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Magnetic Thin Films I

Positive and Negative Anisotropy in Nickel-Iron Films

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Nickel-iron bulk and film alloys annealed or deposited in a dc magnetic field develop twofold uniaxial magnetic anisotropy with the easy axis parallel to the dc magnetic field (positive anisotropy). Furthermore, in films, a small portion of the material may have a twofold easy axis at right angles to the applied dc field (negative anisotropy). Negative anisotropy has been deduced from Bitter pattern studies of locking (counter-rotation of $\boldsymbol{M}$ when reversing fields $\boldsymbol{H}$ are applied). The existence of locking in cases where $\boldsymbol{H}$ makes a large angle to the easy axis implies large-angle dispersion in the orientation of $\boldsymbol{M}$; however, the easy-axis orientation can independently be shown to have only small angular dispersion. These apparently contradictory facts are resolved by assuming regions of negative anisotropy which themselves have small angular dispersion. The origin of positive anisotropy is directed pairs of Ni or Fe atoms; the origin of negative anisotropy is not known at this time although certain film preparation variable can be specified which lead to films with negative anisotropy.

1. Introduction

Nickel-iron films made by vacuum deposition, cathodic sputtering or electrodeposition are found to possess twofold uniaxial magnetic anisotropy. Furthermore, the direction of the easy axis can be controlled by deposition in a *dc* magnetic field $\boldsymbol{H}$. The anisotropy of the major part of the film has an easy axis which is parallel to the depositing field and is described by $E_k = K\sin^2\theta$ where θ is the angle between $\boldsymbol{H}$ and the magnetization $\boldsymbol{M}$ and K is a positive number; hence the designation positive anisotropy. In addition, it is found that for a small fraction of the film the easy axis of the local uniaxial anisotropy may be at right angles to the depositing field; this can be described by taking K as a negative number, and hence the designation negative anisotropy.

Strictly interpreted negative K implies that the entire plane normal to a hard axis is an easy plane. This has not been demonstrated from the experiments, which only show that there are local easy axes in the film plane which are perpendicular to the average easy axis.

Positive anisotropy is principally due to directed-pairs of Ni or Fe atoms, and the present status of this problem will be summarized in Section 2. The presence of negative anisotropy has been deduced from "high-angle locking" experiments which are described in Section 3; the origin of negative anisotropy is not known at this time although the preparation parameters which tend to lead to films having negative anisotropy are also specified in Section 3.

2. Positive Anisotropy

The theory of directed-pair ordering for binary alloy has been given by Néel[1] and Taniguchi[2]; a twofold uniaxial anisotropy K is predicted which is proportional to $(T_C - T_A)$ $c^2(1-c)^2$, where T_C is the Curie temperature, T_A the temperature of anneal, and c the concentration of either alloy component. Experimental polycrystalline data for K as a function of composition for vacuum evaporated films (after Takahashi, Watanabe, Kono and Ogawa[3]) and bulk (after Ferguson[4]) Ni-Fe alloys are shown in Fig. 1-a. Since the maximum of $c^2(1-c)^2$ and T_C occur at 50% and 70% Ni, respectively (Fig. 1-b), maximum K occurs at intermediate compositions. For $T_A = 450°C$, the time for equilibrium in bulk material is ≈ 166 hours so that data for lower T_A cannot be obtained; the curve for $T_A = 350°C$ was obtained by linear extrapolation

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from the data of Ferguson. The relation between the bulk annealing temperature T_A and the film substrate temperature T_s is not clearly defined. Thus if bulk activation energies were required during film formation no uniaxial anisotropy should develop due to the low value of T_s and the short time for deposition (≈100sec). However, the effective film activation energy is not so low that final equilibrium is reached, since the film anisotropy for $T_s=300°C$ is less than the extrapolated bulk anisotropy for $T_A=350°C$ (see Fig. 1-a).

Although a general understanding of uniaxial anisotropy in film and bulk alloys has been obtained, major problems remain. Outstanding among these are: 1) The non-zero anisotropy of pure Ni film (Fig. 1-a);[5,6] 2) the low temperature (<300°C) at which annealing with $\boldsymbol{M}$ in the hard-axis direction can cause changes in K[7]; 3) the detailed behavior of bulk and film single crystals;[8] 4) the role of oxygen which must be present in a few parts per million in order for uniaxial anisotropy to develop in bulk material[9]; 5) the presence of local regions with negative anisotropy.

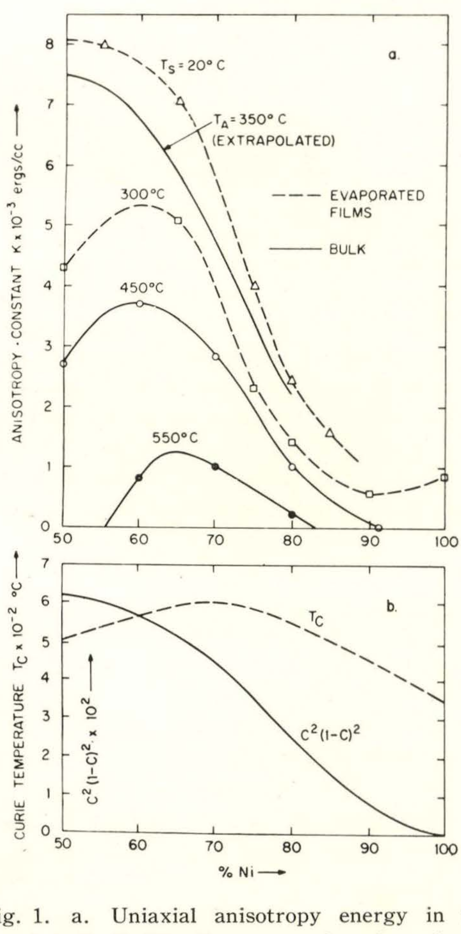

Fig. 1. a. Uniaxial anisotropy energy in film and bulk Fe-Ni alloys as a function of composition. After Takahashi, Watanabe, Kono, and Ogawa [3] and Ferguson [4]. b. Curie temperature T_c and pair-probability factor $c^2(1-c)^2$ as a function of composition.

3. Negative Anisotropy

Bitter pattern studies have led to the assumption of small local regions of negative anisotropy in a wide variety of Ni-Fe films[10]. The clearest case is for films near the composition for zero magnetostriction (81-19) for which the coercive force H_W is greater than the uniaxial anisotropy field $H_K=2K/M$. The condition $H_W>H_K$ implies positive inhibition or locking of the uniform rotation which should occur in an applied field equal to H_K. The Bitter pattern of a film in the "locked" state is shown in Fig. 2-a in which pairs of wall segments perpendicular to the easy axis can be seen. A postulated flux pattern of counter-rotating regions of $\boldsymbol{M}$ is shown in Fig. 2-b; complete rotation is inhibited by magnetostatic interaction and hence H_W can exceed H_K. From this model the initial stages of locking should be reversible, which is in accord with experiment.

Counter-rotation of $\boldsymbol{M}$ might be attributed to small-angle ripple in $\boldsymbol{M}$ generated by a corresponding small-angle dispersion in the orientation of the local positive anisotropy. The existence of ripple has been shown directly by the electron microscope pictures of Fuller and Hale[11] in which the contrast is due to direct interaction between $\boldsymbol{M}$ and electrons passing through the film. Small-angle dispersion can also be measured by study of the remanent state of a film after saturation near the hard axis: If δ is the angular spread in easy-(or hard) axis orientation, then for a film saturated in a direction exceeding δ from the hard axis the remanent state will be single domain pointing in the nearest easy-axis direction; otherwise a multi-domain remanent state will be found. Measurement of δ by this method for the film of Fig. 2 gives $\delta \approx 4°$.

Thus, as far as small-angle locking is concerned, there is no need to introduce the idea of negative anisotropy. However, the

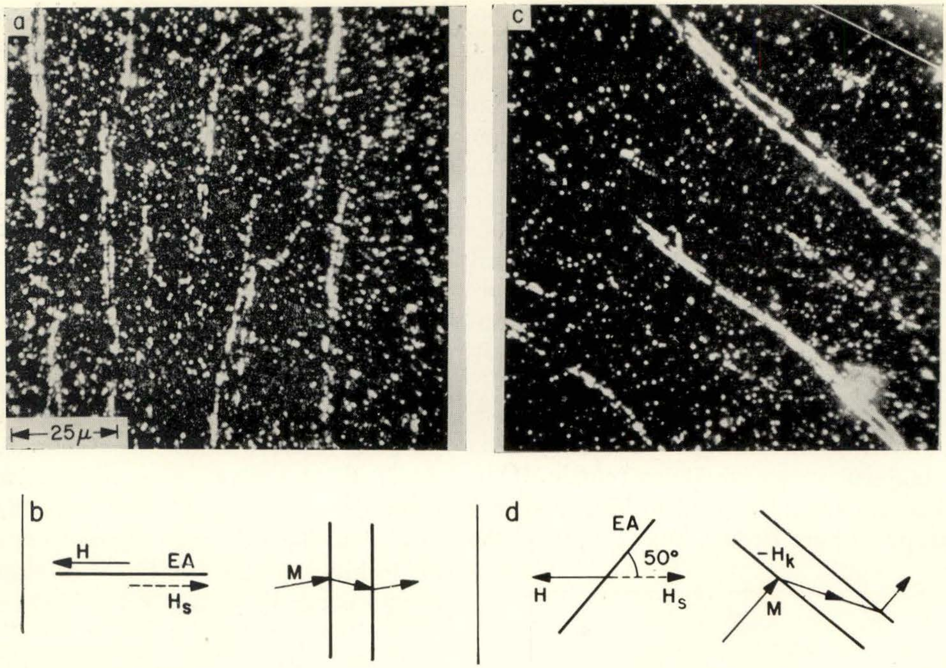

Fig. 2. Negative-H_k regions inferred from high-angle locking (%Ni=85; T_s=300°C; t ≈1100 Å; R ≈ 30 Å/min; H_w=7.0 Oe, H_k=5.6 Oe, h_w=1.3): a. Locking near the easy axis; b. Postulated flux pattern for (a); c. High-angle locking; d. Postulated negative-H_k region to explain (c).

experiment of Fig. 2-c shows the phenomenon of high-angle locking to be present in the same film. In this case the film is saturated at a large angle to the easy axis (55° in Fig. 2-c); when a field in the opposite direction is applied a locking pattern again appears which is oriented *exactly the same* as for small-angle locking (Fig. 2-c). Clearly, if this is to be explained in terms of easy-axis dispersion, δ must be at least 55°; but δ has already been determined as only ≈4° as described above. This inconsistency can be resolved by assuming that regions of negative anisotropy exist which themselves have a dispersion only of ≈4°. Such regions can cause large local kinks in $\boldsymbol{M}$ (Fig. 2-d) in zero field, and also not influence the fall back of M in either easy-axis direction after hard-axis saturation during the measurement showing small δ.

High-angle locking has only been observed in films for which $H_W > H_K$; however, high-angle locking does not always occur when $H_W > H_K$. In some cases a different type of locking may occur, namely the locking pattern may be oriented with respect to the saturation direction instead of the easy axis, a situation which can be described as isotropic locking; as would be expected in such films, δ is no longer small and the results of measurement of δ by the usual methods become ambiguous.

Preparation of films which exhibit high-angle locking is difficult to do reproducibly. In general, it is found that H_W increases and high-angle locking can be observed when $H_W > H_K$ as the following preparation variables, which are being studied over the ranges indicated, are changed as indicated: decreasing rate of deposition (1000 Å/min to 50 Å/min), increasing substrate temperature (25°C to 300°C), increasing thickness (200 Å to 2000 Å), compositions with increasing negative magnetostriction constant λ (75% to 90% Ni). If any of these variables are changed beyond characteristic and interdependent limiting values, a rapid transition of film properties occurs which results in: a drastic increase in H_W, hysteresis loops of a unique shape, and "mottled" Bitter patterns[12]. The Bitter pattern of Fig. 2 is an example of a slight amount of mottling.

Although high-angle locking is more predictable for compositions having negative magnetostriction, it does not seem to necessarily follow that negative H_K is due to strain since high-angle locking patterns similar to Fig. 2 have been observed in films with positive λ (78% Ni); in this case the mottling which is characteristic of negative λ films is absent. The reproducibility of making high-angle locking films from compositions with positive λ is quite low.

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DISCUSSION

S. Methfessel: It might be of interest to mention that we have published the observation of patterns similar to the labyrinth patterns described here already in June 1960 in an IBM Research Report. I would like to ask the following questions:

1) In the first part of the paper, a negative anisotropy has been introduced and in the second part, the picture of the ellipsoids has been given for labyrinth switching. Are these two models consistent with each other, or have they to be applied to different types of film?

2) The labyrinth model has been applied to pulse switching. Does this mean that such labyrinths are developed during the pulse switching process?

D. O. Smith: 1) As discussed in the talk, the two concepts of negative anisotropy and labyrinth switching have been considered as separate phenomena occurring in different films which represent limiting cases in which these phenomena are respectively dominant. Such an approach has the merit of conceptual simplicity. In many films, however, the presence of both effects must be considered simultaneously.

2) If labyrinth switching is important in pulse switching, then of course it is assumed that labyrinths are formed and propagate under pulse conditions. Conceptually this does not seem to me to present any unusual difficulties. It might be mentioned that in some present experiments a film has been partially switched by a fast pulse and the resulting remanent state examined by the Bitter technique. The usual type of labyrinth were observed in this remanent state, lending support to the validity of extending the *dc* labyrinth processes to pulse switching.