Presence of Magnetic Surface Anisotropy in Permalloy Films

G. C. Bailey and C. Vittoria
Naval Research Laboratory, Washington, D.C. 20375
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The existence of uniaxial magnetic surface anisotropy in Permalloy films has been deduced by comparing the theoretical and experimental spin-wave-resonance spectra of several films with different thicknesses. The anisotropy decreases as the film thickness decreases and, for the thinnest film, becomes comparable to that given by Néel's theory (+0.08 erg/cm^2).

INTRODUCTION

Although a number of recent[1–5] (as well as not so recent) papers have dealt with the problem of surface magnetic anisotropy of thin films, there have been no direct definitive experimental measurements of the phenomenon in metal films since Néel first postulated the existence of such an effect many years ago. There are two factors which make possible the present description of magnetic surface effects: (i) The films studied here are of good quality with narrow ferromagnetic-resonance (FMR) linewidths and are evaporated in very high vacuum[6] with the result that the film surfaces are relatively clean. (ii) The macroscopic calculations[7] of the FMR field, linewidth, and intensity of magnetic metals are complete to the point where surface effects are included in the calculations. Films with poor surface "quality" can mask intrinsic surface effects and therefore cannot be readily described theoretically.

Recently,[8] it has been shown that, by *assuming* a surface uniaxial anisotropy K_s, a good fit to an observed perpendicular spin-wave-resonance spectrum could be obtained. However, to critically test the hypothesis of a surface anisotropy and to substantiate the uniaxial nature of the anisotropy, another independent experiment is needed. One crucial test of our hypothesis is to use the *same* values of K_s obtained from perpendicular resonance to fit the parallel-resonance spectrum *completely*. We choose to study the parallel-resonance spectrum instead of a perpendicular spectrum at a different frequency, say, because the parallel spectrum is quite different from the perpendicular spectrum both from the point of view of line intensity as well as line position (see Fig. 1). In the present paper we show that (i) the parallel spectrum of a given film can be fitted very well using the same parameters and the same value of K_s which are used to fit the perpendicular spectrum of that film, (ii) both perpendicular- and parallel-resonance fits for a number of films with various thicknesses show that K_s is a function of film thickness, in direct contrast to work by others,[4] and (iii) for the thinnest film, K_s approaches the value given by the Néel surface-anisotropy theory.[9]

THEORY

The spin boundary conditions used previously[8] were those appropriate to a uniaxial surface anisotropy with the anisotropy axis perpendicular to the film plane, namely,

$$A\frac{\partial m_{x,z}}{\partial n} - K_s m_{x,z} = 0\,, \tag{1}$$

where A is the exchange stiffness parameter, m_x and m_z are the transverse planar components of the rf magnetization, and n is the direction of the outward normal to the film plane, along the y axis. For parallel resonance, a uniaxial surface anisotropy requires that the spin boundary conditions be

$$\frac{\partial m_x}{\partial n} = 0\,, \tag{2}$$

$$A\frac{\partial m_y}{\partial n} - K_s m_y = 0\,, \tag{3}$$

where m_y is the rf magnetization normal to the film plane. Comparing Eq. (1) and Eqs. (2) and (3), it is readily seen that for perpendicular resonance both of the planar components of m "feel" a constraint on their motion,[10] while for parallel resonance, only the normal component is constrained. Furthermore, if the anisotropy axis were in the film plane, m_x and m_y would be pinned for parallel resonance,[11] but only m_z would be pinned (and m_x would be free) for perpendicular resonance.

The detailed boundary conditions with the hard axis normal to the film plane at the two surfaces of the film and for the case of parallel resonance are

$$\sum_{i=1}^{3}\left[A_i e^{-jk_i d} + B_i e^{jk_i d}\right] = h_d\,, \tag{4}$$

$$\sum_{i=1}^{3}(A_i + B_i) = h_0\,, \tag{5}$$

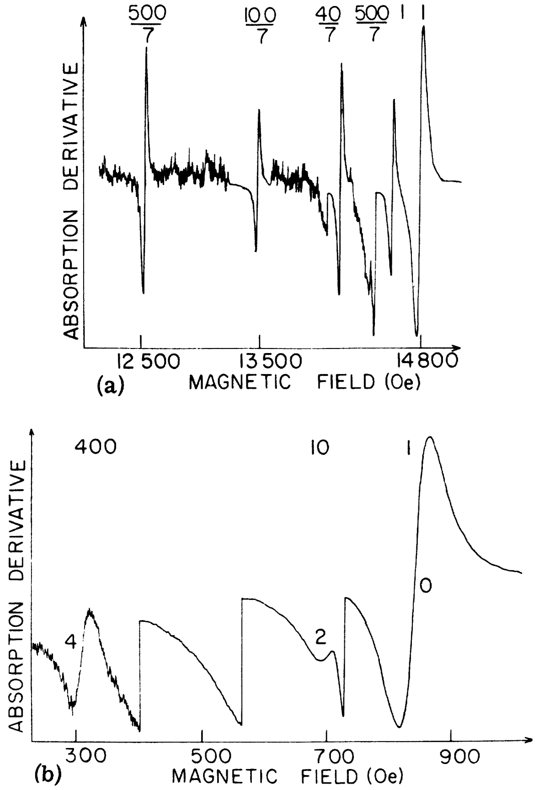

FIG. 1. (a) Spin-wave-resonance absorption derivative for a 2700-Å-thick 75-at.%-Ni–25-at.%-Fe film with the dc magnetic field perpendicular to the plane of the film (from Ref. 8). The numbers above the lines are the gain settings used. (b) Same as (a) except that the dc magnetic field is parallel to the film plane.

$$\sum_{i=1}^{3} Q_i \mu_i \left(\frac{K_s^{(d)}}{A} (A_i e^{-jk_i d} + B_i e^{-jk_i d}) + jk_i (A_i e^{-jk_i d} - B_i e^{jk_i d}) \right) = 0\,, \tag{6}$$

$$\sum_{i=1}^{3} Q_i \mu_i \left(\frac{K_s^{(0)}}{A} (A_i + B_i) + jk_i (A_i - B_i) \right) = 0\,, \tag{7}$$

$$\sum_{i=1}^{3} Q_i jk_i (A_i e^{-jk_i d} - B_i e^{jk_i d}) = 0\,, \tag{8}$$

$$\sum_{i=1}^{3} Q_i jk_i (A_i - B_i) = 0\,, \tag{9}$$

where

$$\mu_i = \frac{H + M/Q_i - 2Ak_i^2/M}{j\omega/\gamma + \lambda(H/M + 4\pi)/\gamma}$$

and $Q_i = 1 - \frac{1}{2} j\delta^2 k_i^2$.

Equations (4) and (5) state that the tangential component of the rf magnetic field is continuous across the film surfaces; Eqs. (6)–(9) describe the pinning conditions of the rf magnetization at the surfaces [see Eqs. (2) and (3)]. The quantities k_1, k_2, and k_3, which are the propagation constants of the linearly polarized magnetic fields, can be obtained from the secular equation.[7] The six unknowns A_i and B_i ($i = 1, 2, 3$) represent the field strengths of the resonant linearly polarized waves; h_0 and h_d, $K_s^{(0)}$ and $K_s^{(d)}$ are the magnetic rf fields and the surface-anisotropy constants, respectively, at the two surfaces; δ is the classical skin depth $c/(2\pi\sigma\omega)^{1/2}$, and d is the film thickness. Finally, H is the internal static field and $\gamma = ge/2mc$. The FMR field H and linewidth ΔH are obtained by the same procedure as outlined in Ref. 8 with the exception that here three k values[7] must be used instead of two.

RESULTS

The experiments were carried out at room temperature and at 9.44 GHz using 400-Hz field modulation. Other experimental details and film-preparation techniques have already been described.[8]

The experimental and calculated parallel-resonance results are compared in Table I for the 2700-Å film. It is to be noted that exactly the same parameters, $K_s^{(0)} = 0.435$ erg/cm^2 and $K_s^{(d)} = 0.55$ erg/cm^2, were used to obtain the parallel-resonance spectrum as were used to fit the perpendicular-resonance spectrum previously reported.[12] The calculated resonance data were obtained using both positive and negative values of K_s for each of two choices of the spin boundary conditions. That is, taking m_x free and m_y partially pinned corresponds to the uniaxial-surface-anisotropy case considered in Eqs. (2)–(9) with the anisotropy axis normal to the film plane and with the hard axis either normal to (K_s positive) or in the plane of the film (K_s negative). In addition, both normal and transverse components were taken as partially pinned corresponding to uniaxial anisotropy, but with the anisotropy axis in the film plane. It is seen that the calculated data for K_s positive and with the anisotropy axis normal to the film plane agree remarkably well with the observed resonance fields, linewidths, and intensities. Moreover, no good over-all agreement is obtained with any other choice of boundary conditions or signs of K_s. Specifically, the line positions and the intensities calculated for the case of the anisotropy axis in the film plane (C and D of Table I) show large disagreement with the experimental values. Although the intensities which are calculated using a negative value of K_s with the axis normal to the film plane (B of Table I) agree moderately well with experiment, the calculated line positions are not as close to the experimental ones as are those which are obtained using K_s positive. Furthermore, using a negative value of K_s for the perpendicular spectrum of Ref. 8 gives results which are in strong disagreement with the experimental

TABLE I. Parameters for in-plane resonance ($d = 2707$ Å).

H_n(Oe)					ΔH_n (Oe)					I_n [a]				
Expt.	Theory				Expt.	Theory				Expt.	Theory			
	A	B	C	D		A	B	C	D		A	B	C	D
837	835	824	780	804	48	48	51	55	56	100	100	100	100	100
692	687	700	424	604	24	25	b	20	21	0.35	0.58	0.17[c]	18	22
307	306	292	132	415	26	19	b	b	b	0.05	0.08	0.05[c]	0.05[c]	0.02[c]
				171					18					6.1

	$K_s^{(0)}$ (erg/cm²)	$K_s^{(d)}$ (erg/cm²)	Spin boundary conditions
A	0.435	0.55	$\frac{\partial m_x}{\partial n} = 0$ and $A\frac{\partial m_y}{\partial n} - K_s m_y = 0$
B	−0.435	−0.55	(A, B)
C	0.435	0.55	$A\frac{\partial m_x}{\partial n} - K_s m_x = 0$ and $A\frac{\partial m_y}{\partial n} - K_s m_y = 0$
D	−0.435	−0.55	(C, D)

[a] All intensities are normalized to the main line intensity and are multiplied by $\Delta H_n/\Delta H_0$.
[b] The line intensities were too small to give reliable values of ΔH_n.
[c] Assuming $\Delta H = 20$ Oe.

values (see Table II). Thus, the surface anisotropy is uniaxial and has a hard axis normal to the film plane.

Similar results are shown in Table III for films with thicknesses of 2020, 1300, and 790 Å. In view of the results for the 2700-Å film, the fit for all cases listed in Table III was obtained by taking K_s positive and the uniaxial axis normal to the film plane. In addition the values of λ (7.5×10^7 Hz), σ ($0.7\times10^5\ \Omega^{-1}\,\mathrm{cm}^{-1}$), g (2.10), A (1.143×10^6 erg/cm), and h_d/h_0 (1) were assumed to be thickness independent. For each case, K_s and M_s were varied to obtain the fit of both the parallel- and perpendicular-resonance spectrum. The agreement between theory and experiment for the line position, which is the parameter which can be measured quite accurately, is quite good. The linewidth and intensity comparison cannot be made in some cases because of the nonresolution of the calculated lines. Nevertheless, where comparisons can be made of I_n and ΔH_n the agreement is good. The calculated and observed perpendicular-resonance parameters for the films considered in Table III show agreement which is as good as or better than that given previously for the 2700-Å film (see Table IV). The cause of the apparent disagreement between theory and experiment for the higher-order linewidths can be ascribed to a small thickness variation across the plane of the film as discussed before.[8] The values of K_s obtained for the films are shown in Fig. 2 as a function of film

TABLE II. Parameters for perpendicular resonance ($d = 2707$ Å).

H_n (Oe)			ΔH_n (Oe)			I_n		
Expt.	Theory		Expt.	Theory		Expt.	Theory	
	A	B		A	B		A	B
14780	14780	14757	52	59	57	100	100	100
14569	14580		22	21		24	23	
14390	14392	14400	21	18	20	0.03	0.02	19
14151	14147		22	16		5.2	6.8	
a	13840	13747	a	14	14	a	0.03	6.3
13481	13466	13317	21	13	12	1.3	3.3	0.04
a	13028	12820	a	12	10	a	0.03	4.14
12536	12525	12254	21	9	9	0.44	2.7	0.04
a	11955	11620	a	7	7	a	0.06	9.7

	$K_s^{(0)}$ (erg/cm²)	$K_s^{(d)}$ (erg/cm²)
A	0.435	0.55
B	−0.435	−0.55

[a] The line intensities were too small to give reliable values for this parameter.

TABLE III. Parameters for in-plane resonance.

H_n (Oe)		ΔH_n (Oe)		I_n	
Expt.	Theory	Expt.	Theory	Expt.	Theory
$d = 2023$ Å					
865	863	32	35	100	100
617	607	23	a	0.13	0.12[b]
316	308	24	a	0.007	0.007[b]
$K_s^{(0)} = 0.15$, $K_s^{(d)} = 0.4$, $M = 887.4$					
$d = 1299$ Å					
857	855	26	25	100	100
709	710	a	a	≈ 0.016	a
280	264	25	19	0.07	0.045
$K_s^{(0)} = 0.2$, $K_s^{(d)} = 0.35$, $M = 899.1$					
$d = 790$ Å					
865	862	25	21	100	100
480	468	24	18	0.018	0.05
$K_s^{(0)} = 0.075$, $K_s^{(d)} = 0.325$, $M = 892.6$					

[a] The line intensities were too small to give reliable values for I_n or ΔH_n. Note: The units for K_s and M are erg/cm² and Oe, respectively.
[b] Assuming $\Delta H = 20$ Oe.

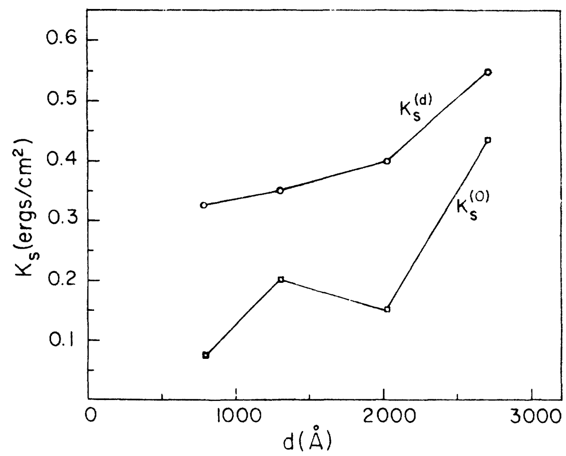

FIG. 2. Magnetic surface anisotropy as a function of film thickness.

thickness. It is seen that $K_s^{(0)}$ decreases much more rapidly with thickness than does $K_s^{(d)}$. Our results are in marked contrast to previous work,[4] which implied that K_s was thickness independent.

DISCUSSION

We now discuss some general features of the magnetic surface anisotropy of these films. There are at least five possible sources for the surface anisotropy, three of which may lead to a thickness dependence for K_s. (i) It has often been suggested that an antiferromagnetic layer on the film surface could effectively pin the spins at the surface. However, the exchange coupling at the surface between ferromagnetic and antiferromagnetic spins should not be thickness dependent (at least, in the range of thicknesses studied here). Furthermore, it is unlikely that an antiferromagnetic layer would give the correct spin-pinning dependence on angle as required by a uniaxial surface anisotropy (that is, an anisotropy energy E_s varying as $K_s \cos^2\theta$, where θ is the angle measured from film normal). (ii) Surface roughness may give rise to a surface field as well as a bulk field.[13] For a film with $d = 2700$ Å and a 200-Å crystallite size,[14] we estimate the effective surface-anisotropy constant from this source to be 0.05 erg/cm^2 but proportional to $1/d$, which is contrary to our observations. (iii) Nonuniformities of the demagnetizing field near the surface may give rise to a uniaxial-surface-anisotropy constant of the order $K_s \approx 4\pi M \Delta M l$, where ΔM is the change in the magnetization M over the distance l. It has been shown that as the film thickness is decreased below 20 Å, the magnetization changes from its bulk value.[1] If we assume that M decreases from its bulk value to 25% of its bulk value within 20 Å of the film surface, we obtain $K_s \approx 0.6$ erg/cm^2. This value is in very good agreement with the observed value for the thickest film. (iv) The qualitative difference in the thickness dependence of K_s on the two sides of the film may be a result of one side of the film being on a substrate and consequently strained differently than the other, free side. The uniaxial-surface-anisotropy constant arising from a stress acting over a distance Δd from the surface of the film is $K_s \sim \frac{3}{2}\sigma\lambda\Delta d$. If typical values of σ and λ (namely, $\sigma = 5\times10^9$ dyn/cm^2 and $\lambda = 5\times10^{-6}$) are assumed, K_s is approximately 0.1 erg/cm^2 if $\Delta d = 200$ Å.[15] This estimated value is of the same order as the observed values of $K_s^{(d)} - K_s^{(0)}$ between 0.1 and 0.3 erg/cm^2. Although both of the two previous sources [items (iii) and (iv)] give reasonable values for K_s, a necessary consequence of the observed thickness dependence of K_s would be a thickness dependence of l or Δd. Certainly, the variety of reported stress distribution across the thickness of a thin film are so numerous[16] that such a variation of Δd is imaginable. On the other hand, it is unlikely that l should change drastically with thickness. Furthermore, *both* Δd and l could remain constant and K_s could change with thickness as a result of a possible de-

TABLE IV. Parameters for perpendicular resonance.

H_n (Oe) Expt.	H_n (Oe) Theory	ΔH_n (Oe) Expt.	ΔH_n (Oe) Theory	I_n Expt.	I_n Theory
$d = 2023$ Å					
14331	14331	33	39	100	100
14224	14220	26	a	0.72	0.63[b]
14018	14018	21	20	7.1	8.0
13687	13695	a	17	≈ 0.08	0.50
13260	13255	22	15	1.4	1.75
a	12692	a	12	a	0.23
12004	12006	24	9	0.31	1.13
$K_s^{(0)} = 0.15$, $K_s^{(d)} = 0.4$, $M_s = 887.4$					
$d = 1299$ Å					
14450	14449	24	26	100	100
14216	14222	19	18	0.49	0.57
13761	13747	20	16	4.0	4.5
a	12993	a	12	a	0.11
11947	11947	24	8	0.46	2.22
$K_s^{(0)} = 0.2$, $K_s^{(d)} = 0.35$, $M_s = 899.1$					
$d = 790$ Å					
14346	14347	24	21	100	100
13804	13830	24	16	2.9	3.3
12596	12592	30	11	2.6	1.6
$K_s^{(0)} = 0.075$, $K_s^{(d)} = 0.325$, $M = 892.6$					

[a] The line intensities were too small to give reliable values for this parameter. Note: The units for K_s and M are erg/cm^2 and Oe, respectively.
[b] Assuming $\Delta H = 20$ Oe.

pendence of K_s on the scattering of the spin waves at the sample surface. For example, as the thickness decreases and the spin-wave wavelength decreases, the scattering from surface inhomogeneities may be quite different, and hence K_s, which reflects the scattering, may change. (v) An estimation of the surface-anisotropy constant from Néel's model gives

$$K_s = -(V/2Na^2)[c_{44}\lambda_{111} - (c_{11} - c_{12})\lambda_{100}],$$

where V is the molar volume, N is Avogadro's number, $a = 3.54$ Å, $c_{44} = 1.1 \times 10^{12}$ dyn/cm^2, $\lambda_{111} = 4 \times 10^{-6}$, $c_{11} - c_{12} = 0.92 \times 10^{12}$ dyn/cm^2, and $\lambda_{100} = 17 \times 10^{-6}$. Assuming a random distribution of (100) and (111) crystallite planes at the film surfaces, we obtain $K_s = +0.08$ erg/cm^2 for our polycrystalline films as compared to the value observed for $K_s^{(0)}$ of $+0.075$ erg/cm^2 for the thinnest films.[17] It is to be expected that the thinnest film may show the intrinsic surface anisotropy.[1] It is difficult to estimate from the other[3] theoretical calculation of surface anisotropy what K_s should be for our films, because the calculations were concerned with film thickness only up to 17 atomic layers.

In conclusion, one can reasonably ascribe the bulk of the magnetic surface anisotropy in the thicker films to a nonuniformity of the demagnetizing field near the surface. The difference in K_s at the two surfaces, for any thickness, may be due to an enhanced magnetic surface field induced on one side of the film magnetostrictively by evaporating the film on a substrate. Although the observed thickness dependence of K_s is not explained well at the present time, certainly with additional experiments, this point may be cleared up. For example, it would be quite useful to study spin-wave spectra on very thin films (100 or 200 Å) at higher frequencies in order to observe more lines. A study of single-crystal films with spin-wave resonance would be helpful for a direct comparison with Néel's model. Finally, studies of films with different nickel-iron composition and on different substrates in order to check the magnetostriction effect at the surfaces would be helpful.

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[12] In Ref. 8, although $K_s^{(d)}$ was positive, it was written as negative because of the way the computer program was written. The convention used here is that positive (or negative) values for $K_s^{(0)}$ or $K_s^{(d)}$ mean that the hard axis is perpendicular (or parallel) to the plane of the film. In addition, we have rewritten the boundary conditions in Eqs. (1)–(9) so that the previous values of K_s must be divided by two to be consistent with the usual definition of K_s.

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[15] The quantity Δd is not necessarily equal to l. We used a reasonable value of Δd obtained from E. Klokholm and B. S. Berry [J. Electrochem. Soc. **115**, 823 (1968)] for iron films.

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[17] The very close agreement may be partially fortuitous particularly in light of the value of the observed anisotropy (0.3 erg/cm^2) on the other side of this film.