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## An hysteretic Behavior of Evaporated Uniaxial $^{19}\text{Fe}^{81}\text{Ni}$ Thin Films

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(9. V. 68)

*Abstract.* AC field an hysteretic magnetization  $M_a(H)$  and remanence  $M_{ar}(H)$  were measured at 20°C on flat uniaxial  $^{19}\text{Fe}^{81}\text{Ni}$  thin films in directions parallel to the plane of film. The films were evaporated at substrate temperatures between 100° and 400°C, and in vacua better than  $10^{-6}$  Torr.

The states  $M_a(H)$  above the saturation remanence  $M_r(\infty)$  are shown to be thermodynamic equilibrium states from which the anisotropy constant  $K_u$  may be obtained.

Below  $M_r(\infty)$ ,  $M_a(H)$  and  $M_{ar}(H)$  are essentially determined by magnetic interaction fields arising from the dynamic domain structures present during the an hysteretic process. The dynamic domain structures are in equilibrium, the static structures at zero fields are not. From these facts a theory of the an hysteretic susceptibilities  $\chi_a$ ,  $\chi_{ar}$  is derived, and the results are shown to agree well with experiment.

### Dedication

This work is dedicated to Professor Georg Busch who, by the high standards and originality of his own work, has educated and encouraged the younger generations of physicists over more than three decades.

### I. Introduction

Anhyseretic quantities as acquired by either thermal cycling or in an *ac* field or by ultrasonic treatment may be considered to a generally well definable extent as equilibrium properties in the sense of thermodynamics. Therefore, in principle, they may be understood easier than any of the hysteresis properties, i.e. coercivity etc.

*ac* field anhyseretic measurements were first introduced by STEINHAUS and GÜMLICH [1] in 1915 as a method to determine the geometric demagnetization factor  $D_g$  of open magnetic circuits. However, soon it became evident [2] that the 'demagnetization factor' determined in this way did not depend on the shape of the sample alone but also on structural properties of the material.

More recently it was recognized [3, 4] that certain anhyseretic properties, such as the anhyseretic initial remanence susceptibility, are exclusively determined by magnetostatic interactions of the ferromagnetic material with its own stray fields that arise, for example, at all sorts of surfaces, external or internal. This means that (i) anhyseretic measurements will yield the effect of sample shape,  $D_g$ , only in cases where no internal sources of stray field exist, and (ii) that such measurements may be a sensitive tool in studying structural properties of materials, once  $D_g$  is eliminated.

Subject (ii) has been reviewed recently by one of the authors [5], and it has been shown for various types of materials how their *ac* anhyseretic behavior may be correlated to their characteristic structural features. Furthermore, it became evident in this work that, also with respect to anhyseretic properties, thin films are understood least of all material types. Therefore, respective studies on thin films were continued, and some of the results will be presented in the following.

### II. Anhyseretic Quantities

The *anhyseretic magnetization*,  $M_a(H)$ , is the magnetization acquired in a *dc* field  $H$ , after an *ac* field  $H'$  superimposed on  $H$  has been slowly decreased to zero from an initial amplitude  $H_1$  chosen such that the total net field is larger than the largest nucleation field in the sample for irreversible magnetization reversal in that direction. When finally, at  $H'_1 = 0$ ,  $H$  is reduced to zero, one obtains the *anhyseretic remanence*,  $M_{ar}(H)$ . The corresponding *initial susceptibilities*,  $\chi \equiv \lim_{H \rightarrow 0} (dM/dH)$ , are denoted by  $\chi_a$ ,  $\chi_{ar}$ , respectively. Later on, the reduced quantities  $j_a(H) \equiv M_a(H)/M_s$ ,  $j_{ar}(H) \equiv M_{ar}/M_s$  ( $M_s$  = saturation magnetization) will be used.

The role of the *ac* field  $H'$  in the process is twofold: (i)  $H'$  serves as a catalyst to eliminate hysteresis, i.e., to overcome energy barriers that prevent irreversible magnetization changes to occur in  $H$  alone, and (ii)  $H'$ , together with  $H$ , gives rise to structure dependent dynamic interaction fields that, in part, determine  $M_a$ ,  $M_{ar}$  [5]. Therefore, the form of the *ac* field (sinusoidal, rectangular) will have presumably little effect on  $M_a$ ,  $M_{ar}$ , whereas the frequency may be of influence [6]. Furthermore,

one expects from (ii) that  $M_a$ ,  $M_{ar}$  will depend on the direction of  $\mathbf{H}'$  relative to  $\mathbf{H}$ , and in anisotropic samples also on the directions of  $\mathbf{H}'$ ,  $\mathbf{H}$  with respect to sample orientation [5].

In this work  $\mathbf{H}'$  is taken always parallel to  $\mathbf{H}$ , and  $M_a$ ,  $M_{ar}$  denote always the components of magnetization parallel to  $\mathbf{H}$ , and hence are treated as scalars.

The effect of sample shape is usually described in terms of a demagnetizing field which is uniform in uniformly magnetized spheroids only, and is there given by

$$\mathbf{H}_D = D_g \mathbf{M},$$

where  $D_g$  is the demagnetization factor. Generally,  $D_g$  is a tensor and is mathematically determined by the ratios of the principal spheroid axes. For uniformly magnetized spheroids, the components  $D_{gi}$  of  $D_g$  may be determined also experimentally from  $\chi_{ai}$  along the principal axes,

$$\chi_{ai}^{-1} = D_{gi}. \quad (1)$$

For non uniformly magnetized spheroids (heterogeneous materials, for example), on the other hand, it has been found experimentally that

$$\chi_a^{-1} = D > D_g, \quad (2)$$

and the difference,  $\Delta D = D - D_g$ , has been attributed to 'internal interaction effects' arising from the structure of the material (c.f. for example NEEL et al. [7]). Unfortunately,  $\Delta D$  is not a constant, characteristic of a material, but depends also on  $D_g$  [8] as one would expect since both the structural and the shape effect have the same physical cause, namely magnetostatic interactions. By a rather general calculation of  $\chi_a(D_g)$  for a given constant value  $\chi_a(0)$  at  $D_g = 0$ , not regarding the physical origin of  $\chi_a(0)$ , NEEL [8] obtained

$$\chi_a^{-1}(D_g) = D_g / \{1 - \exp[-D_g \chi_a(0)]\}, \quad (3)$$

which formula was found [7] to agree well with experimental data on permanent magnet materials. From (3) follows that the structural effect,  $\Delta D(D_g)$ , is largest at  $D_g = 0$ , where  $\Delta D(0) = D = \chi_a^{-1}(0)$ , and decreases rapidly as  $D_g$  increases. This is important to keep in mind, since, if  $\chi_a^{-1}(0)$  is small, no structural effect at all may be detected if  $D_g$  is large at the same time. Actually, there are materials for which  $\chi_a^{-1}(0)$  is exactly zero [5], and, with respect to (3), this fact cannot be derived directly from measurements of  $\chi_a$ , because in practice it is impossible to obtain  $D_g = 0$  exactly.

Materials with  $\chi_a^{-1}(0) > 0$  are called type-I materials, those with  $\chi_a^{-1}(0) = 0$  type-II materials [5]. This classification has been made primarily with regard to applications in magnetic recording. A type-I material may be suitable for *ac*-bias analogue recording, whereas with a type-II material only digital recording is possible.

### III. Experimental Results and Discussion

#### 1. Experimental

$M_a(H)$  and  $M_{ar}(H)$  were measured at 20°C on evaporated flat films with composition 19Fe 81Ni (atomic per cent) in fields  $\mathbf{H}$ ,  $\mathbf{H}'$  parallel to each other and parallel to the film plane, using a self constructed Foner-type vibrating sample magnetometer.

The films were evaporated at vacua better than  $10^{-6}$  Torr in a magnetic field parallel to the film plane to produce uniaxial anisotropy in that plane. Substrate temperatures were kept constant during each evaporation, and for different films were varied between  $130^\circ$  and  $400^\circ\text{C}$ . The films were about  $1200 \text{ \AA}$  thick, and had diameters of 1.4 cm. All of the films behaved as normal, i.e.,  $H_c/H_K < 1$  ( $H_c =$  coercive field,  $H_K \equiv 2 K_u/M_s =$  anisotropy field).

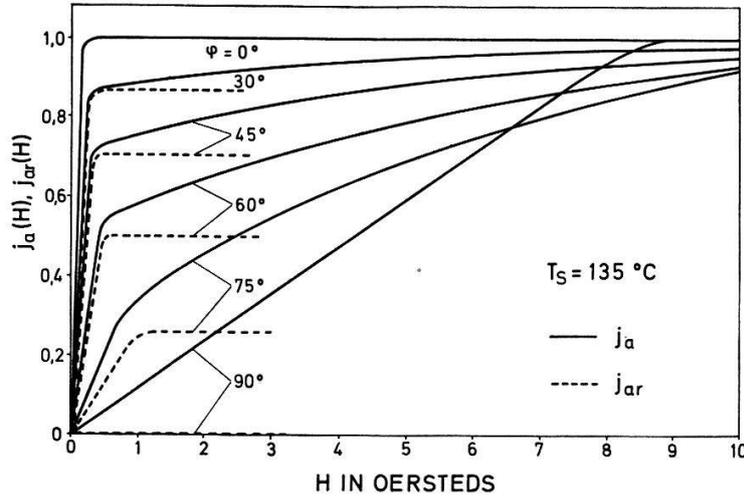


Figure 1

Reduced anhysteretic magnetization  $j_a(H)$  and remanence  $j_{ar}(H)$ , measured at  $20^\circ\text{C}$  on a  $1200 \text{ \AA}$  uniaxial film of  $19\text{Fe}81\text{Ni}$  evaporated at  $T_s = 135^\circ\text{C}$  on a glass substrate. Parameter is the angle  $\varphi$  between field and easy axis of film.

In Figure 1, a representative set of curves  $j_a(H)$ ,  $j_{ar}(H)$  is reproduced, as they were measured at various angles  $\varphi$  between the easy axis and  $\mathbf{H}$ ,  $\mathbf{H}'$  on a film that was evaporated at  $135^\circ\text{C}$ . For all of the other films, some of which were supplied by other laboratories and some were made in our own laboratory, the results are completely analogous. There was not a single exception from this simple, uniform pattern. This is one of the great advantages when one deals with equilibrium states.

## 2. Anhysteretic Magnetization, $M_a(H)$

Consider  $M_a(H, \varphi)$  at a given angle  $\varphi$ . Assume that the film is in thermodynamic equilibrium everywhere along  $M_a(H, \varphi)$  i.e., the Gibbs' free enthalpy  $G$  is at a minimum in any state  $M_a$ . As  $M_a(H, \varphi)$  increases, reversible work would then be done against the magnetic anisotropy forces which, at this film composition, stem almost entirely from the induced uniaxial anisotropy characterized by the free energy expression  $F = K_u \sin^2\varphi$ , and at the same time the magnetostatic self energy of the film,  $D_g M_a^2/2$ , would be increased. Hence, one would expect that the slope of  $M_a(H, \varphi)$  below a value slightly above the saturation remanence  $M_r(\infty, \varphi)$  is  $\chi_a \simeq D_g^{-1}$  and independent of  $\varphi$ , according to Equation (1), and that the total area  $A_a(\varphi)$  (c.f. Fig. 2a) bordered by the curves  $H/D_g$ ,  $M_a \equiv M_s$  and  $M_a(H, \varphi)$  is given by

$$A_a(\varphi) = A'_a(\varphi) + A''_a(\varphi) = \int_0^{M_s} [H - D_g M_a(H, \varphi)] dM_a(H, \varphi) = K_u \sin^2\varphi. \quad (4)$$

Experimentally, it was found that  $\chi_a$  is generally much smaller than  $D_g^{-1}$ , and depends strongly on  $\varphi$ . Furthermore, using  $K_u$  as calculated from torque curves that were measured on the same samples, it was found that, for  $\varphi = 90^\circ$ , Equation (4) holds within experimental error, whereas, for any  $0 \leq \varphi < 90^\circ$ ,

$$K_u \sin^2 \varphi = A'_a(\varphi) < A_a(\varphi), \tag{5}$$

where the area  $A'_a(\varphi)$  is defined in Figure 2a. These results mean that:

(i) all states  $M_a$  along that section of  $M_a(H, \varphi)$  which borders the area  $A'_a(\varphi)$  are equilibrium states corresponding to reversible rotation in unison of  $M_s$  against  $K_u$ , and

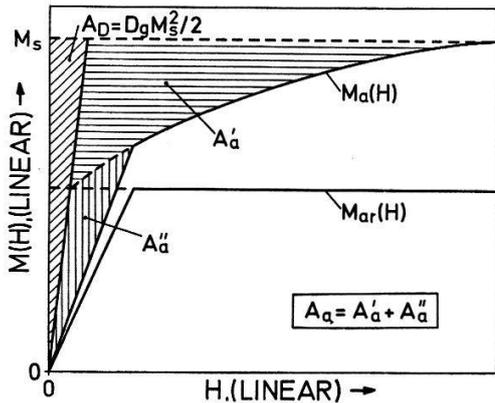


Figure 2a

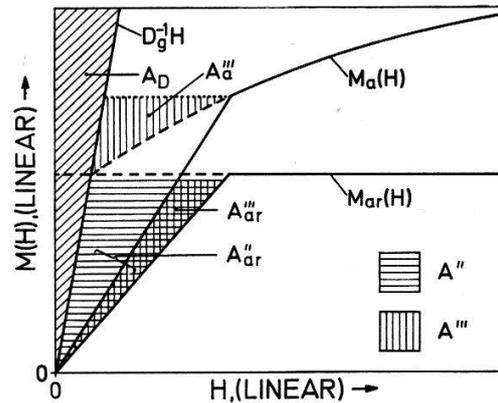


Figure 2b

Energetic analysis of anhyseretic magnetization  $M_a(H)$  and remanence  $M_{ar}(H)$ . For explanation, see text.

(ii) the states  $M_a$  along the lower section of  $M_a(H, \varphi)$  bordering the area  $A''_a(\varphi)$  (defined in Fig. 2a) are produced essentially by a different mechanism which must be identical to that which determines the slope of  $M_{ar}(H, \varphi)$  below  $M_{ar}(\infty, \varphi)$  because the transition  $M_a(H, \varphi) \rightleftharpoons M_{ar}(H, \varphi)$  is completely reversible as  $H$  is switched on and off, i.e., the areas  $A'''_a$  and  $A'''_{ar}$ , as defined in Figure 2b, are equal.

### 3. Anhyseretic Remanence, $M_{ar}(H)$

In the case of thermodynamic equilibrium,  $M_{ar}(H, \varphi)$  would increase from zero to the saturation remanence  $M_{ar}(\infty, \varphi) \equiv M_r(\infty, \varphi) = M_s \cos \varphi$  with a nearly constant slope  $\chi_{ar} \simeq \chi_a = D_g^{-1}$  that is independent of  $\varphi$ , according to Equation (1).

Experimentally it was corroborated that  $M_r(\infty, \varphi) = M_s \cos \varphi$  (c.f. Fig. 1). In fact, it turned out that a measurement of  $M_r(\infty, \varphi)$  is the most accurate way to determine the orientation  $\varphi$  of a film in the sample holder. Thus, the  $M_{ar}(\infty, \varphi)$  states are equilibrium states.

On the other hand, Figure 1 shows that  $\chi_{ar}$  is generally much smaller than  $D_g^{-1}$  and does depend strongly on  $\varphi$ , like  $\chi_a$ . Hence it is concluded from Equation (2) that  $\chi_{ar}$ ,  $\chi_a$  are mostly determined by a structural effect, metallurgical or domain, which now remains to be explained.

### 4. Initial Susceptibilities, $\chi_a, \chi_{ar}$

The decision between metallurgical and domain structure as cause of the structural effect does not seem quite trivial. The following experimental results may provide

some pertinent evidence: In the region  $0 < M_{ar}(H, \varphi) < M_r(\infty, \varphi)$ , domains must be present. Kerr optical observations of domain structures in  $M_{ar}$  states within this region show long, snake like domains. The domain walls run, on average, parallel to the easy axis  $x$  of a film with the mean magnetization directions parallel to the easy axis and opposite in adjacent domains. Let  $+x$  be the easy direction closest to  $+M_{ar}$ . For  $M = 0$  it seems reasonable that the mean widths  $d^+$ ,  $d^-$  of respectively (+)- and (-)-domains are equal. Yet  $d$  decreases rapidly as  $\varphi$  increases. These are well known results. However, as  $M_{ar}$  increases at a given  $\varphi$ ,  $d^+$  increases, whereas  $d^-$  remains constant, and the number of (-)-domains decreases as  $M_{ar}$  increases. These are rather strict rules whence it may be concluded that one has to do here with pseudo-equilibrium domain structures which result from the anhysteretic process. Therefore, it is conjectured that it is primarily the domain structure, or, more specifically, the cooperative mechanisms of reversible and irreversible magnetization changes during the anhysteretic process that determine  $\chi_a$ ,  $\chi_{ar}$ , rather than the metallurgical (grain) structure.

The mechanisms of magnetization changes in 19Fe 81Ni films have been described by MIDDELHOEK [9]. Depending on the angle  $\varphi$  between field and easy axis,  $x$ , the following mechanisms were observed:

(A)  $0^\circ \leq \varphi \leq 10^\circ$ . Motion of domain walls nucleated at the edge of the film.

(B)  $10^\circ < \varphi < \sim 55^\circ$ . Formation of 'bands' by partial rotation, followed by motion of the walls separating reversed from nonreversed bands, finally followed by motion of domain walls nucleated at the edges of the film.

(C)  $55^\circ < \varphi < 90^\circ$ . Formation of 'bands' by partial rotation, followed by motion of walls between bands only.

According to Figure 1 the investigated films are clearly a type-I material as defined at the end of Section II. For a certain class of such material (anisotropic solidified suspensions), one of the authors [5] has recently developed a theory of the anhysteretic process based on a model structure which is, in principle, quite similar to the domain structures in films during magnetization reversal as described by MIDDELHOEK [9]. On the basis of this theory,  $\chi_{ar}$  has been calculated from the condition that the component parallel to  $\mathbf{H}$  of the total field,

$$\mathbf{H}_{\text{tot}} = \mathbf{H} + \mathbf{H}' + \mathbf{H}_M,$$

integrated over one cycle of the  $ac$  field  $H'$ , is zero at that  $ac$  field amplitude  $H'$  where the magnetization freezes, i.e., where  $H_{\text{tot}} \simeq H_c$ .  $H_M$  is the dynamic interaction field [5].

The results are, for mechanism (A),

$$\chi_{ar} = \frac{2 \cot \theta}{N (\sin \phi^- - \sin \phi^+)}, \quad (6)$$

for mechanism (B),

$$\chi_{ar} = \frac{2 \cos \varphi}{N \sin(\varphi - \alpha') [\sin(\phi^- + \alpha') - \sin(\phi^+ - \alpha')]}, \quad (7)$$

and for mechanism (C),

$$\chi_{ar} = \frac{2 \cos \varphi}{N \sin(\varphi - \alpha) [\sin(\alpha + \phi^-) + \sin(\alpha - \phi^+)]}. \quad (8)$$

The symbols in the Equations (6)–(8) have the following meanings (c.f. also Fig. 3):  $\Theta$  is the mean angle of dispersion.  $\phi^+$ ,  $\phi^-$  are the angles between magnetization and easy axis,  $x$ , by which the magnetization is rotated reversibly just before nucleation of an irreversible magnetization change; for rotation in unison, these angles are given by STONER and WOHLFARTH [10], and for curling (or similar inhomogeneous reversal mechanisms) by SHTRIKMAN and TREVES [11]; the  $\phi$ 's depend on  $\Theta$  or  $\varphi$ , respectively, and on the reduced nucleation field,  $h_c \equiv H_c/H_K$ .  $\alpha$  and  $\alpha'$  denote the angles, respectively, between band walls or domain walls and the easy axis; both  $\alpha$  and  $\alpha'$  depend on  $\varphi$  (c.f. MIDDELHOEK [9]).  $N$  is the mean demagnetization factor of a domain or a band parallel to the plane of the film and perpendicular to the long axis of the domain or band, and is of the order of  $10^{-5}$ . Thus, according to Equations (6)–(8),  $\chi_{ar}$  is expected to depend essentially on  $\Theta$  or  $\varphi$ , respectively, and on  $h_c$ , and should be of the order of  $10^3$  to  $10^4$ , in agreement with experiment (c.f. Figures 1, 4, 5).

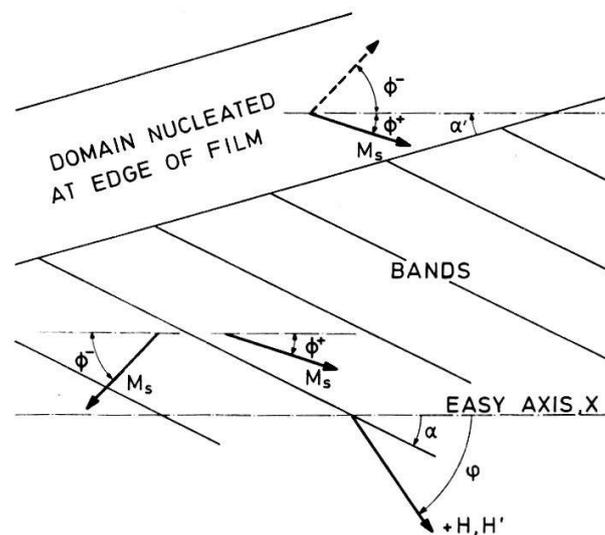


Figure 3

Definition of structural elements and angles that characterize the dynamic domain structure during the anhysteretic process.

In Figures 4 and 5, representative experimental results are shown for  $\chi_{ar}(\varphi)$ , and for  $\chi_{ar}(h_c)$  at constant  $\varphi$ , respectively. In essence, the results are quite well described by the theoretical curves calculated from Equation (7) or (8), depending on the range of angle  $\varphi$  where mechanism (B) or (C) applies. In the calculation, experimental [9] values were used for  $\alpha$ , and  $\alpha'$  was taken as approximately equal to zero.

From the comparison of theory with experiment, it is seen especially that the mechanisms must change at some angle  $\varphi$  between  $45^\circ$  and  $60^\circ$ , as was established experimentally by MIDDELHOEK [9]. The deviations in Figure 5 of some of the experimental data points from the theoretical curves are explained as follows: For a given angle  $\varphi$ , the data are taken on different films in order to vary  $h_c$ . Since  $\chi_{ar}$  depends not only on  $\varphi$  and  $h_c$ , but also on  $N$  and, especially at small  $\varphi$ , also on the ripple angle  $\theta$ , the data for a given  $\varphi$  would fall on one and the same theoretical curve only if  $N$  and  $\Theta$  were the same for all films. This is certainly not the case. This view is supported by the results in Figure 4. Here  $h_c$  also varies, as  $\varphi$  is varied, because the angles

$\phi$  depend on  $\varphi$  and  $h_c$ . Still, there is only little deviation of experimental data from the theoretical curves, because these data were taken on one and the same film.

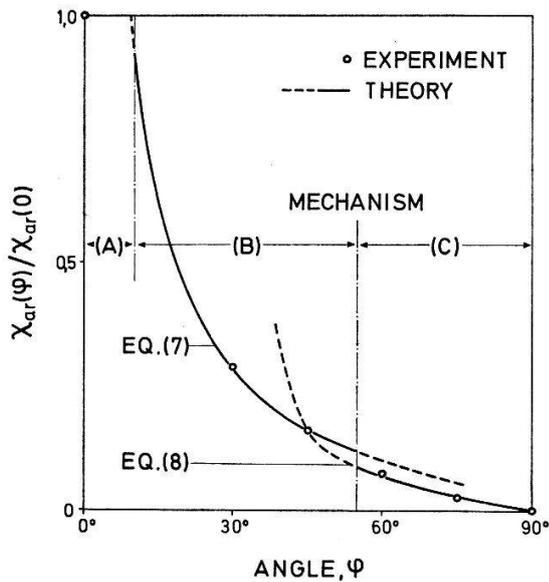


Figure 4

Anhyseretic susceptibility  $\chi_{ar}$  as a function of the angle  $\varphi$  between field and easy axis, measured at 20°C on the same film as in Figure 1. For this film,  $h_c \simeq 0.5$  below  $\varphi = 45^\circ$ , and  $h_c = h_0$  ( $h_0 =$  critical field for reversal by rotation in unison as given by STONER and WOHLFARTH [10] above  $\varphi = 45^\circ$ ). With these  $h_c$ , and with  $\alpha' = 0$ , Equations (7) and (8) give the same value of  $\chi_{ar}$  at  $\varphi = 45^\circ$ . At this point, the theoretical curves were adapted to the experiment.

Thus it appears that the anhyseretic process in uniaxial 19Fe 81Ni films is quite well understood by the present theory. Accordingly, the characteristic quantity  $\chi_{ar}(\varphi, h_c)$  is determined, apart from a very small shape effect ( $D_g \simeq 8.7 \times 10^{-5}$ ), by dynamic stray fields that arise from the domain structure which is in equilibrium while, with decreasing  $ac$  field  $H'$ , the total field  $H_{tot}$  goes through the range of the critical fields  $H_c$  of the film. As  $H_{tot}$  decreases below  $H_c$ , this domain structure freezes in, and is therefore no longer in equilibrium after  $H'$  and  $H$  have been reduced to zero. Therefore, the areas  $A_a''$  and  $A_{ar}''$  in Figure 2 do not constitute energies that are stored in the film in, respectively, the states of saturation and remanence, and hence are not explained by magnetization work, as was stated already in Sections III, 2. and 3.

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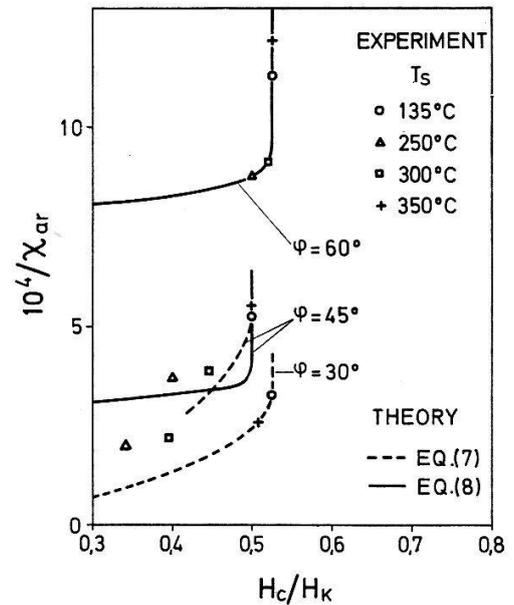


Figure 5

Reciprocal anhyseretic susceptibility  $\chi_{ar}^{-1}$  at 20°C of various 19Fe 81Ni films evaporated at different substrate temperatures  $T_s$  as a function of  $h_c \equiv H_c/H_K$ . Parameter is the angle  $\varphi$  between field and easy axis. The theoretical curves are adapted with one and the same factor.

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## Group Theory of Angular Correlations Related to Diatomic Rotators in Liquids and Solids

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*Abstract.* Angular correlations of diatomic rotators ( $\text{OH}^-$ ,  $\text{OD}^-$ ) in crystals (KCl) have recently been measured with the aid of vibration-rotation spectra. Here we examine by group theory the relations between the different angular correlation functions of diatomic rotators in liquids and solids as determined by electric dipole transitions and magnetic resonances.

*Résumé.* Nous avons récemment mesuré les corrélations angulaires de rotateurs diatomiques ( $\text{OH}^-$ ,  $\text{OD}^-$ ) dans les cristaux (KCl) en observant les spectres de vibration-rotation. C'est pourquoi nous étudions maintenant à l'aide de la théorie des groupes les fonctions de corrélation de ces rotateurs dans les liquides et les solides. Les résultats sont appliqués aux transitions dipolaires et aux résonances magnétiques.

*Zusammenfassung.* Kürzlich haben wir Winkelkorrelationen von zweiatomigen Rotatoren ( $\text{OH}^-$ ,  $\text{OD}^-$ ) in Kristallen (KCl) mit Hilfe von Rotations-Vibrationspektren studiert. Zum besseren Verständnis der experimentellen Resultate untersuchen wir in der vorliegenden Arbeit die Winkelkorrelationsfunktionen solcher Rotatoren mit gruppentheoretischen Methoden. Dabei werden besonders jene Korrelationsfunktionen berücksichtigt, welche bei elektrischen Dipolübergängen und magnetischen Resonanzen auftreten.

### Introduction

Random rotation of molecules in liquids strongly influences the line shapes observed in nmr, esr, vibrational and even Raman spectra. Considerable effort has