JOURNAL OF THE PHYSICAL SOCIETY OF JAPAN VOL. 17, SUPPLEMENT B-I, 1962
PROCEEDINGS OF INTERNATIONAL CONFERENCE ON MAGNETISM AND CRYSTALLOGRAPHY, 1961, VOL. I

Magnetic Properties of Thin Films Evaporated in Ultrahigh Vacuum

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A NiFe alloy with 80% Ni was evaporated onto heated, polished substrates in a vacuum of 10^{-9} mm Hg and in a technical system of 10^{-5} mm Hg. Comparison of the properties of both film types proves the influence of the residual gas on both the wall motion coercive force H_c and the anisotropy field strength H_K.

H_c is interpreted as being determined by two contributions: A roughness part with small temperature dependence and an impurity part with the same temperature dependence as reported for bulk material.

The temperature dependence and the annealing behavior of H_K in films evaporated in ultrahigh vacuum are identical to those of bulk alloys. Films produced in the technical system have an additional gas induced anisotropy which can be changed reversibly by low temperature annealing.

Introduction

Investigations on the magnetic properties of thin films so far reported in the literature, are concerned mostly with NiFe films with a composition of ~80% Ni because of their possible application to computer technology. These "technical permalloy films" are usually evaporated in commercial equipment from recrystallized Al_2O_3 crucibles at a rate of about 100 Å/sec (≈30 atom layers per second) onto glass substrates kept at about 300°C. Evaporation takes place in the presence of a magnetic field in order to establish a uniaxial anisotropy for the magnetization. The residual gas pressure is in the order of 10^{-5} mm Hg. Since at this pressure the number of residual gas atoms striking the substrate per second is sufficient to build up 10 monolayers of residual gas atoms, one should expect to find a gas content in the order of 30 at % in technical films. This amount of gas might not be included completely in the films because of the small sticking coefficient of the gas atoms. Also, the gas might be partially expelled during the growth of the metal crystals. Nevertheless, the influence of the residual gas on the magnetic properties of evaporated films still remains a problem of utmost importance.

We tried to identify the residual gas influence on the uniaxial anisotropy and the coercive force; these being the two parameters which characterize the mode of magnetization reversal. For this purpose, the properties of films evaporated in a ultrahigh vacuum (UHV) system (at 10^{-9} mm Hg residual gas pressure) are compared with those of films evaporated from the same melt in a commercial system at 10^{-5} mm Hg. Since residual gas pressure and deposition rate are closely related with regard to their consequences on gas contents and film structure, a UHV system has been developed which allows "technical" deposition rates of about 100 Å/sec in a 10^{-9} mm Hg vacuum. The amount of residual gas included in a metal evaporated under such conditions can be expected to be of the order of one part in 10^4 or lower.

Experimental Details

For deposition and investigation of films in UHV we used a double-walled water-cooled pyrex glass tube (Fig. 1) which was connected through a liquid air trap to the Hg-pumps of an all-glass UHV system. The metal was melted in a small thoriumoxide crucible inserted into a molybdenum pot, which in turn was heated by stabilized electron bombardment from a surrounding filament. All metal parts can be heated up to 2000°C by electron bombardment or current flow through the supporting double tungsten wires. After consecutive cycles of flashing the metals and baking out the whole system for 12 hours at 450°C, a residual gas pressure of $6\cdot10^{-10}$ mm

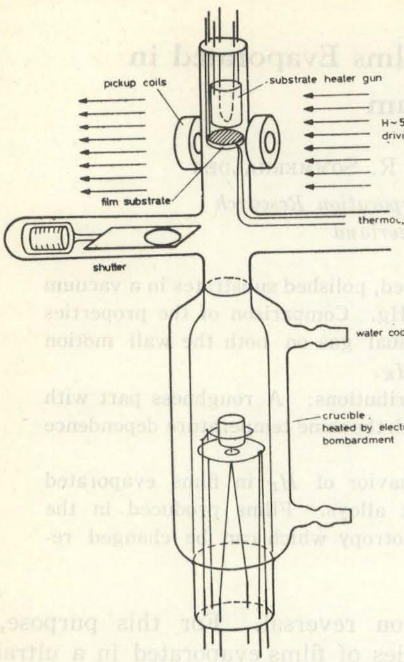

Fig. 1. Tube for evaporation and investigation of NiFe films in ultrahigh vacuum.

Hg was measured in the Alpert ionization gauge. During the evaporation process the pressure rises to the range of 10^{-9} mm Hg. A polished plate of quartz or artificial sapphire was used as film substrate which can be heated from the back by stabilized electron bombardment. The precleaning of the substrate took place at temperatures between 800 and 1000°C.

During the growth of the film a homogeneous 50 cps field was applied by a pair of Helmholtz coils. The induced magnetic flux reversal of the film, which is picked up by two coils from the outside of the vacuum system, was integrated, amplified and displayed as a hysteresis loop on the screen of an oscilloscope. Afterwards the film was left in the vacuum in order to measure the temperature dependence and the annealing behavior of its properties.

The evaporated metal is a technically pure NiFe alloy prepared by sintering in order to keep the amount of additional components such as Mn or Si small.* The composition as well as the thickness of the films were determined by X-ray fluorescence methods after opening the vacuum system.

Some first results obtained on NiFe films with 76% wt Ni are reported here in order to give a qualitative survey rather than a quantitative description of the residual gas influence on H_c and H_K.

The Coercive Force H_c

Fig. 2 shows in log. scales the dependence of H_c on film thickness D measured during the relatively slow deposition (147 Å/min) of 76% NiFe onto an optically flat sapphire substrate at 200°C in UHV. The relative variation in film thickness was determined from the evaporation time at constant rate. The latter was checked by the linear increase of the magnetic flux with time. In agreement with some results obtained on electroplated[1)] layers and on evaporated and subsequently etched films[2)] our results fit the D^{-1} law much closer than the $D^{-4/3}$ law predicted by Néel[3)].

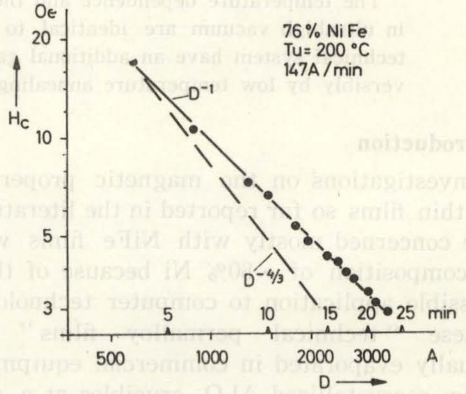

Fig. 2. The decrease of coercive force H_c with thickness D measured during evaporation in UHV (··) compared with D^{-1} (—) and $D^{-4/3}$ (- - -) laws. T_u=substrate temperature.

Measurement of H_c parallel to the easy axis of the magnetization yields the threshold field for Barkhausen jumps. For $H=H_c$ the domain walls are just able to overcome the impediment to their motion along given by the local variation in their energy per unit wall length $E(x)$:

$$2J_sH_cD=\frac{dE(x)}{dx}$$

(J_s=saturation magnetization).

Therefore H_c prop. D^{-1} means that $dE(x)/dx$ is independent of film thickness. If it is as-

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* We thank Dr. Assmus, Vacuumschmelze AG, Hanau, Germany, for his willingness to prepare these alloys for us.

sumed with Néel that the local variations in $E(x)$ are due to local variations in the film thickness with an amplitude $h(x)$:

$$E(x)=\gamma D(x)=\gamma(D_0+h(x)) ,$$

then it follows from $dE(x)/dx=\gamma(dh(x)/dx)=$ const., that for constant energy density γ of the domain walls the surface roughness $dh(x)/dx$ does not vary with film thickness. For the observed thickness range between 500 and 3000 Å this seems to be a plausible assumption.

Fig. 3 shows H_c vs. temperature measured on the film of Fig. 2. After heating up to about 500°C the original, high H_c value of 2.5 oe decreases irreversibly to 1.4 oe. Another film of ~3000 Å thickness evaporated from the same melt at the rate of about 5000 Å/min onto a substrate at 400°C shows already from the beginning $H_c=1.1$ Oe and the threshold field does not show any response to annealing up to 600°C. (Fig. 4). Comparison of these results with Néel's theory indicates that the surface roughness depends on substrate temperature and evaporation rate and that it can be smoothed by annealing. In contrast to this behavior we found that the substrate roughness (*e.g.* of an unpolished molybdenum surface) and its contribution to H_c are characterized by their stability against annealing.

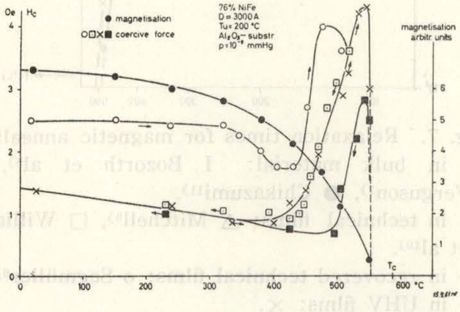

Fig. 3. Temperature dependence of magnetization and coercive force H_c of the same film as in Fig. 2, measured during two thermal cycles. T_u=substrate temperature during evaporation.

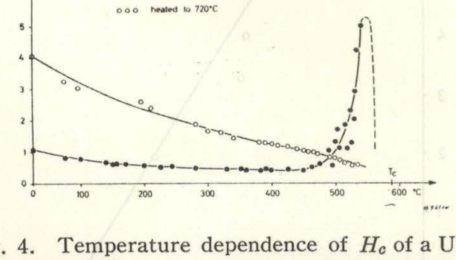

Fig. 4. Temperature dependence of H_c of a UHV film evaporated at higher substrate temperature T_u than the film of Fig. 2. Measurement immediately after evaporation (· – ·) and after annealing at 720°C for a few minutes (o – o).

Annealing the film in UHV for about 10 min at 720°C increases the threshold field H_c irreversibly (Fig. 4). A much stronger temperature dependence of H_c is now observable, which agrees quite well with the results obtained on bulk 75% NiFe alloys[4)5)] (Fig. 5). Therefore, we assume that during the high temperature treatment, the diffusion of impurities from the substrate into the NiFe layer raises the intrinsic* value of H_c so much that it surpasses the contribution of the roughness to the threshold field.

The H_c vs. temperature curve shown in Fig. 6 was obtained on a 3400 Å thick film

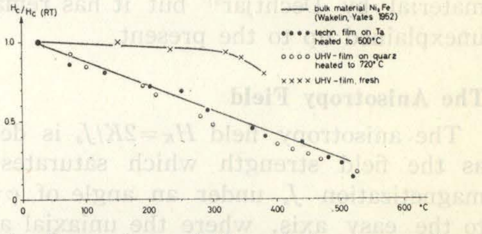

Fig. 5. Comparison of the relative temperature dependence of H_c for bulk Ni_3Fe and different types of evaporated films. H_c (RT)=H_c at room temperature.

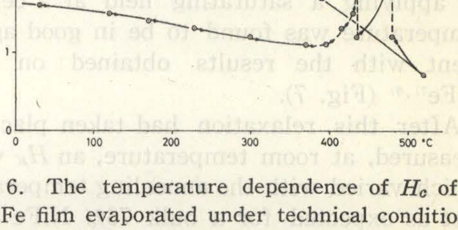

Fig. 6. The temperature dependence of H_c of a NiFe film evaporated under technical conditions at T_u=300°C substrate temperature. Measurement during $1\frac{1}{2}$ thermal cycles.

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* i. e., characteristic for bulk material.

evaporated from the same melt but in a 10^{-5} mm Hg vacuum system under technical conditions onto an electropolished tantalum sheet. The procedure of measurement and the results typical for such "technical" films have already been described in an earlier paper[6]. Comparison with the properties of a UHV film which was obtained with roughly the same deposition rate and substrate temperature, indicates that the pressure difference between 10^{-5} and 10^{-9} mm H_g causes a noticeable influence on H_c only at substrate temperatures higher than 350°C. Above this temperature the strong increase of the intrinsic threshold field takes place (Figs. 6, 5) within minutes. For reasons we do not yet understand, the increase of the H_c in technical films at higher temperatures can be avoided by the addition of a few percent of copper or molybdenum to the NiFe alloy.

The maximum in the H_c vs. temperature curve observed around 500°C in both UHV and technical films is also reported for bulk material by Dechtjar[5] but it has remained unexplained up to the present.

The Anisotropy Field

The anisotropy field $H_K=2K/J_s$ is defined as the field strength which saturates the magnetization J_s under an angle of $\varphi=90°$ to the easy axis, where the uniaxial anisotropy energy $E=K\sin^2\varphi$ of the magnetization reaches its maximum value. We have derived H_K as usual from the initial permeability measured under 90° to the easy axis.

The influence of the residual gas on H_K is much more striking than on H_c as, on UHV films, we never observed the low temperature annealing effect which has been described by A. Segmüller[6] for technical films. The relaxation time necessary to rotate the easy axis into another direction by applying a saturating field at a certain temperature was found to be in good agreement with the results obtained on bulk NiFe[7),8)] (Fig. 7).

After this relaxation had taken place we measured, at room temperature, an H_K value which varied with the annealing temperature just as expected for a bulk 76% NiFe alloy from the interpolation of Ferguson's results[7] (Fig. 8). Also the dependence of H_K on the temperature of the films during the measurement agreed fairly well with the results of Chikazumi[11] for bulk NiFe (Fig. 9). Consequently, the uniaxial anisotropy in the UHV permalloy films proved to be identical with the annealing anisotropy of bulk binary alloys.

Unfortunately, the original H_K value induced during evaporation in an external

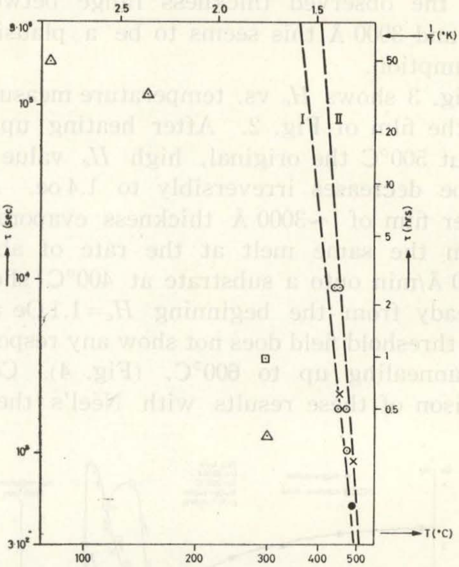

Fig. 7. Relaxation times for magnetic annealing
- in bulk material: I Bozorth et al[8], II Ferguson[7], ● Chikazumi[11].
- in technical films: △ Mitchell[9], □ Williams et al[10].
- in recovered technical films: o Segmüller[6].
- in UHV films: ×.

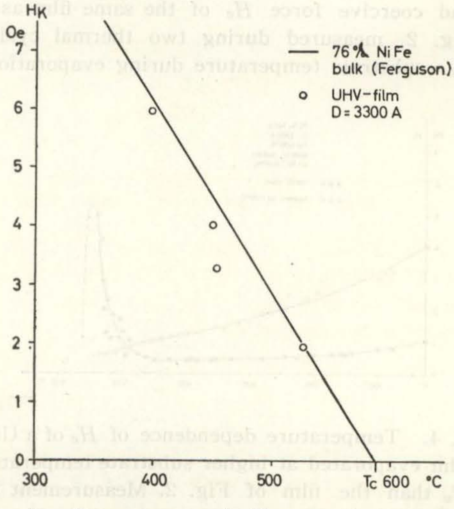

Fig. 8. Dependence of H_K on the annealing temperature for UHV films and bulk material.[7]

field cannot be derived from bulk material data unless the evaporation time is longer than the relaxation time at the substrate temperature. Not even rough estimates can be made because of the lack of quantitative data on the surface processes taking place during the deposition of metal atoms. We measured $H_K=2.2$ Oe after fast evaporation at a substrate temperature of 400°C and $H_K=4.5$ Oe after slow evaporation at 200°C.

A technical film evaporated from the same melt at 300°C substrate temperature in the vacuum of 10^{-5}mm Hg displayed an original H_K value of 5.3 Oe. It showed typically low temperature annealing behavior: After heating to 300°C the H_K value depends reproducibly on whether the field was applied parallel or perpendicular to the easy axis during cooling (Fig. 10). This behavior indicates that two mechanisms of uniaxial anisotropy are superimposed in technical films. The normal bulk material anisotropy remains at 300°C with $H_{K1}=3.7$ Oe unchanged in the old direction, while the additional anisotropy $H_{K2}=1.6$ Oe, which is induced by the residual gas, already rotates at this temperature. The gas induced anisotropy must probably be identified with the uniaxial anisotropy found by Malek[12] and Graham and Lommel[13] in evaporated layers of pure Ni, Fe and Co.

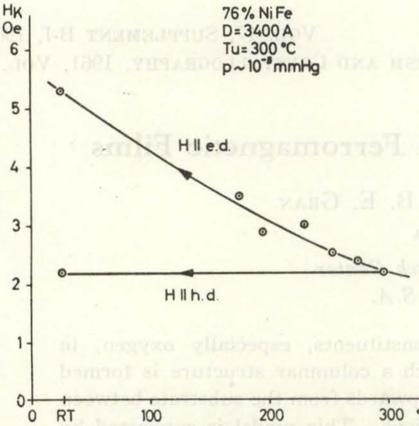

Fig. 9. Relative temperature dependence of H_K for bulk material[11]) and a UHV film.

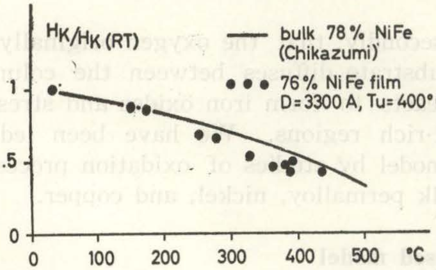

Fig. 10. Low temperature annealing behavior for a technical film, with the field H applied parallel to the easy (e. d.) or hard (h. d.) direction during cooling.

Acknowledgments

We thank Professor W. Döring for helpful discussions, also Messrs. H. Seitz and S. Dunn for their assistance during the measurements.

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DISCUSSION

C. D. GRAHAM, JR.: I have two questions.

1) How reproducible are your results? That is, if several films are made under conditions as nearly identical as you can make them, how much scatter will there be in the values of H_c, K_u and their temperature dependence?

2) The fact that you plot relaxation times for the magnetic annealing process implies that you have followed the magnetic annealing anisotropy as a function of time. What is the form of the curve of K_u *vs.* time, and is the form the same at all temperatures and for all films?

S. METHFESSEL: 1) The reproducibility of our results is indicated by the scattering of the measured values in the diagrams which I have shown.

2) We followed the anisotropy during the annealing treatment and derived from this measurement the relaxation time for turning the easy axis by an angle of 90°. However, the accuracy of the H_K values obtained from the measurements of the initial susceptibility in the hard direction is not very high, and small deviations from exponential law in the K_u *vs.* time curve cannot be detected in ultrahigh vacuum films.

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Influence of Minor Constituents in Ferromagnetic Films

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Studies have been made on the effect of minor constituents, especially oxygen, in permalloy thin films. A model is proposed in which a columnar structure is formed during deposition. The oxygen is assumed to diffuse upwards from the substrate between the columns to form iron oxide and nickel-rich regions. This model is supported by electron photomicrographs of thin-film cross sections which have been prepared by etching. The model permits an interpretation of the origin of *M*-induced anisotropy. Other experiments which have been carried out to test this model are discussed.

Introduction

The manner in which the uniaxial anisotropy is produced in permalloy thin films has been the subject of much controversy. Recently, evidence has been presented which indicates that this anisotropy is ***M***-induced rather than field induced[1,2,3]. It has been suggested[4] that the elongation of the crystallites produces an easy axis which specifies the direction of the magnetization ***M***. Models of this nature are not entirely convincing inasmuch as the mechanism producing this elongation is not considered.

We have re-examined this question and have arrived at a different model for the origin of the *M*-induced anisotropy[5]. The two main features of this model are first, the assumption that the growth proceeds from the substrate in the form of columns, and secondly, that the oxygen originally on the substrate diffuses between the columns and reacts to form iron oxides and stressed nickel-rich regions. We have been led to this model by studies of oxidation processes in bulk permalloy, nickel, and copper.

Proposed model

In the oxidation study of bulk permalloy, the more reactive element (in this case iron) is oxidized first by an internal oxidation mechanism. Iron oxide is formed within the metal matrix until a nickel-rich phase (almost pure nickel) remains at the metal surface to be subsequently oxidized[5]. When the cross sections of oxidized alloy samples are examined with the metallurgical microscope, the presence of the iron oxide along the grain boundaries is clearly evident. This indicates that