M_s)S$,[38] that relates the static and dynamic properties of the system. The observation in our model system of a discontinuous variation of barrier height with applied field implies the divergence of the activation volume. Gaunt's theory,[35] however, considers only the transitions from a single stable state in a bistable system, whereas the observed discontinuity in our model system occurs only when the final state becomes metastable. It is evident that the probability of a transition back to the initial state and to other stable states must also be considered, so that a modification in the theory[35] is required to determine unambiguously the physical implications in this case.

A common difficulty in micromagnetic studies of hysteresis[1,36] is the determination of the precise value of the field that causes an irreversible transition. The main reason is the symmetry of the systems studied that prevents magnetic reversal in the absence of any perturbation from equilibrium. The equilibrium state has therefore been tested by application of some intuitive choice of a perturbation, for instance adding a helicity[1] to the magnetization configuration or applying a small transverse field.[36] The results of such computations may be dependent on the form chosen for those perturbations, so it is not surprising when small differences are observed.[36] The precise value of the switching field, however, is dependent on the thermal fluctuations of the system[16] that provide a natural mechanism of perturbing the equilibrium state. For this reason, the introduction of thermal agitation may help to harmonize the results of micromagnetic studies and allow comparisons to be made.

Our model system is probably rather simple to represent a real system, although the fanning mode of reversal[30] remains of interest[15,17,39,40] in theoretical studies of the switching field of elongated fine particles. Some of the observations in our study, however, are probably also valid for more complex systems. For instance, the modes of thermal activation, despite being collective, may result in the magnetic reversal of only a small part of the system. The implication is that the switching field H_{sw} when the first irreversible magnetic change occurs may be different than the coercive field H_c (the field that makes the magnetization vanish). The magnetization reversal in this case does not occur in a single jump. Measurement of the hysteresis loop of a single barium ferrite crystal[33] has shown that the reversal occurs in a two-stage process. An analogous phenomenon is the transition from an anticurling to a curling state in ferromagnetic cubes.[16,34] The calculation of the separatrix of the energy surface may be useful to determine the conditions when such complications arise.

The possibility of existence of many stable structures implies that in a general treatment of the hysteresis behavior, it is necessary to consider the finite probability of transition to many different states from a given metastable state. These complications do not arise at zero temperature where there is always a single transition that is possible that can be evaluated by standard micromagnetic methods.[1] For our model system that transition is to the state of negative saturation. It is evident that, in principle, the mode of magnetic reversal and the final state may be different at finite temperatures. The implication is that the mode of magnetic reversal that is determined by application of the nucleation theory[27] is not necessarily the only mode possible and different modes may be activated at finite temperatures. In the simplest case, when the separation in the barrier height of different reversal modes is sufficiently large, only the mode of maximum likelihood is at any stage active so that magnetic reversal proceeds by a well defined sequence of jumps, i.e., the size of the jumps does not exhibit a statistical distribution (as in Fig. 1 of Ref. 33). It would be of interest to examine the possibility that the existence of distinct activation processes with different barrier heights may provide an interpretation to the recent observation[41] that the reversal probability of a single permalloy particle cannot be described by a unique relaxation time.

ACKNOWLEDGMENT

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