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MAGNETIC PROPERTIES OF SPUTTERED MULTILAYERS OF Mo/Ni

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We have studied the d.c. magnetization of Mo/Ni artificial superlattices in the temperature range 5–300 K and in magnetic fields of up to $10^7/4\pi$ A m^{-1} (10 kG). The saturation magnetization and the Curie temperature behavior are consistent with expectations based on thin film effects. However, there are indications of ferromagnetic coupling across the normal metal and the coercive field shows an unexpected peak at a layer thickness of 25 Å.

1. INTRODUCTION

Magnetic thin film materials are of considerable importance in uses related to magnetic recording especially magnetic recording material for magnetic disks[1–3]. Applications include parallel and perpendicular magnetic recording as well as erasable magneto-optical disks. In particular, multilayered thin film materials have been used for a number of years in a variety of magnetic disk applications. There is currently a need for the development and characterization of new thin film materials for these applications. By preparing superlattices it is hoped that it will be possible to engineer new useful properties into materials or to fine tune the properties that are of importance.

We present in this paper the d.c. magnetization of artificially produced Mo/Ni multilayers. Earlier X-ray studies[4] have shown that the structure of these multilayers consists of crystalline layers that are coherent in the growth direction and have been commonly designated in the literature as superlattices. Many of the results obtained in this study are as expected because of simple thin film effects[5–7]. However, we also find indication for ferromagnetic coupling across the normal (*i.e.* non-magnetic) metal (molybdenum), a peak in the coercive field *versus* layer thickness and a marked increase in the anisotropy as the layer thickness is decreased.

2. PREPARATION AND STRUCTURAL CHARACTERIZATION

The films used in this study were prepared and characterized as described earlier[4]. Briefly, the samples were prepared on a variety of substrates that were

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attached to a temperature-controlled rotating platform. Two vertical shielded beams of molybdenum and nickel were prepared by sputtering and the substrates were alternately moved above each of the beams. Typical sputtering parameters were 1.2 kW and 1.0 kW into the nickel and molybdenum guns respectively and an argon atmosphere kept at a dynamic pressure of 10 mTorr. The sputtering rates were controlled with an accuracy to better than 1% by controlling the power into the sputtering guns and the rotation speed of the platform was controlled with a stepping motor with an accuracy to better than 1%. In this fashion, layer thicknesses from about 5.0 to 2500 Å could reliably and reproducibly be prepared on 90° sapphire substrates. The substrate was heated solely by the sputtering process and its temperature was typically 200 °C.

The samples (approximately 1 μm thick) were characterized using standard X-ray diffraction in the directions both perpendicular (z) and parallel (x–y) to the layers. In the perpendicular direction the existence of up to 11 superlattice reflections implies that the layers are crystalline and coherent in the z direction with a coherence length greater than 300 Å. The x–y plane structure was obtained (after removal from the substrate) by orienting the layers so that the scattering vector was in the plane of the film. Below a critical nickel or molybdenum thickness of about 8 Å the samples are found to be highly disordered, consisting of small intermixed nickel and molybdenum crystallites or an amorphous Ni–Mo mixture. Above a thickness of 8 Å the samples consist of Ni(111) layers stacked on Mo(110) layers. Except for one interfacial atomic plane, the layers are found to be well segregated into essentially 100% Mo and 100% Ni layers respectively. The crystallite size in the x–y plane of the film is found to be roughly 150–200 Å. The layer thicknesses obtained from X-ray measurements were found to be in good quantitative agreement with thicknesses obtained from the total thickness and the total number of revolutions as well as with calculations obtained from the sputtering rate and the time the substrate spends over the gun.

The crystalline-to-disordered transition has been shown earlier[4] to manifest itself also as a transition from positive to negative in the high temperature (above 20 K) coefficient of resistivity and as an anomalous softening of the shear elastic modulus.

3. MAGNETIZATION

The d.c. magnetization measurements were performed using an SHE VTS10 superconducting quantum interference device magnetometer in the temperature range 5–300 K and in magnetic fields of up to $10^7/4\pi$ A m^{-1}. The samples were removed from the mica substrate by floating them off in water. The samples were weighed with an accuracy to better than 1% and the nickel mass was obtained by scaling to the appropriate volume obtained from the X-ray measurements. The magnetization (per gram of nickel) of the films was high enough that the correction that had to be applied because of the finite magnetization of the sample holder was typically less than 5%. In all figures the accuracy is better than the size of the experimental points.

Figure 1 shows the saturation magnetization at 5 K for three types of sample:

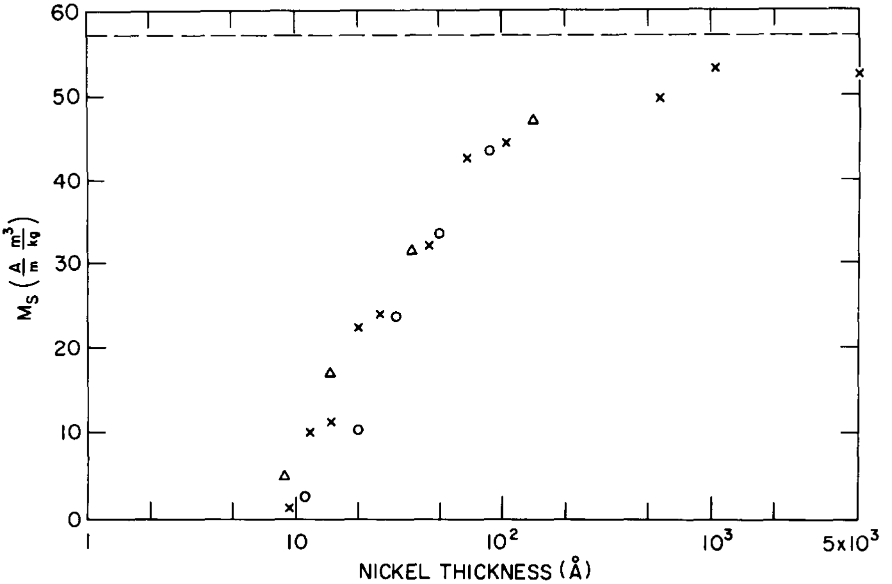
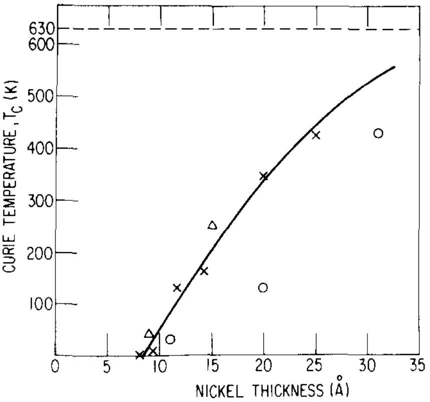


Fig. 1. Saturation magnetization as a function of the nickel thickness: △, $D_{Ni} = 3D_{Mo}$; ×, $D_{Ni} = D_{Mo}$; ○, $D_{Mo} = 3D_{Ni}$. The total thickness is 1 μm for each sample.

$D_{Ni} = 3D_{Mo}$ (triangles), $D_{Ni} = D_{Mo}$ (crosses) and $D_{Mo} = 3D_{Ni}$ (circles). Overall, the saturation magnetization drops to zero around $D_{Ni} \approx 9$ Å and approaches asymptotically the bulk nickel magnetization at large nickel thicknesses. However, it is manifest in this graph that below $D_{Ni} \approx 50$ Å the samples with thinner molybdenum layers consistently show a higher M_s value than those with thicker molybdenum layers. This is a strong indication for some magnetic coupling across the molybdenum layers. The same conclusion is also drawn from the changes in the Curie point to be discussed next.

The plots of Curie temperature T_c *versus* nickel thickness shown in Fig. 2 were obtained from the equation of state or so-called Arrot plots[8] (Fig. 3). Again, the transition temperature T_c is below 5 K at $D_{Ni} \approx 9$ Å and the samples with thinner molybdenum layers (triangles) consistently show the highest T_c for a fixed nickel thickness. There are three possibilities which could give rise to this effect: (a) ferromagnetic coupling across the normal metal, (b) Ruderman–Kittel–Kasuya–Yosida (RKKY) coupling across the normal metal or (c) percolation, due to shorts, across the molybdenum layers. Using a.c. susceptibility measurements in Cu/Ni superlattices Zhou *et al.*[5] have claimed that the increase in T_c for decreasing copper thickness is evidence for RKKY coupling across the normal metal. At this time we feel that from our data for Mo/Ni it is not possible to rule out a percolation ("shorts across the molybdenum") as opposed to ferromagnetic or RKKY coupling which could also give a similar effect.

A typical hysteresis curve at 5 K is shown in Fig. 4. Unexpectedly the coercive field H_c (Fig. 5) exhibits a rather sharp peak as a function of layer thickness around $D_{Ni} = 25$ Å for films of equal layer thickness. Since the T_c value of a sample with $D_{Ni} = 25$ Å is close to room temperature (about 425 K) and since the coercive field is



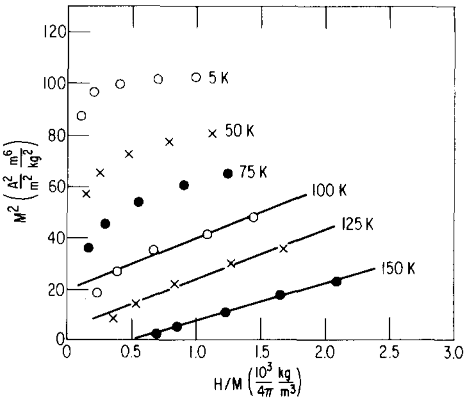


Fig. 2. Curie temperature *vs.* nickel thickness: △, $D_{Ni} = 3D_{Mo}$; ×, $D_{Ni} = D_{Mo}$; ○, $D_{Mo} = 3D_{Ni}$. The total thickness is 1 μm for each sample.

Fig. 3. Typical Arrot plots at various temperatures for an Mo/Ni sample with layer thickness $\Lambda/2$ of 11.7 Å ($T_c \approx 125$ K).

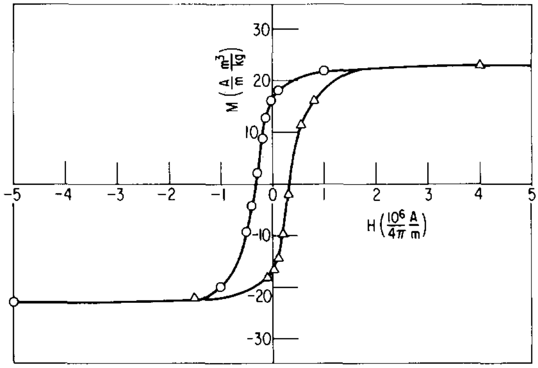


Fig. 4. Hysteresis curve for an Mo/Ni sample with a layer thickness $\Lambda/2$ of 20 Å at 5 K: △, increasing field in one direction; ○, field increasing in the opposite direction.

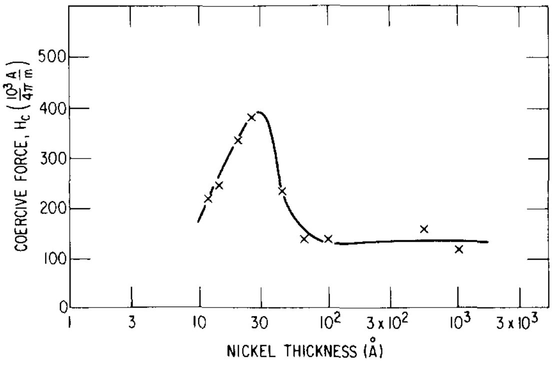


Fig. 5. In-plane coercive force H_c of Mo/Ni samples as a function of nickel thickness for samples of equal layer thickness.

quite high (approximately $10^5/\pi$ A m^{-1} or 400 G) this sample is uniquely suited for such applications such as Curie point writing. In addition, by reducing the film thickness to $D_{\mathrm{Ni}} \approx 22$ Å it is possible to lower the Curie point to slightly above room temperature (about 375 K) and still keep the high coercive force.

A typical plot of magnetization *versus* field in the parallel and perpendicular directions with respect to the film is shown in Fig. 6. In a magnetic field perpendicular to the film the anisotropy field H_a can be extracted from the intercept H_K between the perpendicular and parallel magnetization curves by using the relationship

$$H_K = 4\pi M_s + H_a \qquad (1)$$

As shown in Fig. 7, H_K increases with decreasing D_{Ni}. In addition, as shown earlier (Fig. 1) the M_s value decreases with decreasing D_{Ni}. Therefore, the anisotropy field H_a is greatly enhanced at low thickness, and this again is a desirable feature for a magnetic thin film recording material. The relationship (if any) between the

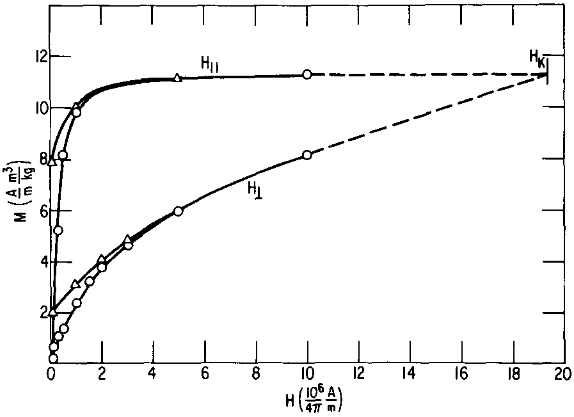


Fig. 6. The magnetization for H parallel and H perpendicular to the film plane for Mo/Ni sample with $D_{\mathrm{Ni}} = 14.3$ Å: ○, increasing field; △, decreasing field.

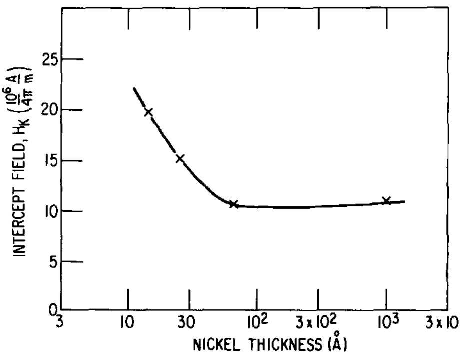


Fig. 7. Intercept field H_K as a function of nickel thickness.

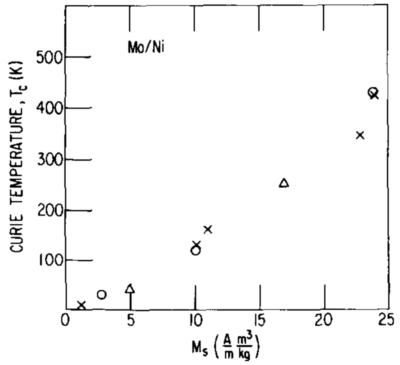


Fig. 8. Curie temperature T_c *vs.* saturation magnetization M_s: △, $D_{\mathrm{Ni}} = 3D_{\mathrm{Mo}}$; ×, $D_{\mathrm{Ni}} = D_{\mathrm{Mo}}$; ○, $D_{\mathrm{Mo}} = 3D_{\mathrm{Ni}}$.

increased anisotropy at low thicknesses and the peak in the coercivity is not understood at present and is the subject of current investigations.

We should like to mention at this point that, in contrast with what was claimed earlier[7] for Cu/Ni superlattices, in the Mo/Ni superlattice the low temperature saturation magnetization is *not* independent of the Curie temperature as shown in Fig. 8. However, the earlier T_c measurements for Cu/Ni were obtained from exponential extrapolation in an M_s *versus* T curve as opposed to Arrot plots used in this work. Because of this we cannot ascertain whether the behavior found earlier for Cu/Ni is fundamentally different or just a consequence of the different experimental procedure to obtain T_c.

4. CONCLUSIONS

We have measured the d.c. magnetization of Mo/Ni superlattices for layer thickness in the range from about 5 to 2500 Å, in the temperature range 5–300 K and in fields of up to $10^7/4\pi$ A m^{-1} (10 kG).

The experimental results are as follows:

(1) The low temperature saturation magnetization and the Curie temperature decrease monotonically to zero at a nickel thickness of about 9 Å. This is roughly the same thickness as that at which the material is found to undergo a transition to a disordered state in the X-ray scattering, resistivity and elastic constant measurements[4]. The exact nature of the disordered state (amorphous or randomly mixed small nickel and molybdenum particles) has not been clarified and is the subject of further studies.

(2) As expected, the Curie temperature and low temperature saturation magnetizations are dependent on each other contrary to what was claimed from earlier measurements for Cu/Ni superlattices.

(3) The saturation magnetization and Curie temperature show some evidence for coupling across the non-magnetic molybdenum layers, *i.e.* the thinner the molybdenum layer the higher are the saturation magnetization and the Curie temperature. The mechanism for the coupling (RKKY coupling, ferromagnetic coupling or percolation) is not uniquely determined by these measurements.

(4) Unexpectedly, the coercive force shows a peak around a layer thickness of 25 Å.

(5) The anisotropy field is also found to increase with decreasing layer thickness, contrary to expectations.

The facts that the coercive force is peaked around a nickel thickness of 25 Å and that the anisotropy field is also high at that point make this sample uniquely suited for such applications as permanent Curie point writing.

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