JOURNAL OF APPLIED PHYSICS VOLUME 83, NUMBER 11 1 JUNE 1998

The field-space perspective on hysteresis in uniaxial ferromagnets

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A procedure for the analysis of hysteresis in the **H** space of a uniaxial ferromagnet with higher-order anisotropy is put forward. The formulation is valid to any order n in the anisotropy expansion. The critical boundaries separating stable from metastable states are cast in a formally decoupled parametric way as $H_x = H_x(M_x)$, $H_z = H_z(M_z)$. The analytic expressions provide the basis for the construction of generalized astroids to any order. For $n > 1$, new features are found and interpreted in their relation to rotational hysteresis and possible spin-reorientation transitions in uniaxial materials. The shape and symmetry of the critical boundaries depend crucially on up to $n-1$ independent ratios of the anisotropy constants against a suitable normalizing quantity; the normalizer can be any from among the set of constants or any linear combination thereof. Self-crossing of an astroid indicates the existence of additional extrema and, hence, of complicated hystereses. © *1998 American Institute of Physics.* [S0021-8979(98)23911-9]

I. INTRODUCTION

Hysteresis is a complex nonlinear phenomenon of delicate sensitivity to the past states of the system.[1] In ferromagnets whose anisotropy could be reasonably well characterized by a phenomenologic free energy expansion $F(\theta,\phi)$ in the angular variables of the saturation magnetization **M**, hysteresis holds place even if one allows for homogeneous rotation of **M** only.[2] There are certain advantages with the analysis of a uniaxial system in its field space. For instance, one can determine the boundaries of stability of states corresponding to different minima of the anisotropy energy for *any* field direction and not only for the two principal configurations with field parallel or perpendicular to the axis of symmetry. The case with only the lowest term in the anisotropy expansion has been systematically discussed and implemented.[1,3] The only exceptions dealing with astroids in higher orders are Refs. 4 and 5 concerning a system with two anisotropy constants. In the following, we demonstrate that this type of analysis is easily extended to *any* order in the anisotropy expansion due to a special feature of the problem at hand, so that the first two orders are deduced as particular cases. The procedure gives rise to field-space diagrams which will be referred to as generalized astroids. Self-crossing of the boundaries of stability, found already in the second order,[4,5] arises over large domains of values of the anisotropy constants and is the most pronounced feature of this generalization. It signals the existence of complicated hysteresis behavior due to the emergence of additional competing energy minima.

II. GENERALIZED ASTROIDS: ANALYTICAL AND EXAMPLES

One carries out the field-space analysis with the phenomenological thermodynamic potential of the type $g_A = f_A + \epsilon_Z$, where $\epsilon_Z = -\mathbf{M}\cdot\mathbf{H}$ is the Zeeman term, while

$$f_A(\theta;\{a_j\}) = \sum_{k=1}^{\infty} a_k \sin^{2k}\theta \tag{1}$$

is the full expansion for the anisotropy energy density of a uniaxial ferromagnet.[3] The $\{a_j\}$'s are the anisotropy constants of order j, while θ is the angle between **M** and the crystallographic axis of cylindrical symmetry **n**.

In the assumed symmetry, all three relevant vectors **n**, **H**, and **M** lie in the same plane, since there is no torque to drive the magnetization out of the plane (**H,n**). The remagnetization processes are confined to this plane. This makes all results directly applicable to situations, typical of thin ferromagnetic films, where the magnetization vector is confined within the plane of the film because of the strong dipolar contribution.[6] We choose the z axis along **n** with $\mathbf{M} = (M_x, 0, M_z)$ and $\mathbf{H} = (H_x, 0, H_z)$. To find the stable directions of **M** under applied field, one has to look for extrema of $g_A(\theta)$ by solving $g'(\theta) = 0$ and requiring that $g''(\theta) \geqslant 0$ at the eventual solutions of the extremal equation. In zero field, there is always more than one stable solution, while in sufficiently high fields, the Zeeman energy favors conforming alignment of **M** and **H**. So the boundaries between perfectly aligned and competing equilibrium states are at some finite magnitude of field which may vary with field direction. It is determined by the simultaneous consideration of the conditions $g'(\theta) = g''(\theta) = 0$ by inserting the expansion from Eq. (1). One observes that this system of two equations is linear in the components of the field, so that, solving for H_x and H_z, one obtains

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$$H_x=\frac{2}{M}\sum_{k=1}^{\infty} ka_k \sin^{2k-1}\theta[2(1-k)+(2k-1)\sin^2\theta],$$

$$H_z=\frac{2}{M}\sum_{k=1}^{\infty} ka_k(1-2k)\sum_{p=0}^{k-1}\binom{k-1}{p}(-1)^p\cos^{2p+3}\theta. \tag{2}$$

This is obviously a parametric solution of the type $H_x=H_x(\theta)$, $H_z=H_z(\theta)$ which describes the boundaries of stability in the field space of the uniaxial system to arbitrary order. A seemingly redundant complication has arisen in passing from $\sin\theta$ to $\cos\theta$ in Eqs. (2). This was motivated by the desire to cast the parametric solution in a formally decoupled way as $H_x=H_x(M_x)$, $H_z=H_z(M_z)$. The form serves to identify correspondence rules between the particular principal cases of fields applied along **n** or perpendicularly to it. The rules come about by comparing identical powers in M_x and M_z in the two equations for the respective field components. They save half of the labor in exploring the phase diagrams in **H** space, since features of given symmetry for a given set of anisotropy constants $\{a_k\}$ must be identical, up to permutation of H_x and H_z axes, for the *corresponding set* of constants. This is in fact a symmetry argument which has escaped attention in the earlier field-space (astroid) studies; here, one deduces it from the general solution to arbitrary order.

A further general observation, valid to any order, is borne out by looking in turn at the two principal configurations $\mathbf{H}\|\mathbf{n}$ and $\mathbf{H}\perp\mathbf{n}$. In the first case, the conforming solution ($\mathbf{H}\|\mathbf{M}$) is $\sin\theta=0$ and is stable, *to any order*, for $H\geqslant H_{A1}$ with the anisotropy field $H_{A1}=2|a_1|/M$ depending on a_1 alone despite of considering the contributions from all orders. In the second case, the conforming solution is $\cos\theta=0$ and is stable for $H\geqslant H_{A2}$ with $H_{A2}=2|\Sigma_{k=1}^{\infty}k\cdot a_k|/M$. As a very interesting consequence, in a system with a spin-reorientation transition, which would occur in a given anisotropic ferromagnet if $a_1\rightarrow 0$ under variation of some parameter like temperature etc., the anisotropy in the direction of the symmetry axis **n** becomes soft ($H_{A1}\rightarrow 0$) which gives rise to a number of peculiarities. Either H_{A1} or H_{A2} may serve as natural normalizing quantities for the generalized astroids to arbitrary order, but the choice is not restricted and may be varied, should particular considerations of simplicity hold in a particular system. Generally, the precise shape of the boundaries in **H** space depends on up to $n-1$ constitutive ratios to order n. In principle, the part of a normalizing quantity may be played by any linear combination of anisotropy constants.

The predictive power of the **H**-space analysis to higher orders depends crucially on the fact that the tangent construction[1] for the determination of the hysteresis along any route in field space remains valid even with anisotropy constants of higher orders. It is indeed a substitute for the numerical solution of a high-degree polynomial equation. The existence of more competing states as one goes to higher orders in the anisotropy energy gives rise to rather complex generalized astroids whose generation by Eqs. (2) can, however, be performed pretty easily.

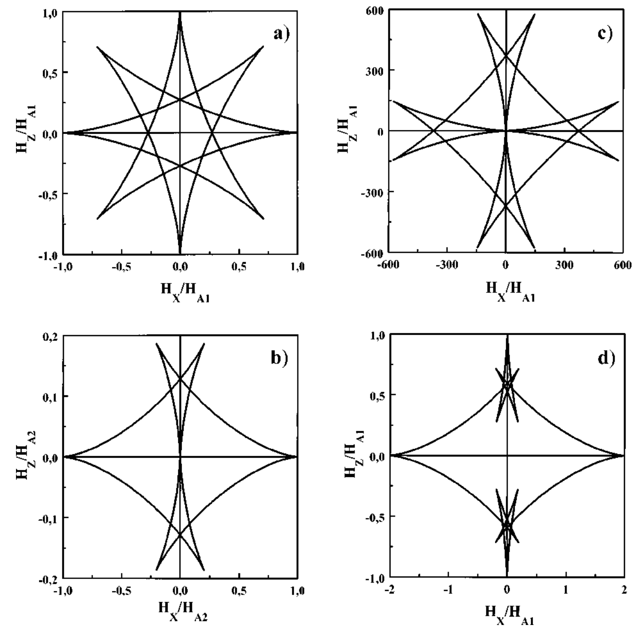

FIG. 1. (a) The octopole astroid ($a_1=-a_2$, $a_3=0$). The exceptionally high eightfold symmetry derives from the exchange of stability along this special line in the anisotropy space of the system (Ref. 8). (b) Swallow tails due to softening of the anisotropy axis **n** for a system with a spontaneous orientational transition. (c) Generalized astroid for a system with both anisotropy fields gone soft ($a_1\approx 0$, $a_1+2a_2+3a_3=0$). Note the squarelike aspect of the ''mainframe.'' (d) A generalized astroid with all $|a_{1,2,3}|$ equal ($a_{1,3}<0$, $a_2>0$). Self-crossings of this type imply complicated secondary hystereses.

We proceed to illustrate the general method by several examples up to order $n=3$ in the anisotropy expansion. For bulk systems, the necessity of considering three orders in the expansion for a uniaxial system arises in the analysis of the behavior of the rather important group of highly anisotropic rare-earth–transition-metal compounds.[7] The examples are indicative of the variety of nontrivial cases which are in store when higher-order anisotropies are non-negligible. Consider the case with $a_3=0$ first. A symmetric astroid results when $a_1=-a_2$ [Fig. 1(a)]. This condition corresponds to the line in the anisotropy space along which a stable canted solution exists ($a_1<0$) or there is a pair of coexisting minima of equal depth ($a_1>0$).[3] The symmetry is exceptionally high with the vertices of the octopolelike astroid lying on a circle.[8] All cases which are of the self-crossing type may be viewed as resulting from continuous deformations of the most symmetric case upon variation of the ratio a_2/a_1. Generalizing these observations, the most symmetric astroids to any given order of the expansion will be realized on the manifolds where, in zero field, the eventual minima transform into each other continuously or exchange stability, while coexisting. Now consider the case with $a_2\neq 0$, $a_1=a_3=0$. This is a two-constant approximation where, additionally, the lowest order contribution goes to zero as would be the case in systems exhibiting orientational transitions in zero field. The behavior in the principal field configurations has been elucidated in great detail.[9] The astroid in Fig. 1(b) sheds light on the behavior of such a system under an applied field of *arbitrary* direction. One observes the ''softening'' of the anisotropy field H_{A1} in that the swallow tails adjacent to the **n** axis run smoothly down to the origin. By the same token, the direction perpendicular to **n** will become soft whenever the linear combination of constants defining H_{A2} goes to zero. Thus, swallow tails along the H_x axis will run to the origin on the hyperplanes in the anisotropy space defined by $\Sigma_{k=1}^{n} k \cdot a_k = 0$ in order n. This is corroborated by Fig. 1(c) where the condition $a_1 + 2a_2 + 3a_3 = 0$ is imposed. Since a_1 is intentionally very small for this plot, the same figure typifies also the situation at an orientational transition [unlike Fig. 1(b), here $a_3 \neq 0$]. Now both H_{A1} and H_{A2} are zero, hence, the symmetric outlook of the cross between the two complete swallow tails. Note that the ''mainframe'' of the astroid in Fig. 1(c) is very nearly square-shaped. This last feature comes up also with sets of constants which do not lead to self-crossing; we have observed it, e.g., with $a_2/a_1 \approx -1/8$, $a_3/a_1 \approx 1/2$. The outcome of all constants being equal is presented in Fig. 1(d) ($|a_1| = |a_2| = |a_3|$, $a_{1,3} < 0$, $a_2 > 0$). Complicated self-crossings imply nontrivial secondary hystereses as one already knows from the lower-order treatments.[4,5]

III. DISCUSSION

The method described above can be used to study the possibility of eliminating undesirable hysteresis in uniaxial materials by applying stress in a suitable direction. The idea has been put forward in the context of cubic magnetostrictive materials,[10] but it applies to any symmetry, in principle. The advantage of the uniaxial setting is that there is a single angular degree of freedom, while in cubic symmetry two such degrees are relevant and the analysis relies on an expansion in small deviations from the symmetry axis. The analysis can be performed most easily for stresses along or perpendicular to **n**. Since the stress-induced contribution is always of the lowest order, it shifts a_1 by an amount $\tilde{a}_1$. For stress σ along **n**, the shift is $\tilde{a}_1 = -\sigma \lambda^{\alpha,2}$ for both tetragonal and hexagonal symmetry; here, $\lambda^{\alpha,2}$ is the magnetostrictive coefficient for the mode of change of length along **n**.[11] Note that the sign of the shift can be controlled, for a given material (i.e., for a given $\lambda^{\alpha,2}$) by applying tensile or compressive stress. The hysteresis for driving fields along one of the two principal directions can be diminished and, eventually, eliminated if the anisotropy field along the other (conjugate) axis is much smaller than along the direction of the driving field. That is, make one of the axes (say, **n**) anisotropically soft by applying stress; then the axis perpendicular to **n** is automatically the (relatively) harder one; hence, remagnetization along it would be as closely hysteresis-free as the **n**-axis is soft. Since there are two options for the quantities to be modified by stress, H_{A1} and H_{A2}, and two further options for the sign of $\tilde{a}_1$, one recognizes four generic possibilities as a framework for selecting suitable material parameters for prospective applications.

The study of hysteresis by the described generalized-astroid construction is readily applicable to ultrathin ferromagnets as well. In these, there is a strong enhancement of the lowest-order anisotropy due to broken crystallographic symmetry at the surface whereby the anisotropy per surface atom is typically about an order of magnitude larger than in the bulk.[12] In both bulk and thin-film uniaxial systems, the consideration of generalized astroids in applied field may offer valuable insights, especially when a spin-reorientation transition is to be analyzed [cf. Figs. 1(b) and 1(c)]. In the vicinity of such transition points, higher-order anisotropies come into play.

Apart from the prospective applications described in the above paragraphs, the general solution to arbitrary order given in Eq. (2) provides for an overview on the whole problem by: (i) reducing the calculation in any particular order to a *deductive* procedure whereas the $n=1$ and $n=2$ solutions appear as special, *ad hoc* constructions independent of each other; (ii) leading straightforwardly to correspondence rules valid to any order by virtue of the formally decoupled solutions $H_x(M_x)$ and $H_z(M_z)$; and (iii) uncovering to any desired order the possibility for, and the mechanism of, making one of the principal axes anisotropically soft and thus allowing the identification of hysteresis-free conditions. Finally, the field-space perspective should be viewed as complementary to stability analyses in the anisotropy space of the system[13] whereby only the union of both approaches may yield as detailed description as possible and/or required.

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