Spin-Reorientation Transitions In Ultrathin Ferromagnetic Films Under Applied Field

Y. T. Millev
Max Planck Institute of Mathematics in the Sciences, Inselstr. 22-26, D-04103 Leipzig

H. P. Oepen and J. Kirschner
Max Planck Institute of Microstructure Physics, Weinberg 2, D-06120 Halle, Germany

***Abstract*—The influence of external field on thin ferromagnetic films with spin-reorientation transitions (SRTs) is explored in the presence of different orders of thickness-dependent uniaxial anisotropy. The two principal configurations are addressed and the first two anisotropy contributions are examined. A simple and natural representation of the field-induced SRTs has been found. It preserves the linearity of the thickness-driven trajectories even with field. The crosspoints of these trajectories with the phase borderlines correspond to crossover thicknesses of a linear type for positive second-order anisotropy contributions and of both linear and nonlinear types for negative second-order anisotropies. In the latter case, the nonlinear boundaries correlate with the coexistence of phases which brings about first-order-like reorientations and concommittant hysteresis effects. Practical schemes are proposed for the determination of the complete set of anisotropy parameters from measured field dependences of the crossover thicknesses.**

Index Terms—**Anisotropy, ultrathin films**

I. Introduction

Systems undergoing spin reorientation transitions (SRTs) under variation of some parameter have been the object of considerable interest for several decades now [1]–[4]. Recently, SRTs in quite a number of thin and ultrathin ferromagnetic systems have been intensively studied because of the fundamental and technological issues which are at stake here [5]. A systematic discussion of thickness and temperature driven spontaneous (zero-field) SRTs in ultrathin films of uniaxial symmetry has been given very recently [6]. Here, we analyze the most important features of the behavior of such systems in an applied field within the framework of a general phenomenologic description of the competing effects. The thickness dependence of the anisotropy energy provides for an additional degree of freedom which makes possible the study of SRTs at fixed temperature and is best exploited in wedge-shape geometry.

II. Source representation in applied field

Spontaneous SRTs occur whenever the lowest-order contribution to the magnetic anisotropy energy goes to zero under variation of some parameter like temperature, concentration of a component, thickness, etc. At the zero point, the behavior of the system is stabilized by the higher-order contributions. So far as only the angular dependence is concerned, the free enthalpy

$$g_A(\theta) = a \cdot \sin^2\theta + b \cdot \sin^4\theta - \mathbf{H}\cdot\mathbf{M} \qquad (1)$$

provides the common basis for the analysis of bulk and thin film systems on equal footing. Here, the last term is the unidirectional Zeeman contribution, favoring parallel (conforming) alignment of magnetization $\mathbf{M}$ and applied field $\mathbf{H}$, which must be added to the intrinsic uniaxial anisotropy contributions described by the first two terms; θ is the angle between $\mathbf{M}$ and the crystallographic axis $\mathbf{n}$ of uniaxial symmetry. The quantities g_A, a, b have the dimension of energy per unit volume. a and b are the first and second anisotropy constants. Standard analysis of stability of possible equilibrium orientations, or phases, requires that $g_A(\theta)$ be minimized: $g_A'(\theta) = 0$, $g_A''(\theta) \geq 0$. The boundaries of stability of a given minimum are found by simultaneous examination of the equalities in the last expressions. In zero field, given the presence of higher anisotropies, one finds coexistence of phases over large portions of the anisotropy space of the system. This circumstance gives rise to first-order SRTs and to the related hysteresis effects upon variation of the driving parameter causing the change of the anisotropy constants. When a field is applied, continuity considerations suggest that coexistence of phases would persist in some form and to some extent. This expectation is proven in the following for the two principal orientations of the applied field that are typically used in the experiment, the coaxial one ($\mathbf{H}||\mathbf{n}$) and the in-plane one ($\mathbf{H} \perp \mathbf{n}$). It is in these configurations that the minimization problem is analytically solvable. More precisely, defining $x = M_H/M$ where M_H is the magnetization component along the field in either configuration, one has to solve the cubic $x^3+px+q=0$ in order to find the minima. We are only interested in situations where the angle between $\mathbf{H}$ or $\mathbf{M}$ and $\mathbf{n}$ does not exceed $\pi/2$, hence, $0 \leq x \leq 1$. The coaxial case is recovered with the identities $p = -1-a/2b$, $q = -HM/4b$, whereas the in-plane case results in $p = a/2b$, $q = -HM/4b$. The fact that in such normalization one gets the mathematically canonic form suggests that the (p,q)-representation is a source representation for the problem at hand. The results of the stability analysis in this representation are given in Fig. 1. Two different types of solution exist, a conforming and a canted one. The important borderlines are $q = 0$, $p+q+1=0$, and $p^3/27+q^2/4=0$ $(0 \leq q \leq 2)$. The most intriguing feature is the coexistence of these phases within the curvilinear triangle OCT. OC is a line

Manuscript received October 15, 1997.
Y. T. Millev, millev@mis.mpg.de, on leave from the CPCS Lab, Institute of Solid State Physics, Bulgarian Academy of Sciences, Sofia; H. P. Oepen, oepen@mpi-halle.mpg.de.

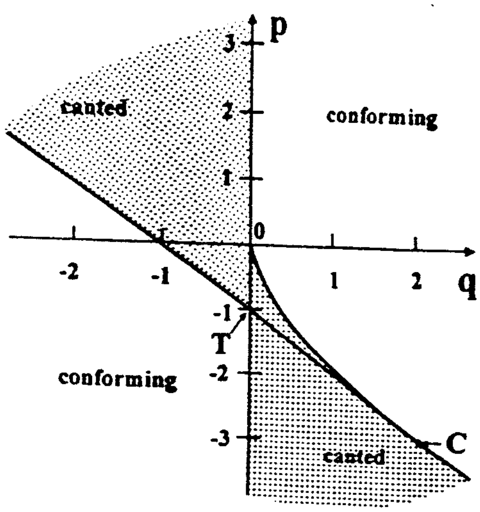

Fig. 1. Source representation ((p,q)-diagram) valid for both principal field configurations. Dotted regions correspond to a canted solution for the equilibrium magnetization. The remaining portions admit a conforming solution only (**M** along **H**). Both phases coexist within the curvilinear triangle OCT.

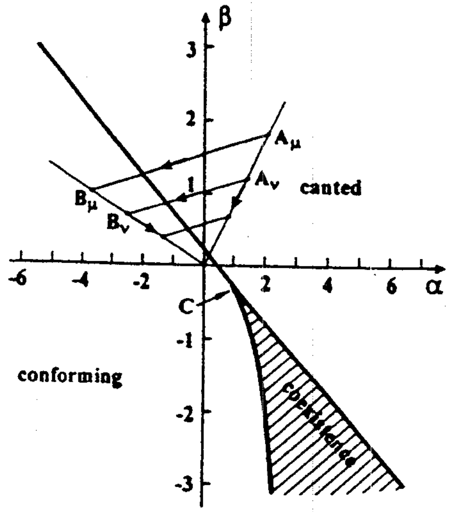

Fig. 2. Linear trajectories for thickness-driven SRTs in applied field. Here, $\alpha = a/HM$, $\beta = b/HM$ and the explicit form is given by (3). The borderlines for the different phases are obtained by transforming from (p,q) to film-specific variables according to (2).

of discontinuous transitions, while C ($p = -3$, $q = 2$) is a multicritical point where critical lines of continuous (CT) and discontinuous (OC) transitions merge.

III. Specification to ultrathin films

A. Internal structure of the anisotropy constants

In order to describe the behavior of ultrathin films with a SRT, one needs more detailed information about the structure of the anisotropy constants a and b. We address thickness-driven SRTs only and suppress the temperature dependence whose analysis is a complicated problem on its own both in the bulk [7] and in thin films [8]. For the thickness dependence, there is enough evidence both on the theoretical [9], [10] and on the experimental side [5], [11] that in quite a number of very thin films inverse proportionality of the interface anisotropy contribution to the thickness d holds and that this contribution is additive to the bulk one. More specifically,

$$a = -\Delta + K_{1s}/d,\ b = K_{2b} + K_{2s}/d, \qquad (2)$$

where $\Delta \equiv -K_{1b} + (N_z - N_x)M^2/2$, K_{ib} and K_{is} with $i = 1$ or 2 are the effective bulk and surface contributions to the magnetocrystalline anisotropy, while N_z and N_x are the demagnetization factors along the axis **n** and perpendicular to it [12]. An explicit determination of $a(d)$ and $b(d)$ in ultrathin films of $Co/Au(111)$ is given in [13] for the annealed case and in [14] for the non-annealed case.

On the analytical side, (2) underpins a nonlinear, though tractable, transformation from the canonic (p,q) variables of the previous section to film-specific variables.

B. Trajectories describing the system under applied field

Now we introduce a representation which is characterized by linear trajectories of the thickness-driven evolution of the ultrathin system under field. To this end, we normalize (2) against the Zeeman-term amplitude HM. Let us define $\alpha \equiv a/HM$ and $\beta \equiv b/HM$. Following the procedure given in [6] for the zero-field case, we eliminate the thickness d from the resulting relations, i.e. we pass over from a parametric representation $\alpha(d)$, $\beta(d)$ to an explicit relation $\beta = \beta(\alpha)$ which turns out to be linear just as in the zero-field case [6]:

$$\beta = (\kappa_{2s}/\kappa_{1s})\alpha + (\delta \cdot \kappa_{2s} + \kappa_{2b} \cdot \kappa_{1s})/\kappa_{1s}\ . \qquad (3)$$

Here, $\delta = \Delta/HM$ and $\kappa_\mu \equiv K_\mu/HM$ ($\mu = 1b, 2b, 1s, 2s$). Note that $\kappa_{2s}/\kappa_{1s} = K_{2s}/K_{1s}$, hence, the slope of the linear trajectories is given, even with field, by the ratio of the surface constants.

The structure of the $(\alpha,\ \beta)$-space is easily deduced by transforming the equations for the borderlines from the $(p,\ q)$ representation and is depicted in Fig. 2 for the in-plane field configuration together with a family of illustrative trajectories under thickness variation. There are considerable advantages in using the (α,β)-representation with regard to both simplicity and usefulness. First, as noted above, the slope of any trajectory is independent of the field and equals the ratio K_{2s}/K_{1s}. Therefore, trajectories corresponding to different field magnitudes are parallel to each other in either field configuration. Second, for a given system the intercept of any trajectory with the ordinate is inversely proportional to the field in each of the field configurations. For $H \to 0$, the intercept and the trajectory itself go to infinity in accordance with the fact that the zero-field case cannot be "observed" in this representation. For $H \to \infty$, the intercept goes to zero from above or below depending on its sign. Like the slope mentioned above, the sign of the intercept is independent of the field. Hence, for a given system the sign of the intercept and the slope of a trajectory are invariants of this representation and do not depend on the field strength. While a more detailed description of the whole construction will be given elsewhere, it may suffice to summarize here that the isolines of constant thickness are represented by the family of rays going into the origin with increasing field, while the isolines of constant field are given by the family of parallel segments $\{A_\mu B_\mu\}$ (Fig. 2). These are the eventual thickness-driven trajectories.

C. *Crossover (critical) thicknesses for SRT in field*

A crossover thickness corresponds to the point where a given trajectory crosses a given phase boundary. There are three nontrivial types of trajectory distinguished by one, two, or three cross points. For the determination of the crossover thicknesses, one notes that these relate to two distinct conditions, a *linear (nonlinear)* one for a crosspoint X_L (X_N) of a trajectory with a linear (nonlinear) phase boundary in the $(\alpha - \beta)$ representation.

For the *coaxial* field configuration, the defining equation for the crossover thickness d_L at a crosspoint of type X_L is $\alpha(d_L) = -1/2$, while d_N at a crosspoint of type X_N is given by the expression $\alpha(d_N) = -2\beta(d_N) + (3/2)\,[\beta(d_N)]^{1/3}$.

For the ***in-plane*** field configuration, d_L is defined by $\alpha(d_L) + 2\beta(d_L) = 1/2$, while d_N is given by $\beta(d_N) + 8\alpha^3(d_N)/27 = 0$.

It is especially instructive to analyze the expressions for the linear crossover thicknesses d_L. One finds that

$$\begin{aligned} \text{coaxial}: 1/d_L &= 1/d_1 - [M/2K_{1s}]H\,; \qquad (4) \\ \text{in}-\text{plane}: 1/\mathrm{d_L} &= 1/d_2 + [M/2(K_{1s} + 2K_{2s})]H, \end{aligned}$$

where $d_1 = K_{1s}/\Delta$ and $d_2 = (K_{1s} + 2K_{2s})/(\Delta - 2K_{2b})$ are the characteristic zero-field thicknesses found in [6]. Note that in the coaxial configuration (H||n) d_L is sensitive to the first-order contributions only. Should both dependences (4) be determined experimentally, they provide for four conditions (two slopes and two intercepts) binding together five, generally unknown, quantities ($\{K_\mu\}$ and M), hence, one may proceed with the estimation of the set of anisotropy constants. Even more importantly, the understanding of the expected shifts in the critical thicknesses upon variation of field is crucial to the correct interpretation of experiments. Under field, the reorientation thickness always deviates from the zero-field situation (Fig. 3). For systems exhibiting coexistence in zero field [6], the critical thicknesses separating the competing phases diverge upon increasing |H| in either configuration. For systems exhibiting the true canted phase in zero field [6], the crossover thicknesses move closer upon increasing |H|, get equal at $H = H_{cross} = 2(\Delta \cdot K_{2s} + K_{2b} \cdot K_{1s})/M(K_{1s} + K_{2s})$, and diverge for $H > H_{cross}$. Thus, the zero-field behavior, which is determined by the intrinsic anisotropy parameters, leaves its fingerprint and predetermines the tendency of the shift in the critical thicknesses upon variation of field (Fig. 3). Moreover, a single measurement leading to, e.g., $d_L(in - plane) > d_L(coaxial)$ at a given value of field is definitely inconclusive as it cannot tell on the intrinsic behavior of the system and should be interpreted with care, if at all.

IV. Summary

We have analyzed the consequences of the thickness dependence of anisotropy in ultrathin films for the SRT behavior in applied field for the two major field configurations. A linear-trajectory representation for the thickness-driven evolution in field has been introduced and exploited for the determination of the relevant crossover thicknesses d_L. The tendency of the shifts in d_L with change of field has been traced back to the intrinsic behavior of the system. The developed method makes possible to extract information on the anisotropy parameters from the dependences $d_L(H)$ and $d_N(H)$ or, additionally, from the shape of the magnetic profiles $M_H(H)$ in wedge-shape geometry. This latter method will be discussed at length separately.

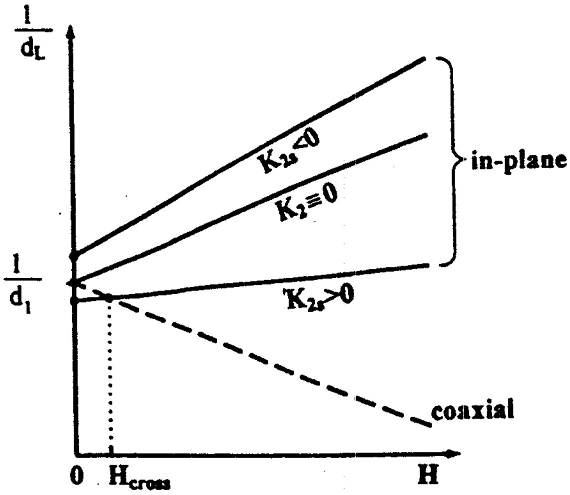

Fig. 3. Tendency of shift of critical thickness with field (cf (4-5)). Coaxial (dashed): Independence of higher-order contributions. In-plane (thick lines): Effect of K_{2s} is appreciable. The intrinsic behavior ($H = 0$) is as characteristic as a fingerprint. Depending on the zero-field behavior, the $1/d_L(H)$ lines taken alternately in the two configurations may or may not have a crosspoint. ($K_{1s} > 0$ is assumed, since this sign is consistent with the existence of a SRT.)

References

[1] R. M. Bozorth, *Ferromagnetism*, New York: IEEE Press, 1993.

[2] L. D. Landau and E. M. Lifshitz, *Electrodynamics of Continuous Media*, Oxford: Pergamon, 1960, ch. 5.

[3] M. Darby and E. Isaac, *IEEE Trans. Magn.* **10**, 259 (1974).

[4] G. Asti, in *Ferromagnetic Materials*, vol. V, K. H. J. Buschow and E. P. Wohlfarth, Eds., Amsterdam: Elsevier, 1990, pp. 398 - 464.

[5] U. Gradmann, in *Handbook of Magnetic Materials*, vol. VII, K. H. J. Buschow, Ed. Amsterdam: North-Holland, 1993, pp. 1-96.

[6] Y. Millev and J. Kirschner, *Phys. Rev.* **B54**, 4137 (1996).

[7] Y. Millev and M. Fähnle, *IEEE Trans. Magn.* **32**, 4743 (1996).

[8] P. J. Jensen and K. H. Bennemann, *Solid State Comm.* **100**, 585 (1996).

[9] L. Néel, *J. Phys. Rad.* **15**, 225 (1954).

[10] M. N. Barber, in *Phase Transitions & Critical Phenomena*, C. Domb and J. L. Lebowitz, Eds. London: Academic Press, 1983, pp. 145-266.

[11] K. Baberschke and M. Farle, *J. Appl. Phys.* **81**, 5038 (1997).

[12] W. F. Brown, Jr., *Magnetostatic Principles in Ferromagnetism*, Amsterdam: North-Holland, 1962, pp. 104-112.

[13] H. P. Oepen, M. Speckmann, Y. T. Millev, and J. Kirschner, *Phys. Rev.* **B55**, 2752 (1997).

[14] H. P. Oepen, Y. T. Millev, and J. Kirschner, *J. Appl. Phys.* **81**, 5044 (1997).