Reorientation transitions in ultrathin ferromagnetic films by thickness- and temperature-driven anisotropy flows

Yonko Millev and Jürgen Kirschner
Max-Planck-Insitut für Mikrostrukturphysik, Weinberg 2, D-06120 Halle, Germany
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A unified thermodynamic treatment is presented for reorientation transitions in ultrathin ferromagnetic films, driven by temperature or thickness variations. Since the physical mechanism underlying such transitions is the competition between surface, bulk, and shape anisotropy, the natural and proper scenery to examine the problem is the anisotropy space of the corresponding system. The recently developed anisotropy-flow concept is then applied to detect and characterize the possible thickness- and temperature-driven reorientation transitions, consistent with the standard, though not universally valid, assumption for the additive separation of the bulk and surface contributions. Three generic types of transition are easily identified with a number of subcases each. Each generic scenario is seen to come about as a simple consequence of the route which the system follows in the anisotropy space under variations of the driving parameter. The relevant characteristic thicknesses are easily identified. We find that, as a rule, there exist *two* borderlines in the ferromagnetic part of the *T-d* diagram of the system; these should lead in principle to a much richer structure of the diagram in question than has been reported so far. [S0163-1829(96)07329-8]

I. INTRODUCTION

Some ferromagnetic layers of a thickness of a few monolayers (ML) exhibit a remarkable transition accompanied by a reorientation of the direction of the spontaneous magnetization.[1,2] At fixed temperature below the Curie temperature T_C, the increase in thickness leads to the reorientation of the magnetization from a perpendicular into an in-plane direction starting at some critical thickness. At a fixed and very small thickness d, the increase of temperature from values much smaller than the corresponding T_C triggers the same crossover starting at some critical (reorientation) temperature. Reports on the thickness-driven transition come mostly from the experimental side,[2] whereas the temperature-driven transition has been analyzed theoretically as well.[3–5] In the first case the reorientation appears to be continuous, while both continuous and discontinuous variations have been found under variation of temperature. As to the range in the relevant parameter (thickness or temperature) over which the reorientation is accomplished, experimental and theoretical evidence agree on a remarkably small range in the thickness-driven case, but disagree in the temperature-driven case, where experiment favors relatively large ranges on the temperature scale[2,6,7] in contrast to restricted theoretical evidence.[5]

Qualitatively, the reorientation transition (RT) in ultrathin films has been traced back to the competition between bulk, surface, and dipolar (shape) anisotropies. Only very thin films stand the chance of generating perpendicular anisotropy which could override the effect of the ultimately large demagnetization factor ($N_z=1$), characteristic for the direction perpendicular to a flat specimen.

Quite recently, the notion of *temperature-driven anisotropy flows* has been put forward in the study of the variation of the bulk magnetic anisotropy of single-ion origin.[8] Basically, the idea boils down to combining the results of a standard thermodynamic (TD) analysis of the stability of a bulk system with anisotropies of different orders with the statistical-mechanical calculation of the variation of the phenomenological anisotropy constants. The microscopics are valid for a whole class of theories and are underpinned by a newly found parametric method. The consideration of anisotropy constants of different orders was but a natural ingredient of the flow analysis in the anisotropy space.

There is little evidence to date of a detailed quantitative clarification of the microscopic contributions to the different anisotropies and, hence, of the details of the RT in ultrathin films. Still, an exhaustive TD analysis of the direction-dependent part of the free energy can give astonishingly abundant information under rather minimal assumptions. In fact, both types of transition are put on equal and general footing if one analyzes the problem in terms of *thickness-driven* or *temperature-driven* anisotropy flows (trajectories), characteristic for the anisotropy phase diagram of the respective thin film. While the idea to implement phenomenological free-energy expressions is not new in the thin-film context,[9–11] the formulation we suggest has several advantages and offers insights which should become obvious in the following and are summarized in the final section.

II. GENERAL FRAMEWORK

Consider a very thin flat film whose easy axis for the magnetization $\mathbf{M}$ is collinear with the normal $\mathbf{n}$ to the surface. Defining θ as the angle between $\mathbf{M}$ and $\mathbf{n}$ and assuming that there is no external magnetic field, one writes down the phenomenological expression for the direction-dependent part of the free-energy density as

$$F_0=K_1\sin^2\theta+K_2\sin^4\theta+\tfrac{1}{2}\,\mu_0M^2\cos^2\theta. \qquad (1)$$

The first two terms involve the first and second anisotropy contributions in standard notation, while the last term gives

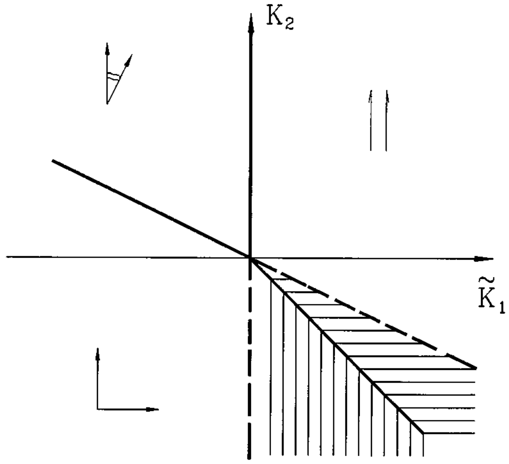

FIG. 1. Possible phases for the free energy of Eq. (3). Thick lines separate different phases. Metastability domains are hatched (fourth quadrant). See also Eqs. (6)–(10) in the text.

the demagnetization energy with the extreme demagnetization factor of unity in the perpendicular direction. More complicated angular dependences arising from non-negligible inplane anisotropies and involving an additional independent angular variable can be treated along the lines described below. It proves very advantageous to redefine the zero of the free energy:

$$F \equiv F_0 - \tfrac{1}{2}\mu_0 M^2 = \tilde{K}_1 \sin^2\theta + K_2 \sin^4\theta, \tag{2}$$

where now the reference state of zero free energy is the one with $\mathbf{M}||\mathbf{n}$ and we have defined

$$\tilde{K}_1 \equiv K_1 - \tfrac{1}{2}\mu_0 M^2. \tag{3}$$

Thus, the depolarizing effect of the dipolar self-energy has been absorbed into an expression which is formally completely analogous to the bulk phenomenological free energy with first- and second-order uniaxial anisotropies in the absence of applied field.

A. Structure of the anisotropy space

Minimization of the free energy of Eq. (2) with respect to θ reveals the possibility for three different TD phases which are to be distinguished by the corresponding equilibrium values of θ. The calculation appears so trivial that it is practically always forgotten to mention the analysis of the *relative stability* of the possible phases. We summarize briefly the results of the exhaustive TD analysis (cf. also Fig. 1): (i) phase 1 (perpendicular or magnetization-up phase), $\theta=0$; (ii) phase 2 (in-plane phase), $\theta=\pi/2$; (iii) phase 3 (''canted'' phase), $\sin^2\theta=-\tilde{K}_1/2K_2$, i.e.,

$$\theta = \arcsin\sqrt{-\tilde{K}_1/2K_2} \quad (0 \leqslant -\tilde{K}_1/2K_2 \leqslant 1). \tag{4}$$

There is a territorial dispute in the phase diagram between the first two phases in that there is an overlapping domain where both of them are minimal. Their relative stability is decided upon by considering the depths of the respective minima:

$$\Delta F_{21} \equiv F(\theta=\pi/2) - F(\theta=0) = \tilde{K}_1 + K_2. \tag{5}$$

Thus, the phase boundaries are given by the thick lines in Fig. 1, which are defined as follows:

$$1 \leftrightarrow 2 \quad K_2 = -\tilde{K}_1 \quad (\tilde{K}_1 > 0), \tag{6}$$

$$2 \leftrightarrow 3 \quad K_2 = -\tfrac{1}{2}\tilde{K}_1 \quad (\tilde{K}_1 < 0), \tag{7}$$

$$3 \leftrightarrow 1 \quad \tilde{K}_1 = 0 \quad (K_2 > 0). \tag{8}$$

There are, however, two wedges in the fourth quadrant, defined by

$$-\tilde{K}_1 \leqslant K_2 \leqslant -\tfrac{1}{2}\tilde{K}_1 \quad (\tilde{K}_1 > 0) \tag{9}$$

and

$$K_2 \leqslant -\tilde{K}_1 \quad (\tilde{K}_1 > 0), \tag{10}$$

where each neighbor is *metastable* across the boundary (hatched regions in Fig. 1).

B. Anisotropy-flow concept

Quite generally, if the anisotropies are functions of $n+1$ parameters, then any one of them, say, x, would drive a corresponding evolution or flow with the remaining n parameters, say, $y_1, y_2, \ldots, y_n$, being held fixed:

$$\tilde{K}_1 = \tilde{K}_1(x; y_1, \ldots, y_n), \tag{11}$$

$$K_2 = K_2(x; y_1, \ldots, y_n). \tag{12}$$

Varying the flow parameter x between an initial and a final state will force the system to evolve along a specific trajectory between these two extreme states in the phase diagram. Provided that $\tilde{K}_1(x)$ and $K_2(x)$ are monotonic functions at fixed $y_p (p=1,2,\ldots,n)$, no horizontal or vertical line (i.e. no straight line parallel to any of the anisotropy axes) may be crossed more than once. While this property of the flow seems rather obvious by simple examination of the diagram, its consequences are very stringent. Independently, further severe restrictions on the basin of flow arise, if the sign of one or both anisotropy constants is conserved under the x-driven evolution of the system. The picture described in this paragraph is not restricted to the anisotropy analysis of thin films only and represents a systematic extension of the anisotropy-flow analysis of Ref. 8.

For the reorientation in thin films, up to this point we have profited from incorporating the demagnetization effect into an anisotropic free energy expression with a shifted zero point and a redefined first anisotropy constant $\tilde{K}_1$. Expression (2) is, however, deceptively simple and already the metastability phenomenon is an indication of the underlying complications. The difficulty comes in with the fact that the relevant TD variables are the thickness d of the film and its temperature T; hence, $\tilde{K}_1 = \tilde{K}_1(T,d)$ and $K_2 = K_2(T,d)$ (at that, we have assumed a zero external field). Quite generally, any of these two TD parameters can serve as the driving parameter for the anisotropy flow of the system in the phase diagram of Fig. 1. The experimental situation corresponds to either fixing d and varying T or fixing T and varying d (the variation of d should not be taken too literally, of course). In the first case, one finds a temperature-driven anisotropy flow, while in the second a thickness-driven anisotropy flow occurs. The situation can be suitably approached starting with a $T-d$ diagram which has, however, been delineated in a few experimental cases only such as Fe/Ag(100) (Ref. 12) and Fe/Cu_3Au(001).[13] In view of the experimental difficulties involved, these diagrams cannot be expected to be very detailed. Nevertheless, they provide for abundant information in a condensed form.

In order to move forward with the description of the RT's by means of analyzing the anisotropy flows in the anisotropy space which is now structured by virtue of the TD analysis of the stability of phases of different easy axes, one has to know more about the structure of the quantities $\widetilde{K}_1(d,T)$ and $K_2(d,T)$.

C. Phenomenologic ansatz for the dependence of anisotropy on *d* and *T*

At this point we introduce the usual phenomenological assumption about the structure of the anisotropy constants K_1 and K_2:

$$K_1(d,T)=K_{1b}(T)+\frac{2K_{1s}(T)}{d}, \tag{13}$$

$$K_2(d,T)=K_{2b}(T)+\frac{2K_{2s}(T)}{d}. \tag{14}$$

The subscripts b and s stand for bulk and surface, respectively. While, admittedly, this is a plausible assumption, there are at least three groups of arguments in favor of its large domain of validity. *Intuitively,* this is the simplest possible dependence which leads to a correct dimensionality of the anisotropy as well as to a correct limiting value for very large thicknesses where the bulk and the dipolar anisotropy contributions are the only ones. *Experimentally,* there is a large number of thin-film ferromagnetic (FM) materials where the correctness of this assumption is held up.[2,14] *Theoretically,* general considerations about the thermodynamics of systems in restricted geometry backed up by statistic-mechanical investigations on general microscopic models and by finite-size scaling studies support the assumption (13) and (14) with respect to both the additivity of bulk and surface contributions *and* to the $1/d$ form of the latter (cf. Ref. 15 and references therein).

Note that the thickness dependence thus defined is of the monotonic type and would bring about the type of general restriction discussed above. Second, the more exotic thickness dependence is presented in a very simple *explicit* way. This is in contrast to the temperature dependence which is *implicit* even for the bulk case.[16] A parametric method for the computation of $K_{1b}(T)$ and $K_{2b}(T)$ which is as good as an explicit one has very recently been propounded for a whole class of untrivial theories.[8,17] Even then the temperature dependence of K_{1s} and K_{2s} remains unknown and no effective theoretical framework has ever been suggested. This difference (explicit vs implicit) brings about a corresponding difference in the possibilities for deriving definite results within the framework of the TD analysis by anisotropy flows. Finally, going one level deeper into the structure of bulk and surface anisotropy constants as given in Eqs. (13) and (14), one may discuss the different sources which are known by now. Since this point is not directly relevant to the procedures which follow, we refer to available detailed discussions.[2,9,18–20] Still, it is worth emphasizing that whatever source contributes to K_1 must, in principle, contribute to K_2 as well for *both* bulk and surface anisotropy constants. It is, therefore, amazing that no one has attempted to estimate the *second* surface magnetocrystalline contribution in the manner of Néel.[18]

III. REORIENTATION TRANSITIONS BY THICKNESS-DRIVEN ANISOTROPY FLOWS

We now suppose that the temperature is held fixed and examine the anisotropy flows induced by varying the thickness d of the ultrathin film. With the simple d dependence of Eqs. (13) and (14), one can immediately find the trajectories in the $(\widetilde{K}_1,K_2)$-anisotropy plane. Eliminating d from Eqs. (13) and (14), one finds that the trajectory is a (segment of a) line:

$$K_2=a\widetilde{K}_1+b, \tag{15}$$

where the slope and the intercept are given by

$$a=\frac{K_{2s}}{K_{1s}}, \tag{16}$$

$$b=\frac{1}{K_{1s}}(K_{2b}K_{1s}-K_{2s}K_{1b}+\tfrac{1}{2}\,\mu_0K_{2s}M^2). \tag{17}$$

The anisotropy flow is completely specified if one knows the initial and final states. The simple relation (15) with the accompanying definitions (16) and (17) constitutes the basis for an exhaustive classification as will now be explained.

At this stage, it is advisable to recall the characteristic features of the RT's in ultrathin ferromagnetic films in order to be able to extract the relevant information without exhausting all theoretical possibilities. For very small thicknesses, the surface contribution to anisotropy is very large and dominating. For large thicknesses, the surface contribution is negligible and the magnetization lies within the plane. Hence, the initial anisotropy state must be in the domain of phase 1 in Fig. 1, while the final state is within phase 2. If K_{1s}, K_{2s}, K_{1b}, K_{2b}, and M are known, the initial and final states can be exactly positioned in the diagram. The thickness-driven anisotropy flow is then the segment of the line between these two points. In fact, the domain where the final state belongs can be defined fairly exactly, since to do so one is entitled to neglect the difficult-to-measure surface anisotropy at the final stage. The final point thus lies within a relatively small rectangular target $\mathcal{Z}$ of dimensions $2K_{1b}\times 2K_{2b}$ within the left half of the diagram in Fig. 2, whose vertices have the coordinates $(-\frac{1}{2}\mu_0M^2\pm|K_{1b}|,\pm|K_{2b})$. As to the initial point, *depending on the sign of* K_2, it may be either within the first or within the portion of the fourth quadrant which belongs to phase 1. A glimpse at the phase diagram indicates that the sign of the intercept b in Eq. (15) is immediately relevant to the question whether the anisotropy flow traverses the *origin*,

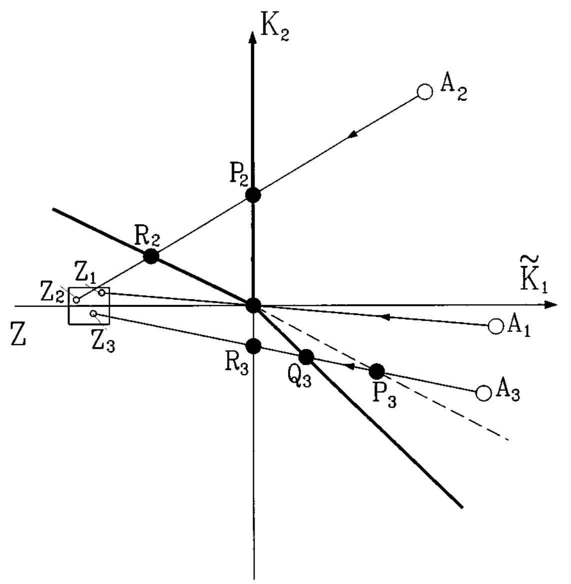

FIG. 2. Generic scenarios for the thickness-driven flows: (1) via the origin, (2) with a positive intercept, (3) with a negative intercept. In each generic case, only one out of three possible subcases is illustrated. Z is the small target where the anisotropy evolution terminates.

the *canted* phase in the second quadrant, or the region with the *metastable* phases in the fourth quadrant. In other words, *three generic cases arise corresponding to whether b is zero, positive, or negative.* In each of these, three subcases arise according to the sign of the slope a. Thus, the anisotropy-flow concept combined with the explicit thickness dependence of $\tilde{K}_1$ and K_2 provides for a simple computational scheme. The route in the anisotropy space tells one what phases will be traversed, while the cross points of the trajectory with the borderlines delineating the different phases lead to simple analytic expressions for the characteristic critical thicknesses in the problem.

The elementary elimination of the thickness in the above derivation suggests a considerable generalization: The thickness-driven flow would be linear for any dependence of the form

$$K_1(d,T)=K_{1b}+2K_{1s}f(d), \tag{18}$$

$$K_2(d,T)=K_{2b}+2K_{2s}g(d), \tag{19}$$

provided that $f(d)\equiv g(d)$. In other words, if that additive part of K_1 and K_2 which depends on the thickness is of the *same functional form*, the anisotropy flow is just as simple as with the inverse-d dependence $[f(d)=g(d)=1/d]$ and is linear of the form (15). This leads to the important consequence that the existence of *three* generic types is guaranteed with an arbitrary thickness dependence.

A. Anisotropy flow traversing the origin

By Eq. (15), such a flow would occur if and only if $b=0$ [Eq. (17)] at the given temperature $T<T_C$ we are considering. Note that the condition $b=0$ involves implicitly the temperature and this makes possible, at least in principle, that the generic type of thickness-driven anisotropy evolution varies for different fixed temperatures so that $b=0$ is attained at some particular temperature. Clearly, this case is not so exceptional and restrictive, since it involves a whole temperature-parametrized manifold of opportunities.

In the present case A, the thickness-driven RT occurs when the trajectory crosses over, upon increasing d, from the domain of phase 1 to the domain of phase 2 via the origin. As no intermediate stable or metastable phases are being traversed, the transition is reversible and abrupt. Analytically, it takes place at $\tilde{K}_1(d_c,T)=0$ and by Eq. (13) this corresponds to a critical thickness given by

$$d_c(T)=\frac{2K_{1s}}{\frac{1}{2}\,\mu_0M^2-K_{1b}} \quad (T<T_C). \tag{20}$$

If one can neglect K_{1b} (or, more precisely, if $|K_{1b}|\ll\mu_0M^2/2$), which is a rather well-satisfied condition for the first-order bulk anisotropy constant, the critical thickness takes on an especially simple form $d_c(T)\approx 4K_{1s}/\mu_0M^2$. This quantity sets a first characteristic scale in the problem. One comes to recognize that its smallness and, hence, the smallness of the characteristic thickness of the ferromagnetic film that would exhibit a RT are dictated by the relative smallness of K_{1s} with respect to the dipolar anisotropy energy.

B. Anisotropy flow with a positive intercept $(b>0)$

All the three subgeneric cases, corresponding to $a>0$, $a=0$, or $a<0$, can be treated on equal footing with the relevant *specifying inequalities* kept in mind [cf. Eqs. (13) and (14) and Fig. 2). Thus, the first subcase which is the most likely to occur in practice is specified by the conditions $K_{2s}/K_{1s}>0,(K_{2b}K_{1s}-K_{2s}K_{1b}+\mu_0K_{2s}M^2/2)/K_{1s}>0$. Now the anisotropy flow A_2Z_2 in Fig. 2 traverses the domain of TD stability of the canted phase. As there are no metastable phases in these precincts, the RT occurs via a continuous change of the canting angle θ. The entrance into and the exit out of phase 3 upon increasing the thickness are at the cross points P_2 and R_2 in Fig. 2, respectively, i.e., at $\tilde{K}_1(d_1,T)=0$ and $K_2(d_2,T)=-\tilde{K}_1(d_2)/2$, where d_1 and d_2 correspond to the onset and completion of the reorientation process, respectively. By Eqs. (13) and (14),

$$d_1(T)=d_c(T)=\frac{2K_{1s}}{\frac{1}{2}\,\mu_0M^2-K_{1b}}, \tag{21}$$

$$d_2(T)=\frac{2(2K_{2s}+K_{1s})}{\frac{1}{2}\,\mu_0M^2-(K_{2b}+2K_{1b})}. \tag{22}$$

The thickness at the entrance to phase 3 has the same formal appearance as the critical thickness d_c of case A, since it is given by the same formal condition $(\tilde{K}_1=0)$. Note, however, that the specifying inequalities are different in both cases. There are further obvious physical restrictions on d_1 and d_2, namely, $0<d_1<d_2$, which, by virtue of Eqs. (21) and (22), are inequalities involving K_{1s}, K_{2s}, K_{1b}, K_{2b}, and M. By Fig. 2, varying the thickness between d_1 and d_2 would drive the system across the canted phase between the points P_2 and R_2. The width of the RT is given by the quantity $\Delta(T)\equiv d_2-d_1$.

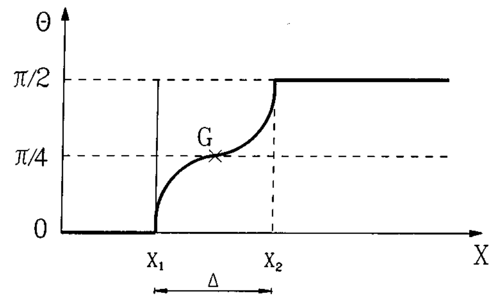

FIG. 3. Evolution of $\theta(x)$ (continuous regime). x is the driving parameter (d or T). The slopes at both ends of the RT are infinite. In the thickness-driven case, the curve is centrosymmetric with respect to the point $G=[(d_1+d_2)/2,\pi/4]$ only when the bulk contributions can be neglected.

If now it may be assumed that $|K_{1b}|\ll\frac{1}{2}\mu_0M^2$, $|2K_{2b}+K_{1b}|\ll\frac{1}{2}\mu_0M^2$, then Eqs. (21) and (22) simplify to $d_1(T)\approx4K_{1s}/\mu_0M^2$, $d_2(T)\approx4(2K_{2s}+K_{1s})/\mu_0M^2$. The width of the RT is, correspondingly, $\Delta\approx8K_{2s}/\mu_0M^2$. The last relation defines a *second* characteristic thickness which is proportional to the *second* surface constant K_{2s}. Since $\Delta>0$, one finds an additional condition $K_{2s}>0$ which must be examined together with the inequalities formulated above. The experimental findings in different systems seem to favor the conclusion that both d_c and Δ are of the order of a few ML's. Even if separate estimates for the two surface constants might appear somewhat risky, there is no doubt that the relation $K_{1s}/K_{2s}\approx2d_c/\Delta$ is very well founded under the conditions of negligible bulk anisotropies. *Vice versa*, if this relation is found to be significantly violated, this might be due to bulk contributions, including eventual large magnetoelastic contributions.

Let us now examine the way in which the canting angle $\theta(d)$ evolves along the linear trajectory of the thickness-driven flow. By the relations specifying thermodynamically phase 3, $\theta(d)$ may be recast as

$$\theta(d)=\arcsin\sqrt{\frac{1-\alpha-d_c/d}{\beta+\Delta/d}}, \tag{23}$$

where the dimensionless quantities α and β are proportional to the first and second bulk anisotropy constants, respectively: $\alpha\equiv2K_{1b}/\mu_0M^2,\beta\equiv4K_{2b}/\mu_0M^2$. The dependence in Eq. (23) would be completely specified if the characteristic quantities α, β, d_c, and Δ are known at the given fixed temperature. The qualitative appearance of a typical curve $\theta(d)$ is known[10,11,5] (cf. Fig. 3). However, one can derive more precise analytic information. In the first place, one finds from the general expression (23) that $\theta(d)$ has an infinite slope at the onset d_1 and at the end d_2 of the transition. Since the coefficients of proportionality (the amplitudes) for the diverging slopes are not equal in general, we have an indication of a certain asymmetry of the curve $\theta(d)$. Indeed, the curve is centrosymmetric with respect to the point $G=((d_1+d_2)/2,\theta=\pi/4)$ in Fig. 3, only if α and β can be safely set to zero, i.e., provided the bulk anisotropy contributions are much smaller than the dipolar one. In this case, the centrosymmetric property is readily established, since $\theta(d)\approx\arcsin\sqrt{(d-d_c)/\Delta}$. The last expression depends on the characteristic lengths d_c and Δ only. This higher symmetry can also be recognized in the equality of the amplitudes of the diverging slopes at d_1 and d_2 in the limit $\alpha,\beta\to0$. Altogether, should it be possible to establish the centrosymmetric property of $\theta(d)$ in a given experiment, one immediate consequence would be that α and β are really very small. On the contrary, the detection of a considerable asymmetry signals substantial bulk anisotropy contributions. Such should inevitably arise from strain-induced anisotropy, if large misfits, inherent to each specific interface, are present.[2,21] Mismatches of the order of 10% correspond to a very large strain on the microscopic scale when compared with typical values, characteristic of bulk strains. Under such extreme circumstances, the strain-induced anisotropy can compete by itself with the dipolar anisotropy or, else, act synchronously with it towards dragging the magnetization direction into the plane of the film, depending on the sign of the relevant magnetostriction constant characterizing the strain-induced anisotropy. Moreover, nonlinear elastic effects will have to be accounted for.

C. Anisotropy flow with a negative intercept ($b<0$)

In this generic case, the anisotropy flow traverses domains of metastability in the phase diagram between the initial (magnetization-up) and final (in-plane magnetization) states. The canted phase is now out of play and the reorientation must take place abruptly. In contrast to the abrupt transition of case A, this transition is irreversible. A system which evolves, upon *increasing* the thickness, from a state with perpendicular magnetization is caught by the local free-energy minimum and would not leave it until it becomes absolutely unstable, i.e., until the minimum disappears at the line $\widetilde{K}_1=0$. Such a reorientation scenario has been detected very recently for Co/Au ultrathin films.[22] The effective variation of thickness has been achieved by using wedge-shape films. Another scheme which would allow in principle a controllable variation in thickness is the electrochemical deposition method.

The three possible subgeneric cases with $a>0$, $a<0$, or $a=0$ can be treated on equal footing. We describe in greater detail only the first subgeneric scenario ($b<0$ with $a<0$; cf. A_3Z_3 in Fig. 2). The defining inequalities become $K_{2s}/K_{1s}<0$, $(K_{2b}K_{1s}-K_{2s}K_{1b}+\frac{1}{2}\mu_0K_{2s}M^2)/K_{1s}<0$. The crossover through the domains of metastability is given by the segment P_3R_3, while the phase boundary is crossed at the point Q_3 upon increasing the thickness. By metastability, the flop of the magnetization direction into the plane would occur at R_3, rather than at Q_3, at the thickness d_2 which is formally given by the same expression as d_1 of the previous generic case A [Eq. (21)], because of the formally identical defining condition $\widetilde{K}_1(d_2)=0$. Note that we keep the subscript 2 for the greater of two characteristic thicknesses which arise in both generic cases. On decreasing the thickness, the in-plane phase would persist up to the point P_3. The thickness d_1 at which the abrupt RT should occur is given by the expression for d_2 of the previous case A, i.e., formally, $d_1=d_2,d_2=d_1$ [cf. Eqs. (21) and (22)]. The thickness d_m which corresponds to the point Q_3 at the phase boundary is defined by the condition $K_2(d_m)=-\widetilde{K}_1(d_m)$. Hence,

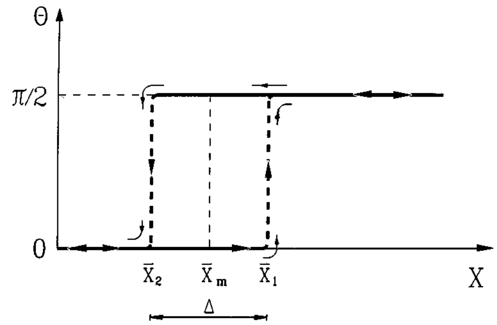

FIG. 4. Evolution of $\theta(x)$ (metastable scenario). The diagram is valid for both d- and T- driven RT's with $x=d$ or $x=T$, respectively. x_m corresponds to the value of d or T at the borderline between the phases involved (cf. Fig. 1).

$$\overline{d_m}(T)=\frac{2(K_{1s}+K_{2s})}{\frac{1}{2}\mu_0 M^2-(K_{1b}+K_{2b})}. \tag{24}$$

The initial defining inequalities are now supplemented by the natural requirements $0<d_1<d_m<d_2$.

In the limit of negligible bulk anisotropy, the width of the metastable region is given by $\Delta=d_2-d_1=-8K_{2s}/\mu_0 M^2$. The thickness d_m corresponding to the borderline between the two phases is precisely the arithmetic mean of the two characteristic lengths: $d_m=(d_1+d_2)/2$. Thus, the limit of negligible bulk anisotropy exhibits once again a higher symmetry of the transition. Altogether, one finds that in the generic scenario with metastability effects the same small characteristic thicknesses which were found in the previous case control the size of the RT, this time, however, with a different physical interpretation.

The evolution of the canting angle $\theta(d)$ is discontinuous at the reorientation points (cf. Fig. 4). The possibility for three generic scenarios for the variation of $\theta(d)$ in the thickness-driven case with a $1/d$ thickness dependence has been recognized in Ref. 11. In view of the generalization of the last paragraph before Sec. III A above, three generic types of variation of $\theta(d)$ will persist with any thickness dependence of the anisotropy constants, provided it is of the same functional form for both K_1 and K_2.

IV. REORIENTATION TRANSITIONS BY TEMPERATURE-DRIVEN ANISOTROPY FLOWS

Now the thickness is assumed to be held fixed and very small, while the temperature is the driving parameter for the flow in the anisotropy space. Experimental examples can be found in Refs. 2, 6, and 7.

As already discussed at the end of Sec. II, the principal difficulty in analyzing the temperature-driven transitions lies with the *implicit* character of the temperature dependence. That is, one cannot describe the anisotropy trajectories as

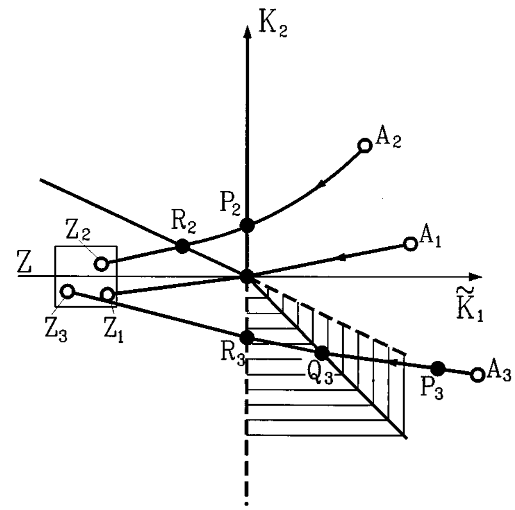

FIG. 5. Generic scenarios for temperature-driven RT's. The trajectories are exemplary and are drawn under the assumption of monotonic variation with T.

parametrized by T. However, in view of the discussion in the opening sections and with the help of the analysis of the anisotropy diagram, one can still predict rather generally some important features of the reorientation in this case. The evolution for the cases of interest (transitions from the magnetization-up to the in-plane states) starts at low temperatures in the domain of phase 1 and finds its way to the domain of phase 2 via either of three possible ways: via the origin, via phase 3, or via the mine fields of metastability in the fourth quadrant. One can safely assume that the variation of K_{1s}, K_{2s}, K_{1b}, K_{2b}, and M with temperature is monotonic. The general considerations of monotonicity put forward in Sec. II B are then valid. Three typifying trajectories under the assumption of monotonicity are given in Fig. 5. The continuity of the flow between the initial and final points of the temperature-driven evolution gives rise once again to *three generic types* of evolution according to the sign of the intercept, i.e., the sign of $K_2(T)$ at the point of change of sign of $\tilde{K}_1(T)$. For the sake of brevity, we do not consider each subgeneric case for the temperature-driven anisotropy flow separately. Rather, we concentrate on some general predictions which are possible even without knowing the temperature dependence of the anisotropy constants.

First, one can get insights into the $\theta(T)$ dependence for the generic scenario via the canted phase (positive intercept) by using the TD information about the cross points in the phase diagram where a given trajectory crosses the boundaries of phase 3 (Fig. 5). One can prove, for instance, that the derivatives of $\theta(T)$ at the entrance T_1 and at the exit T_2 of the RT diverge. The derivation holds in its general form for the case of the thickness-driven transition as well where we proved the same property by using the explicit thickness dependence. Indeed, by Eq. (4),

$$\frac{\partial}{\partial x}[\theta(x,y)]_{|y=\text{ const}}=\sqrt{-\frac{2K_2}{\tilde{K}_1}}\frac{1}{1+\tilde{K}_1/2K_2}\frac{1}{4(K_2)^2}\left(K_2\frac{\partial\tilde{K}_1}{\partial x}-\tilde{K}_1\frac{\partial K_2}{\partial x}\right)_{|y=\text{const}}. \tag{25}$$

We already did explicitly the case with $x \equiv d$ and $y \equiv T$. The same formal expression (25) holds with $x \equiv T$ and $y \equiv d$. In both cases the denominators in the first two factors get to zero at the entrance point into and at the exit point out of the canted phase by virtue of the defining equations for the TD boundaries of the neighboring phases involved and regardless of whether the transition is driven by thickness or temperature variation. One thus has a proof, based on general arguments of stability of phases, that the variation of the angle in either of both cases is very steep with an infinite slope in the relevant variable at the onset and at the end of the reorientation. A typical curve for $\theta(T)$ in this temperature-driven case would look like the corresponding curve for $\theta(d)$ in the thickness-driven case (Fig. 3), as far as the qualitative shape and the infinite slopes at T_1 and T_2 are concerned. Otherwise, the arcsine curve $\theta(T)$ would typically be asymmetric.

Second, one could proceed to analyze the range ΔT over which the RT is accomplished. One could proceed along the following lines in order to determine the conditions which would bring about a small range ΔT, consistent with theoretical predictions.[5] By now, we observed that a small ΔT means that the portion P_3R_3 in Fig. 4 is covered at a large speed or rate of change. Exploiting the implied mechanical analogy to the full, one may formulate the following criterion: Any portion of a particular trajectory $K_2(\tilde{K}_1)$ will be covered "quickly," if the rate of change as given by the expression

$$v(T) = \sqrt{(\dot{K}_2)^2 + (\dot{\tilde{K}}_1)^2} \tag{26}$$

is large along this portion, i.e., $v(T) \gg v_0(T)$, where $v_0(T)$ is some reference rate of change. In Eq. (26) and below, the dot above a given quantity denotes the derivative with respect to the driving parameter (here T). A self-suggesting choice for the reference speed would be to consider the quantity, corresponding to the physical situation under discussion but neglecting the surface terms. Imposing a high rate of change by the inequality $v^2 \gg v_0^2$, one can derive the following sufficient conditions for the smallness of ΔT: $|\dot{K}_{1s}|/d \gg \mu_0 M |\dot{M}|, |\dot{K}_{2s}|/d \gg |\dot{K}_{2b}|$. That is to say, an expectation of a small ΔT would be met if the rates of change of the first and second surface anisotropy energies are larger separately than the rates of change of the dipolar anisotropy energy and the second bulk anisotropy energy, respectively. All inequalities involving rates of change could be expressed in terms of the corresponding derivatives with respect to magnetization, if one assumes, like in the bulk case, that the temperature dependence enters the various anisotropy terms only via the magnetization, i.e., $K_{1s} = K_{1s}(m(T))$, $K_{1b} = K_{1b}(m(T))$, etc.[23]

Third, we comment briefly on the connection of this type of analysis with existing theoretical predictions based on statistic-mechanical calculations. In a series of interesting papers,[5] Moschel and Usadel investigate the temperature-driven RT in a system of a few ML's from a statistical-mechanical point of view. These studies involve the mean-field analysis of a quantum model of interacting spins, whereby dipolar and single-ion anisotropies are explicitly examined. A temperature-driven reorientation was found to take place only in a narrow window of the microscopic parameter values. The range ΔT where the reorientation occurs was also found to be very narrow. These authors determine $\theta(T)$ in the region of the reorientation for various relative magnitudes of the single-ion anisotropy in the different layers. Both untrivial generic cases of RT's via the canted phase of via the metastable region were detected. The first-order (irreversible) transition was found to occur as an exception only if all (microscopic) anisotropy parameters had the same value. It is obvious from Ref. 5 that the overall, macroscopic anisotropy constants for their model can be determined, since these authors dispose of the free energy as a function of the canting angle $\theta(T)$.[24] It would be of much value to analyze the temperature dependence of the phenomenological anisotropy constants $\tilde{K}_1$ and K_2 from the computed free-energy dependences and thus give a microscopic support for the TD analysis or else delineate its shortcomings. It is, however, beyond doubt that the sign of the intercept of the trajectory in the $(\tilde{K}_1, K_2)$ plane must correlate with the type of RT (positive for the reversible, negative for the irreversible case). This would then mean that a uniform distribution of the single-ion microscopic anisotropies as given and treated in their microscopic model invariably produces a negative second-order anisotropy energy K_2.

V. DISCUSSION

We have demonstrated that the anisotropy-flow concept combined with the proper account of the structure of the anisotropy space provides for a unified understanding and classification of both thickness- and temperature-driven RT's. In both cases, there are three generic types of RT depending on the route of the corresponding system in the anisotropy space.

In the thickness-driven case, the trajectories are explicitly seen to be linear in the anisotropy space under the usual assumption for the $1/d$ variation of the anisotropy constants. Thus, three generic scenarios are identified and described according to the sign of the intercept. Physically, a zero intercept corresponds to a cross point from the perpendicular into the in-plane phase which coincides with the origin, and a positive intercept induces a continuous crossover via the canted phase, while a negative intercept corresponds to an anisotropy flow via the domains of metastability in the fourth quadrant (Fig. 1). In each of the generic cases, there are three subcases according to the sign of the slope of the linear trajectory. Each particular regime is specified by a set of inequalities involving K_{1s}, K_{2s}, K_{1b}, K_{2b}, and M which constitute an indispensable part of the analysis. These are supplemented by further natural restrictions for the pertinent characteristic thicknesses. The necessary condition for an untrivial scenario to occur is thus a nonzero intercept (an effectuation of the "trivial" scenario does not require that K_2 be identically zero; it suffices that K_2 be zero at the crossover point only). In both the continuous and the metastable untrivial cases, *two* characteristic thicknesses are identified by examining the cross points into and out of the intermediate phases (the canted phase for the continuous, the metastable domain for the discontinuous case). These thicknesses are, conveniently, the critical thickness d_c at which the reorientation begins and the width Δ of the RT. Furthermore, the evolution of the canting angle $\theta(d)$ has been analytically described and it has been found that it has divergent slopes at both ends of the transition. Provided that the bulk contributions can be neglected, the characteristic lengths take on an especially simple form controlled by K_{1s} and K_{2s}, respectively, with the ratio of the first and second surface anisotropy constants being simply expressed as twice the ratio of d_c and Δ. In the same limit of negligible bulk contributions, the RT exhibits a higher symmetry: (i) In the continuous regime, the $\theta(d)$ curve is centrosymmetric and has equal amplitudes for the divergences at both ends (Fig. 3); (ii) in the discontinuous regime, the limits of absolute instability of the neighboring phases are symmetric with respects to the phase boundary dividing them (Fig. 4). This information can be used in both directions: If it is found in experiment that the said symmetries exist, then the bulk contributions should be very small; if a substantial deviation from the above symmetries is detected, the bulk contributions are considerable and non-negligible. The natural formulation in terms of flows in the anisotropy space allows for a substantial generalization. Indeed, the same three generic types of behavior will occur with any thickness dependence of K_1 and K_2 (and not only with the usual inverse-*d* ansatz), given the natural assumption that the dependence is of the same functional form for both K_1 and K_2.

In the temperature-driven case, the temperature dependence of the anisotropies involved is not known with sufficient degree of certainty, which is why the anisotropy flows can be only qualitatively discussed. Still, very general conditions such as monotonicity of the temperature dependence or the conservation of the sign of one or more anisotropy constants under the anisotropy flow may serve to locate the domain in the anisotropy space where the RT takes place. One finds once again three generic types of behavior corresponding to the intermediate region in the anisotropy space which is being crossed between the initial and final states. In the continuous reorientation regime and despite the implicit character of the temperature dependence, we prove by general arguments that the slopes of $\theta(T)$ at both ends of the RT are infinite. The derivation is valid in its general form for the thickness-driven case as well which is a further generalization beyond the $1/d$ phenomenological assumption. Since experimental and theoretical evidence diverge on the question of the relative width ΔT of the temperature-driven RT's, we derive sufficient conditions for the eventual smallness of ΔT by evoking a self-suggesting mechanical analogy.

There is another set of important conclusions which are related to the construction of the (T,d) diagram of systems exhibiting a RT. Because of the experimental difficulties, only one borderline, dividing the perpendicular and the in-plane phases, can be seen in reported diagrams.[12,13] Strictly speaking, this implies that $K_2 \equiv 0$ for all T, so that only the critical line $d_c(T)$ matters. However, as we have seen in Secs. III B and III C above, there are *two* such critical lines, namely, the pairs $d_1(T), d_2(T)$ and $\tilde{d}_1(T), \tilde{d}_2(T)$ for the continuous or discontinuous scenarios, respectively. The first pair defines the boundaries of the canted phase, while the second pair delineates the domain of metastability in the (T,d) representation. In the lack of knowledge of the explicit temperature dependences, one is free to speculate educatedly about the relative positioning of the curves within each pair. A brief thought indicates that both the continuous and the discontinuous scenarios would occur for *the same ultrathin system* but in different domains of its (T,d) diagram, if the critical curves defined above cross at some point (T_0, d_0). This very point is defined by the conditions $\tilde{K}_1(T_0,d_0)=0$, $K_2(T_0,d_0)=0$. One might expect especially intriguing behavior in the vicinity of this point, should it happen to occur in a given particular system. It is precisely at such a point that there is *no anisotropy* in the system (K_2 is zero, while K_1 is compensated by the dipolar anisotropy). The (T,d) diagram of a system with such a point would be split into a paramagnetic phase, an in-plane ferromagnetic phase, a perpendicular phase, a canted phase, and a domain of metastability. It is clearly seen that both types of presentations [the one with the anisotropy flows and the one in the (T,d) diagram] are complementary and can be used rather successfully for the prediction of new features. This issue will be further discussed separately.

The anisotropy-flow concept can be further elaborated in several respects without significant increase in mathematical complexity. First, ''bilinear'' angular dependences as found in the pioneering paper by Néel[18] for different orientations of surfaces of bulk cubic materials may be included in the phenomenological free energy. Even in the most complicated case this would amount to a dependence on one more angular variable which must then undergo minimization. This is equivalent to the inclusion of in-plane anisotropies. Second, the lucid presentation allows one to come to the natural conclusion that whatever source of surface or bulk anisotropy contributes to K_1 would inevitably contribute to K_2 as well. In this context, it seems astonishing that the Néel contribution to K_{2s} has not been found theoretically by now. As the estimate $K_{1s}/K_{2s} \approx 2d_c/\Delta$ indicates, if $d_c \approx \Delta$, which has indeed been found for a number of systems with a RT, then K_{2s} must be comparable to K_{1s} and, hence, the same should be true for the magnetocrystalline (Néel) contributions to K_{1s} and K_{2s}. Third, the free-energy expression might be extended to include the third-order anisotropy constant with the corresponding bulk and surface additive contributions. This is not an academic option only as can be seen by examination of certain *ab initio* results (cf. e.g., Fig. 2.7 on p. 46 of Ref. 25). Fourth, the influence of an externally applied field on the RT (field-induced RT) can be systematically studied. Finally, it is well known that a complicated domain structure arises in ultrathin ferromagnetic films and that its description is especially difficult precisely in the region where the RT takes place.[3,6,7,10,26] The motivation and the results of the present paper are especially appealing in the perspective of understanding the domain structure in systems with a RT as completely as possible. Apart from the identification of the peculiar point in the (T,d) diagram, which is bound to induce peculiarities in the domain structure because of the effectively nonexistent anisotropy there, one has to understand the underlying anisotropy-related phenomena before attempting to explain the much more complex domain structure.

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