The Concept of Anisotropy Flows in Thickness- and Temperature-Driven Reorientation Transitions in Ultrathin Ferromagnetic Films

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Abstract—**The anisotropy-flow concept is applied to analyze in detail the reorientation transitions, driven by either thickness or temperature variations. The analysis brings to the fore the common underlying features in both cases. Three generic scenarios for the occurrence of the reorientation are predicted and described in both cases on general thermodynamic grounds. The method is especially effective when underpinned by the usual assumption about the explicit thickness dependence of the first two anisotropy constants.**

I. Introduction

Ultrathin ferromagnetic (FM) layers exhibit a remarkable transition accompanied by reorientation of the magnetization from a perpendicular to an in-plane direction. This may happen at fixed temperature T with variation of thickness d or at fixed d with variation of T below T_c [1]–[4]. Both types of transition can be suitably presented, in principle, in a (T, d)-diagram [5], [6]. Qualitatively, the reorientation transition (RT) is traced back to the competition between bulk, surface, and shape anisotropies. Below, both types of transition are treated on equal footing by applying the concept of anisotropy flows, put forward in the context of bulk single-ion magnetic anisotropy (MA) [7]. The anisotropy free energy in zero applied field is cast as in the bulk two-constant approximation: $F = \tilde{K}_1 \sin^2\theta + K_2 \sin^4\theta$, where θ is the angle between the magnetization $\mathbf{M}$ and the normal $\mathbf{n}$ to the film surface, the reference zero state is the one with $\mathbf{M} \parallel \mathbf{n}$, and $\tilde{K}_1 \equiv K_1 - \mu_0 M^2/2$. Denoting with $1, 2$, and 3 the perpendicular, in-plane, and canted phases, respectively, one finds in a standard analysis of thermodynamic stability that the phase boundaries in the anisotropy space are

$$1 \leftrightarrow 2 \quad K_2 = -\tilde{K}_1 \quad (\tilde{K}_1 > 0); \tag{1}$$

$$2 \leftrightarrow 3 \quad K_2 = -\tilde{K}_1/2 \quad (\tilde{K}_1 < 0); \tag{2}$$

$$3 \leftrightarrow 1 \quad \tilde{K}_1 = 0 \quad (K_2 > 0). \tag{3}$$

The boundaries are given by the thick lines in Fig. 1. Besides, there are two wedges in the fourth quadrant, defined by $-\tilde{K}_1 \leq K_2 \leq -\tilde{K}_1/2$ $(\tilde{K}_1 > 0)$ and $K_2 \leq -\tilde{K}_1$ $(\tilde{K}_1 > 0)$, where the neighbors 1 and 2 are *metastable* across the common border (hatched regions in Fig. 1).

II. The anisotropy flow concept

Quite generally, if the anisotropy constants are functions of $n + 1$ parameters, any one of them would drive a corresponding evolution (flow) with the remaining n parameters $(y_1, \ldots y_n)$ held fixed: $\tilde{K}_1 = \tilde{K}_1(x; y_1, \ldots, y_n)$, $K_2 = K_2(x; y_1, \ldots, y_n)$. Varying the flow parameter x reversibly between an initial and a final state will force the system evolve along a specific trajectory. Any general additional information (monotonicity, conservation of sign of K_i, etc.) leads to a considerable reduction in the allowed basins for the anisotropy flow. In the present context, the usual phenomenologic assumption is made [1], [8], [9]:

$$K_1(d, T) = K_{1b}(T) + 2K_{1s}(T)/d, \tag{4}$$

$$K_2(d, T) = K_{2b}(T) + 2K_{2s}(T)/d. \tag{5}$$

Thus bulk (b) and surface (s) contributions are treated as additive. Besides, the d-dependence is *explicit*, while the T-dependence is practically unknown.

III. Thickness- and temperature-driven RTs

A. *Thickness-driven anisotropy flows*

In the thickness-driven case, with T held fixed and eliminating d from (4), one finds that this type of flow is simply a segment of a straight line

$$K_2 = a \cdot \tilde{K}_1 + b, \tag{6}$$

where the slope a and the intercept b depend on $K_{1s}, K_{2s}, K_{1b}, K_{2b}$, and M. The flow is completely specified if one knows the initial and final states. An immediate generalization results by noting that the thickness-driven flow would be *linear* whenever the d-dependent part of K_1 and K_2 is of the same functional form for both K_1 and K_2: $K_1(d, T) = K_{1b} + 2K_{1s}/f(d)$, $K_2(d, T) = K_{2b} + 2K_{2s}/f(d)$ with *any* $f(d)$. Three generic cases of thickness-driven flows can immediately be specified as corresponding to whether the intercept b is zero, positive, or negative. In

Manuscript received February 28, 1996.
This work was supported by the Max Planck Society and the NSF No. Φ 560.

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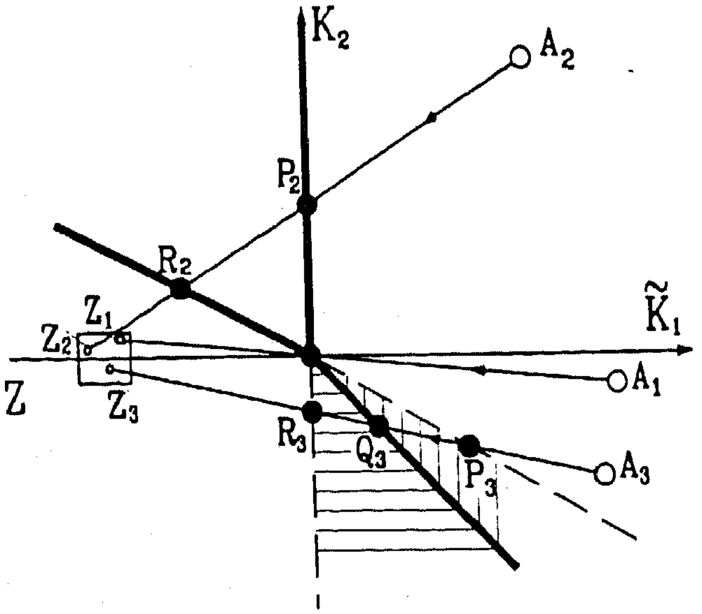

Fig. 1. Anisotropy space for the effective two-constant free energy. Thick lines: phase boundaries; thin lines: generic thickness-driven flows; hatched regions: domains of metastability of a neighboring phase. For any RT, the evolution starts from within phase 1 and flows into the small rectangle Z belonging to the in-plane phase.

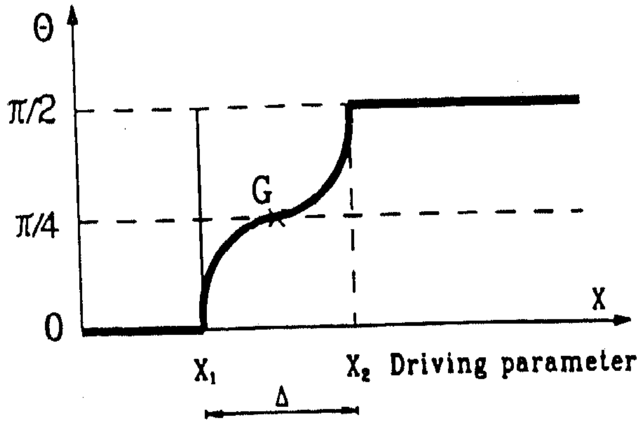

Fig. 2. Evolution of the canting angle θ driven by x ($x = d$ or T). The slopes at x_1 and x_2 are infinite by general considations (cf. (9)). The curve is centrosymmetric with respect to the point G only if the bulk contributions are negligible.

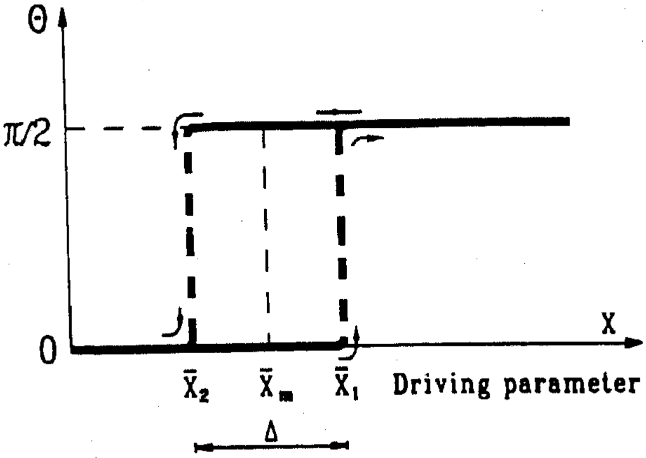

Fig. 3. Hysteresis features of θ for flows driven by x ($x = d$ or T) and proceeding via the metastable domains of Fig. 1. The abrupt RT takes place at $\bar{x}_1$ ($\bar{x}_2$) for increasing (decreasing) values of the driving parameter. The critical values correspond to the crosspoints of the trajectory A_3Z_3 with the thick dashed lines in Fig. 1. $\bar{x}_m$ corresponds to Q_3 in Fig. 1.

each of them, there are three possible subcases according to the sign of the slope a. The full description thus involves a set of defining inequalities involving the K's and M. In Fig. 1 each generic case $b = 0$, $b > 0$, or $b < 0$ is represented with the corresponding segment A_iZ_i for a single subgeneric case only. All anisotropy flows end up within the rectangular target Z of dimensions $2K_{1b} \times 2K_{2b}$ whose vertices have coordinates $(-\frac{1}{2}\mu_0 M^2 \pm |K_{1b}|, \pm|K_{2b}|)$.

For $b = 0$, the system crosses over from phase 1 to phase 2 via the origin (A_1Z_1 in Fig. 1). The condition for the occurence of this scenario is the lack of higher-order anisotropy *at the point of RT only*. Hence, a necessary condition for a nontrivial scenario to occur is $K_2 \neq 0$. Since no intermediate phases are involved, the RT is reversible and abrupt and takes place at a critical thickness d_c given by the conditon $\tilde{K}_1(d_c, T) = 0$. Provided that $|K_{1b}| \ll \mu_0 M^2/2$, one finds that $d_c(T) \approx 4K_{1s}/\mu_0 M^2$. This sets the first characteristic thickness in the problem. One may recognize that the smallness of the critical thickness of a film that would exhibit a RT is dictated by the relative smallness of K_{1s}.

For $b > 0$, the flow traverses the canted phase (A_2Z_2 in Fig. 1). The RT occurs via a continuous change of θ. The entrance into, and the exit out of, phase 3 upon increasing d are at the points P_2 and R_2, respectively, i.e. at $\tilde{K}_1(d_1, T) = 0, K_2(d_2, T) = -\tilde{K}_1(d_2)/2$, where d_1 and d_2 correspond to the onset and completion of the reorientation process. For bulk contributions much smaller than the shape-anisotropy contributions, one finds

$$d_1(T) \approx \frac{4K_{1s}}{\mu_0 M^2}, \; d_2(T) \approx \frac{4(2K_{2s} + K_{1s})}{\mu_0 M^2}. \qquad (7)$$

It is suitable to take as a *second* characteristic thickness $\Delta(T) \equiv d_2 - d_1 \approx 8K_{2s}/\mu_0 M^2$. Obviously, the width of the RT is controlled by the relative smallness of K_{2s}. The onset and the width of the RT are experimentally well-defined quantities. One thus comes to the estimate

$$K_{1s}/K_{2s} \approx 2d_c/\Delta. \qquad (8)$$

While the system evolves along the segment P_2R_2 in the canted phase, $\theta(d)$ varies between 0 and $\pi/2$ with infinite slopes at d_1 and d_2. For negligible bulk contributions, $\theta(d) \approx \arcsin\sqrt{(d - d_c)/\Delta}$ and the curve $\theta(d)$ in phase 3 is centrosymmetric with respect to the point $G = [(d_1 + d_2)/2, \pi/4]$ (cf Fig. 2).

For $b < 0$, the anisotropy flow proceeds via the metastable states in the fourth quadrant (A_3Z_3 in Fig. 1). Hence, the RT is discontinuous with the typical features of a first-order transition [10] and with the thickness as a driving parameter (Fig. 3).

The characteristic thicknesses are once again determined by the crosspoints P_3, Q_3, and R_3 of the linear flow with the phase boundaries. The characterization proceeds under the restrictions of the full set of defining inequalities ($b < 0$ with $a > 0$, $a < 0$, or $a = 0$) plus additional obvious restrictions on the thicknesses.

B. Temperature-driven RTs

The explicit determination of $K_1(T)$ and $K_2(T)$ for thin films exhibiting RTs is far from resolved [11], [12]. Consequently, the trajectories are not explicitly known. However, with the realistic assumption of monotonicity of variation with T, the continuity of the reversible flow between the initial and final points gives rise once again to *three general types* of (not necessarily linear) evolution according to the sign of $K_2(T)$ at the point of change of sign of $\tilde{K}_1(T)$. The conditions defining the crossover points to neighboring phases are now implicit. However, the same considerations about how the RT proceeds in each generic case hold true. Furthermore, a general proof of the infinity of the slopes of $\theta(T)$ at the entrance and exit of the canted phase may be given by noting that

$$\frac{\partial}{\partial x}\left[\theta(x,y)\right]_{|y=const} = \sqrt{-\frac{2K_2}{\tilde{K}_1}\cdot\frac{1}{1+\frac{\tilde{K}_1}{2K_2}}\cdot\frac{1}{4(K_2)^2}} \cdot\left(K_2\frac{\partial \tilde{K}_1}{\partial x}-\tilde{K}_1\frac{\partial K_2}{\partial x}\right)_{|y} . \quad (9)$$

For either $x \equiv d$, $y \equiv T$ (subsection A) or $x \equiv T$, $y \equiv d$ (this subsection), the denominators in the first two factors become zero at the crosspoints with the phase boundaries.

As regards the range in the relevant driving parameter over which the RT is accomplished, experimental and theoretical evidence agree on a remarkably small range in the d-driven case, but disagree in the T-driven case, where experiment favors relatively large ranges on the T scale [3], [4] in contrast to restricted theoretical evidence [12]. In an attempt to determine the conditions which would bring about a small range ΔT, consistent with theoretical predictions, one may observe that a small ΔT means that the corresponding portion of the trajectory in the anisotropy space is covered at a large speed or rate of change. Exploiting the implied mechanical analogy, one comes to the criterion that any portion of a particular trajectory $K_2(\tilde{K}_1(T))$ will be covered "quickly", if the rate of change as given by

$$v(T) = \sqrt{(\dot{K}_2)^2 + (\dot{\tilde{K}}_1)^2} \quad (10)$$

is large along this portion, i.e., if $v(T) \gg v_0(T)$, where $v_0(T)$ is a reference rate of change corresponding to the physical situation under discussion but neglecting the surface terms. The dots in (10) denote differentiation with respect to the driving parameter (here: T). In the lack of knowledge of the explicit T dependence of the quantities involved, one could still derive sufficient conditions for the smallness of ΔT:

$$|\dot{K}_{1s}|/d \gg \mu_0 M|\dot{M}|, |\dot{K}_{2s}|/d \gg |\dot{K}_{2b}|. \quad (11)$$

That is to say, an expectation of a small ΔT would be met if the rates of change of K_{1s} and K_{2s} are larger than the rates of change of the dipolar anisotropy energy and K_{2b}, respectively.

IV. Discussion

The anisotropy-flow concept provides far-reaching and systematizing insights into both types of RT in ultrathin FM films. It allows a clear presentation of the problem in the anisotropy space. In the thickness-driven case, one identifies two characteristic thicknesses. The onset of the RT is controlled by K_{1s}, whereas K_{2s} controls the width of the RT. A simple estimate of their ratio in terms of the experimentally measured quantities d_c and Δ is given. Furthermore, the curves $d_1(T)$ and $d_2(T)$, although not explicitly known, are definitely connected with more complicated (T,d) phase diagrams than has been possible to detect by now. In principle, for any given system they may lead to the subdivision of the FM portion of this diagram into up to four further subdomains. The crosspoint of these two curves, if it exists, would correspond to zero effective anisotropy in the system which might lead to a rather peculiar behavior of the system. Further complications (magnetoelastic effects, roughness, in-plane anisotropy, etc.) can be incorporated into the scheme without loss of generality and without considerable increase of mathematical complexity.

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