Vol. A42 (1972) ACTA PHYSICA POLONICA No 4

# INTERACTION IN MULTI-LAYER FILMS

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*(Received January 18, 1972)*

The paper presents the technology of depositing thin multi-layer magnetic films with perpendicular easy axes, and the determination of the dependence of the interaction energy on the angle $\alpha$ between the magnetization directions of the ferromagnetic components in the case of a film of Ni-Fe alloy. The dependence of the interaction energy on the angle $\alpha$ is found to be of the type $a\alpha+b\sin^2\alpha+c\sin^2 2\alpha$ when the ferromagnetic layers are in direct contact, and of the type $a\alpha+b\cos\alpha$ when they are separated by a non-magnetic layer.

## 1. Introduction

Multi-layer magnetic systems are of interest both from the point of view of the possibility of studying the structural and dynamical properties of thin films and their application in miniature electronic circuits. Such circuits permit a low coercive field to be applied, and increase the reproducibility of magnetic parameters. Moreover, double films also double the amount of information available for computer storage. The potential energy of such systems is the sum of uniaxial anisotropy energy, the energy of interaction with external field, and the energy of interaction between ferromagnetic constituents. The determination of the functional behaviour of the interaction energy is an essential problem and has been discussed in many papers [1, 2, 3]. Quantitative results for double layer films have been obtained by Goto [1] on the basis of the following assumption: the magnetization vectors of both components lie in the plane of the film; the interaction is strong, of the type of exchange interaction; the thicknesses, magnetizations and anisotropy fields of both constituents are the same. In such a case the interaction energy $E_c$ is given by the formula

$$E_c = h(\varphi_1-\varphi_2)^2 \tag{1}$$

where $h$ is a constant depending on the parameters of the sample, and $\varphi_1$, $\varphi_2$ are the angles formed by the respective magnetization vectors of the constituents with the mean

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easy axis. The model of coupling fields described by Yelon [2] and Bruyer [4] permits the correlation between the magnetization vectors of the two constituents to be explained both in the case of layers directly deposited on one another and in the case of layers separated by a nonmagnetic material.

In the case when the thicknesses, the uniaxial anistotropy fields and the compositions of both layers are identical, the interaction energy is given by

$$E_c = -H_i M \cos(\varphi_1 - \varphi_2) \tag{2}$$

where $H_i$ is the coupling field produced by the $i$-th layer of the film.

Such a form of interaction energy for films with magnetic layers separated by a non--magnetic one has also been obtained in Ref. [5] under the assumption of the model of magnetostrain interaction due to stray fields originating from the ripple effect.

The validity of the model of Goto [1] and that of Yelon [2] is restricted to the case of strong interaction (where the angle between the magnetization vectors is small in the remanence state). From the point of view of pure research, however, it is essential to know the behaviour of the interaction energy in a wide range of angles between the magnetization vectors.

The present paper presents the dependence of interaction energy on the angle between the magnetization vectors obtained on the basis of experimental data concerning the behaviour of susceptibility as a function of external field.

## 2. *Theory*

The method of determining the dependence of the interaction energy on the angle $\alpha$ between the magnetization vectors in both ferromagnetic layers can be applied for a wide range of $\alpha$ in the case of double films as well as in the case of magnetic films separated by a non-magnetic thin layer [6]. This means that it is not restricted to strong coupling only. The method can be used under the following assumptions:

*i*) the thicknesses, magnetizations and the uniaxial anisotropy fields of both constituents are the same;

*ii*) the angle between the easy axes of the two constituents is $\pi/2$;

*iii*) the vertical magnetization component $M_z = 0$.

Taking into account the uniaxial asymmetry energy of both magnetic constituents as well as their interaction energy — which we assume to be isotropic [6] — we obtain that the total energy density $E$ is given by the formula

$$E(\alpha, \varphi) = \frac{K_u}{2}\left[-\cos 2\varphi \sin\alpha - 4h\cos\frac{\alpha}{2} + 2\frac{E_c(\alpha)}{K_u}\right] \tag{3}$$

where $h$ denotes the external magnetic field (in normalized units) applied in the direction of the average magnetization determined by the angle $\varphi$; $\alpha$ is the angle shown in Fig. 1, and $K_u$ is the uniaxial anisotropy constant. The easy directions of the first and second layers make a 45-degree angle with the $x$-axis.

For slowly varying fields the minimization of the total energy with respect to $\alpha$ yields

$$\frac{1}{K_u}E'_c(\alpha) = \frac{1}{2} + \left(\frac{h}{2}\right)^2 - \left(\frac{h}{2} + \sin\frac{\alpha}{2}\right)^2 \quad (4)$$

where the mean magnetization angle $\varphi$ is equal to zero, as the films were saturated in this direction before the measurements. Thus, the coupling energy density $E_c(\alpha)$ may be obtained from Eq. (4) by numerical integration. Therefore, the problem resides in finding

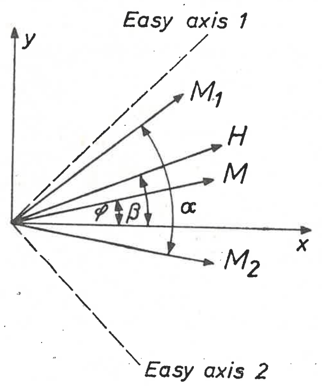



Fig. 1. Reference frame. The film plane is $x0y$. $M_1$ and $M_2$ are mean magnetizations in the first and second layers, respectively. The film was initially saturated along the $x$-axis

the relationship between the angle $\alpha$ and the magnetic field $h$ from experimental data. The dependence of the angle $\alpha$ on the field $h$ can be determined by analysing the experimental behaviour of the reversible susceptibility $\kappa(H)$. By applying the formula for the reversible susceptibility given in Ref. [6] we obtain that the reciprocal susceptibility $\frac{1}{\kappa}$ is given by the formula

$$\frac{1}{\kappa} = \frac{2\mu_0 H_k}{M}\left[\tan\frac{\alpha}{2} + \frac{h}{2}\frac{1}{\cos\frac{\alpha}{2}}\right] \quad (5)$$

where $H_k = \frac{2K_u}{M}$ is the anisotropy field.

The use of Eq. (5) permits the determination of the angle between the magnetization vectors for every value of stationary magnetic field $h$ from the experimentally determined relation $\kappa(H)$. As the values of susceptibility are taken from experimental data, we can find the unknown angle $\alpha$ for each value of field $h$.

## 3. *Experimental results*

### 3.1. Technology

The measurements were made for double films and for mangetic films separated by a thin non-magnetic layer. The double films, composed of two layers of alloy composed of about 78% Ni and 22% Fe, were deposited by vacuum evaporation at $5\times10^{-5}$ torr with normal incidence. The substrate temperatures were 453 K for the first layer and 423 K for the second layer. In order to obtain magnetic films separated by a thin non--magnetic layer with perpendicular easy axes of the magnetic constituents, the first ferromagnetic layer containing 75% Ni + 25% Fe was deposited on the substrate at 523 K, the non-magnetic layer at 473 K, and the second ferromagnetic constituent (81% Ni + + 19% Fe) at 453 K (all temperatures relating to the substrate). The uniaxial anisotropy was induced by a field of $1.6 \times 10^4 \frac{\mathrm{A}}{\mathrm{m}}$ during evaporation. As a result of several tests it was found that the above mentioned conditions must be fulfilled rather rigorously, in particular the substrate temperature should be maintained constant to within 5 K and the temperature during the deposition of the second magnetic layer must be adequately lower [8]. In order to induce perpendicular easy axes on the specimen one should rotate it after the first deposition by an angle of $\pi/2$ with respect to the external magnetic field with considerable accuracy ($\pm 2°$). After evaporation the samples should be cooled in vacuum to 373 K in the presence of a magnetic field applied along the direction of the second easy axis, and then to 323 K without external magnetic field. The temperature at which the external field is switched off is crucial since, on the one hand, the second easy direction should be fixed and, on the other, the first easy direction should not be allowed to rotate.

### 3.2. Results

The measurements were made only with those films which satisfied the assumptions of the model described in the preceding text. The following selection criteria were used.

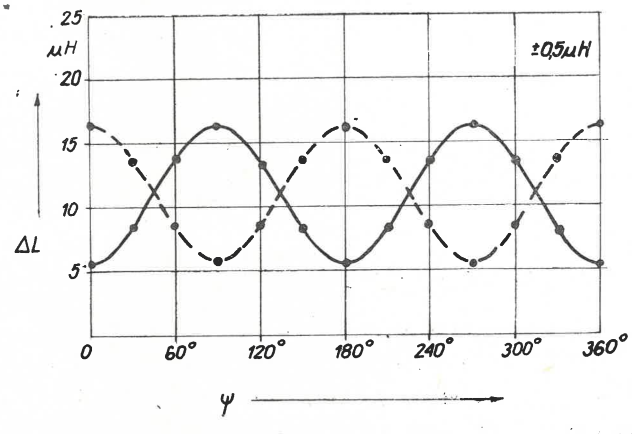
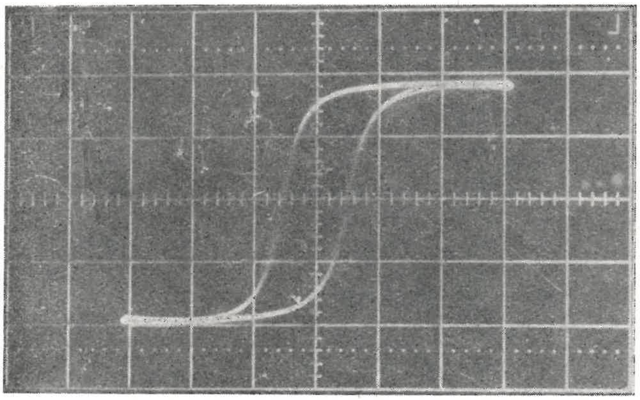



Fig. 2. Dependence of inductance $\Delta L \sim \kappa$ on $\psi$. The field which saturates the thin film was applied: a) along the $x$-axis — solid line, b) along the $y$-axis — dashed line

1. Measurement of the initial susceptibility as a function of the angle $\psi$ at which the varying magnetic field $H$ is applied. The shape of the dependence of the initial susceptibility $\kappa$ on $\psi$ should be the same after saturation of the sample along the $x$-axis well as along the $y$-axis (Fig. 2); it should be only shifted by $\pi/2$ [7].

2. Determination of the hysteresis loop of the film. The hysteresis loops obtained when the magnetizing field is applied in the $x$ and $y$-direction should be the same (Fig. 3).

3. Measurement of the reversible susceptibility as a function of the field, $\kappa_{\perp}(H)$. The experimentally determined dependence of the reversible susceptibility $\kappa_{\perp}$ on $H$ should



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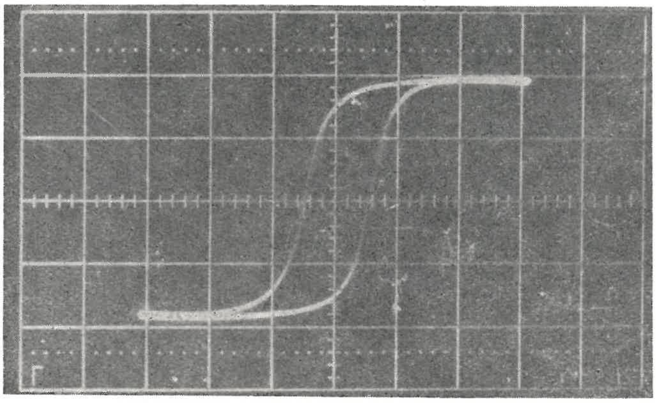

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Fig. 3. Hysteresis loop for a film whose ferromagnetic layers interact and easy axes are mutually perpendicular. The magnetizing field is applied: a) along the $x$-axis, b) along the $y$-axis

be the same when the magnetic field is applied in the direction of $\pm x$ and in the direction of $\pm y$ (Fig. 4).

The measurements aimed at a determination of the dependence of the interaction energy $E_c$ on $\alpha$ were made only for those samples which satisfied the above-mentioned criteria.

As it is more convenient to use $\frac{1}{\Delta L_{\perp}}$ than $\Delta L_{\perp}$ in the calculations, the plots are presented as $\frac{1}{\Delta L_{\perp}}$ *vs* $H$. Figure 5, 6, and 7 show the dependence of reciprocal inductance

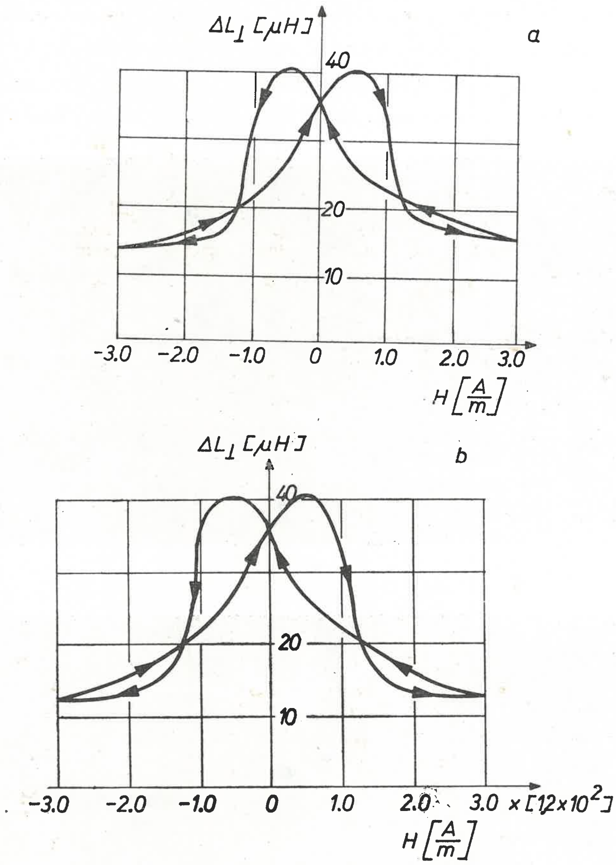



Fig. 4. Dependence of $\Delta L_{\perp} \sim \kappa_{\perp}$ on magnetic field $H$. Sample saturated before measurement: a) along the $x$-axis, b) along the $y$-axis

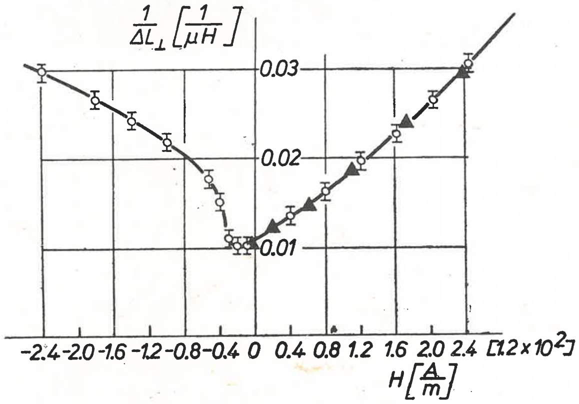



Fig. 5. Dependence of reciprocal inductance $\frac{1}{\Delta L_{\perp}}$ on external magnetic field $H$ for film No 2 whose magnetic layers were deposited immediately one upon the other

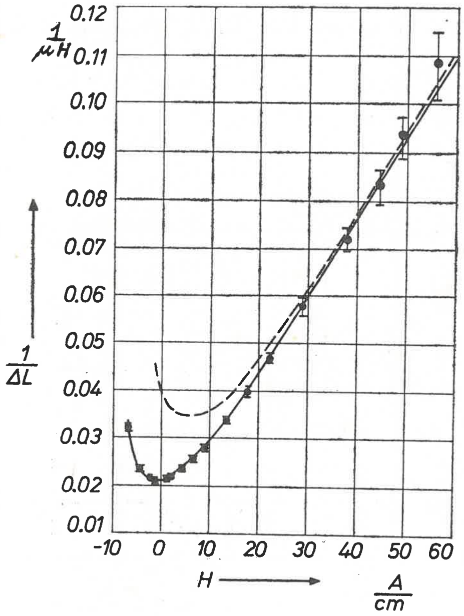



Fig. 6. Dependence of reciprocal inductance $\frac{1}{\Delta L}$ on external magnetic field for film No 843 whose magnetic layers are separated by a non-magnetic layer

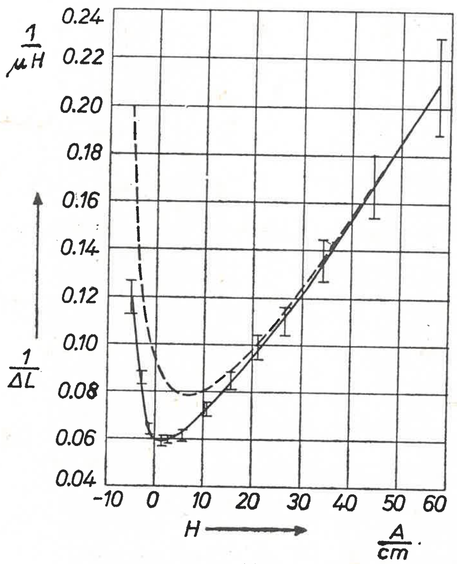



Fig. 7. Dependence of reciprocal inductance $\frac{1}{\Delta L}$ on external magnetic field $H$ for film No 834

$\frac{1}{\Delta L_{\perp}}$ as a function of external magnetic field $H$, obtained in the present experiment for double films and for those separated by a non-magnetic layer. The experimental points are mean values from measurements in both easy directions $x$ and $y$, and saturation along the direction $\pm x$ and $\pm y$.

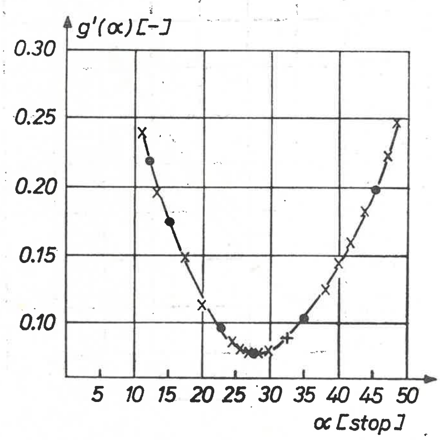



Fig. 8. Dependence of the derivative of interaction energy $g'$ on the angle $\alpha$ for double films (sample No 2)

The shape of the dependence of the expression $\frac{E_c'(\alpha)}{K_u}$ on $\alpha$, where $E_c'(\alpha)$ is the derivative of interaction energy density with respect to $\alpha$, was determined with the use of Eqs (3) and (4) and from the experimental relation $\frac{1}{\Delta L_{\perp}}(H)$. The result $g'(\alpha) = \frac{E_c'(\alpha)}{K_u}$ for double films is shown in Fig. 8.

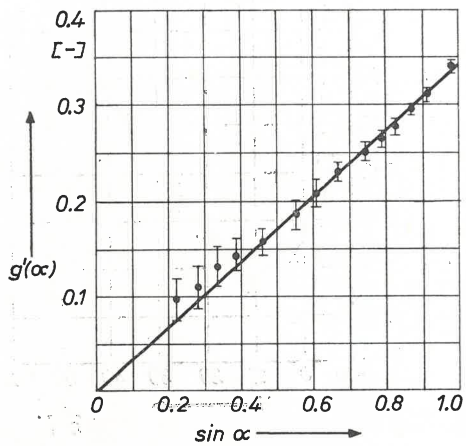



Fig. 9. Dependence of the derivative of interaction energy $g'$ on $\sin\alpha$ for sample No 843 whose magnetic layers are separated by a non-magnetic layer ($t > 100$ Å)

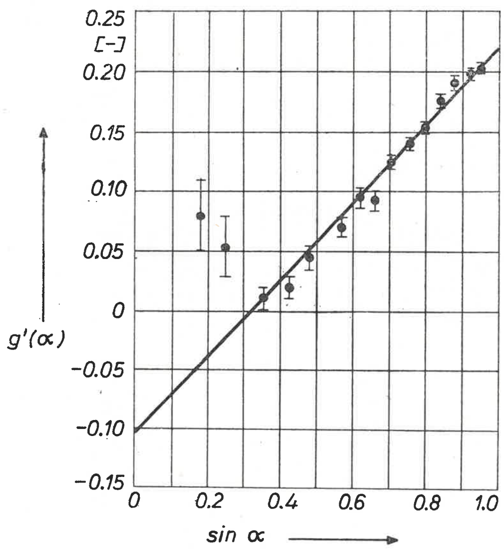



Fig. 10. Dependence of the derivative of interaction energy $g'$ on $\sin\alpha$ for sample No 834 with non-magnetic layer thickness $< 100$ Å

For films containing a non-magnetic layer the results are presented in terms of the dependence of $g'$ on $\sin\alpha$, to check the validity of the linear relation $g'(\sin\alpha)$ predicted by the models applied so far (Figs 9, 10). The thickness of the non-magnetic layer in sample No 843 was greater than 100 Å while that of No 834 was less than 100 Å.

## 4. *Discussion of results*

Let us first consider double films with no separating non-magnetic layer. In this case the expression for interaction energy density may contain several terms which for symmetry reasons are periodic functions of $\alpha$. On the other hand, the term corresponding to exchange energy is not periodic since it should be a monotonous function of $\alpha$. We therefore expect that the interaction energy can be described as a sum of some non-periodic function of $\alpha$ and periodic functions of the expansion into a Fourier series, but only with those terms which remain after consideration of symmetry arguments. The periodic part of the expression for the energy should be an even function of $\alpha$ and invariant when $\alpha \to \to \alpha+\pi$ (symmetry with respect to a change of sense of the magnetization vector). Thus, the expression for the energy may contain terms of the type $\cos 2\alpha$ and $\cos 4\alpha$. By making use of Fig. 8 it is possible to find that the interaction energy density which satisfies the above conditions can be written as

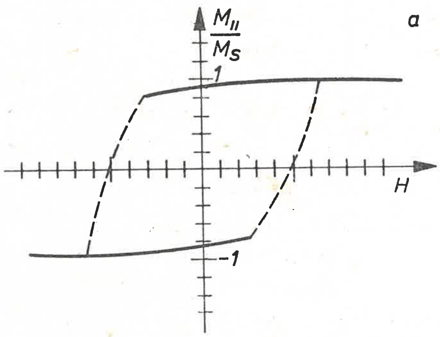
$$g(\alpha) = a|\alpha| + b\sin^2\alpha + c\sin^2 2\alpha. \tag{5}$$

The coefficients $a$, $b$, $c$ for the film for which the experimental results were shown in Fig. 8 are

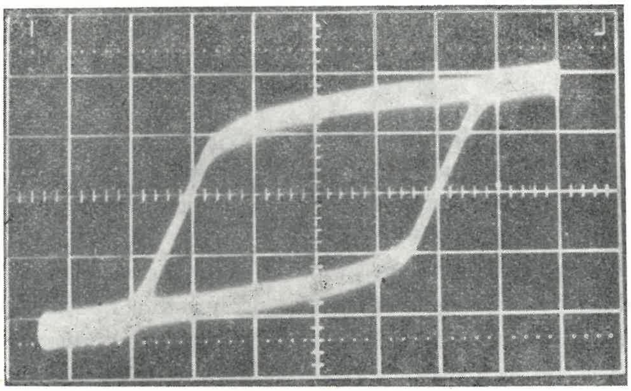
$$a = 0.455, \quad b = -0.255, \quad c = -0.177 \quad \text{(for sample No 2).}$$

An analysis of the term $b \cdot \sin^2 \alpha$ has been made in Refs [8, 9], while the term $a|\alpha|$ will be discussed in a later paper. No simple physical interpretation, however, could be found for the term $c \sin^2 2\alpha$. This term is a natural consequence of the expansion into a Fourier series and is probably due to anisotropic stress or may be the result of the influence of terms of order higher than fourth appearing in the crystallographic energy.

The correctness of the expression for interaction energy (Eq. (5)) has been checked by comparing the theoretical loop of longitudinal hysteresis in the easy direction with the experimental loop. The knowledge of the angle $\alpha$ for each value of $h$ permits $\cos \frac{\alpha}{2}$ to




a



b

Fig. 11. Hysteresis loops measured in easy direction for sample No 2: a) theoretical (calculated on the basis of Eq. (5)), b) experimental

be calculated, which in turn enables us to plot the hysteresis loop in $\frac{M_{||}}{M}$ units. Examples of hysteresis loops for sample No 2 are given in Fig. 11.

In the case of multi-layer films with a non-magnetic layer in between, the dependence og $g'$ on $\sin \alpha$ (Figs 9 and 10) can be regarded to be linear, if one allows for the experimental errors made in the determination of the points, and can be expressed by means of the formula

$$g'(\alpha) = a \sin \alpha + b. \qquad (6)$$

The deviations of the experimental points from the linear dependence seen in the figures both in the small and the large $\alpha$-angle range have been discussed by Lubecka [11]. It can be concluded from Figs 9 and 10 that the type of energy of interaction between the ferromagnetic layers of the film depends on the thickness of the non-magnetic layer. For sample No 843, in which the thickness of the non-magnetic layer was greater than 100 Å, the parameter $b$ in Eq. (6) vanishes. In such a case the energy of interaction between the ferromagnetic layers is equal to the energy of the coupling fields, and the energy density can be expressed as

$$g(\alpha) = -2h_i \cos \alpha \qquad (7)$$

where the relationship between $a$ and $h_i$ ($a = 2h_i$) is obtained from a comparison of the theoretical (1) and experimental (7) formulae.

In sample No 834 the thickness of the non-magnetic layer was less than 100 Å. For this film the parameter $b \neq 0$ and the interaction energy density, being the sum of the exchange and coupling field energy densities, can be expressed as

$$g(\alpha) = -2h_i \cos \alpha + b. \qquad (8)$$

The constant $b$ and the value of the coupling field $H_i$ for films for which the results are shown in Figs 9 and 10, are

$H_i = 1.894$ A/cm and $b_i = 0$ for sample No 843,

$H_i = 2.135$ A/cm and $b_i = -0.105$ for sample No 834,

where $H_i = h_i H_k$.

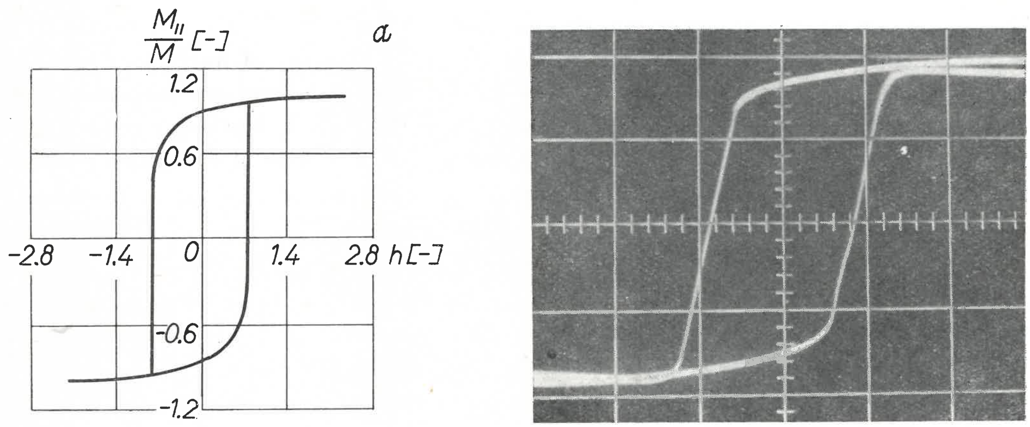
For the investigated films the values of the coupling field are positive, *i.e.*, the interaction between the magnetic layers of the film is of the ferromagnetic type. In the case of these films it is also possible to confirm the correctness of the expression for the interaction energy by comparing the theoretical hysteresis loop with the experimental longitudinal one. The value of coercive field $h$ can be determined from Eqs (3) and (6) and the condition

$$\frac{\partial h}{\partial\left(\cos\frac{\alpha}{2}\right)} = 0.$$

On the other hand, the remanent induction point must satisfy the relation

$$\frac{M_r}{M} = \left[\frac{16h_i^2+1+2b-4h_i\sqrt{16h_i^2+1-4b^2}}{2(16h_i^2+1)}\right]^{\frac{1}{2}}. \qquad (9)$$

Figures 12a and 13a show the ratio $\frac{M_{||}}{M}$ as a function of the field $h$ for samples Nos 843 and 834. For the sake of comparison Figs 12b and 13b show the hysteresis loops obtained experimentally. It can be seen that the agreement is satisfactory. There is also




a b

Fig. 12. Hysteresis loop in the easy direction for film No 843: a) theoretical, b) experimental.

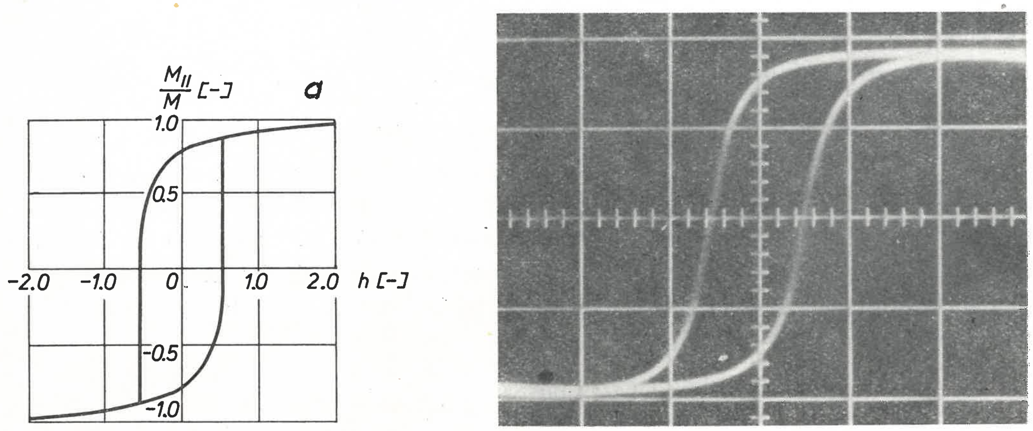



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Fig. 13. Hysteresis loop in the easy direction for film No 834: a) theoretical, b) experimental

reasonable agreement, within the limits of error, between the experimental behaviour of the initial susceptibility and that calculated on the basis of the obtained dependence of interaction energy on the angle $\alpha$ [10].

## *5. Conclusions*

The interaction energy was determined quantitatively using the experimental method residing in the determination of the dependence of interaction energy on the angle $\alpha$ described in Ref. [6].

The problem of determining the interaction energy, which is of great importance in investigations of multi-layer films has not yet been solved satisfactorily in the literature, because:

a) in all cases the authors confined the assumptions to strong interactions only, when the angle $\alpha$ is very small:

b) they assumed a definite type of dependence $E_c \sim \alpha^2$.

The method described in the present paper can be applied also to larger $\alpha$ angles and does not make any definite assumption about the interaction energy. On the basis of the results obtains for double films it was found that the behaviour of the interaction energy is considerably different from that proposed by other authors [1, 2] who considered above all the exchange energy. The term responsible for the exchange energy was rigorously considered in the framework of the variational method [11]. The term $b \sin^2 \alpha$ is associated with magnetoelastic energy [8] due to additional stress produced during magnetostrictive processes. The factor $b$ depends on the chemical composition of both layers. It is characteristic that $b$ may be negative for compositions near to 81 % Ni + 19% Fe only, which gives rise to a decrease of the effective interaction energy. For compositions definitely different from 81% Ni + 19% Fe the factor $b$ is always positive and quite high, which results in an increase of interaction energy. This observation seems to explain why other authors were unable to obtain coupling of layers for typical permalloy, but obtained strong coupling for materials with high cobalt contents.

In the case of films with a thin non-magnetic layer, however, it was found that:

1. The interaction energy between ferromagnetic layers is just the energy of the coupling field (Eq. (7)) if the thickness of the non-magnetic component is greater than 100 Å;

2. The interaction energy is the sum of the coupling field energy and exchange energy (Eq. (8)) when the nonmagnetic layer thickness is less then 100 Å.

The experimental method of determining $g'(\alpha)$ used in the present work leads to a confirmation of the data given in the literature and establishes the validity of the model of interaction by coupling field proposed in Ref. [5], as its consequence is coupling of the ferromagnetic type. For interaction of this type:

1. The values of the coupling field are positive, and

2. The angle between the magnetization vectors in the remanence state is smaller in films in which there is ferromagnetic coupling than in those in which there is no coupling.

The present paper was restricted to the study of films of composition near to 81% Ni + + 19% Fe because of their possible application in technology (films of this composition are characterized by optimum parameters: $H_k$, $H_c$, dispersion, *etc.*). It should be pointed out that by using this technique it is possible to obtain films which can be used as four state elements if the vacuum evaporation process is fully controlled. In particular, this concerns the evaporation rate and the substrate temperature during the deposition process, as composition fluctuations of the various layers are probably due to variations in the deposition rate.

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