

Magnetic Anisotropy of Evaporated Films

MINORU TAKAHASHI, DENJIRO WATANABE, TETSUO KONO
AND SHIRO OGAWA

*The Research Institute for Iron, Steel and Other Metals
Tohoku University, Sendai, Japan*

The origin of the anisotropy in evaporated iron and nickel-iron films was investigated. The evaporation was done by normal incidence. The temperature dependence of the induced magnetic anisotropy, the relations of the anisotropy to film thickness, substrate temperature, substrate material and the relaxation phenomena of the anisotropy during isothermal annealings with and without magnetic field were observed. The temperature dependence of the anisotropy constant is not explained by the crystal anisotropy. The observed relations between the anisotropy and film thickness etc. suggest that the stress induced in the interior of the film is rather responsible for the anisotropy than the stress of limited parts of the film unless the anisotropy due to the atoms pair orientation is responsible. The former stress may be caused by the structure defects formed during evaporation. The relaxation phenomena of the anisotropy in the alternate isothermal heat treatment reveal that the anisotropy is partly originated from a directional alignment of such imperfections as vacancies, dislocations and impurities. This directional alignment is possibly, closely related to magnetostriction, although the details are not yet clear.

The origin of the uniaxial magnetic anisotropy in evaporated films of ferromagnetic metals and alloys is not yet clear, apart from the anisotropy caused by oblique incidence. Few systematic studies have been done on the influence of condition of film preparation upon the magnetic anisotropy. The present paper describes a systematic and quantitative investigation on the influence of evaporation condition upon the uniaxial anisotropy. This investigation was made in order to clarify the origin of this anisotropy in evaporated iron and iron-nickel films.

Some of the following three factors will be possible as the origin: (i) Trace of fibrous structures or anisotropic aggregation of crystallites and anisotropic shape of crystallites,¹⁾ (ii) stress localized on the film surface,²⁾ (iii) stress induced by lattice imperfection.³⁾

The induced uniaxial anisotropy was measured by a torque magnetometer. The torque curves change gradually from two-fold to four-fold symmetry with increasing measuring field, H_m , as shown in Fig. 1 (a). The

rotational hysteresis loss, $W_r = \frac{1}{2} \cdot \int_0^{2\pi} (\vec{L} - \vec{L}') d\theta$,

has its maximum in the neighborhood of the anisotropy field (Fig. 1 (b)). In the expression of W_r , θ is the angle between the direction

of easy magnetization and that of the magnetization, and $\vec{L}$ and $\vec{L}'$ are torques measured in clockwise and counter-clockwise direc-

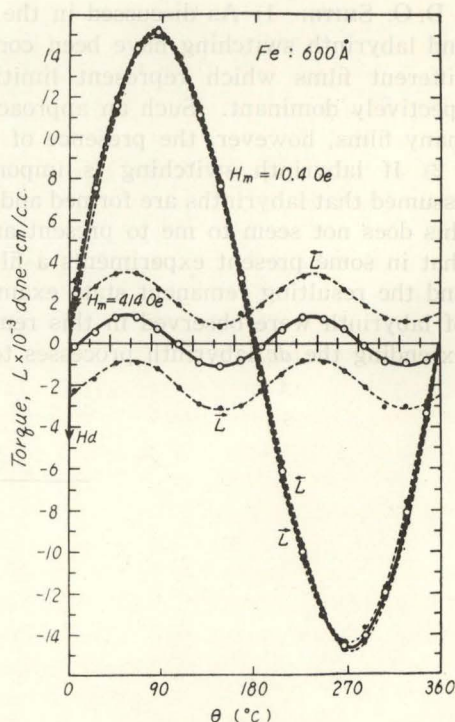


Fig. 1 (a). Some examples of the torque curves for an iron film formed at 20°C. The solid curves are means of $\vec{L}$ and $\vec{L}'$ curves.

tions, respectively. A maximum field attainable by a Helmholtz coil used in the present study is 773 Oe, and so the coefficient of $\sin 2\theta$ obtained by Fourier analysis of the torque curve was not saturated in some cases. (cf. Fig. 1 (b)). The anisotropy

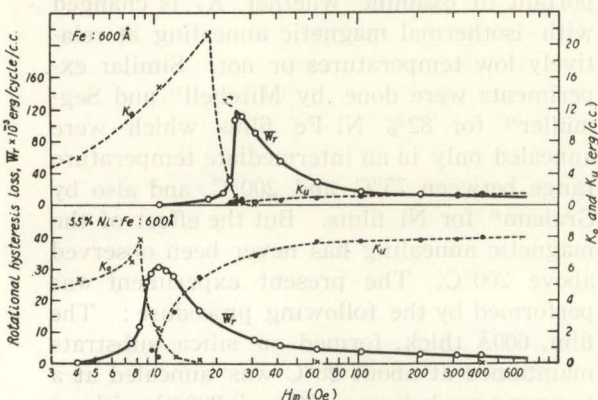


Fig. 1 (b). The relations between the rotational hysteresis loss and the measuring field. K_0 and K_u are the coefficients of $\sin \theta$ and $\sin 2\theta$, respectively.

constant, K_u , was determined by interpolation of the coefficient of $\sin 2\theta$ into $H_m \rightarrow \infty$ by $1/H_m$. In order to investigate the above three factors, (1) the temperature dependence of K_u , (2) the relation of K_u to film thickness, substrate temperature and substrate materials, and (3) the change of K_u with isothermal magnetic annealing have experimentally studied.

Results obtained:

(1) Change of K_u with specimen temperature

Any appreciable fibrous structure was not detected by electron diffraction. Even if a trace of this structure undetected by diffraction exists, it possibly causes the anisotropy which may sufficiently be observed. Then the temperature dependence of K_u should correspond to the one of the crystal anisotropy,⁴⁾ that is, it must be proportional to M_{ST}^2 for iron and to $(1-1.74 T/T_c)(M_{ST}^0)^2$ for iron-nickel film, respectively. M_{ST} is the saturation magnetization at T and T_c is the Curie temperature. The observed temperature dependence of K_u in the range between 200°C and -180°C, as shown in Fig. 2, however, cannot be explained by the crystal anisotropy. The atom pair orientation theory⁵⁾ may partly be responsible for the

origin of K_u in iron-nickel films, because, by annealing at 350 or 450°C for several hours, the temperature dependence of K_u approaches what is expected by this theory, i.e., $K_{uT} \propto M_{ST}^2$ where K_{uT} and M_{ST} are the anisotropy constant and saturation magnetization, respectively, at absolute temperature T . (Fig. 2). (2) Relation of K_u to film thickness, and substrate temperature³⁾ and substrate material.

First, if the stress localized on the film surface causes the magnetic anisotropy, K_u should depend on film thickness. The former decreases with increasing thickness, since K_u is a volume effect, while the stress is a surface effect. The results obtained are shown in Figs. 3 (a) and (b). The values of K_u vary considerably from experiment to experiment in both iron and iron-nickel films formed on the substrates maintained at about 20°C, while it is certain in the case of the

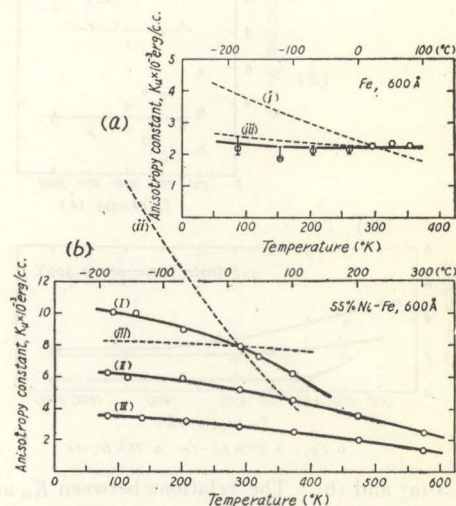


Fig. 2 (a) and (b). Temperature dependence of the induced anisotropy for Fe (a) and 55% Ni-Fe film (b).

(1) curve: $K_{uT} = K_{uTR} (M_{ST}/M_{STR})^{10}$

(2) curve: $K_{uT} = K_{uTR} (1-1.74 T/T_c)/(1-1.74 T/TR) \cdot (M_{ST}/M_{STR})^{10}$

(3) curve: $K_{uT} = K_{uTR} (M_{ST}/M_{STR})^2$

(I): Substrate temperature is 20°C.

(II): The substrate was held at 300°C during evaporation. The film was annealed at 350°C for 5 hours in a magnetic field immediately after the evaporation.

(III): The substrate was held at 300°C during evaporation. The film was annealed at 450°C for 5 hours in a magnetic field immediately after the evaporation.

substrate maintained at 300°C that K_u decreases with increasing thickness in the range from 200 to 2000Å. Second, K_u is remarkably decreased with increasing substrate temperature, as shown in Fig. 4. Third, silica plates were used as substrate in the above cases, but almost the same results were obtained with glass plates, as substrate, except in the case of the film 200Å thick formed at 300°C.

The above results suggest that a very small part of the uniaxial anisotropy might be caused by the surface stress, but a greater part is originated from some other origins.⁶⁾

(3) The change of K_u with isothermal magnetic annealing

The structural defects formed during evaporation, that is, such imperfections as vacancies, dislocations, impurities etc., may

be the most powerful factors for the origin of the anisotropy, if these imperfections are directionally aligned. The reason why such an alignment is formed cannot be given at present. If such imperfection are assumed to be the origin of the anisotropy, it is important to examine whether K_u is changed with isothermal magnetic annealing at relatively low temperatures or not. Similar experiments were done by Mitchell⁷⁾ and Segmüller⁸⁾ for 82% Ni-Fe films which were annealed only in an intermediate temperature range between 75°C and 200°C, and also by Graham⁹⁾ for Ni films. But the effect of the magnetic annealing has never been observed above 200°C. The present experiment was performed by the following procedure: The film, 600Å thick, formed on silica substrate maintained at about 20°C was annealed at a temperature between 100 and 300°C without magnetic field for a long time sufficient to stop the decrease of K_u . Then the film underwent a long time annealing at the same temperature in a magnetic field applied along

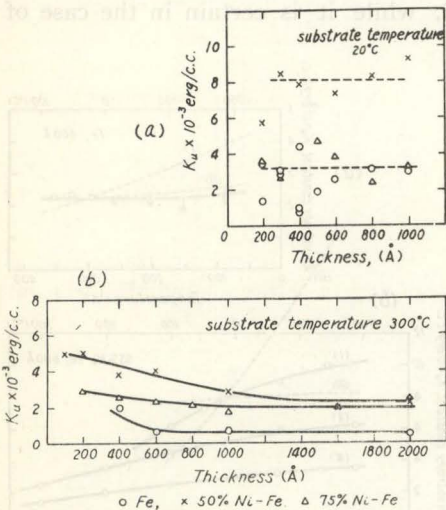


Fig. 3 (a) and (b). The relations between K_u and film thickness for Fe and Ni-Fe films formed at 20°C and 300°C, respectively.

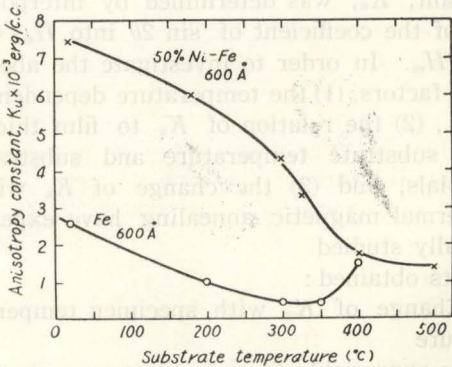


Fig. 4. The relation between K_u and substrate temperature for Fe and 50% Ni-Fe film with 600Å thickness.

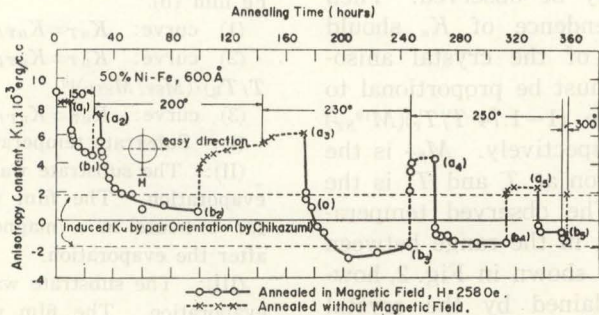


Fig. 5. The anisotropy constant as a function of the isothermal annealings with and without a magnetic field.

the direction of difficult magnetization. The specimen was succeedingly annealed at the same temperature or a slightly higher one without the magnetic field. The measurement of K_u was always done at room temperature. The results are shown in Fig. 5. There are two distinct relaxation processes in the curve. K_u is gradually decreased in the first process (with the magnetic field), (a) $\rightarrow$ (b), and, at last, the direction of easy magnetization is completely exchanged with that of difficult magnetization, but K_u is abruptly recovered as soon as the second process (without the magnetic field, (b) $\rightarrow$ (a), begins. The explanation of this effect may tentatively be given by assuming that the anisotropy in question consists of both a part due to the atom pair orientation and a part due to the orientation of imperfections.

References

- 1 D. O. Smith, M. S. Cohen and G. P. Weiss: *J. Appl. Phys.* **31** (1960) 1755; D. O. Smith: *J. Appl. Phys. Suppl.* **32** (1961) 70S.
- 2 S. Chikazumi: *J. Appl. Phys. Suppl.* **32** (1961) 81S.
- 3 E. W. Pugh, E. L. Boyd and J. F. Freedman: *I. B. M. J. Research Develop.* **4** (1960) 163; M. Prutton and E. M. Bradley: *Proc. Phys. Soc.* **75** (1960) Part 4, 557.
- 4 W. J. Carr, Jr.: *J. Appl. Phys.* **29** (1958) 436.
- 5 L. Néel: *J. Phys. Radium* **15** (1954) 225; S. Taniguchi: *Sci. Rep. Res. Inst. Tohoku Univ.* **A7** (1955) 269.
- 6 W. Andrä, Z. Málek, W. Schüppel and O. Stemme: *J. Appl. Phys.* **31** (1960) 442.
- 7 E. N. Mitchell: *J. Appl. Phys.* **29** (1958) 286.
- 8 A. Segmüller: *J. Appl. Phys. Suppl.* **32** (1961) 89S.
- 9 C. D. Graham, Jr. and J. M. Lommel: *J. Appl. Phys. Suppl.* **32** (1961) 83S.

DISCUSSION

P. BRISSONNEAU: Directional ordering energy due to pairs of substitutional atoms in general case does not depend on temperature so simply than M_s^2 . For some binary alloys, in bulk materials, experiment gives a more rapid decrease with temperature than M_s^2 .

A. A. HIRSCH: Normally the coercive force is treated as a field at which an irreversible transition of the magnetization vector occurs between two stable states. In a paper presented at the conference on thin films in Louvain (1961) we have shown that in some cases more than two equilibrium positions of the magnetization are possible. In the same particle one may therefore expect either a small or a large coercive force. We have supposed that energy barriers introduced by different kinds of imperfections concentrated in the volume of the particles can give a preference for a large or a small coercivity.

K. HOSELITZ: We observe during magnetic annealing experiments on thin permalloy films that the relaxation time for changes in the easy direction is shorter for films which are freshly evaporated than for films which have been annealed. This may confirm the proposal by the authors of the present paper that some part of the anisotropy is due to imperfections, especially lattice vacancies.