Thermally Activated Magnetization Reversal in Elongated Ferromagnetic Particles

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Elongated ferromagnetic single-domain particles are believed to exhibit a high stability of their remanent magnetization. However, here it is shown that for elongated particles thermal fluctuations lower the coercivity much more drastically than predicted by current theories. The rate for magnetization reversal is calculated for a classical model of a ferromagnet that allows for a spatially nonuniform magnetization distribution along the sample. The prefactor of the Arrhenius factor is explicitly evaluated and analytical results are obtained in the experimentally important limit of external magnetic fields close to the anisotropy field and for moderate damping.

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Thermally activated magnetization reversal in ferromagnetic single-domain particles has widespread implications ranging from rock magnetism [1] to the limits of storage density in magnetic media [2]. This field has been neglected theoretically, while the closely related topic of macroscopic quantum tunneling sees a rapidly growing interest both experimentally [3] and theoretically [4].

Small single-domain particles typically exhibit two equivalent equilibrium configurations which are separated by an anisotropy barrier. Applying an external magnetic field antiparallel to the magnetization, this barrier height is reduced and, for small enough particles, thermal fluctuations eventually lead to a magnetization reversal.

A complete theory for this effect is due to Néel [1] and Brown [5] and has recently been extended [6] to include the regime of small dissipation. This theory is based on the picture that the magnetization of the particle acts rigidly and hence the energy barrier corresponds to a magnetization configuration which is uniform over the sample. The Arrhenius factor therefore leads to an exponential suppression of thermal effects in the particle volume. While this picture is adequate for small particles with moderate aspect ratios, it fails to describe elongated particles: Considering the conceptual limit of an infinitely long cylindrical particle of arbitrarily small cross section, the theory predicts a complete suppression of thermal effects. However, it is intuitively clear that the formation of a spatially localized "nucleus" leads to a finite energy barrier and thus to a finite reversal probability (cf. Fig. 1). In fact, experiments tend to show a lower coercivity than theoretically predicted. This has been explained within the Néel-Brown theory by the *ad hoc* introduction of an "activation volume" which is smaller than the actual particle volume. However, this concept ignores the exchange energy which arises at the boundary of the activation volume. Recent theoretical efforts have then focused on a discussion of different candidates for spatially nonuniform saddle points [7] in bulk ferromagnets or in small spheres [8]. However, the evaluation of the switching rate from first principles is an important unsolved problem.

It is the purpose of this Letter to provide for the first time a rigorous theory of the magnetization reversal rate in an elongated, defect free particle via a spatially nonuniform reversal mode. Starting from a model energy density, a nonuniform saddle point structure is derived that has to be overcome during magnetization reversal. In contrast to the Néel-Brown treatment, the energy barrier is shown to be proportional to the cross-sectional area rather than the volume of the particle. The reversal rate is then calculated by solving the Fokker-Planck equation, which describes the stochastic magnetization dynamics near the nonuniform saddle point. In the experimentally important limit of an external field close to the anisotropy field the rate is evaluated in closed form. The resulting coercivity is shown to be considerably lower than that of the Néel-Brown theory. Furthermore, the angular dependence of the coercivity is shown to be asymmetric unlike the Stoner-Wohlfarth theory. Both of these results are in qualitative agreement with recent measurements on a single elongated particle [9].

The present treatment is based on the statistical mechanical theory [10] of the decay of a metastable state. It has been used [11] to describe the decay of supercurrents in thin wires as well as the propagation of dislocations. In contrast to those treatments which focused on overdamped situations, we consider the case of moderate

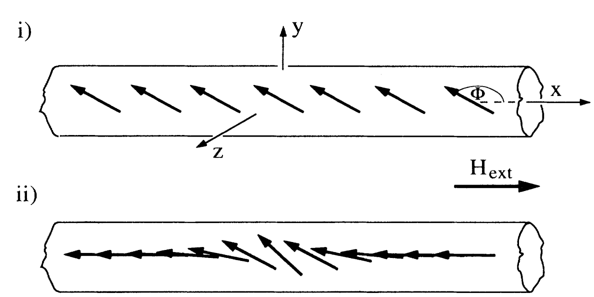

FIG. 1. Nuclei for (i) uniform (Néel-Brown) and (ii) nonuniform [Eq. (3) for $R = 0.3$] magnetization reversal.

damping, since dissipation in magnetic systems is small.

In order to keep the problem theoretically tractable, I shall consider a sample of constant cross section $\mathcal{A}$. The length L should be considerably larger than a domain wall width. The particle diameter is assumed to be small enough in order to justify the assumption of transversal symmetry over the sample cross section [12]. I focus here on bulk properties and discard any effects occurring at the particle ends which depend in a subtle way on the sample shape. The energy per unit area of a magnetization configuration $\mathbf{M} = \mathbf{M}(x,t)$ of constant magnitude $|\mathbf{M}| = M_0$ is then given by

$$\mathcal{E} = \int_{-L/2}^{L/2} dx \left\{ \frac{A}{M_0^2} \left[(\partial_x M_x)^2 + (\partial_x M_y)^2 + (\partial_x M_z)^2\right] + \frac{K_h}{M_0^2} M_z^2 - \frac{K_e}{M_0^2} M_x^2 - H_{\text{ext}} M_x \right\}, \tag{1}$$

where $\partial_x = \partial/\partial x$. The first term in (1) is the classical counterpart of the exchange interaction characterized by the exchange constant A. The second term is an easy axis anisotropy directed along the particle axis. The third term is a hard axis anisotropy which breaks the rotational symmetry around the easy axis. This hard axis might arise from an easy axis that is misaligned with the particle axis. K_e, $K_h > 0$ are anisotropy constants including contributions of both crystalline and shape anisotropies. The last term describes the coupling to the external magnetic field. Note that the model is exact if crystalline anisotropies dominate over demagnetizing contributions. For purely demagnetizing induced anisotropy it becomes exact in the limit of configurations that vary slowly compared to the particle diameter. The anisotropy constants in (1) are then related to the components of the demagnetizing tensor.

The dynamics of the magnetization is assumed to obey the dissipative Landau-Lifshitz-Gilbert equations (see, e.g., [2]) which can be rewritten as

$$(1+\alpha^2)\partial_t \mathbf{M} = -\gamma \mathbf{M} \times \mathbf{H}_{\text{eff}} - \frac{\alpha\gamma}{M_0} \mathbf{M} \times [\mathbf{M} \times \mathbf{H}_{\text{eff}}], \tag{2}$$

where $\gamma > 0$ denotes the gyromagnetic ratio and α is a dimensionless damping constant. The first term on the right-hand side describes the conservative precession in the effective field $\mathbf{H}_{\text{eff}} \equiv -\delta\mathcal{E}/\delta\mathbf{M}$ while the second term describes the dissipative relaxation of the magnetization towards the effective field.

It is convenient to use dimensionless units defined by $[x] = \sqrt{A/K_e}$, $[\mathcal{E}] = 2\sqrt{AK_e}$, and $[t] = M_0(1+\alpha^2)/2\gamma K_e$ corresponding to the domain wall width, half the domain wall energy per area, and the precession period in the anisotropy field, respectively. The reduced external field is given by $h = H_{\text{ext}} M_0 / 2K_e$ and the dimensionless ratio of the anisotropy constants is denoted by $Q = K_e/K_h$. To incorporate the constraint $|\mathbf{M}| = M_0$ we write $\mathbf{M} = M_0(\sin\theta\cos\phi, \sin\theta\sin\phi, \cos\theta)$.

The energy density (1) exhibits two minima which correspond to a spatially uniform magnetization configuration. The metastable state ($\phi_m = \pi, \theta_m = \pi/2$) and the stable state ($\phi_0 = 0, \theta_0 = \pi/2$) are directed antiparallel and parallel to the external magnetic field, respectively.

For external fields smaller than the anisotropy field, the Euler-Lagrange equations of (1) have a nonuniform solution which describes a localized excursion from the metastable state. Expressed in spherical coordinates and assuming $L/\sqrt{A/K_e} \gg 1$, this nucleus reads

$$\phi_s = 2 \arctan \left(\frac{\cosh\left(\frac{x-x_0}{\delta}\right)}{\sinh R} \right), \quad \theta_s = \pi/2, \tag{3}$$

which is formally equivalent to the planar saddle-point solution [7] in bulk ferromagnets. The nucleus (3) may also be viewed as a superposition of two π domain walls with opposite sense of twist which are centered at $x - x_0 = \pm R\delta$. R and δ are related to the external field as

$$\operatorname{sech}^2 R = h, \quad \delta = \coth R. \tag{4}$$

In the sequel, it is useful to use R instead of h as an independent parameter. The degeneracy of (3) with respect to x_0 gives rise to a zero mode in the excitation spectrum of the nucleus. For convenience, we shall put $x_0 = 0$ in the sequel. For $h \to 1$, i.e., close to the anisotropy field, the nucleus (3) represents a slight deviation from the uniform metastable state whereas for $h \to 0$ the nucleus consists of an already reversed domain which is delimited by two π-domain walls. The energy per area of the structure (3) with respect to the metastable state is the sum of deformation and Zeeman energy, respectively,

$$\mathcal{E}_s = 4\tanh R - 4R\operatorname{sech}^2 R. \tag{5}$$

To ensure that Eq. (3) is indeed a physically relevant nucleus for magnetization reversal for elongated particles, I shall show that this structure has exactly one unstable mode and that once this configuration is reached, the magnetization may be reversed without further expense in energy. Performing a linearization around the nucleus, i.e., $\phi(x,t) = \phi_s(x) + \varphi(x,t)$, $\theta(x,t) = \pi/2 - p(x,t)$ with φ, p small, an expansion of the energy per area up to second order yields

$$\mathcal{E}^{(2)} = \mathcal{E}_s + \frac{1}{2}\int_{-L/2}^{L/2} dx\, \varphi \mathcal{H}^{s\varphi} \varphi + \frac{1}{2}\int_{-L/2}^{L/2} dx\, p \mathcal{H}^{sp} p. \tag{6}$$

The operators in (6) are for large L given by

$$\mathcal{H}^{s\varphi} = -\frac{d^2}{dx^2} + \delta^{-2} V_-\left(\frac{x}{\delta}, R\right), \tag{7}$$

$$\mathcal{H}^{sp} = -\frac{d^2}{dx^2} + \delta^{-2} V_+\left(\frac{x}{\delta}, R\right) + Q^{-1}, \tag{8}$$

with the potentials

$$V_\pm(\xi, R) = 1 - 2\operatorname{sech}^2(\xi + R) - 2\operatorname{sech}^2(\xi - R) \pm 2\operatorname{sech}(\xi + R)\operatorname{sech}(\xi - R). \tag{9}$$

The same operators surprisingly also occur [13] in the description of fluctuations around a 2π Bloch wall [14]. The corresponding eigenvalue problems are denoted by $\mathcal{H}^{s\varphi}\chi_n^{s\varphi} = E_n^{s\varphi}\chi_n^{s\varphi}$ and $\mathcal{H}^{sp}\chi_n^{sp} = E_n^{sp}\chi_n^{sp}$. The index n denotes either bound states which describe localized deformations of the nucleus or spin wave excitations.

In order to determine the unstable modes of (3), we employ the zero modes of the nucleus and the 2π Bloch wall. The ground state $\chi_0^{sp} \propto \text{sech}(x/\delta - R) + \text{sech}(x/\delta + R)$ has energy $Q^{-1} > 0$, and $\chi_1^{s\varphi} \propto d\phi_s/dx \propto \text{sech}(x/\delta - R) - \text{sech}(x/\delta + R)$ has zero energy. Since the latter is antisymmetric with one node [a consequence of the opposite relative sense of twist of the two domain walls in (3)], there is one negative eigenvalue of $\mathcal{H}^{s\varphi}$. Since χ_0^{sp} is nodeless, I conclude that there is *exactly one unstable mode* of the nucleus, i.e., $(\varphi, p) = (\chi_0^{s\varphi}, 0)$.

An explicit sequence of configurations which connects the metastable with the stable state via the saddle point is obtained by (3) for fixed $\delta = \coth R_0$ ($h = \text{sech}^2 R_0$ is the value of the external field) and arbitrary R. The energy $\mathcal{E} = 4\tanh R - 4R\,\text{sech}^2 R_0$ monotonically increases with R until the saddle point is reached at $R = R_0$ and then decreases monotonically until the magnetization is reversed. Our proposed mechanism of magnetization reversal is therefore not obstructed by the existence of intermediate energy barriers and it leads over the lowest barrier of the model (1). Note that the above properties have not been shown for any other proposed mechanism of nonuniform magnetization reversal.

We are now in a position to calculate the rate of magnetization reversal within the framework of the statistical mechanical theory of the decay of metastable states [10]. The deterministic dynamics (2) of the magnetization is near the saddle point (3) given by

$$\partial_t\varphi = -\mathcal{H}^{sp}p - \alpha\mathcal{H}^{s\varphi}\varphi, \quad \partial_t p = \mathcal{H}^{s\varphi}\varphi - \alpha\mathcal{H}^{sp}p, \tag{10}$$

with φ, p defined as above. Since Eqs. (2) and (10) do not contain random forces and thus are ultimately not consistent with the fluctuation-dissipation theorem, they are not able to describe the thermally induced formation of critical nuclei. Within a Langevin description, random forces can be included by fluctuating magnetic fields [5] in (2) and (10), respectively. Here, I choose the equivalent Fokker-Planck description [15] in which the time dependence of φ and p is shifted to the probability functional ϱ. Assuming white noise [5,6], the dynamics of ϱ in the vicinity of the saddle point is given as follows:

$$\partial_t\varrho = -\int dx \sum_i \frac{\delta J_i}{\delta\psi_i(x)}. \tag{11}$$

The right-hand side of (11) is a functional divergence of the probability current

$$J_i = -\sum_j M_{ij}\left[\mathcal{H}_j\psi_j(x) + \frac{1}{\beta\mathcal{A}}\frac{\delta}{\delta\psi_j(x)}\right]\varrho. \tag{12}$$

Here a compact notation has been used where $\psi(x) = (\varphi(x), p(x))$ and $(\mathcal{H}_1, \mathcal{H}_2) \equiv (\mathcal{H}^{s\varphi}, \mathcal{H}^{sp})$. The matrix M is defined by $M_{11} = M_{22} = \alpha$, $M_{12} = -M_{12} = 1$, and $\beta^{-1} = k_B T$. The Fokker-Planck equation (11), (12) is constructed in such a way that (i) $\varrho_{\text{eq}} \propto \exp(-\beta\mathcal{A}\mathcal{E}^{(2)})$ with $\mathcal{E}^{(2)}$ as in (6) a currentless equilibrium distribution and (ii) the thermal expectation values $\langle\varphi\rangle$ and $\langle p\rangle$ obey the deterministic equations (10). Integrating J_i transverse to the unstable mode [10] (leaving details to Ref. [16]), I obtain the switching rate

$$\Gamma = \lambda_+\mathcal{L}\sqrt{\frac{\beta\mathcal{A}}{2\pi^3}}\sqrt{\frac{\det\mathcal{H}^{m\varphi}}{\det'|\mathcal{H}^{s\varphi}|}}\sqrt{\frac{\det\mathcal{H}^{mp}}{\det\mathcal{H}^{sp}}}e^{-\beta\mathcal{A}\mathcal{E}_s}. \tag{13}$$

Equation (13) separates into a prefactor and the Arrhenius factor. The prefactor includes a factor of 2 due to the existence of two equivalent saddle points (ϕ_s and $-\phi_s$). The barrier energy $\mathcal{A}\mathcal{E}_s$ is assumed to be much larger than $k_B T$ and is given by (5). The dynamics of the system only enters in the form of the nucleus decay frequency λ_+ which is defined as the positive eigenvalue of the right-hand side of (10). The factor $\mathcal{L} = \sqrt{\mathcal{E}_s}L$ arises from the integration over the zero mode expressing the fact that the nucleus may appear anywhere on the long axis of the sample. $\mathcal{H}^{s\varphi}$ and $\mathcal{H}^{sp}$ are given by (7), (8) and the operators $\mathcal{H}^{m\varphi} = -d^2/dx^2 + \delta^{-2}$, $\mathcal{H}^{mp} = \mathcal{H}^{m\varphi} + Q^{-1}$ govern the fluctuations around the uniform metastable state. "det" or "det||" denotes the product of all eigenvalues or their modulus, respectively. The prime indicates omission of the eigenvalue zero. The prefactor is evaluated [16] by means of the Jacobi method [17] or via scattering phase shifts. The results reveal that the prefactor varies by 3 orders of magnitude between external fields $h = 0.1$ and $h = 0.9$. This is in sharp contrast to the Néel-Brown case, where the prefactor is almost constant, a fact which gave rise to the notion of a constant "attempt frequency." I focus on two interesting limiting cases.

For large hard-axis anisotropy, the "mass" Q^{-1} leads to a suppression of out of easy-plane fluctuations and consequently the ratio of the p determinants equals 1. In this limit and for large α, the rate is given by

$$\Gamma = \alpha|E_0^{s\varphi}|\frac{4}{\pi^{3/2}}L\sqrt{\beta\mathcal{A}}\,\tanh^{3/2}R\,\sinh R\,e^{-\beta\mathcal{A}\mathcal{E}_s}, \tag{14}$$

where $E_0^{s\varphi}$ is the ground state energy of $\mathcal{H}^{s\varphi}$. This result also describes the creation rate of a kink-antikink pair in the overdamped double sine-Gordon model.

Experimentally important is the limit of an external field close to $h = 1$, i.e., small R. For $\alpha \ll R \ll 1$, the switching rate (13) is given by

$$\Gamma = L\sqrt{\beta\mathcal{A}}\frac{4}{\pi^{3/2}}\sqrt{3Q^{-1}}R^{7/2}e^{-\beta\mathcal{A}\mathcal{E}_s}, \tag{15}$$

and for $R \ll \alpha \ll 1$ we have

$$\Gamma = L\sqrt{\beta\mathcal{A}}\frac{12}{\pi^{3/2}}\left(\alpha + \frac{1}{\alpha}\right)R^{9/2}e^{-\beta\mathcal{A}\mathcal{E}_s}. \tag{16}$$

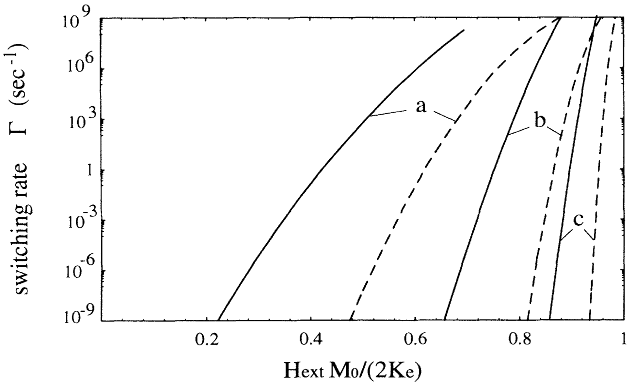

FIG. 2. The total switching rate is shown as a function of the external field h for particle diameters of (a) 100 Å, (b) 200 Å, and (c) 400 Å. For comparison, the dashed lines indicate the results of the Néel-Brown theory for an assumed particle aspect ratio of 1:15.

Note that these results are expressed in dimensionless units. Reinstating units, we recognize that for small damping the prefactor becomes independent of α and merges into the "transition state" value [10]. For large α, the prefactor becomes proportional to α^{-1} in accordance with other overdamped theories [10]. Note that the present theory relies on the concept of diffusion in configuration rather than energy space. It is therefore not applicable to arbitrarily small values of α. To ensure the applicability of the present theory, the energy loss per (approximate) period near the saddle point should exceed $k_B T$. For small R, I obtain (in dimensionless units) the following condition for the validity of (13)

$$8\pi Q^{-1/2}\beta\mathcal{A}\alpha R^2 > 1. \qquad (17)$$

For quantitative estimates, I assume the following material parameters of a typical magnetic recording particle $A = 5\times10^{-7}$ erg/cm, $K_e = 7\times10^5$ erg/cm^3, $M_0 = 480$ Oe, $\gamma = 1.5\times10^7$ Oe^{-1}s^{-1}. For $T = 300$ K, $Q^{-1} = 0.2$, $R = 0.2$, and $\alpha = 0.05$, the condition (17) is satisfied down to particle diameters of 70 Å.

With these parameters, the switching rate Γ (13) is shown in Fig. 2 for various sample diameters. For arbitrary values of h, the prefactor has been evaluated numerically. The result is compared with the Néel-Brown theory as in [6] for an assumed aspect ratio of 1:15 which is characteristic of such particles as CrO_2. Note that the coercivity for the 100 Å particle can be as low as 1/3 that of the Stoner-Wohlfarth value $H_{\rm ext}M_0/2K_e = 1$.

As will be discussed in detail elsewhere, an external field applied perpendicular to the sample will only allow for a spatially uniform barrier. Therefore the present theory predicts, as opposed to current theories, an asymmetry in the angular dependence of the switching field. In addition, the coercivity reduction from the Stoner-Wohlfarth value is maximal for fields which are directed along or perpendicular to the particle. Both properties are in agreement with recent experiments [9] on an isolated γ-Fe_2O_3 particle.

In conclusion, I have shown that in elongated particles, a nonuniform saddle point yields a considerable lower coercivity than current theories. Consequently one would also expect a corresponding lowering of the blocking temperature, a fact which is important in geophysical applications.

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