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THE EFFECT OF Pd-OVERLAYERS ON THE MAGNETIZATION OF CHROMIUM FILMS

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Abstract. — Measurements with a high vacuum torque magnetometer showed that the magnetic moment of thin Cr-films with Pd- or Ag-overlayers deposited on mica sheets was smaller than the magnetic moment of the mica sheet, similar to results found previously for pure Cr-films. This may mean that the average susceptibilities of the films are negative, and of the order of -10^{-4} , or that small permanent moments form pointing perpendicular to the surface.

Introduction

Room temperature measurements with a high vacuum torque magnetometer showed that polycrystalline chromium films deposited on mica or on NaCl covered mica sheets had a smaller magnetic moment than the mica sheet [1]. This result could mean that the thin chromium film is strongly diamagnetic, or that very minute permanent dipole moments form, lined up perpendicular to the surface.

In contrast, experimental and theoretical studies by Klebanoff *et al.* [2, 3] showed that (001) chromium single crystals have a large ferromagnetic surface moment (3 electrons per atom), with its spin polarization parallel to the (001) surface. Since our polycrystalline films should have some grains with this orientation, these films are observed to markedly differ from Cr(001) single crystals in their magnetic properties. In earlier work, we found [4] that a chromium film deposited on a polycrystalline iron film decreased the magnetic moment at the interface between chromium and iron, indicating that each moment at the Cr-surface is lined up antiparallel to the moment of the iron surface. Therefore, it would be surprising if extremely small ferro- or ferrimagnetic volume elements line up their moment perpendicular to the surface. Since the moment is very small, it seems possible that it is associated with surface or surface impurity states. The free surface of the chromium film was exposed for 15 to 30 min to a pressure of the order of 10^{-7} to 10^{-8} Torr, immediately after deposition. This exposure to possible contamination could lead to the formation of a surface layer responsible for the observed magnetic moment. It was felt that covering the chromium surface *in situ* with palladium or silver immediately after chromium evaporation, and measuring the moment of the mica sheet with all these overlayers, would help in the analysis of the observed effect.

Experimental approach

Moments were measured with a torque magnetometer (base pressure $\leq 2 \times 10^{-8}$ Torr after a bake-out

period of about 1 1/2 days) in which a mica sheet whose thickness is approximately 10 μm with length and width of 2 cm was supported by a 5 cm long W-wire of 0.01 mm diameter. The period of oscillation of the mica sheet $\tau = 2\pi/\omega$ can be measured reproducibly within 0.1 %. Uncertainties in the magnetic field H should lead to random errors of about 0.1 %. First, the frequency $\omega = \omega_0$ of the mica sheet (with or without a NaCl overlayer) was measured in the magnetometer at zero field or at applied fields of up to 2 500 Oe, then the Cr-film was deposited by evaporation "in situ", immediately followed by the deposition of the Pd- or Ag-overlayer, and the new frequency $\omega = \omega_1$ was measured. The latter measurement was performed 15 to 30 min later to avoid heating effects.

Auger electron spectroscopy studies showed that chromium films covered the mica or NaCl substrate completely. No contamination could be detected at the interface between the palladium and chromium films. Chromium films deposited on NaCl covered mica sheets and protected by a palladium overlayers were removed from the substrate by dissolving NaCl in glycerin. The electron diffraction pattern showed only rings associated with polycrystalline chromium and palladium. The films were continuous.

Results and discussion

The torque due to gradients in the magnetic fields was large, compared with the torque due to demagnetizing fields. The effect of demagnetizing fields was therefore neglected.

Single adjacent mica and Cr-volume elements sharing the same surface element are subjected to the same magnetic fields and field gradients, the ratio of the torques should be independent of both field and field gradients. This gives (as shown in Ref. [1]) the relation:

$$\frac{I_1 \omega_1^2(H) - I_0 \omega_0^2(H)}{I_0 \omega_0^2(H) - I_0 \omega_0^2(H=0)} = \frac{\chi_{\text{Cr}} \cdot V_{\text{Cr}} + \chi_{\text{Pd}} \cdot V_{\text{Pd}}}{\chi_{\text{mi}} \cdot V_{\text{mi}}} \quad (1)$$

I_i is the moment of inertia of the mica sheet and $\omega_i(H)$ is its angular frequency of oscillation: $i = 0$

Table I. - $d(\text{Me})$: thickness of Me-film, calculated with the assumption of isotropic evaporation, in nm (Me = Cr, Pd or Ag); T : test temperature. Errors given in table I are due to uncertainties in ω , T , χ_{mica} , and H . The Cr-volume is estimated to be about 30 % smaller than the isotropic evaporation value used. Therefore, the diamagnetism is about 40 % larger than given in this table.

Table Ia

Sample	$d(\text{Cr})$	$d(\text{Pd})$	$T(\text{K})$	$\chi(\text{Cr})$ [cgs units]
1	12.6	9.1	367	$-(0.4 \pm 0.07) \times 10^{-4}$
2	14.0	11.2	349	$-(1.5 \pm 0.15) \times 10^{-4}$
3	14.0	11.2	364	$-(1.4 \pm 0.2) \times 10^{-4}$
4	14.3	11.5	373	$-(0.5 \pm 0.06) \times 10^{-4}$

Table Ib

Sample	$d(\text{Cr})$	$d(\text{Ag})$	$T(\text{K})$	$\chi(\text{Cr})$ [cgs units]
1	2.8	10.5	298	$-(2.2 \pm 0.3) \times 10^{-4}$
2	5.5	10.5	298	$-(0.8 \pm 0.1) \times 10^{-4}$
3	10.2	10.3	298	$-(0.3 \pm 0.05) \times 10^{-4}$
4	21.6	10.4	298	$-(0.5 \pm 0.06) \times 10^{-4}$

and 1 corresponds to the situation with and without overlayer respectively. V_k is the volume of the Cr-film ("k" = Cr), the Pd-film ("k" = Pd), or the mica sheet ("k" = mi). The volume susceptibility χ_{mi} was determined experimentally as 4×10^{-6} in cgs units. The volume susceptibility of palladium was used for χ_{Pd} , and the volumes of Cr and Pd were calculated with the assumption of isotropic evaporation (Cr-films with Ag-overlayers were analysed similarly). Therefore χ_{Cr} can be calculated with equation (1). Values are given in table I.

V_{Cr} is estimated to be 30 % less than the true volume of the Cr-film. χ_{mica} is known within 10 %. Errors due to uncertainties in H and ω are less than 10 %. Therefore, our Cr-films are probably about 40 % more diamagnetic than data in table I show. However, even an error by a factor of 2 should not change the essential conclusions.

The Cr-films with a Pd- or Ag-overlayers showed the same behavior as pure Cr-films (Tab. Ia, Ib). Again, the magnetic moment of the system Cr-film + Pd- (or Ag-) overlayer + mica was smaller than the magnetic moment of the mica sheet alone, suggesting that

the reduced moment is associated with the Cr-film, or the interface between Cr-film and mica (or NaCl on mica) substrate. This model is supported by results from preliminary measurements, in which chromium was deposited on silver coated mica. In this case, no noticeable diamagnetism was found.

These results can be explained with the assumption that the Cr-film is strongly diamagnetic, with susceptibilities in the -10^{-4} range. This diamagnetism is stronger than found in any metal, except for superconductors. Resistivity measurements on our films do not indicate any superconductivity. If the Cr-film would contain only small disconnected volume elements which are superconducting, the film would not show macroscopic superconductivity. However, only zero conductivity would be proof of superconductivity. A second model which could explain the results, would require that a fraction of 10^{-3} to 10^{-4} (surface) atoms of the Cr-film form a magnetic phase with a dipole moment perpendicular to the surface. It is difficult to reconcile this with known properties of chromium [2, 3] or Cr-compounds. More work is required before we can understand the observed experimental results.

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