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Hungarian Academy of Sciences

CENTRAL
RESEARCH
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PHYSICS

BUDAPEST

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FERROMAGNETIC NUCLEAR RESONANCE INVESTIGATION OF THE SURFACE MAGNETIZATION IN IRON SHEETS

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ISBN 963 371 280 7
HU ISSN 0368 6330

ABSTRACT

The role of domain structure and domain properties in ferromagnetic nuclear resonance /FNR/ experiments is reconsidered. Using the FNR signal intensity as a measure of surface domain wall volume, it is found that the behaviour of the surface magnetization differs from that of the bulk magnetization of iron sheets. Namely, a "critical" field below which the FNR signal remains unchanged is observed in the surface magnetization. This "lag" of surface domain wall annihilation is sensitive to the given surface conditions and in particular, to the rolling deformation. On the other hand, the pinning introduced by cold working, rolling, or by ion implantation with boron, or by carbide precipitation has only a slight effect on the radiofrequency wall dynamics measured by rotary saturation technique at room temperature. Considering the small skin depth /~2μ/, the FNR as a surface testing method is discussed.

АННОТАЦИЯ

Рассматривается роль структуры и свойств доменов в ЯФР эксперименте. Интенсивность сигнала ЯФР, который характеризуется поверхностной величиной доменных стенок, дала сведение о разнообразном поведении намагничивания в железных пленках. Наблюдалось критическое значение магнитного поля, ниже которого сигнал ЯФР не уменьшается. Это опоздание в уничтожении доменных стенок чувствительно к конкретным свойствам поверхности, в частности к деформации при помощи проката. Напротив этого статистического поведения, было получено только небольшое влияние на радиочастотную динамику доменных стенок /эффект пиннинг/, которая была измерена при комнатной температуре методом насыщения в развертной координационной системе. Пиннинг обуславливался холодной прокаткой, карбидовым выделением и бором, внедренным при помощи ионной имплантации. Благодаря малой скиновой глубине /2 μм/ была рассмотрена возможность использования ЯФР, как метод для исследования поверхностей.

KIVONAT

Ujravizsgáljuk a doménszerkezet és doméntulajdonságok szerepét a ferromágneses anyagokon végzett mag mágneses rezonancia kisérletekben. Az FMR jel amplitudót a felületi doménfalak mennyisége mértékének tekintve kimutatjuk, hogy a felületi és térfogati mágnesezési folyamat teljesen különbözően megy végbe polikristályos vaslemez mintákon. Nevezetesen, a felületi mágnesezést vizsgálva egy "kritikus" tér figyelhető meg, ami alatt az FMR jelamplitudó változatlan. Ez a "késlekedése" a felületi doménfalak annihilizációjának függ a konkrét feluleti feltételektől, különösen a hengerlési deformációtól. Azonban a különböző eljárásokkal: hideghengerléssel, karbid precipitációval, implantált bórral az anyagba bevitt doménfalmozgást gátló pinning csak igen gyenge hatással van a rádiófrekvenciás faldinamikára, amit forgó telités technikával mértünk szobahőmérsékleten.

A kis szkinmélységből /~2μm/ adódóan megvizsgáljuk annak a lehetőségét, hogyan lehetne az FMR-t, mint felületi anyagvizsgáló módszert hasznositani.

Introduction

It is generally accepted that in multidomain ferromagnetic metals with appreciable crystalline anisotropy, the ferromagnetic nuclear resonance /FNR/ signal arises from the domain walls [1] where the oscillation of the internal field produces much larger effective enhanced exciting fields than in the domain. One of the most direct confirmations of this was reported by Bohn et al. [2] who measured the FNR signal intensity of nickel powder as a function of radiofrequency field strength and found two intensity maxima corresponding to the wall and domain resonances.

Due to the small skin depth /$\sim 2\mu$/ at FNR frequency of 45 MHz /comparable to the surface closure domain depth/ the FNR technique is particularly suitable for investigating the surface magnetization process of metallic iron. Thus, FNR measurements complement the information about the magnetization process obtained by more conventional techniques such as permeability spectra measurements [3] which reveal the internal /"bulk"/ domain structure and various visual observation methods [4] which can investigate only the superficial magnetic structure.

As regards the domain structure, two kinds of measurement can be made by FNR technique. First, one can study either the domain wall dynamics at 45 MHz [5] or the structure of the domain wall in the particular case of uniaxial ferromagnets [6] by enhancement factor mesurements. Second, accepting that the FNR signal arises from the surface domain walls, one can study the annihilation of these walls by measuring the decrease in the FNR signal intensity in an external magnetic field.

In this paper both kinds of measurement are described. In both cases continuous exciting technique of nuclear magnetic resonance in polycrystalline iron samples are used. The enhancement factor measured by the so-called rotary saturation technique [7] proved to be insensitive to structure, while the annihilation field measurements revealed a characteristic "lag" of the surface relative to the bulk magnetization. This "lag" proved sensitive to the given surface conditions. The annihilation of surface domain walls, that is, the saturation of surface magnetization could be reached at several hundreds of Oe, this value is comparable to the anisotropy field, and in contrast with the several tenth of Oe, the usual coercive force of bulk magnetization. Similar discrepancy between the surface and bulk magnetizations was observed by H.de Waard et al. [4] who measured the magneto-optical Kerr effect.

Experimental

A frequency modulated, fully transistorized oscillator was developed for these experiments [8]. It is based on a circuit first designed by Robinson [9]. The frequency of the oscillator can be tuned, swept and modulated by varicap diodes. The r.f. voltage on the tank circuit can be changed between 10-500 mV and the frequency stability kept at about 10^{-5} for a short period /15 min./. The main advantage of this detector is that even at high frequencies a tank coil of 8-12 turns on a relatively long /$\sim$ 20 cm/ connecting cable can be used which facilitates the experiments carried out in an external magnetic field and the detection of the PMR signal during mechanical tests.

The sheet samples of different thicknesses /4 - 500 μ/ had a purity of 99,99%. Except for the in situ mechanical deformation measurements, the sample was prepared from several sheets of 18x8 mm², parked in "sandwich" form separated by paper insulators. The sheet samples have the advantage over the generally used powder samples that they permit the surface conditions to be

controlled in metallographic and domain structure investigations. On the other hand quality factor of the tank circuit is reduced due to the higher permeability and the effective filling factor is smaller than for the powder samples. This disadvantage is partly counterbalanced by the fact that a sheet /bulk/ sample has an enhancement factor 3-4 times larger than that of the powder sample [10] .

The effect of mechanical deformation on the FNR signal was studied both under applied tensile strain and after cold rolling in order to investigate both the elastic and plastic deformation ranges.

A surface detection of the FNR signal was achieved with the aid of an open, toroidal sample coil. In this way the surface magnetization of actually bulk samples can be studied in a nondestructive way.

Results and discussion

Wall dynamics measurements

To study the dynamics of domain wall motion at 45 MHz, the enhancement factor distribution was measured by the so-called rotary saturation technique [7] . Rotary saturation means, in fact, a second resonance in the rotating frame arising from the audio-frequency modulation of the r.f. exciting field. This audio-frequency nuclear resonance /Fig.1./ is detected as a change in the FNR signal intensity with the modulating audio-frequency. The mean value of the enhancement factor can be determined from the point of minimum intensity,

$$\eta = \frac{2\omega_{min}}{\gamma H_x}$$

where H_x, the effective amplitude of the r.f. field, can be measured separately by means of a small inductive coil of several turns inserted in the sample coil. In this

way the measurement of the enhancement factor η , and its distribution is reduced to the measurement of audio-frequency.

It is worth mentioning that η , which contains an effective quantity H_x depending on the distribution of the wall orientation relative to the r.f. field direction is not the only parameter of the domain wall dynamics. The wall dynamics are better caracterized by the distribution of the transversal component of the internal field, the actual exciting field, which is reflected precisely by the rotary saturation signal.

In Fig.1. one can compare the dynamic properties of the surface closure domain walls in a powder sample with those of sheet samples of different thicknesses.

There is no great difference in the enhancement factors although the demagnetizing fields of the different samples differ from one another by three orders of magnitude. This indicates that the oscillations of domain walls in iron at 45 MHz and at room temperature are determined mainly by the viscosity term, the stiffness term, which is a fuuction of the demagnetizing field while the pinning of the wall is less important. This means that the displacement of the wall due to the r.f. field must be much smaller than the d.c. value determined by the demagnetizing field.

We have carefully investigated the role of the pinning in radio-frequency domain wall dynamics. Extrapinning was introduced by cold working, rolling, by ion implantation with B and by carbide precipitation. In every case only a slight change was observed in the rotary saturation signal as can be seen e.g. in Fig.2. for carbon precipitation.

These measurements suggest that the enhancement factor cannot be utilized as a structure sensitive parameter. This is in contrast to the drumhead model of Stearns [5] in which a dependence of the enhancement factor on the wall pinning is assumed. According to the Stearns model the impurities and the structural defects by augmenting domain nucleation and thus producing on the average more and smaller domain wall area segments should lead to a decrease in the enhancement factor for less pure samples. Her powder samples however differ not only in purity but in size and shape as well so that the observed differences in the enhancement factors may be due to the size and shape effect as it was found to be the case in the present measurements on powder and sheet samples.

Recently, Velez Gonzales et al. [6] have investigated domain walls in Ba $Fe_{12}O_{19}$ by NMR spin echo technique and found no evidence for a broad distribution of the enhancement factor due to wall pinning effects. The distribution of η is generated primarily by the orientation dependence of the magnetization within the domain walls /the domain wall structure/ which remains almost unchanged in the r.f. field, the oscillation amplitude is negligiable as compared with the width of the domain wall. In this particular case of an uniaxial ferromagnet, where mainly 180° walls exist, the structure of these walls could be studied experimentally by measuring the position dependence of the enhancement factor for nuclei within the wall, the measurement confirmed the predicted structure.

Annihilation field measurements

The second kind of experiment concerning the domain structure is the measurement of the decrease in the FNR signal as a function of the applied magnetic field. Since the FNR signal in the case of ferromagnetic metals originates only from the surface domain walls, one can follow the annihilation of these walls in an external applied field.

In Fig.3. the results obtained for polycrystalline iron sheets of different thicknesses are shown. There exists a thickness dependent "critical" field below which the FMR signal remains undiminished. It can be seen that the surface magnetization starts only above a certain field which is of several 100 Oe for thick /bulk/ samples.

This striking lag of the surface magnetization as compared to the bulk magnetization is in accordance with the Kerr rotation measurements of de Waard et al. [4] obtained for polycrystalline iron sample of 100 /um thickness. The existance of a "critical field" is in agreement also with Mercier's [11] and Herve's [12] but in disagreement with Bennet's [13] FMR results obtained for powder samples.

There are some experimental factors which influence the FMR measurements in an external magnetic field.

One of this is the orientation of the radio-frequency field relative to the external field. In a polycrystalline sample there are domain walls with different orientations relative to the exciting r.f. field H_x of the sample coil. The decrease in the FMR signal as the intensity of the applied field increases is more accentuated in the parallel /$H_x \parallel H_o$/ than in the perpendicular /$H_x \perp H_o$/ configuration because only the field exists in both configurations /Fig.4./.

The enhancement factor varies with H_o both in the powder and in the sheet samples. In Fig.5. the shift of the rotary saturation signal to lower frequencies in an external field of 200 Oe is shown. The decrease the enhancement factor is presumably due to the same processes which affect partly also the magnetic permeability. There is an increased wall stiffness in high fields probably because of the wall being shifted into a region of higher megnetostatic energy 13 .

Furthermore, the role of the demagnetizing factor had to be clarified. Though the packing of the sample influences also the actual demagnetizing factor, in our geometry the demagnetizing field is a very small fraction of the external field up to the saturation of the sample. In order to investigate the effect of the demagnetizing field on the H_o dependence of the FNR signal intensity, toroidal wound strip cores with 12 /um thickness were also measured. The experimental arrangement is shown in Fig.6. In a field of about 10 Oe all domain walls should disappear from the d.c. magnetization curve. However, the FNR signal is only slightly affected in such a low field. In this arrangement we could not apply a higher field than 50 Oe but this was sufficient to show that the surface domain walls persist in a much larger magnetic field than the ordinary saturating field where the internal walls are already suppressed.

This suggests a different annihilation mechanism of the surface domain walls from that of the internal walls, though the demagnetizing factor might influence the actual value of the critical field and may be correlated to the thickness dependence of the latter.

Seeing the peculiar behaviour of the surface magnetization, some of the material factors were also investigated.

The correlation between the domain pattern taken by Bitter technique and the FNR signal amplitude can be seen in Fig.7. The two sheet samples of 200 /um thickness had the same surface area of about 4 cm^2 but the grain sizes were different, $\sim$ 70 /um and 1000 /um respectively. The domain sizes do not differ by an order of magnitude, however, the smaller the grain size the smaller should be the domain size [14], hence a greater signal would be expected according to the greater number of the domain walls where the FNR signal is generated. The Bitter pattern persisted far above the ordinary saturating field in accordance with the data of the FNR annihilation field measurements.

A preliminary investigation of the effects of grain size, radiation damage, impurities on the surface magnetization of pure polycrystalline iron sheets has shown that the annihilation of surface domain walls is not very seriously affected by these factors. For example 600 wppm carbon, presumably in precipitated form, causes an increase in the oritical field at which the surface magnetization starts /Fig.8./. In this measurement the perpendicular configuration was chosen for the fields H_x and H_o where the ciritcal field is higher than in the parallel configuration, hence any variation of it is easier to detect.

It is of interest to see, how the annihilation of surface walls depends on the rolling deformation. One would expect an increase in the threshold field because of the pinning of domain walls on the crystal defects introduced by cold working. However, actually the critical field decreased with decreasing thickness. In Fig.9. one can see the decrease in FNR amplitude in external applied field for sheets with thickness of 25 /um, when the parameter is the final reduction in thickness by cold rolling.

It turned out that the texture of the sample produced by rolling deformation has an important role in the peculiar lag of the surface magnetization.

Finally, it ought to be mentioned that there is no hysteresis in the field dependence of the FNR amplitude. This shows that the pinning mechanism cannot be the dominant factor in the annihilation of surface walls.

To give some physical explanation to this peculiar behaviour of surface magnetization one has to start from the formation of the surface closure domain structure.

The over-riding driving force for domain creation in iron is the reduction of the magnetostatic energy. If the crystal anisotropy is large, as in the case of iron, one would expect a random orientation of the domain magnetization relative to the surface of the sample. The free-pole energy will be reduced by the formation of flux closure structure which effectively compensates the surface magnetostatic energy with the least expenditure of anisotropy, magnetostriction and wall energies.

Bitter technique investigation [15] revealed an increase in surface closure domain density with an increase in the angle of the axis of easy magnetization to the sheet of the sample. When the magnetization lies in the plane of the sheet, only a simple 180° wall structure is observed.

It can be seen that the surface closure domains gradually annihilate as the magnetization is rotated toward the plane of the sheet by increasing applied field. Thus the annihilation of the surface walls is essentially a rotation magnetization process determined by anisotropy H_K, demagnetization H_L, and applied field H_o while the pinning of the surface walls is less important.

Unfortunately, it is not possible to predict the actual surface domain structure in iron simply by a minimization of the total crystal energy including magnetocrystalline energy, magnetostriction, free-pole energy and the energy in an applied field. Nevertheless, considerable insight into this threshold phenomena can be gained by considering the order of magnitude of the dominant terms in the expression for the total energy.

Let us consider a model of randomly oriented grains with uniaxial magnetic anisotropy. The rotation process can be described by the minimization of the sum of energies

contributing to the domain magnetization I_o in the field H_o, H_K and H_D.

The external field is applied parallel to the sheet. The total energy can be written as

$$F = H_o I_o \cos\Theta - \frac{N_I}{\mu_o} I_o^2 \sin^2\Theta - \frac{N_{II}}{\mu_o} I_o^2 \cos^2\Theta + K\cos^2(\Theta_o - \Theta) \qquad /1/$$

where:

N_I and N_{II} are the perpendicular and parallel demagnetization factors relative to the sheet of the sample;

Θ_o the angle of the easy axis to the surface of the sheet;

Θ the angle between I_o and the surface of the sheet.

At a certain small angle Θ, not predictable yet, the surface closure domain structure disappears, and the corresponding annihilation field can be calculated from the condition $\frac{dE}{d\Theta} = 0$ taking the average on the distribution of angle Θ_o, as

$$H_o = \frac{1}{I_o \sin\Theta}\left[-K\sin 2(\Theta - \Theta_o) - (N_I - N_{II})\frac{I_o^2}{\mu_o}\sin 2\Theta\right. \qquad /2/$$

In accordance with the experimental facts, the annihilation field given by /2/ depends on the shape and size of the sample /N_I and N_{II}/ and on the texture of the sample produced by cold rolling which influences the crystallite orientation distribution /Θ_o/.

Of course, a more extensive investigation is needed to clarify the annihilation of surface domain walls and the related problem, the nucleation of magnetic domains. It is advisable to study the problem using a single-crystal

sample and care must be taken to avoid possible complications of the demagnetizing field.

Summarising, the FNR has proved a suitable method for studying the r.f. dynamics and the annihilation of surface domain walls. In Table I. the FNR technique is compared with others used for the study of magnetization processes:

Table I.

Measuring technique	Field of information	Magnetization frequency
Visual observation	the surface only	-
FNR	surface layer of $\sim 2\,\mu m$	45 MHz
Permeability spectra	inside of material	audio-frequency

Because of the relative simplicity of an FNR equipment the FNR technique is thought to be useful as a surface testing method. For convenience an open, toroidal FNR coil /Fig.10./ can be used With such a coil the surface of an actually bulk sample can be studied in a nondestructive way. It permits e.g. the effect of mechanical deformation on the FNR signal linewidth and amplitude to be investigated. A preliminary investigation [16] of pure iron samples revealed a weak contribution from the crystal effects and from the tensile stress to the parameters of the FNR signal. For example a 98 % reduction of sheet thickness by cold rolling does not increase the linewidth by more than 40%.

Acknowledgement

We wish to thank Prof.L.Pál for his continuous interest in this work, Dr.C.Hargitai, Dr.E.Kisdi-Koszó, and Dr.G.Zimmer for helpful discussions. We are indebted to Dr.K.Lázár for preparing the samples and to Dr.F.Tóth for the construction of the measuring apparatus.

Captions to the Figures

Fig. 1. Rotary saturation signal on powder and sheet samples H_x= 12 mOe

Fig. 2. The effect of 600 wppm C impurity on the RSS

Fig. 3. FMR signal as a function of the applied steady magnetic field

Fig. 4. The reduction of FMR measured in the configurations of parallel and perpendicular fields

Fig. 5. The decrease in the enhancement factor due to the applied field as refelected by the shift of the rotary saturation signal

Fig. 6. Sketch of the experimental arrangement for toroidal samples

Fig. 7. The correlation between the domain pattern taken by Bitter technique and the FMR signal amplitude The grain size G: a/~ 70 /um; b/ ~ 1000 /um

Fig.8. Increase in the critical field due to the carbon impurity

Fig. 9. Dependence of the annihilation field on the decrease in thickness ε by cold rolling

Fig.10. Sketch of the surface testing measurement on bulk samples

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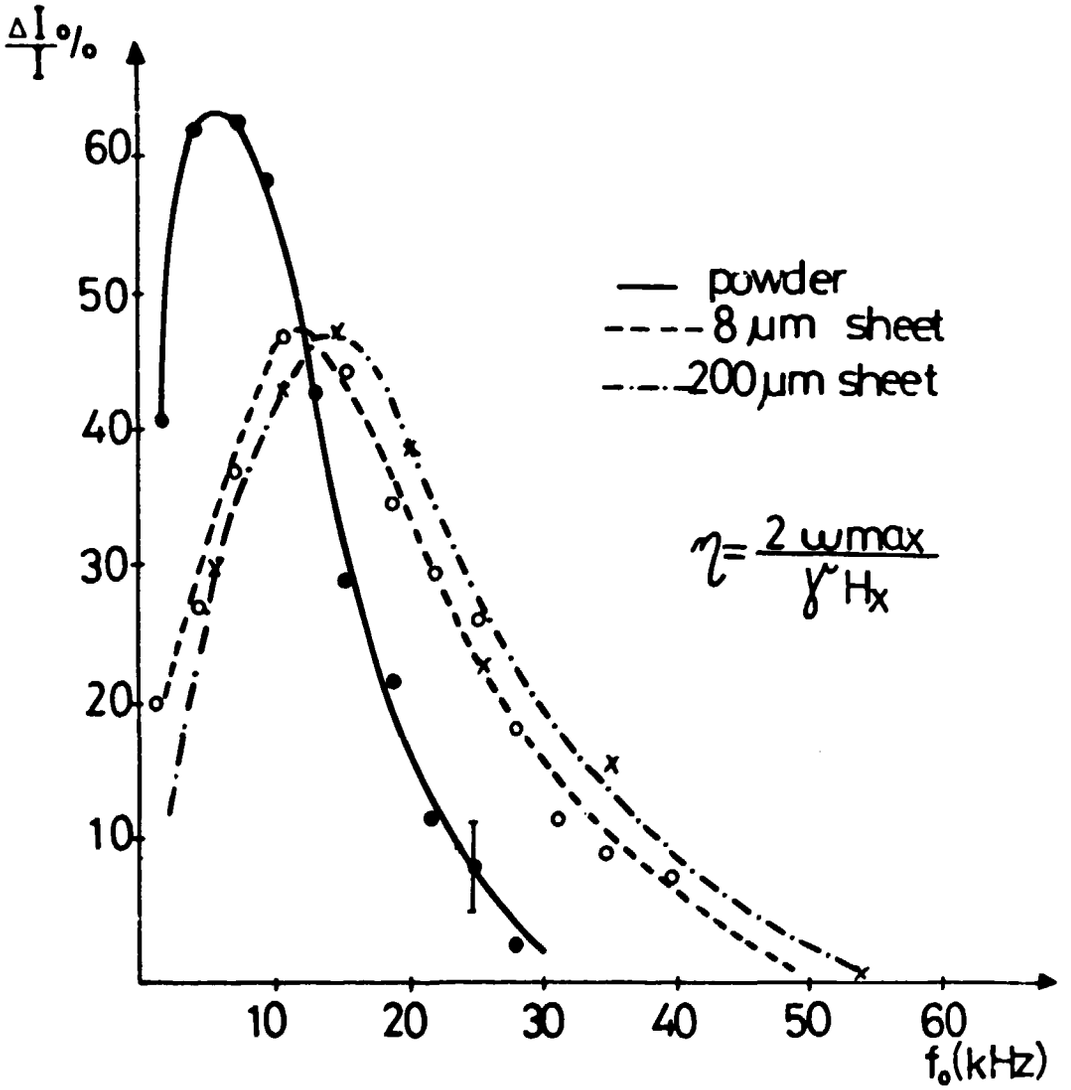
ΔI/I %
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50
40
30
20
10
10
20
30
40
50
60
f₀(kHz)
powder
8 µm sheet
200 µm sheet
η = 2 ωmax / γ Hx

Fig. 1.

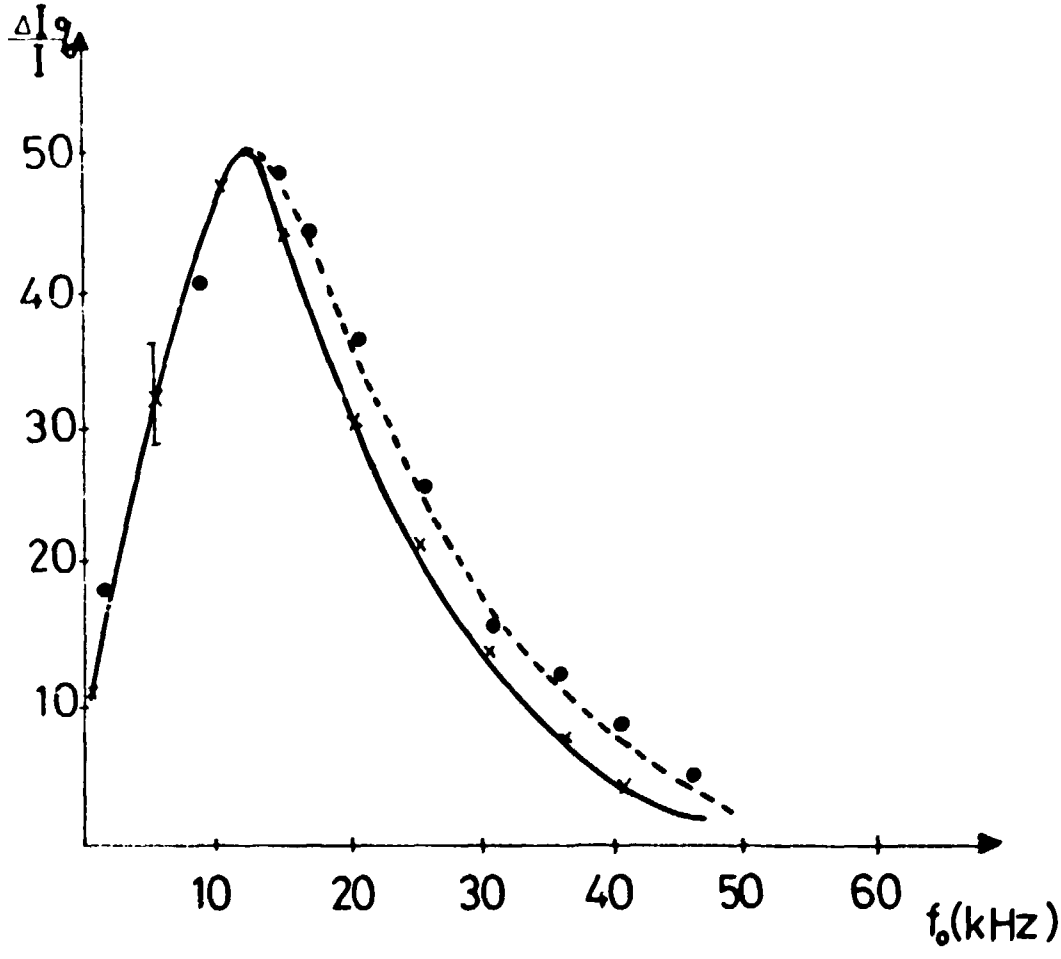
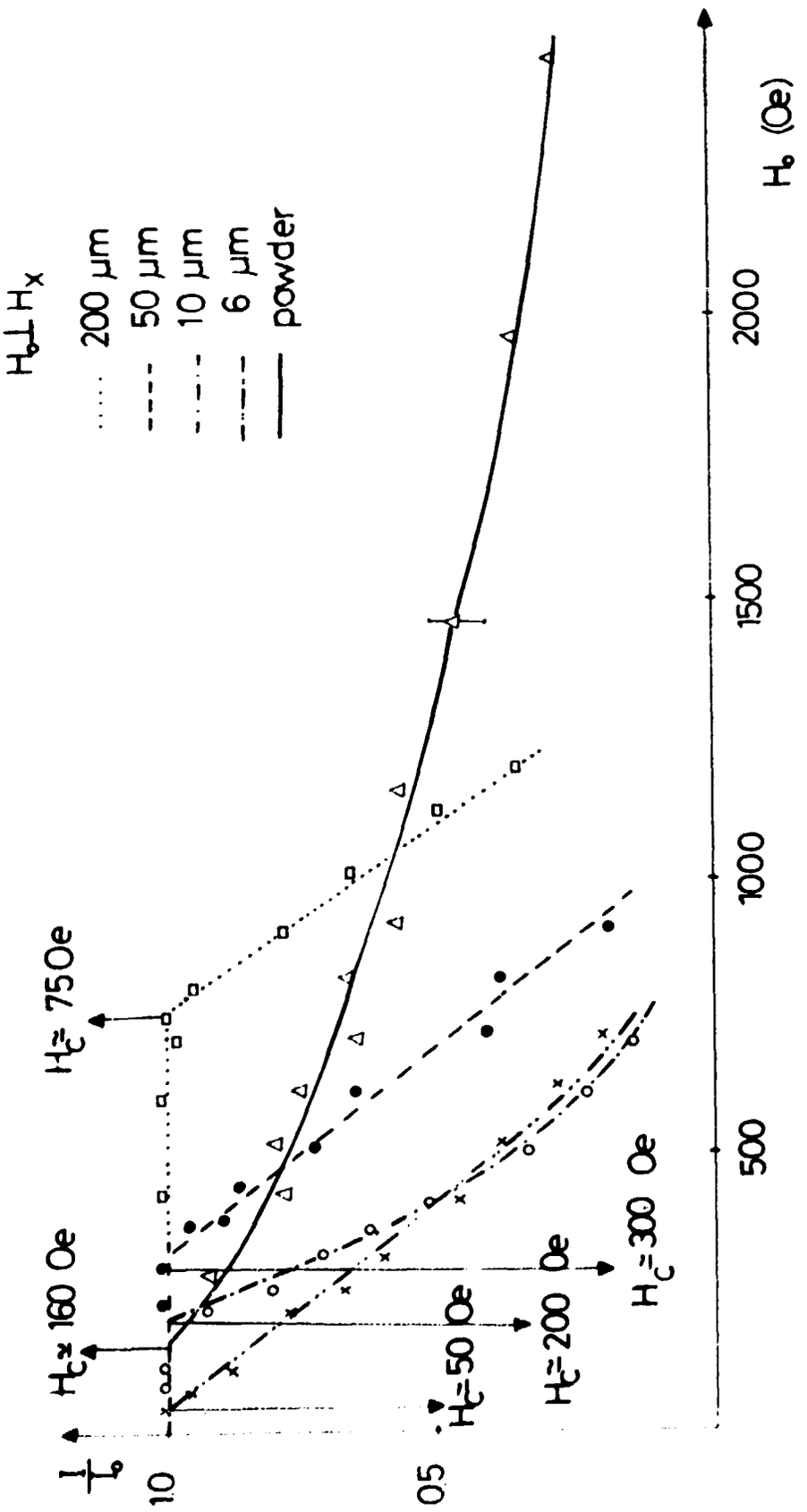


Fig. 2.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50 µm
10 µm
6 µm
powder
H₀ (Oe)
500
1000
1500
2000
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Hc≈ 750e
Hc≈ 160 Oe
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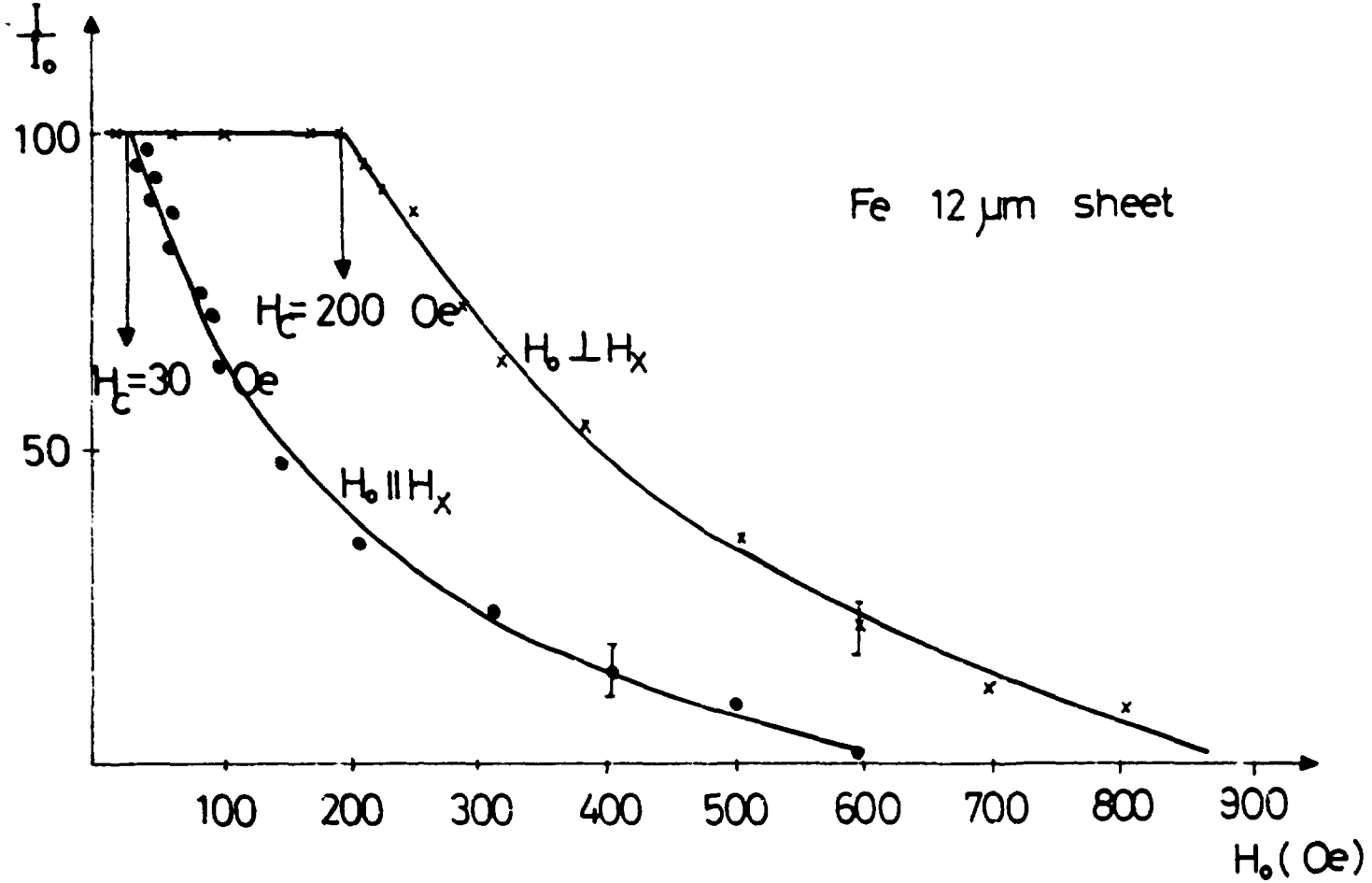
Hc≈200 Oe
Hc≈300 Oe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50
Fe 12 μm sheet
Hc= 200 Oe
Hc=30 Oe
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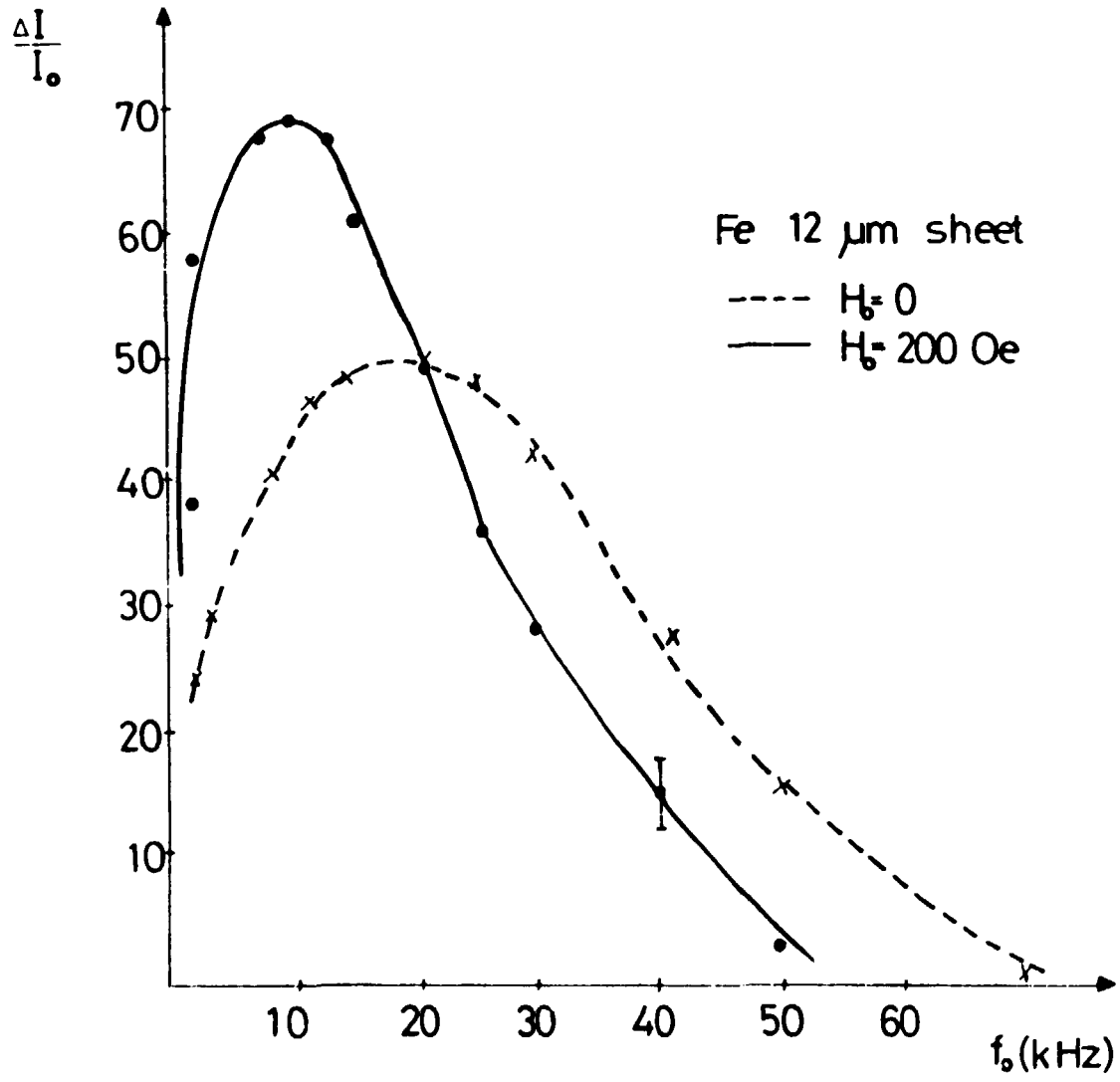
500
600
700
800
900
H₀ (Oe)

Fig. 4.


ΔI/I₀
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50
40
30
20
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H₀= 200 Oe
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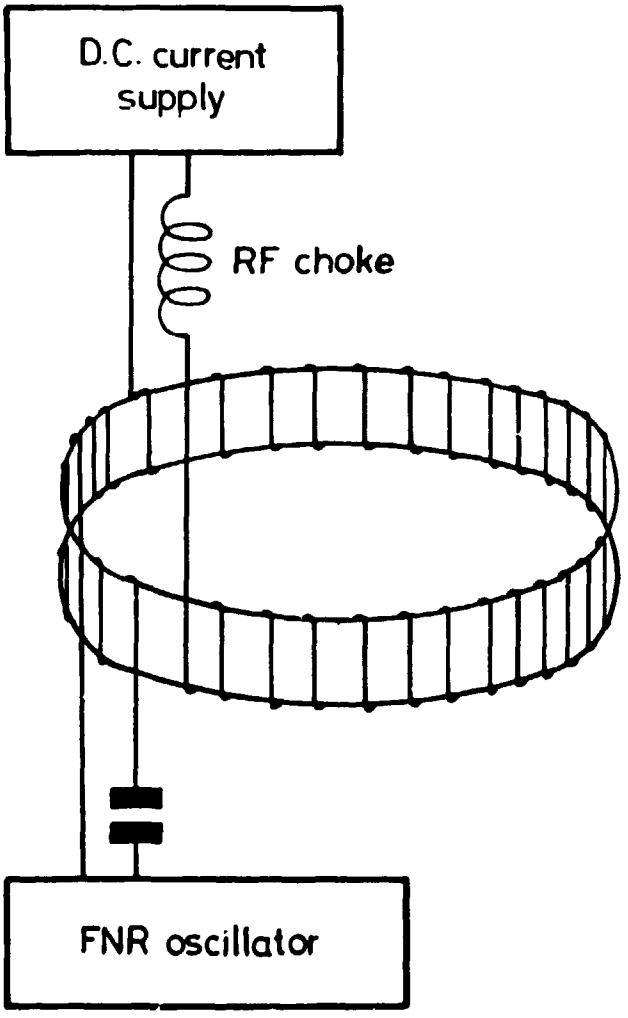
20
30
40
50
60
f₀(kHz)

Fig. 5.



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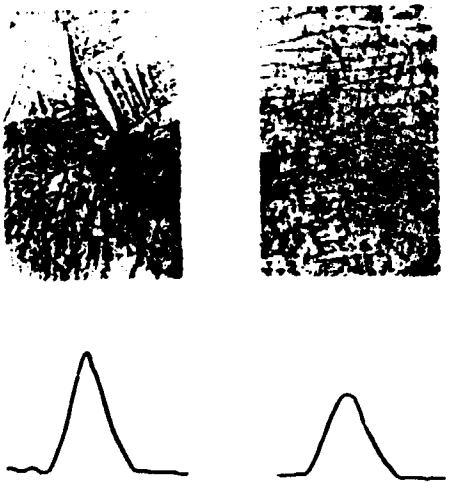
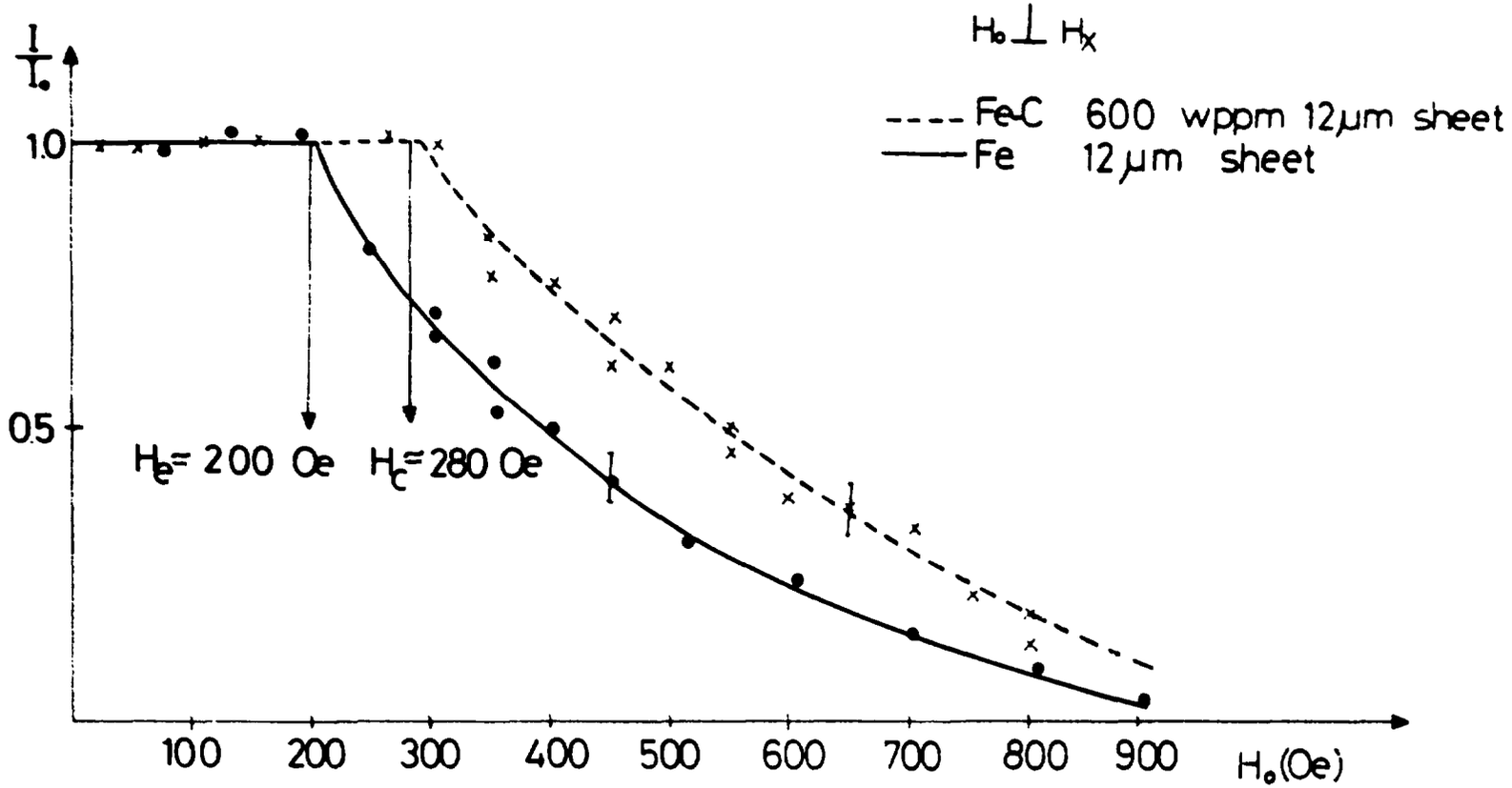


Fig. 7.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H₀ ⊥ Hx
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—— Fe 12μm sheet
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Hc= 280 Oe
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200
300
400
500
600
700
800
900
H₀(Oe)

Fig. 5.

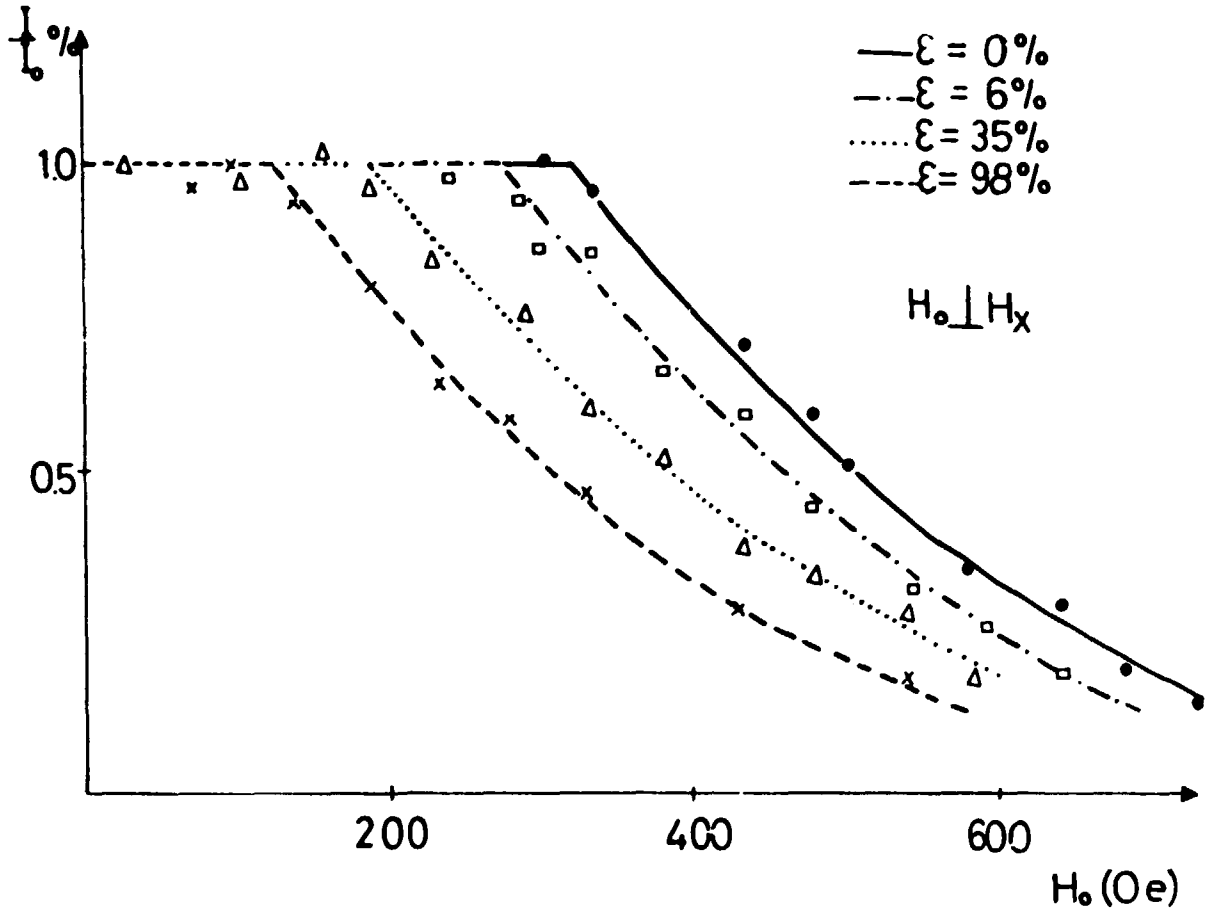

Fig. 9.

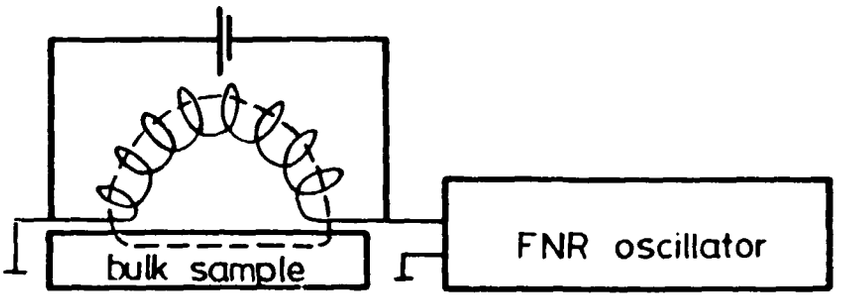

Fig. 10.

Kiadja a Központi Fizikai Kutató Intézet
Felelős kiadó: Vasvári Béla
Szakmai lektor: Harqitai Csaba
Nyelvi lektor: Kovács Jenőné
Példányszám: 300 Törzsszám: 1977-816
Készült a KFKI sokszorosító üzemében
Budapest, 1977. szeptember hó