TWO-DIMENSIONAL "FERROMAGNETISM" IN IRON*

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The magnetization of an iron film deposited on a noble metal in the presence of an external field is measured as the thickness is continually increased atomic layer by layer. The increment of magnetic moment per additional atom of deposited iron approaches the bulk value at three layers, but two of the layers remain nonmagnetic independent of film thickness. No explanation is offered for the existence of the two nonmagnetic layers, but the observed magnetization of the additional layers is explained quantitatively by a spin-wave theory which includes anisotropy energy.

A new technique was used to study magnetization in iron films as the thickness was continually increased, atomic layer by layer, effectively from zero. Figure 1(a) shows an actual recording of the relative magnetization M as iron deposition proceeds by electrolytic means. Figure 1(b) is a simultaneous recording of the increase in internal flux, or the product Ml, for l layers. The ambient field of the order of 1000 Oe in the plane of the film is sufficient to cause saturation. The film thickness is obtained from the electrolytic current (and the time). Magnetization is observed by the change in mutual inductance between two coils, one of which is powered by the line frequency (60 Hz), thus providing the ambient field.

Two observations are immediately apparent from Fig. 1. In Fig. 1(a) note the abrupt change in M beyond two atomic layers: M increases from zero to nearly the bulk value with addition of the third layer. Hence two layers of iron are not ferromagnetic, but the third layer is critical. A critical thickness has been suggested by earlier work[1] in which the individual films of uncertain thickness were studied. In Fig. 1(b) note that the extrapolated linear portion of Ml (i.e., the bulk region) does not intercept the origin (magnetization and thickness, zero). This implies that two layers of iron remain magnetically inactive in spite of the increase in film thickness. Stated in another manner, it is apparently true that all crystalline magnetic iron films have two nonmagnetic atomic layers.

It is tempting to assume that these two nonmagnetic layers are the two surface layers. Except for the fact that the film is deposited on a metallic substrate, this would be a valid conclusion. The nonmagnetic substrate was necessarily metallic to enable electrodeposition. It consisted of a single-crystal plane surface on a polished quartz support. Copper, silver, and gold were used. The fcc copper surface was ascertained to be the (111) plane which has a nearest-neighbor spacing of 2.55 Å. The adjacent iron layer is believed to be the (110) plane with spacing 2.48 Å. This reasonably good match can be expected to provide negligible lattice distortion by the substrate. The use of silver and gold substrates (2.88-Å spacing) yields a further check. Despite the deliberately poor spacing match, the observed magnetic perturbation was negligible, and two nonmagnetic layers were observed independent of film thickness as with copper.

Observations based on films as thin as a few atomic layers are subject to skepticism regarding film uniformity. Film uniformity was established by the following simple method: Iron and copper are electrochemically dissimilar and hence constitute a voltaic cell when immersed in electrolytic solution. Hence an immersed iron film will gradually be consumed whenever a portion of its copper substrate is exposed; the rate

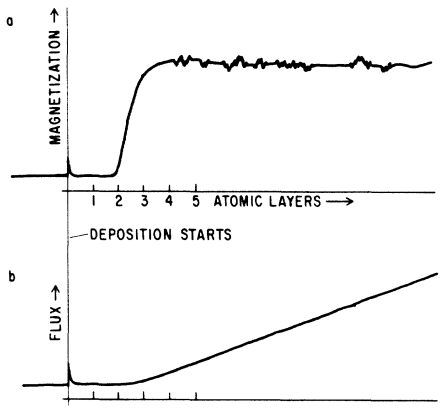

FIG. 1. (a) A recording of the increase in magnetization as the film thickness is increased by electrolytic deposition. The magnetization is zero until two atomic layers have been deposited. The magnetization approaches the bulk value at four layers. (b) A recording of the increase in magnetic flux with film thickness. Note that the flux increases linearly with thickness, but two atomic layers remain nonmagnetic.

of consumption is dependent upon the area of the exposed copper. By calibrating the consumption rate, the effective area of film imperfections or holes can be determined. Figure 2 gives sample data illustrating the method for two-layer films. The abscissa is the relative area of exposed copper; the ordinate is the reciprocal of the film lifetime. (Film lifetime is monitored by the voltaic emf generated between the iron film and an auxiliary copper test electrode.) Three data points in Fig. 2 are for known areas of exposed copper deliberately added to the film. The fourth datum, located on the ordinate axis marked "self," represents self-consumption of the film resulting from its unknown area of imperfections. The abscissa intercept at -17% indicates the imperfection area. Inasmuch as each datum point represents an independent two-layer sample, the value 17% is a determination of the mean imperfection area of these four samples. This imperfection area is considered remarkably small for a film of two atomic layers; although small, a similar correction was introduced in the determination of film thickness.

The influence of the two nonmagnetic layers must by necessity be neglected, as being beyond our present understanding, in order to consider the theoretical implications of these observations. Our speculations will therefore be confined to an attempt to understand the magnetization of the active layers, and in particular, the fact that one active layer ($l=3$) results in a magnetization per atom that is close to the bulk value.

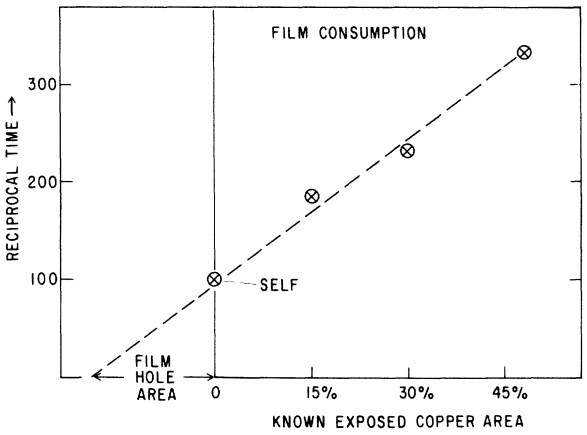

FIG. 2. An illustration of the method for measuring the area of nonuniformity or holes in an iron film 2 atomic layers thick. The reciprocal of the film consumption time is plotted versus area of deliberately exposed copper substrate. In this example the hole area is 17% of the total, indicating a remarkably uniform film.

The simplest theory for the magnetization of a small number of layers in an external field is a noninteracting-spin-wave theory, without anisotropy or dipole interactions. The standard treatment can be applied to our observed bcc iron layer, which appears to be a (110) plane. The result,[2] which is independent of any specific model for the spin waves, is

$$-\frac{\delta M}{M_0}=\frac{1}{4\pi\sqrt{2}(l-2)S}\,\frac{kT}{J}\ln\left(\frac{kT}{g\mu_B H}\right). \qquad (1)$$

Here $-\delta M/M_0$ is the relative demagnetization, J the effective exchange integral, l the number of layers (including the two nonmagnetic layers), H the external field, and T the temperature. Equation (1) is valid for $g\mu_B H \ll kT$ and $l-2 \ll 8\pi^2 J/kT$, the latter condition being the requirement that spins in different layers move together.

Figure 3 shows the predicted magnetization according to Eq. (1) as compared with observation for $H=1000$ Oe. For the purposes of the calculation it was assumed that J is the same as the corresponding quantity in bulk iron, namely 400°K.[3] Note that the predicted magnetism, while appreciable, is lower than is observed by an amount well-exceeding experimental error, estimated at ±5%. The only parameter subject to adjustment in the theory is the assumed bulk value for J. Adjustment appears unreasonable, for this value would need to be increased by a factor 3 in order to conform with observation.

Dipole interactions, thus far neglected, cannot

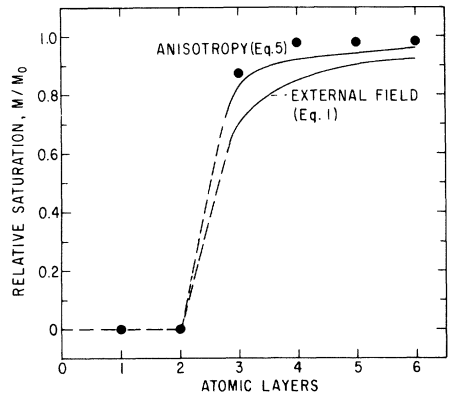

FIG. 3. Comparison of observed magnetization with calculated values. The calculations are made under the assumption that the number of magnetic layers is 2 less than the total number of deposited layers. The external field is taken to be 1000 Oe. The anisotropy energy is assumed to be $J/10$.

be expected to change these results significantly. In a two-dimensional layer the dipole interaction has a short range,[4] and the dipole interaction between neighboring spins is small compared with the exchange interaction.

The finiteness of the planar dimensions of the film can be taken into account[5]; the effect is to add to H in (1) an increment $\sim 10^{-7}$ Oe.

Agreement with observation would be improved if the quantity H in Eq. (3) were replaced by a much larger effective magnetic field. Anisotropy energy is a possible source of a large effective field. Our observations indicate that the magnetization is free to rotate in the plane of the film, thus ruling out an "easy" axis in the plane, or a surface anisotropy of the type considered by Davis and Keffer.[6] We consider instead the possibility that the film plane is an "easy" plane.

Designating the film plane as the yz plane with the z direction defined by H, a model (localized-spin) Hamiltonian is

$$\mathcal{H} = -g\mu_B H \sum_i S_{iz} - \tfrac{1}{2}J \sum_{\langle i,j\rangle} \vec{S}_i \cdot \vec{S}_j + A \sum_i (S_{ix})^2. \tag{2}$$

We assume $S=1$, and $A>0$ for an easy plane. The spin-wave dispersion relation is, using linear spin-wave theory,[7]

$$\hbar\omega_q = [\epsilon_q(\epsilon_q + A)]^{1/2}, \tag{3}$$

where $\epsilon_q = g\mu_B H + Ja^2q^2$. The departure from saturation is not determined by the number of thermal spin waves because the state with $\delta M = 0$ is not the true ground state of the Hamiltonian. We find in the standard way[8]

$$-\delta M/M_0 = (1/2N)\sum_q (\hbar\epsilon_q - \hbar\omega_q + \tfrac{1}{2}A)/\hbar\omega_q + (1/2N)\sum_q [(\hbar\epsilon_q + \tfrac{1}{2}A)/\hbar\omega_q][\exp(\hbar\omega_q/kT) - 1]^{-1}. \tag{4}$$

For $g\mu_B H \ll A$, $(g\mu_B HA)^{1/2} \ll kT$, and $l^2 \ll 8\pi^2 J/kT$, we have from Eq. (4)

$$-\frac{\delta M}{M_0} = \frac{A}{16\pi\sqrt{2}J(l-2)} + \frac{kT}{4\pi\sqrt{2}J(l-2)} \ln \frac{kT}{(g\mu_B HA)^{1/2}}. \tag{5}$$

Of the two terms in Eq. (5) the first is negligible. Figure 3 gives the result assuming an anisotropy energy $A = J/10$, a reasonable maximum value; the value of J is taken to be the bulk value ($J = 400$°K) as before. Note that for a magnetic monolayer ($l=3$) the calculated magnetization is 83% of saturation, which agrees with observation to within experimental error. However, the calculated magnetization approaches the bulk value less rapidly than is observed, possibly because of our assumption of constant J, independent of thickness.

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[4]$\sum' R^{-3}$ converges.

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[7]The anisotropy term leads to complications. If the Holstein-Primakoff transformation is applied to (4), and only the quadratic part of the transformed Hamiltonian is retained, an incorrect result with an unphysical gap in the spin-wave spectrum for $H \to 0$ is obtained. The difficulty can be traced to the fact that this procedure leads to incorrect matrix elements of $[S_{i\pm}, \mathcal{H}]$ between states with less than two spin reversals. We have derived (5) and (6) by the lowest order version of the method of W. Marshal and G. Murray, United Kingdom Atomic Energy Research Establishment Report No. AERE-R4879, and to be published.

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