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Role of thermodynamic fluctuations in magnetic recording (invited)

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The thermal stability of the information stored in magnetic recording media is determined by a complex hierarchy. The leading consideration is the static or zero-temperature magnetization reversal complemented by the intrinsic temperature dependence of the micromagnetic parameters. Thermally activated Arrhenius (or Néel-Brown) processes modify the reversal by realizing paths close to static reversal, whereas “giant fluctuations” corresponding to reversal fields much higher than the nucleation field can safely be excluded. Thermally activated reversal in very thin elongated nanoparticles limits the thermal stability of magnetic recording media but degenerates into coherent rotation as the temperature is lowered, thereby reconciling micromagnetism and thermodynamics. A particularly complicated situation is encountered in alloys, where sublattices containing heavy transition-metal atoms act like earthquakes that modify the energy landscape.

I. INTRODUCTION

Thermally activated magnetization reversal is a key consideration in ultrahigh-density magnetic recording, because the ever-decreasing bit and particle size facilitate thermally activated magnetization reversal.1–3 In a simple picture, the magnetization of particles of volume \( V \) and uniaxial anisotropy \( K_1 \) is stabilized by an energy barrier \( K_1V \), and when \( V \) is very small, then the room-temperature magnetization direction becomes unstable due to thermal activation. The decay of the magnetization is a manifestation of the magnetic aftereffect, which was discovered as early as 1889.4

If \( K_1V \) was the only consideration, then one could achieve virtually diverging areal densities by using long cylinders of volume \( V = \pi R^2L \), where \( R \) and \( h \) are the radius and the length (height) of the cylinders. The closely related and exactly solvable micromagnetic case of long ellipsoids of revolution5 indicates that the coercivity remains finite as \( R \) approaches zero. In a strict sense, this finding is limited to zero temperature, but analyzing the involved Boltzmann factors reveals that the same is true for nonzero but low temperatures.6

However, the finite coercivity predicted by micromagnetic analysis is at odds with the findings of statistical mechanics, which predicts the absence of ferromagnetism in one-dimensional magnets, including infinite wires.7,8 The situation is complicated by the well-known experimental findings that magnetization reversal in nanowires involves volumes much smaller than the wire volume9 and that coercivities are often much lower than predicted from the above-mentioned micromagnetic nucleation modes. Micromagnetism explains the low coercivities and small activation volumes by structural imperfections,5,6 whereas the finite-temperature approach assumes thermal activation.7,8 If the thermal mechanism was the main consideration, then the finite-temperature behavior would be quite different from the zero-temperature limit. This is not observed—typical activation volumes and the coercivities remain small at zero temperature and can be traced to structural imperfections.

The aim of this paper is to reconcile the seemingly contradictory thermodynamic and micromagnetic approaches and to elaborate how magnetization reversal is realized in magnetic particles of interest in magnetic recording. We will see and analyze that the reversal obeys a complicated hierarchy. First, static magnetization processes are largely determined by imperfections and dominate at low temperature. As a crude rule, static magnetization reversal in particles smaller than 5 nm is coherent, whereas larger particles tend to reverse incoherently, by a variety of different mechanisms (Sec. II). Second, the intrinsic temperature dependences of micromagnetic parameters, such as the spontaneous magnetization \( M_s \) and anisotropy \( K_1 \), modify the static behavior (Sec. III). Third, there are corrections due to Arrhenius- or Néel-Brown-type thermal activation over static energy barriers (Sec. IV). Fourth, “giant” thermodynamic fluctuations may lead to reversal paths other than that corresponding to static reversal, or modify the energy landscape.

Throughout the paper, emphasis is on temperatures significantly below the Curie temperature \( T_C \), excluding critical fluctuations and very fast phenomena.10,11 The latter are important for heat-assisted magnetic recording but irrelevant for the long-term stability of stored information.

II. STATIC REVERSAL

Finite-temperature magnetization reversal is almost always based on static magnetization reversal, which implies the vanishing of a metastable energy minimum in a reverse magnetic field. This section summarizes typical mechanisms. A widely known coercivity mechanism is the Stoner-Wohlfarth (SW) reversal or coherent rotation, but there are other well-investigated and important mechanisms. A key
distinction is between nucleation and pinning (Fig. 1). Nucleation is defined as the instability of a magnetization state in a reverse field. Figure 2 illustrates that there are three basic types of nucleation, (a) coherent rotation, (b) curling, and (c) localized nucleation. For coherent rotation and curling, the respective nucleation fields (coercivities) are

\[ H_c = \frac{2K_1}{\mu_0 M_s} + \frac{1}{2} (3D - 1) M_s \]

and

\[ H_c = \frac{2K_1}{\mu_0 M_s} - DM_s + \frac{c(D)A}{\mu_0 M_s R^2}. \]

In the latter equation, \( c \) is 8.666 for spheres \( (D=1/3) \) and 6.678 for needles \( (D=0) \), whereas the hemisphere model of Fig. 2 is described by \( D=1/3 \) and \( c=8.13 \).

Curling costs exchange energy but is magnetostatically favorable due to vortexlike flux closure. In perfect ellipsoids of revolution, there is a transition from coherent rotation to curling for radii larger than about 10 nm. This transition is independent of the anisotropy and unrelated to the single-domain character of the magnet—many or most particles in permanent magnetism and high-density magnetic recording are single domain but reverse incoherently, with a relatively low coercivity. \( \Phi \) Aside from curling effects, this reflects localized nucleation due to “soft” imperfections. An exception are particles smaller than about 5 nm, where reversal is coherent (Stoner-Wohlfarth-like), irrespective of the presence of imperfections and grain boundaries.

Pinning means that imperfections impede the motion of preexisting domain walls and dominates the behavior in strongly disordered magnets. Weak pinning is frequently encountered in soft magnetic materials and refers to the trapping of a wall by ensembles of many pinning centers, whereas strong pinning is realized by a few relatively strong defects, as in Fig. 1. Depending on the domain-wall curvature, one encounters Kersten pinning, \( \Phi \) where the coercivity

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blattice exchange $J'$ and generally more pronounced than the temperature dependence of the magnetization. Simple ferromagnets, such as Fe and Co, are well described by the Callen and and focus on relevant degrees of freedom, such as domain-$this is neither practical nor necessary.

The Callen and Callen approach is a poor approximation for alloys, because it relates the anisotropy to the net magnetization, rather than taking into account that the main anisotropy contribution comes from heavy atoms (4d, 5d, or 4f) electrons, whereas the magnetization is largely due to the 3d elements. For example, rare-earth anisotropy reflects the electrostatic crystal-field interaction of the aspherical 4f charge clouds. Thermal intramultiplet excitations in-plane anisotropy decays as $\propto P_s t$ for the probability $P_s t$ of the 4$^4$s/\H$_{20849}$ and $m=10$ (cubic).

In L$^1_0$ magnets and $m=1$ for actinide compounds. In L$^1_0$ magnets, the temperature dependence of the anisotropy is linked to the collapse of the 4d/5d moment. In all cases, the anisotropy is of the single ion type, as contrasted to Néel-type pair anisotropy.

IV. MAGNETIZATION DYNAMICS

A key aim of the paper is to rationalize the temperature dependence of the magnetization. It is possible to predict the evolution of any physical system from the time-dependent Schrödinger equation $i\hbar \partial \Psi / \partial t = \hat{H} \Psi$, but in most cases this is neither practical nor necessary. Coarse-grained equations abstract from irrelevant heat-bath degrees of freedom and focus on relevant degrees of freedom, such as domain-wall positions. Important examples are master or rate equations, Fokker-Planck equations, and Langevin (or random-force) equations, which are physically largely equivalent. These equations form the basis for the following sections and are therefore briefly explained.

The introduction of transition rates $W(s,s') = W(s' \rightarrow s)$ between states $s'$ and $s$ yields the rate or master equation
\[
\frac{\partial P(s)}{\partial t} = \int \left[ W(s,s') P(s') - W(s',s) P(s) \right] ds' \tag{3}
\]
for the probability $P(s)$. Assuming random but small jumps $\pm \Delta s$ (diffusion) and a deterministic term (drift) yields the Fokker-Planck equation
\[
\frac{\partial P}{\partial t} = \frac{\Gamma_0}{k_B T} \frac{\partial}{\partial s} \left[ \frac{\partial E}{\partial s} P \right] + \Gamma_0 \frac{\partial^2 P}{\partial s^2}, \tag{4}
\]
where the drift is described by the force $\partial E / \partial s$. In some cases, it is possible to find exact solutions. For example, the zero-field magnetization of small platelike particles with zero in-plane anisotropy decays as $M_z(t) = M_z(0) \exp(-\Gamma_0 t)$. In the limit of nonequilibrium states captured in deep potential valleys (activation energy $E_a \gg k_B T$), the Focker-Planck dynamics approaches the Arrhenius limit with the relaxation rate $\Gamma_0 \exp(-E_a / k_B T)$. This regime is also known as Kramers’ escape-rate theory and, in magnetism, as the Arrhenius-Néel-Brown theory. The Landau-Lifshitz precession enters these expressions only indirectly by affecting $\Gamma_0$.

Solving the Fokker-Planck equation yields the probability $P(s,t)$, from which averages such as $\langle s(t) \rangle$ and $\langle s(t)|s'(t') \rangle$ are obtained by integration. The explicit calculation of $P(s)$ can be avoided by using the Langevin equation
\[
\frac{\partial s}{\partial t} = -\frac{\Gamma_0}{k_B T} \frac{\partial E}{\partial s} + \sqrt{2\Gamma_0} \xi(t). \tag{5}
\]
Here the random thermal forces $\xi(t)$ have the character of a delta-correlated white noise, where $\langle \xi(t) \rangle = 0$ and $\langle \xi(t) \xi(t') \rangle = \delta(t-t')$ ensure the equilibrium limit $P \to \exp(-E/k_B T)$.

Kramers’ escape-rate theory yields the above-introduced Arrhenius or Néel-Brown law, which has been used in magnetism since the 1930s. $	au = \tau_0 \exp\left(\frac{E_a}{k_B T}\right). \tag{6}$

where $E_a$ is the activation energy associated with the energy barrier and $\tau_0 = 1 / \Gamma_0$ is an inverse attempt frequency of the order of $10^{-10}$ – $10^{-9}$ s. There is also an activation entropy $S_a$ describing the number of paths over the energy barrier, so that $\tau_0 = \exp[(E_a - TS_a)/k_B T]$, but $S_a$ is conveniently incorporated into $\tau_0$. In Kramers’ theory, $S_a$ increases with decreasing curvature of the energy maximum (saddle point).

Inverting Eq. (6) and assuming a time scale $\tau \sim 100$ s yields the accessible energy barrier $E_a = k_B T \ln(\tau / \tau_0)$, or $E_a = 25k_B T$. This is the well-known 25-kT rule. At room temperature, the surmountable energy barrier is therefore $E_a = 7500$ K, significantly smaller than typical micromagnetic energy barriers of the order of 100 000 K. In magnetic recording, $\tau \sim 10$ y, and the corresponding energy barrier $E_a \approx 40k_B T$ (up to 70kBT for high reliability).

A slightly different equation is the logarithmic magnetic-viscosity law
\[
M(H,t) = M(H,t_0) - S \ln(t/t_0), \tag{7}
\]
where $S$ is the magnetic-viscosity constant. For example, typical permanent magnets lose a small fraction of their magnetization, typically a few 0.1%, within the first hours after production. This Jordan aftereffect is due to energy-barrier distributions naturally occurring in magnetic materials and reproduced by integration over all energy barriers,
\[
M(t) = -M_s + 2M_s \int_{-\infty}^{\infty} P(E) e^{-\Gamma_0 t} \exp(-E/k_B T) dE. \tag{8}
\]
For low temperatures, this yields $M(t) = M(t_0) - 2M_s k_B T P(E) \ln(t/t_0)$.

Figure 5 shows that the energy in Eq. (6) depends on the magnetic field. A frequently used energy-barrier expression is
\[
E_a = K_0 V_0 \left(1 - \frac{H}{H_o}\right)^m, \tag{9}
\]
where parameters $K_0$, $V_0$, $H_0$, and $m$ describe the magnet’s real structure. Zero-temperature or static switching occurs
when $E_a$ vanishes, that is, for $H=H_0$. Substituting $E_a$ into Eq. (6) and equating the field $H$ with $H_c$ yields the Kneller-Sharrock equation:

$$H_c = H_0 \left(1 - \frac{k_B T}{K_0 V_0} \ln(\tau_0)\right)^{1/m}. \quad (10)$$

Equations (9) and (10) mean that thermally activated reversal is facilitated by an external field which reduces $E_a$ until the static switching condition $H=H_0$ is nearly satisfied (Fig. 5). Note that $K_0$ and $H_0 \sim 2K_0/M_s$ are both temperature dependent (Sec. III).

To derive $m$ in Eq. (9), one expands the micromagnetic energy in the vicinity of the saddle point. The inclusion of linear, quadratic, and cubic terms yields $m=3/2$. This exponent was first obtained by Néel in 1950 and describes a variety of coherent and incoherent magnetization processes, such as strong domain-wall pinning and the reversal of misaligned Stoner-Wohlfarth particles. Symmetric energy barriers, the cubic coefficient is zero, and one must include a quartic term. This changes the exponent to $m=2$, as exemplified by aligned Stoner-Wohlfarth particles.

Linear laws ($m=1$) are occasionally assumed in calculations, but their derivation from physically meaningful energy landscapes has remained elusive. Other approaches start from unrealistic or ill-defined energy landscapes. For example, series expansion in the vicinity of $H=0$ reduces $E_a \sim (1/H - 1/H_0)$ to an $m=1$ law, but for $H=0$ it amounts to the unphysical prediction of an infinite energy barrier. Note that linear laws $E_a \sim H_0 - H$ looks like a Zeeman energy, but the Zeeman interaction does not lead to a linear field dependence. However, linear laws can be used to rationalize experimental data, using $E_a \sim M_s V(H - H_0)$ to derive temperature-dependent activation volumes $V^*$.

V. CASE STUDY: COMPOSITE NANOPARTICLES

Before returning to our original problem of reconciling thermodynamics and micromagnetics, let us use an example to summarize Secs. II–IV. In magnetic recording, one tries to combine writability (small $H_c$) with thermal stability (large $E_a$). One possible scenario is to exchange couple hard and soft regions. In the simplest case one considers a hard particle of volume $V_0$ and anisotropy $K(T)$ coupled to soft particle of volume $V_0$ and zero anisotropy. The total energy is

FIG. 5. Field induced and thermally activated magnetization processes. In most systems, thermal activation is a small correction to the leading field-dependent or “static” mechanism.

When $E_a$ vanishes, that is, for $H=H_0$. Substituting $E_a$ into Eq. (6) and equating the field $H$ with $H_c$ yields the Kneller-Sharrock equation:

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VI. GIANT FLUCTUATIONS

At zero temperature, the magnetization reversal is realized by the path with the lowest saddle-point energy. “Giant fluctuations,” which are reminiscent of the thermally activated uphill motion of a big stone, have very small Boltzmann factors $\exp(-E_a/k_B T)$ and can usually be ignored.
However, the activation energy for small particles is \( K_1 V \), and for sufficiently small particles, \( E_a \) become comparable to \( k_B T \). The same is true for thin wires of radius \( R \), where the activation energy scales as \( \pi R^2 \). This makes long and thin wires unsuitable for data storage.

To reconcile statistical mechanics \(^7\) and micromagnetism,\(^5,6\) we use a harmonic approximation. Figure 7 shows that the approximation breaks down when the magnetization angle reaches \( 90^\circ \) (\( M_z = M_x \)). This provides a natural cutoff for the perpendicular magnetization component \( m = M_x / M_z \), and the Langevin equation reduces to

\[
\frac{\partial m(r)}{\partial t} = \left[ 2A\nabla^2 m - (2K - \mu_0 HM_x) m \right] V + \sqrt{2\Gamma_0} \xi(r,t).
\]

(12)

It is easily solved by normal-mode analysis and yields both the lowest lying mode (nucleation mode) and excited modes, including giant fluctuations.

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In one dimension, the energy depends on the radius of the wire. A rough but essentially correct argument is to assume that thermal activation leads to the reversal of a wire segment of length \( 2L \). This is paid by the creation of two domain walls of combined energy \( 2\pi R^2 \gamma \), where \( \gamma = 4(AK_1)^{1/2} \) is the domain-wall energy. Equating this energy to \( 25k_B T \) yields the transition temperature

\[
E = A_d \left[ \sqrt{\frac{\pi}{d}} L \right]^d \left[ \frac{A}{2L^2} + \left( K - \frac{1}{2} \mu_0 M, H \right) \right],
\]

(13)

where \( A_d \sim R^{3-d} \). Figure 9 shows the energy of the zero-field fluctuations as a function of the fluctuation size. The physical realization of the modes is governed by their Boltzmann factors.

Below \( T_0 \), thermal excitations lead to magnetization fluctuations whose range \( L \) is larger than the domain-wall thickness parameter \( \delta_0 = (A/K_1)^{1/2} \). This is basically a random-field problem \(^{40} \) and yields

\[
L = \frac{\delta_0}{\sqrt{1 - \frac{H}{H_0}}},
\]

(15)

In the limit of static magnetization reversal, where \( H = H_0 \), this equation reproduces the coherent-rotation mode (\( L = \infty \)). At small temperatures, the fluctuations obey \( L \sim 1/T \). This reconciles the dynamic behavior with exact nucleation mode.

In two and especially three dimensions it is easy to form small nuclei (Fig. 9), but they rapidly collapse and do not lead to magnetization reversal. In two dimensions, one obtains the field-independent activation energy \( E_a = 2\pi A \gamma \), where \( \gamma \) is the film thickness. This energy corresponds to a cylindrical domain of length \( t' \) and radius \( L' \approx 2\delta_0 \). Taking \( E_o = 25k_B T \) and \( A = 10 \) pJ/m, we obtain room-temperature stability thickness of about 1.5 nm.

**VII. SUBLATTICE INSTABILITY**

Finally, we briefly discuss giant fluctuations related to anisotropy, as contrasted to magnetization fluctuations. For simplicity, we consider rare-earth fluctuations, Fig. 10(a), in a nanoparticle containing \( N \) atoms. Here \( K_1(T) \sim K_0 J^2 / T^2 \), and the average anisotropy energy is \( N K_0 J^2 / T^2 \), but fluctuations \( (K_1)^2 \) play an important role on a local scale. In some regions, the anisotropy is temporarily reduced, similar to the soft region in Fig. 6. This deteriorates the thermal
stability of the stored information. Pictorially, thermal activation does not push the magnetization over the saddle point but changes that height of the mountain range, like an earthquake.

The effect can be quite strong. The local random field exerted by the spins in Fig. 10(a) is of order $J^*$. For a small particle containing $N$ particles, the magnitude of the effect is $N^{1/2}J^*$, and the ratio $\delta E/(E) \sim T^2/J^2K_0N^{1/2}$. When $\delta E$ exceeds $E$, the fluctuations actually dominate the average anisotropy. This is the case for particles smaller than $N_c \sim T^2/J^2K_0^2$. Since $T/K_0 \sim 100$ and $T/J \gg 1$, $N_c \sim 10,000$, corresponding to particle diameters of a few nanometers. This example shows that sublattice effects yield a disproportionately strong contribution to the thermal instability.

VIII. CONCLUSIONS

In summary, we have analyzed how thermal excitations affect the magnetization of magnetically stored information. Thermal activation is embedded in a complicated hierarchy involving static magnetization reversal, intrinsic temperature variations, thermal activation over static energy barriers, and giant fluctuations away from static reversal. The static reversal mode is a solution of a well-defined micromagnetic problem. It usually involves structural imperfections and cannot be postulated on intuitive grounds. In a strict sense, there are no giant fluctuations in magnetism, and even extreme cases, such as thin wires, approach the correct micromagnetic limit at low temperatures. However, these excitations have a big impact on the magnetization reversal for extremely high densities, corresponding to bit sizes of about 2 nm. In this regime, the achievable areal density scales as the ratio of domain-wall energy to temperature.

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