Linear and elliptical magnetization reversal close to the Curie temperature

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Abstract – For further improvement of magnetic information storage density and writing speed, laser-induced writing procedures have been extensively explored recently. Within the framework of the Landau-Lifshitz-Bloch equation of motion, which does not conserve the length of the magnetization vector, we investigate thermally assisted switching analytically. We show that for temperatures close to (but still below) the Curie temperature two reversal modes appear, an elliptical mode and a linear one. We calculate the coercive fields and energy barriers for both elliptical and linear switching. Investigating the dynamics of linear reversal, which is the more relevant case close to the Curie temperature, we calculate the temperature dependence of the minimal time and field needed for thermally assisted switching below and above the Curie temperature.

Experimental studies with femtosecond time resolution are currently casting light on the physics of ultrafast magnetization processes [1–6]. Though the interpretation of these experiments in terms of a picosecond magnetization dynamics was controversially discussed, a rapid decrease and recovery of the magnetization following a laser pulse is now well established [7]. Recent experimental work [8] has even demonstrated the phenomenon of opto-magnetism, in which a circularly polarized laser pulse induced precessional motion in $DyFeO_3$. More recently, the same workers [9] demonstrated that the opto-magnetic effect can give complete magnetization reversal in GdFeCo *on a timescale of only* 1 ps.

While the microscopic details of the energy and momentum transfer from the laser light to the magnetization are still under debate, it was shown that the response of a magnetic system to pulsed heating can be described in terms of an atomistic spin model the dynamics of which is based on the Landau-Lifshitz-Gilbert (LLG) equation with Langevin dynamics [10]. However, less attention has been paid to studies of the magnetization reversal process during laser heating in the presence of a magnetic field [11]. Clearly this is important in understanding the dynamics of magnetization processes in the picosecond timescale, which is of practical importance in relation to heat-assisted magnetic recording (HAMR), which has been proposed as a means of writing information on high anisotropy magnetic media.

In this letter we present a theory of magnetization reversal at elevated temperatures in the presence of an applied magnetic field, applicable close to and even above the Curie temperature T_c, including an analysis of a mechanism which we term "linear reversal". At 0 K, the magnetization of a single domain magnetic nano-particle reverses by circular rotation, with all magnetic spins held parallel by the exchange field. This we term circular reversal. With increasing temperature, the magnetization has been shown to shrink as it moves into the magnetic hard direction [12]. This has an analogy with the onset of elliptical domain walls in magnetic materials [13–16] and as a result we term this elliptical reversal. At temperatures close to T_c, the transverse components of magnetization vanish [17] and we are left with a "linear" reversal mechanism. This type of reversal mode was discussed in [17] for thermally activated switching in zero field. It is a characteristically different reversal mechanism, which does not exhibit the precession expected from the reversal of single domain nano-particles. In the following, we explore

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the characteristic temperature separating the elliptical and linear reversal regimes and derive expressions for the energy barrier of the linear reversal mechanism. We will show that the picosecond reversal [9], which is beyond precessional mechanism, is consistent with linear reversal.

Analytical results for energy barriers and switching field exist so far only in the zero-temperature limit (most prominent here is the Stoner-Wohlfarth theory [18]) or within the framework of Brown's theory [19], where a stochastic LLG equation is used to describe the dynamics of nano-particles under the influence of a thermal field. In these approaches, the thermodynamic behavior of the particle itself is neglected by assuming that the magnetization of the particle is constant in magnitude. In recent atomistic simulations [12], it has been demonstrated that at temperatures approaching the Curie temperature additional effects occur, which cannot be described in this kind of approach: i) the magnetization vector magnitude is not conserved, ii) longitudinal magnetization relaxation occurs, with the longitudinal relaxation time increasing approaching the Curie temperature (critical slowing down), iii) at the same time the transverse relaxation time decreases. However, it has been shown that all these effects are in agreement with single macro-spin dynamics based on the Landau-Lifshitz-Bloch equation (LLB), which was derived by Garanin for classical [20] and quantum [21] average spin polarization. At low temperatures it coincides with the standard LLG equation but it is valid up to and beyond the Curie temperature T_c. The necessity of the longitudinal relaxation to model pump-probe experiments *via* a micromagnetic approach has been noted by several authors [22] who suggested to use for this purpose the Bloch equation. The advantage of the LLB equation resides in the fact that it is a much more consistent approach which has a more rigorous foundation and has been tested against the predictions of atomistic modeling [23].

The LLB equation (see [20] for more details) can be written in the form

$$\begin{aligned}\dot{\mathbf{m}} &= -\gamma(\mathbf{m}\times\mathbf{H}_{\text{eff}}) + \frac{\gamma\alpha_{\|}}{m^2}(\mathbf{m}\cdot\mathbf{H}_{\text{eff}})\mathbf{m} \\ &\quad - \frac{\gamma\alpha_{\perp}}{m^2}\left(\mathbf{m}\times(\mathbf{m}\times\mathbf{H}_{\text{eff}})\right), \qquad (1)\end{aligned}$$

where $\mathbf{m}$ is a spin polarization normalized to its zero-temperature value. It is not assumed to be of constant length and even its equilibrium value, m_e, is temperature dependent. Hence, besides the usual precession and relaxation terms, the LLB equation contains another term that controls longitudinal relaxation.

The LLB equation is valid for finite temperatures and even above T_c though the damping parameters and effective fields are different below and above T_c. $\alpha_{\|}$ and $\alpha_{\perp}$ are dimensionless longitudinal and transverse damping parameters. For $T \leqslant T_c$, they are $\alpha_{\|} = 2\lambda T/(3T_c)$ and $\alpha_{\perp} = \lambda(1 - T/(3T_c))$. For $T \geqslant T_c$ the damping parameters are equal, $\alpha_{\perp} = \alpha_{\|} = 2\lambda T/(3T_c)$. Here, λ is a microscopic damping parameter that characterizes the coupling of the individual, atomistic spins to the heat bath. Note that even assuming λ to be temperature independent, the macroscopic damping parameters of the LLB equation turns out to be temperature dependent [12]. In the limit $T \to 0$, the longitudinal damping parameter $\alpha_{\|}$ vanishes and with $\alpha_{\perp} = \lambda$ the LLB equation goes over to the usual LLG equation.

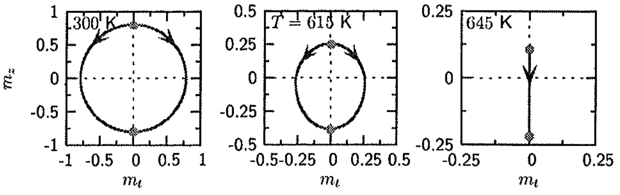

Fig. 1: (Color online) Trajectories for reversal at different temperatures. For 300 K the reversal is nearly circular (Stoner-Wohlfarth type), at 615 K elliptical deviations occur, at 645 K the reversal is linear. The red points represent the stationary points which are the initial condition as well as the final state. In the following, they are referred to as m_0 and m_∞.

For a single-domain particle, the effective field $\mathbf{H}_{\text{eff}}$ is

$$\mathbf{H}_{\text{eff}} = \mathbf{B} + \mathbf{H}_{\text{A}} + \begin{cases} \dfrac{1}{2\tilde{\chi}_{\|}}\left(1 - \dfrac{m^2}{m_e^2}\right)\mathbf{m}, & T \lesssim T_c, \\ -\dfrac{1}{\tilde{\chi}_{\|}}\left(1 + \dfrac{3T_c m^2}{5(T_c - T)}\right)\mathbf{m}, & T \gtrsim T_c, \end{cases} \qquad (2)$$

where $\mathbf{H}_{\text{A}} = -\left(m_x\mathbf{e}_x + m_y\mathbf{e}_y\right)/\tilde{\chi}_{\perp}$ represents the anisotropy field and $\mathbf{B}$ represents an external magnetic field. Here, the susceptibilities $\tilde{\chi}_l$ are defined by $\tilde{\chi}_l = \partial m_l/\partial B_l$ with $l = \|, \perp$. Note, that at low temperatures the perpendicular susceptibility $\tilde{\chi}_{\perp}$ is related to the temperature-dependent anisotropy constant K *via* $\tilde{\chi}_{\perp} = M_s^0 m_e^2/(2K)$ [20], where M_s^0 is the zero-temperature saturation magnetization. In the following we use $\tilde{\chi}_l$ and the reduced zero-field equilibrium magnetization $m_e(T)$ as calculated from a spin model for FePt (for details see [16,24]) for all our calculations. The corresponding Curie temperature T_c is 660 K.

The effective fields of the LLB equation are the derivative $\mathbf{H}_{\text{eff}} = -\frac{1}{M_s^0}\frac{\delta f}{\delta \mathbf{m}}$ of the free-energy density

$$f = -BM_s^0 m_z + \frac{M_s^0}{2\tilde{\chi}_{\perp}}(m_x^2 + m_y^2) + \frac{M_s^0}{8\tilde{\chi}_{\|}m_e^2}\left(m^2 - m_e^2\right)^2, \qquad (3)$$

if we assume the external magnetic field B to point in z-direction. As in the case of the Stoner-Wohlfarth theory, the quasi-static coercive fields can be calculated from consideration of the free energy. However, due to the fact that the LLB equation allows for a variation of the magnetization magnitude different reversal mechanisms are possible (see fig. 1). The zero-temperature limit of the LLB equation is identical to the LLG equation and the reversal is circular as in the Stoner-Wohlfarth model.

With increasing temperature the reversal path becomes more elliptical with a smaller magnetization magnitude along the hard axis. We will call this path elliptical in the following even though, strictly, only the minimal energy path for thermally activated switching in zero field is elliptical [17]. The reversal path becomes oval due to the influence of the external field. Close to the Curie temperature, a reversal without any magnetization transverse to the easy axis sets in, which we will refer to as linear reversal. Note that for pure thermal switching, without magnetic field, these reversal paths were already discussed in [17].

We will start with the case of a linear reversal. During a linear reversal process the magnetization changes its direction along the z-axis only, without any x- or y-components of the magnetization. This reversal is only possible at finite temperatures where the magnitude of the magnetization can shrink to zero followed by a reappearance with opposite direction along the easy axis. The free energy along that path depends only on m_z. The condition $\frac{\partial f}{\partial m_z}=0$ leads to stationary points, either a local minimum, a global minimum, and a maximum for fields smaller than the coercive field or just one global minimum for fields above the coercive field. The corresponding magnetization values are given by the condition

$$B=\left(m_z^3-m_e^2 m_z\right)/\left(2\tilde{\chi}_{\|} m_e^2\right). \qquad (4)$$

Let us call the solutions for the two minima $m_\pm$ and the solution for the maximum m_B. When additionally $\frac{\partial^2 f}{\partial m_z^2}=0$ the free-energy minimum becomes unstable, yielding the coercive field for linear reversal,

$$B_c^l=m_e/(3\sqrt{3}\tilde{\chi}_{\|}), \qquad (5)$$

and the corresponding magnetization value where switching sets in, $m_z^l=m_e/\sqrt{3}$. In other words, starting from a positive magnetization, a negative field will lead to a decreasing magnetization. When the magnetization is reduced by a factor of $1/\sqrt{3}$ the system is no longer in a local free-energy minimum (in z-direction) and a linear reversal sets in. In this regime, the coercive field does not depend on the $\tilde{\chi}_\perp$ and, hence, on the anisotropy constant. Instead only the longitudinal susceptibility determines the coercive field. Note, however, that in general the coercive field for linear reversal is huge unless the temperature approaches the critical region where the longitudinal susceptibility diverges. For fields $B<B_c$, the energy barrier for linear reversal can be calculated as $\Delta f=f(m_B)-f(m_+)$. However, since for the calculation of m_+ and m_B a third-order equation has to be solved (eq. (4)), the results are rather lengthy and will be published elsewhere.

For lower temperatures the system will reverse rather by rotation and in the following we will discuss this more common type of reversal process, where the magnetization follows a (more or less) elliptical path. The first derivative, $\frac{\partial f}{\partial m_x}$ is always zero at $m_x=0$. Let us once again assume

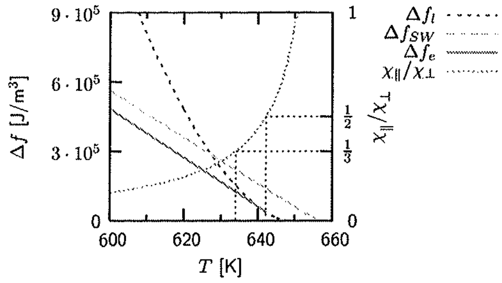

Fig. 2: (Color online) Energy barriers for linear and elliptical reversal as well as the Stoner-Wohlfarth limit. The susceptibility ratio $\tilde{\chi}_\|/\tilde{\chi}_\perp$ is also shown.

that we start with positive magnetization and lower the magnetic field. Then, the second derivative $\frac{\partial^2 f}{\partial m_x^2}$ must be negative for $m_x=0$ and $m_z=m_+$, so that the free energy has a (local) minimum at that point leading to the condition $m_+>m_e\sqrt{1-2\tilde{\chi}_\|/\tilde{\chi}_\perp}$. When with decreasing field and, hence, decreasing m_+ the above condition is violated, elliptical reversal sets in unless the system reverses earlier on a linear path. Note that $\tilde{\chi}_\|/\tilde{\chi}_\perp$ increases with temperature (from 0 at $T=0$ to infinity at $T=T_c$ as can be seen in fig. 2) leading to the fact that the elliptical path vanishes already below T_c, at a temperature T^*, where $\tilde{\chi}_\|/\tilde{\chi}_\perp=1/2$ as discussed in [17] for thermally activated switching in zero field.

Taking into account the limiting magnetization value for linear reversal, elliptical reversal occurs under the condition $m_e/\sqrt{3}\leqslant m_+\leqslant m_e\sqrt{1-2\tilde{\chi}_\|/\tilde{\chi}_\perp}$, which is fulfilled if $\tilde{\chi}_\|/\tilde{\chi}_\perp\leqslant 1/3$. The magnetic field required to lower the magnetization to the critical value where elliptical reversal sets in, $m_z^e=m_e/\sqrt{1-2\tilde{\chi}_\|/\tilde{\chi}_\perp}$, is the coercive field for elliptical reversal,

$$B_c^e=\frac{m_e}{\tilde{\chi}_\perp}\sqrt{1-2\tilde{\chi}_\|/\tilde{\chi}_\perp}, \qquad (6)$$

with the constraint $\tilde{\chi}_\|<\tilde{\chi}_\perp/3$. In the zero-temperature limit $\tilde{\chi}_\|$ vanishes and the Stoner-Wohlfarth limit for circular reversal is recovered.

Transforming eq. (3) into polar coordinates (m,θ) the energy barrier for elliptical reversal can be calculated as well. The saddle point can be found from the conditions $\frac{\partial f}{\partial m}=\frac{\partial f}{\partial \theta}=0$ and the energy difference between saddle point and local energy minimum can be calculated. However, once again third-order equations have to be solved so that the results are rather complicated and will be published elsewhere. Nevertheless fig. 2 shows the energy barriers for linear or elliptical reversal along with $\tilde{\chi}_\|/\tilde{\chi}_\perp$. Close to T_c the ellipticity of the reversal leads to a reduction of the energy barrier as compared to the Stoner-Wohlfarth limit. Furthermore, it can be seen that even below T_c the energy barrier Δf_l for the linear reversal is reduced in relation to Δf_e, for the elliptical reversal. Actually, the linear reversal becomes more favourable

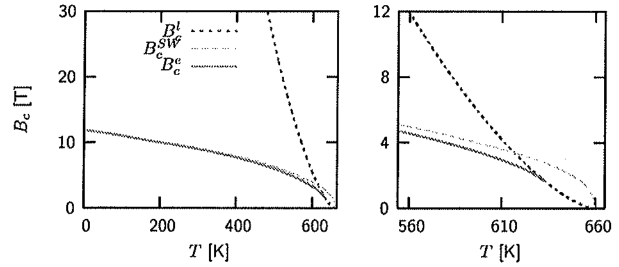

Fig. 3: (Color online) Coercive fields *vs.* temperature. The values for linear, elliptical, and circular (Stoner-Wohlfarth) reversal are shown. The figure on the right-hand side shows the critical region in more detail.

at T^* which is consistent with the earlier result that the elliptical path vanishes at this temperature. This is an important finding in relation to ultrafast laser pump-probe processes and, especially, for HAMR from a practical point of view. For high anisotropy materials such as FePt the critical temperature T^* for linear reversal is below the actual T_c. Here, linear reversal is clearly involved in the switching process by cooling through T_c in the presence of an applied field due to the reduced energy barrier Δf_l of the linear reversal in relation to the circular one as already discussed. Note, however, that the relevance of these findings is not entirely restricted to high anisotropic materials, since linear reversal occurs for all temperatures above the Curie temperature, probably the temperature range where heat-assisted writing will have to take place.

Figure 3 shows the coercive fields required for either linear or elliptic reversal. For comparison, a thermodynamically corrected Stoner-Wohlfarth limit $B_c^{SW} = 2K/(M_s^0 m_e)$ with temperature-dependent magnetization and anisotropy constant is shown as well. For lower temperatures the magnetic field needed for elliptical reversal is close to the Stoner-Wohlfarth limit and much smaller than the one needed for linear reversal. However, for temperatures approaching T_c linear reversal takes over.

The next step is to discuss the dynamics of the reversal process. Linear reversal is along the z-axis only and the LLB equation reduces to a simple one-dimensional differential equation of the form

$$-a\dot{m}_z = m_z^3 + bm_z - c. \qquad (7)$$

The parameters a, b, and c can be identified from the LLB equation and are different above and below T_c (see table 1), but with $a, b \leqslant 0$. This differential equation can be integrated analytically.

Its solution depends on the number of roots of the polynomial $m^3 + bm - c$. Most interesting in the context of thermally assisted switching is the case of only one real root, which occurs when the external field exceeds the coercive field. In this case, only one minimum of the free energy exists and no metastable states. Above T_c, this condition is always fulfilled.

Table 1: Parameters a, b, and c of eq. (7). μ is the atomic magnetic moment and J_0 is the sum over all exchange integrals at a given site.

	$T < T_c$	$T = T_c$	$T > T_c$
a	$2m_e^2 \dfrac{\tilde{\chi}_\parallel}{\gamma\alpha_\parallel}$	$\dfrac{5}{3}\dfrac{\mu}{J_0}\dfrac{1}{\gamma\alpha_\parallel}$	$\dfrac{5}{3}\dfrac{T-T_c}{T_c}\dfrac{\tilde{\chi}_\parallel}{\gamma\alpha_\parallel}$
b	$-m_e^2$	0	$\dfrac{5}{3}\dfrac{T-T_c}{T_c}$
c	$2m_e^2\tilde{\chi}_\parallel B$	$\dfrac{5}{3}\dfrac{\mu}{J_0}B$	$\dfrac{5}{3}\dfrac{T-T_c}{T_c}\tilde{\chi}_\parallel B$

With $m(t=0) = m_0$ and $m(t \to \infty) = m_\infty$ (see also fig. 1), the solution can be written in the form

$$t = -\frac{a}{3m_\infty^2 + b}\left(\ln\left[\sqrt{\frac{(2m_0 + m_\infty)^2 + p^2}{(2m_z + m_\infty)^2 + p^2}} \times \left|\frac{m_z - m_\infty}{m_0 - m_\infty}\right|\right] + \frac{3m_\infty}{|p|}\left(\arctan\left[\frac{2m_0 + m_\infty}{|p|}\right] - \arctan\left[\frac{2m_z + m_\infty}{|p|}\right]\right)\right).$$

Note that for the case of only one real root $p^2 = 3m_\infty^2 + 4b \geqslant 0$. During thermally assisted switching field and temperature have to be applied for a certain time to guarantee that the magnetization will recover along the direction defined by the field. Let us assume that the magnetization is first in negative direction. Then, a rectangular field (positive) and temperature pulse is applied. The magnetization will increase in time. As soon as the magnetization is positive the field (and temperature) can be switched off, and the magnetization will recover in the positive direction, *i.e.*, switching will occur. Hence, the minimum time needed for the field and temperature pulse is given by $m_z(t_p) = 0$. Using the above equation, the minimal pulse time can easily be identified.

In certain limits, simplifications can be found. For $T < T_c$, $b = -m_e^2$ and $a = 2m_e^2\tilde{\chi}_\parallel/(\gamma\alpha_\parallel)$ (see table 1). Assuming $m_0 = -1$, which mimics a low temperature initial condition and overestimates the time for reversal, $m_\infty > 0$, and that close to the coercive field $p^2 = 3m_\infty^2 - 4m_e^2 \ll m_\infty^2$, we can approximate the minimal pulse time as

$$t_p \approx \frac{2m_e^2\tilde{\chi}_\parallel}{\gamma\alpha_\parallel(3m_\infty^2 - m_e^2)}\left(\frac{3\pi m_\infty}{\sqrt{3m_\infty^2 - 4m_e^2}} - \ln[2] - 3\right). \qquad (8)$$

For $T \gg T_c$, $b = 5(T - T_c)/(3T_c)$ and $a = b \times \tilde{\chi}_\parallel/(\gamma\alpha_\parallel)$ (see table 1). Assuming once again $m_0 = -1$ and small $m_\infty \approx \tilde{\chi}_\parallel B > 0$, we can approximate the minimal pulse time as

$$t_p \approx \frac{\tilde{\chi}_\parallel}{\gamma\alpha_\parallel}\left(\frac{\tilde{\chi}_\parallel B}{\sqrt{b}}\arctan\left[\frac{1}{\sqrt{b}}\right] - \ln\left[\tilde{\chi}_\parallel B\sqrt{\frac{1+b}{b}}\right]\right), \qquad (9)$$

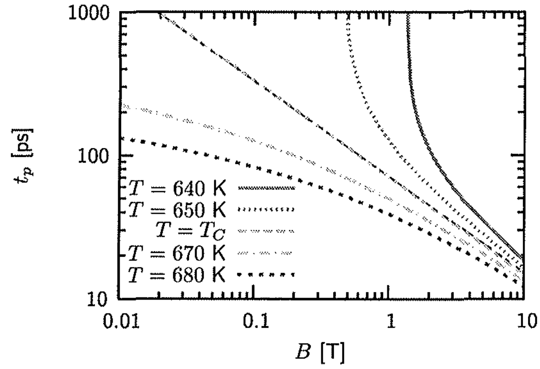

Fig. 4: (Color online) Minimal pulse time *vs.* magnetic field for different temperatures for the linear regime. For $T \approx T_c$, the analytical solution coincides with eq. (10).

where for smaller magnetic fields and higher temperatures the arctan term can be neglected as well.

For $T \approx T_c$, $m_\infty = \sqrt[3]{\frac{5}{9}\frac{\mu}{J_0}B}$. For smaller magnetic fields, *i.e.*, small m_∞, we can expand the equation above, yielding

$$t_p \approx \frac{2\pi}{3\sqrt{3}\gamma\alpha_\parallel}\left(\frac{5\mu}{3J_0B^2}\right)^{1/3}. \qquad (10)$$

In fig. 4 the minimal pulse time is shown *vs.* the magnetic field for different temperatures. The material parameters are once again those for FePt with $\lambda = 0.02$. However, since $t_p \sim 1/\alpha_\perp \sim 1/\lambda$ other values of λ will simply shift the curves. Note that below 630 K elliptical reversal would set in, the dynamics of which cannot be calculated analytically.

In summary, we have investigated a novel and intriguing mechanism of thermally activated reversal, specifically a linear reversal mode in which precession is not involved; reversal is *via* a state of zero net magnetization. Essentially, the reversal mode evolves from coherent or "circular" reversal at zero temperature to elliptical reversal at non-zero temperature, and finally to the linear mode close to T_c. The importance of our findings derives from its significance in relation to ultrafast laser pump-probe processes and, from a practical point of view, HAMR. We find that the transition to linear reversal occurs at a temperature that differs from T_c by an amount *which increases with increasing magnetic anisotropy energy.* The current interest in temperature-assisted magnetization reversal in high anisotropy materials means that the critical temperature for linear reversal is 10–20 K below T_c for materials such as FePt. Consequently, reversal by cooling through T_c in a bias field must involve linear reversal, with its reduced energy barrier relative to circular rotation. Given the scaling of the timescale with $1/\lambda$, it is clear that reversal on a timescale of picoseconds is achievable with fields of ≈ 10 T in materials with large damping (such as GdFeCo [9]). This suggests linear reversal as an important contribution to the optically induced ultrafast reversal.

Our calculations show that writing in FePt with a field of 1 T appears only to be possible very close to or even above T_c (see fig. 3). In this temperature range, the reversal is definitely linear. The minimal pulse time for the writing procedure is of the order of 10–100 ps. Note that this is without the timescale for recovery, which might lead to an overall much slower writing process [10] though slow recovery is likely to be suppressed in nm scale grains. However, in nano-particles another problem arises: the energy barrier for linear reversal is much smaller than expected from a naive Stoner-Wohlfarth type of model (see fig. 2) suggesting that thermal fluctuations play a crucial role during the writing procedure. These fluctuations could lead to a reduction in the written magnetization. This is a potential limit to magnetic recording technology that will be explored in a separate publication.

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