MAGNETIC RESONANCE IN EPITAXIAL IRON FILMS

A. B. VAGANOV and B. HEINRICH[1)]

Institute of Physics Problems, USSR Academy of Sciences

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An investigation was made of the magnetic resonance in epitaxial films of iron evaporated in vacuum on cleaved faces of an LiF single crystal. The measurements were carried out in a magnetic field directed normal ($\lambda \sim 3$ cm) or parallel to the surface ($\lambda \sim 3$ cm and 8 mm). The results indicated the absence of any pinning of the surface spins in the investigated samples.

MUCH work has been done on spin-wave resonance in thin films since the discovery of this phenomenon.[1] The interest in this resonance is due to the fact that it helps us to understand certain problems in ferromagnetism (see Wolf's review[2]). However, the processes occurring during the evaporation of ferromagnetic films are complex and, therefore, the spin-wave resonance spectra of these films (on which most of the work has been done) are often more strongly affected by the technology of film preparation than by the physics of the resonance. Therefore, it would be of great interest to study spin-wave resonance in the physically most perfect samples, i.e., in single crystals or in epitaxial films.

The present authors are aware of two such studies. Waksmann et al.[3] investigated the spin-wave resonance of epitaxial Permalloy films. Epitaxial films of iron, nickel and cobalt were investigated by Rusov,[4] who observed spin-wave resonance in a static magnetic field parallel to the surface of the sample at a wavelength of ~ 8 mm. The absorption line width of the iron films was considerably greater than the line width of bulk single crystals and the existence of up to four absorption peaks demonstrated strong surface pinning of the spins.

The present paper reports an attempt to prepare epitaxial films of iron with magnetic properties close to those of iron single crystals and without surface pinning of the spins. The properties of these films were determined by means of the magneto-optical equatorial Kerr effect and from the magnetic resonance in a static magnetic field perpendicular ($\lambda \sim 3$ cm) or parallel ($\lambda \sim 3$ cm and 8 mm) to the surface of the film.

1. PREPARATION OF THE SAMPLES

Films were prepared by the successful method used in[5] for epitaxial films of iron, nickel, and cobalt. A single crystal of LiF was cleaved in air and placed in a vacuum chamber. Iron of 99.99% purity was evaporated in $\sim 10^{-4}$ mm Hg vacuum, established by a conventional oil diffusion pump. The LiF crystal was placed inside a heater. The substrate temperature was measured with a thermocouple. Before the evaporation was started the substrate was heated to a temperature slightly above 200°C by passing an alternating current through the heater. During the evaporation, this heater was switched off. The change in the substrate temperature during the evaporation did not exceed 10 deg C. The diameters of the films were 4 mm and the distance to the evaporator was 50 mm. The tungsten evaporator was carefully annealed before use. The substrate was screened during the preliminary melting of a charge and after the end of evaporation. The rate of evaporation was ~ 10 Å/sec.

The film thickness, measured by an interference method with an MII-4 microscope, was 700–1600 Å.

2. MAGNETO-OPTICAL MEASUREMENTS

Measurements of the magneto-optical equatorial Kerr effect[6] were used for quality control, the selection of the samples, and the study of the magnetic properties of the films. The magneto-optical signals of all our samples were equal to the signal of bulk iron. The fields corresponding to the magnetization saturation of anisotropic ferromagnets were known to depend on the orientation of the external field relative to the crystallographic axes. Therefore, we measured the amplitude of the hysteresis loop of the films during switching in an alternating field. This amplitude was determined as a function of the angle of rotation of a sample relative to the magnetic field (Fig. 1). The magneto-optical signal exhibited a 90° anisotropy in fields $\gtrsim 10$ Oe and the ratio of the magnetizations along the easy and difficult axes ranged from 0.63 to 0.75 (depending on the sample) in a field of ~ 20 Oe. The corresponding ratio for bulk single crystals of iron was known to be 0.73.[7] The demagnetization factor was estimated from the magnetization curves, which were deduced from the magneto-optical measurements. The estimate gave $\sim 10^{-3}$, in agreement with the

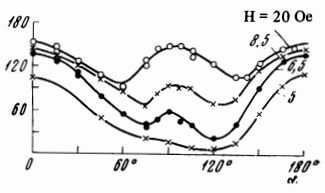

FIG. 1. Dependence of the magneto-optical signal (mV) on the angle of rotation of sample 1 in an external magnetic field.

[1)]Physics Institute, the Czechoslovak Academy of Sciences.

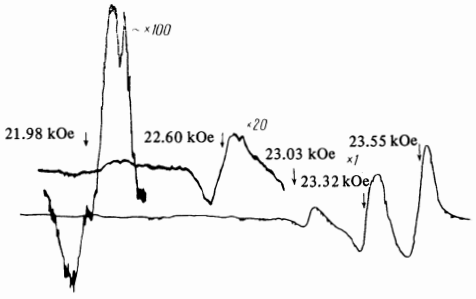

FIG. 2. Spin-wave resonance spectrum of sample 1 in the perpendicular configuration at f = 9.25 GHz. The arrows indicate the resonance peak positions.

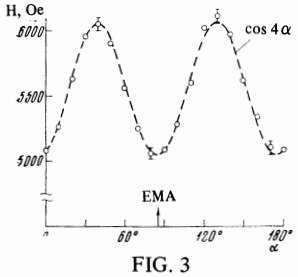

FIG. 3

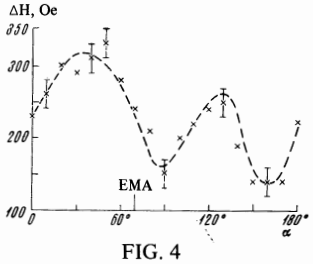

FIG. 4

FIG. 3. Angular dependence of the resonance field of sample 1 at f = 35.9 GHz. EMA is the easy magnetization axis.

FIG. 4. Angular dependence of the width of the resonance absorption line of sample 3 at f - 34.26 GHz. Film thickness L = 1640 ± 70 Å.

measurements carried out on cleavage steps in the substrate.

The coercive force of the investigated films ranged from 5 to 14 Oe, in agreement with the results of other workers.[8]

X-ray diffraction studies showed that the easy magnetization axes of the films coincided with the [011] axes of the substrates. The same orientation of the easy axes was reported in[5].

3. RESONANCE MEASUREMENTS

All the measurements of the resonance absorption were carried out using a conventional microwave spectrometer with a modulated external field. The signal reflected from the resonator was isolated by a selective amplifier and a phase-discriminating detector. A film was pressed against the outer wall of the resonator, which had an aperture of 2 mm diameter. The width of the absorption line was taken to be the difference between the fields corresponding to the maximum and the minimum of the derivative of the absorption signal.

Frequency was measured by means of standard wavemeters with an error of $\lesssim 10^{-3}$. The magnetic field was measured to within ± 50 Oe.

The magnetic resonance in the samples was determined for the perpendicular configuration at a frequency of 9.25 GHz. Figure 2 shows the resonance spectrum of sample 1, which exhibited the largest number of the absorption peaks. One other sample had two peaks but all the other samples had only one peak. The width of the principal peak was 90—200 Oe. The maximum variation of the resonance field from one sample to another did not exceed 0.3 kOe.

The resonance absorption in the parallel configuration was measured at wavelengths of 8 mm and 3 cm. In the 8-mm range, only one absorption peak was observed in every sample (Fig. 3). The line width depended on the angle of rotation of the sample relative to the static magnetic field (Fig. 4).

The measurements at 10 GHz yielded results similar to those reported in[9,10] for reasonably perfect epitaxial films, i.e., two absorption peaks were found when the external field was applied along the difficult magnetization axis (these peaks were within ± 8° measured from this axis).

4. DISCUSSION OF RESULTS

A. Quality of Samples

The magneto-optical measurements show that the investigated samples have a 90° anisotropy and that the structure-sensitive quantities, such as the coercive force and the ratio of the magnetizations along the easy and difficult axes in a field of ~20 Oe, are close to the values reported for reasonably perfect epitaxial iron films and for iron single crystals. The satisfactory agreement of the angular dependence of the resonance field with the cos 4α law in the case of the 8-mm results and those 3-cm results which were obtained in the parallel configuration also shows that the epitaxial structure of our films is reasonably perfect.

The characteristic widths of the resonance absorption lines for iron single crystals are $\Delta H(f \sim 9\ \text{GHz}) \sim 30$ Oe[11,12] and $\Delta H(f \sim 36\ \text{GHz}) \sim 150$ Oe,[12,13] whereas the widths for epitaxial films of iron are $\Delta H(f \sim 9\ \text{GHz}) < 40$ Oe.[9] The widths of the principal peak of our samples are $\Delta H(f \sim 9\ \text{GHz}) = 90-200$ Oe and $\Delta H(f \sim 35\ \text{GHz}) = 90-300$ Oe. The smallest value of the line width in the 8-mm range (parallel configuration) is observed for each sample along a direction which coincides approximately with the easy magnetization axis (EMA in Figs. 3 and 4). This width is approximately equal to the line width in the perpendicular configuration although the frequencies for the two configurations differ by a factor of nearly 4. The line width for polycrystalline iron films is approximately the same, and it also varies little with the frequency.[14] The frequency dependence of the line width may be a reflection of the conditions during preparation of the films.

The dependence of the line width on the angle of rotation of a sample in an external field is also reported by Rossing and Bjork[15] for epitaxial films of Permalloy, and by Vittoria et al.[16] for nickel single crystals. The cause of this dependence is not yet understood but it may be related to the regular distribution of oxygen in the crystal lattice (see[17,18]). Additional investigations on more perfect samples may furnish information on this point.

Nevertheless, the close agreement between the line widths of iron single crystals and those of our films in the 8-mm range demonstrates that the films have a reasonably perfect structure in the sense of homogeneity and the absence of internal stresses.

All these results show that the bulk magnetic properties of our samples should be identical with the corresponding properties of iron single crystals.

B. Pinning of Spins at the Surface

One of the most important problems in the interpretation of spin-wave resonance spectra is the behavior of of the magnetic moment at the film surface.[2] The method of preparation of the films and the agreement between their magnetic properties and those of iron single crystals show that the magnetic moment at the film-substrate interface changes in a layer whose thickness does not exceed several atoms. The thickness of the layer in which the magnetic moment varies rapidly at the film-air interface is at least equal to the thickness of the oxide which forms in air on the film surface. It is known that the steady-state thickness of the oxide layer on polycrystalline iron may reach ~50 Å.[19] There are grounds for assuming that the thickness of the oxide layer on single crystals is less than this value. Therefore, we may regard our films as uniform in respect of the magnetic moment with the exception of a surface layer ~ 50 Å thick.

Sparks[20] showed that the pinning of the spins at the surface of a thin film and, therefore, the spin-wave resonance depend on the ratio of the thickness ϵ of the layer in which the magnetic moment changes and a certain characteristic distance $\epsilon_{cr} = \sqrt{A/M_S^2}$, where A is the exchange interaction constant, which is $A = 2 \times 10^{-6}$ erg/cm for iron,[11] and M_S is the magnetic moment. The magnetic moment of iron is $M_S = 1.7 \times 10^3$ G and the corresponding critical distance is $\epsilon_{cr} \sim 80$ Å. If $\epsilon \lesssim \epsilon_{cr}$, the pinning of the spins at the surface is very weak, and it is less for the parallel than for the perpendicular configuration. These conditions are fully applicable to our samples. The variations in the degree of pinning from sample to sample may be due to the slower rate of evaporation toward the end of the cycle, i.e., due to variations in the composition of the surface layer. This effect was small for the technique used to prepare our samples. The best means for checking the constancy of the rate of evaporation is provided by the radiofrequency microbalance method.[21]

Thus, we may assume that the principal peak obtained in the perpendicular configuration corresponds to uniform precession. The resonance measurements and the generally accepted formulas for the frequency dependence of the resonance field of cubic crystals[11,22] can be used to determine the magnetic moment M_S, the anisotropy field H_K, and the gyromagnetic ratio γ. These values are $M_S = 1600$ G, $H_K = 0.53$ kOe, $\gamma = 1.87 \times 10^7$ rad·sec^{-1}·Oe^{-1} and the scatter of these values for different samples is within $\lesssim 10\%$. For iron single crystals, the corresponding values are $M_S = 1700$ G, $H_K = 0.50$ kOe, and $\gamma = 1.82 \times 10^7$ rad·sec^{-1}·Oe^{-1}.

In the absence of the surface spin pinning the absorption peaks should be distributed along the field axis in accordance with the quadratic dependence of the resonance frequency on the wave vector of the spin waves, which assumes the values $k_n = n\pi/L$ (n = 0, 1, 2, 3, 4), where L is the film thickness.[2] This distribution is observed for sample 1, which is the only one which has more than two peaks (Fig. 5). The value of the exchange constant $A = (1.7 \pm 0.5) \times 10^{-6}$ erg/cm calculated for this sample agrees, within the limits of the experimental error, with the value assumed for iron in[11].

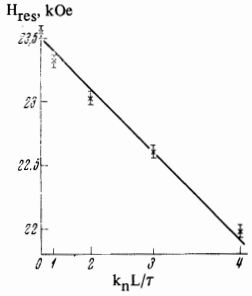

FIG. 5. Resonance fields for various spin-wave modes in sample 1 (f = 9.25 GHz, M = 1600 G, L = 1240 ± 70 Å.

C. Intensities of Absorption Peaks

We shall now consider the problem of the intensities of the spin-wave absorption peaks of thin films. The number of such peaks and their intensities depend on the spin pinning at the surface and on the ratio of the film thickness to the skin depth under resonance conditions. All these quantities are influenced by the technology used in the preparation of the films. Basically, the situation is as follows. As pointed out in[2], the power absorbed by a spin-wave mode is

$$P_n \propto \left(\int_0^L h m_n dz \right)^2, \qquad (1)$$

where $h = h_0 \exp(-z/\delta)$ is the high-frequency field in a film under resonance conditions; $\delta = \delta_0/\sqrt{\mu}$ is the skin depth under resonance conditions; $m_n = \cos k_n z$ in the absence of spin pinning at the surface, or $m_n = \sin k_n z$ in the case of total pinning. It follows immediately that, if the film thickness is $L \sim \delta$, few peaks are observed if spins are pinned at the surface, i.e., few peaks are observed for films which are not sufficiently homogeneous because pinning is usually due to a change in the saturation moment in the surface layer. If $L > \delta$, few absorption peaks are observed for homogeneous and inhomogeneous films because the high-frequency field is nonuniform within a film. In the absence of spin pinning in a film, the peaks are more closely spaced and their intensity decreases more rapidly than in the case of films exhibiting spin pinning. Therefore, films with no pinning exhibit a few strong peaks only in a certain range of thicknesses and Landau-Lifshitz damping constants.

Figure 6 shows the relative intensities of various spin-wave modes in the absence of surface pinning. These intensities are calculated using Eq. (1), on the assumption that the skin depth and the absorption line width do not differ from peak to peak. The spin-wave resonance spectra of our samples can be described quite satisfactorily in this way. Integration of the curves for sample 1 in Fig. 1 gives the intensity ratio 1 : 0.7 : 0.21 : 0.057 : 0.013. If these values are superimposed on Fig. 6, it is found that $L = 1240$ Å $= (3 \pm 0.3)\delta$, and hence $\delta = 400 \pm$ Å, which corresponds to $\mu = 1600 \pm 300$.

Kaganov[23] obtained an expression for the surface impedance of thin films in the absence of pinning in the

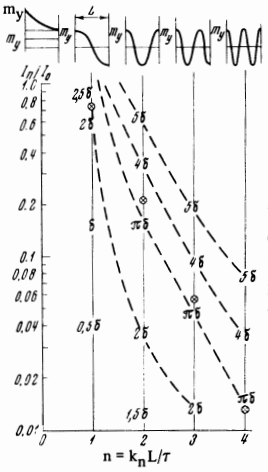

FIG. 6. Relative intensity of the excitation of various spin-wave modes in films of different thicknesses in the absence of the surface pinning of spins. The film thickness is given in terms of the skin depth under resonance conditions.

be perpendicular configuration. On the other hand, the surface impedance is known to obey

$$Z = (-\mu\omega / i \cdot 4\pi\sigma)^{1/2},$$

where σ is the electrical conductivity of the sample. Equating the two expressions for the impedance and substituting our value of μ, we find that $\lambda = 2.5 \times 10^8$ rad/sec, which is of the same order of magnitude as the value of λ deduced directly from the line width.

The other samples used in the present investigation were either too thick, so that the gap between the principal and the next peak along the field axis was less than the width of the principal peak, or they were too thin, so that $L < \delta$ and the intensities of the higher peaks were too low.

The present authors are aware of only one other study of samples exhibiting no pinning which could be used to check the suggested intensity scheme: this study is concerned with the sample described in[3]. An epitaxial Permalloy film, L = 850 Å thick, is reported in that paper to have three absorption peaks at 9.3 GHz in the perpendicular configuration. The distribution of these peaks along the field axis obeys the square law with n = 0, 1, and 2. The ratio of the intensities of the two extreme peaks is $I_2/I_0 \sim 10^{-2}$. Figure 6 gives $L = 1.4\delta$, i.e., $\delta \sim 600$ Å, which means that $\mu \sim 730$. An estimate based on this value of μ gives $\lambda = 5 \times 10^7$ rad/sec. The value calculated from the line width is $\lambda = 7 \times 10^7$ rad/sec.

D. Line Width

The absorption line width, ΔH, can be represented in the form

$$\Delta H = \Delta H_\sigma + \Delta H_\lambda;$$

ΔH_σ is due to the electrical conductivity of the film and ΔH_λ is due to the Landau-Lifshitz damping. In the case of thin films (see[23]), we have

$$\Delta H_\sigma = \frac{1}{3\sqrt{3}} \left(\frac{L}{2\delta_0}\right)^2 \cdot 4\pi M_S, \qquad (2)$$

where δ_0 is the skin depth. If we assume that the electrical conductivity of the films is the same as that of bulk iron, we find that—for $\mu = 1$—the skin depth is $\delta_0 = 1.62 \times 10^{-4}$ cm at 9.25 GHz and $\delta_0 = 0.85 \times 10^{-4}$ cm at 35 GHz. Soohoo[24] gives the following expression for ΔH_λ:

$$\Delta H_\lambda = 2\lambda\omega / \sqrt{3}\gamma^2 M_s, \qquad (3)$$

where λ is the Landau-Lifshitz damping constant. The damping constant is a structure-sensitive factor and its value for iron single crystals ranges from 0.4×10^8 to 1.0×10^8 rad/sec.[11]

Calculations based on these formulas give $\lambda \gtrsim 7 \times 10^8$ rad/sec for our samples at 9.25 GHz. In the 8-mm range, we find that $\lambda \sim 2.5 \times 10^8$ rad/sec, if we use the smallest value of the line width, and $\lambda \sim 6 \times 10^8$ rad/sec, if we use the largest value of this width.

CONCLUSIONS

Thus, the method described can be used to prepare epitaxial iron films whose properties are close to those of single crystals and which exhibit no pinning of the surface spins, i.e., in which the surface layer with a different value of the magnetic moment is $\lesssim 80$ Å thick. Samples exhibiting no pinning and having strong higher absorption peaks were obtained for the first time. Spin-wave resonance is sensitive to the magnetic properties in such thin films. This is useful in experiments in which the surface-layer magnetization plays a decisive role, for example, in investigations of the polarization of photoelectrons emitted from transition metals.[25]

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