Magnetic moments in Cr thin films on Fe(100)

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The magnetism at the surface of a Cr film grown epitaxially on a Fe(100) whisker is observed as a function of Cr thickness by scanning electron microscopy with polarization analysis. Use of a wedge-shaped film of linearly increasing thickness allows the magnetism to be followed continuously for 75 Cr layers. Over the temperature range measured from just below the Néel temperature of bulk Cr, T_N, to 1.8 T_N, the surface magnetic moment is seen to persist and change direction with each additional Cr layer, but there are phase slips in this antiferromagnetic ordering. These are consistent with an incommensurate spin density wave (SDW) in the Cr film having a wavelength of 40 layers at T_N. An irregularity in the antiferromagnetic stacking order in the first four layers is discussed and the behavior of the moments at the subsequent phase slip is examined. The limitations which prevent the determination of the surface magnetic moment from the spin polarization of secondary electrons are discussed.

Chromium is a spin density wave (SDW) antiferromagnet with many interesting properties which have been intensively studied.[1] Because of the limited understanding of interlayer exchange coupling between Fe layers separated by a Cr space layer, we recently investigated[2] the properties of bare Cr thin films on Fe (100). We found a surface magnetic moment and SDW antiferromagnetic order within the film even well above the bulk Cr Néel temperature, $T_N=311$ K. This behavior is due to the presence of the Fe substrate which has a very different influence than a nonmagnetic substrate would have on the Cr and causes much different behavior than exists in bulk Cr where thermal fluctuations destroy the antiferromagnetic order above T_N. It was observed that while usually the direction of the surface moment changes with each additional Cr layer added to the film, there are exceptions to this regular stacking order at certain thicknesses. The first of these occurs within the first four layers. The others occur with every additional 20 layers of Cr where there is a phase slip in the spin density wave.

It is the purpose of this paper to elaborate on three aspects of our previous investigation. First, we discuss the stacking irregularity observed in the first four layers and the discrepancy between our results and the results of Walker *et al.*[3] Second, we analyze the behavior of the polarization of secondary electrons emitted from the surface of films with thicknesses at which the phase slip takes place. And third, we discuss the limitations that prevent the definitive measurement of the Cr moment from the secondary electron spin polarization data.

A continuous range of Cr film thickness was obtained by evaporating a film of linearly increasing thickness, that is a wedge of Cr, by moving a piezo-controlled shutter in front of the substrate. The substrate is a very high quality single crystal Fe whisker prepared as described previously.[4] Scanning electron microscopy with polarization analysis (SEMPA) was used to measure the spin polarization of secondary electrons generated in the SEM and thus obtain a direct measure of the net spin density near the specimen surface as a function of Cr film thickness. The Cr films were evaporated onto the Fe substrate at a temperature of 250 to 350 °C. Reflection high energy electron diffraction (RHEED) intensity oscillations recorded as a function of thickness along the wedge demonstrated layer by layer growth and provided a measure of the Cr film thickness on an atomic scale as described previously.[2,4] The relative film thickness is known to ±0.1 layers and the absolute film thickness to ±0.2 layers. Details of the SEMPA experimental technique are presented elsewhere.[5,6]

The polarization of the secondary electrons emitted from the Cr surface, P(Cr), measured[2] as a function of Cr thickness as the SEM beam was scanned along the Cr wedge, is shown in Fig. 1. When the Cr film is very thin the highly polarized secondary electrons from the Fe dominate as seen in Fig. 1(a). An average attenuation length l for electrons in Cr at the energies collected for the SEMPA measurement is obtained by fitting an exponential to the polarization decrease and found to be $l=0.55\pm0.04$ nm. After subtracting the observed exponential decrease in the Fe polarization one obtains the Cr polarization P(Cr), shown magnified by a factor of four in Fig. 1(b).

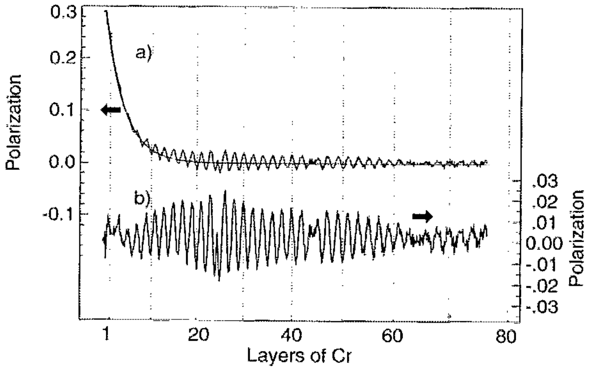

FIG. 1. (a) The polarization P(Cr) of the secondary electrons emitted from Cr as a function of the Cr wedge thickness. (b) Data of (a) on an expanded scale after subtracting the exponential shown. The SEMPA data in this and the following figures were measured at room temperature.

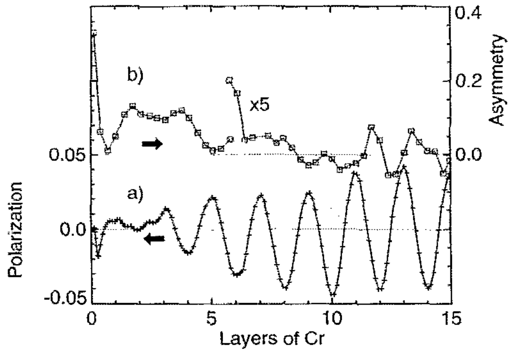

FIG. 2. An irregularity in the antiferromagnetic layer ordering already evident in P(Cr) from Fig. 1 is also apparent in a lower noise data run shown on an expanded scale in (a) and contrasted to the inelastic scattering asymmetry measured by Walker *et al.*[3] shown in (b).

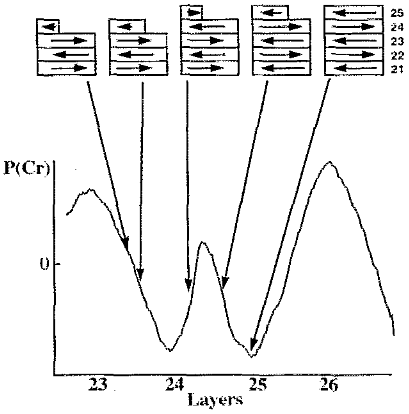

FIG. 3. The abrupt change in the phase of the oscillation of P(Cr) by π between 24 and 25 layers is caused by an additional node in the SDW and accompanying change in the layer stacking which is shown schematically.

The first question we address is the regularity of the stacking of the first few layers of Cr. Spin polarized photoemission measurements[7] of the $3p$ core levels of Fe and Cr showed that the first layer of Cr on Fe has magnetic moments opposite to those of Fe. Thus, since P(Cr) is dominated by the top layer of Cr, we would expect P(Cr) to be opposite in sign to Fe for the first layer and subsequent odd layers. P(Cr) for the first several layers of Cr from somewhat lower noise data than in Fig. 1(b) is shown on an expanded scale in Fig. 2(a). The oscillation of P(Cr) seen above four layers is less clear in the first few layers, but the negative excursion at four layers clearly indicates a stacking contrary to expectations. The downward excursion at two layers went negative in some data sets; this region is being studied further. Our thickness scale has been corrected for the nonlinearity below two layers caused by the penumbra arising from the finite source of the Cr evaporator and the shutter to sample spacing. We have independently verified on a very shallow wedge for which the penumbra effect was less than 1/10 layer that the result of Fig. 2(a) is reproduced.

There is a discrepancy between our results and the asymmetry in the inelastic scattering of a polarized electron beam at a loss energy of 1.9 eV measured by Walker *et al.*[3] in Fig. 2(b). There are similarities in the overall shape of the curve in the first few layers but the thickness scale appears expanded such that the measured asymmetry is opposite to that of Fe at five layers, an odd numbered layer as expected for regular antiferromagnetic stacking and antiparallel coupling at the interface. Both types of stacking order in thin Cr films on Fe, with and without a stacking irregularity, have been considered theoretically.[8,10] We speculate that the origin of the discrepancy between the experiments lies in the very different substrates. Walker *et al.*[3] used a polished Cr(100) crystal covered with 25 ML of Fe while we used an Fe single crystal whisker. If the initial coverage of Cr is tied up in defects, the onset of the asymmetry oscillations could be shifted to higher coverage as observed by Walker *et al.*

The second irregularity which we examine occurs between 24 and 25 layers and is repeated 20 and 40 layers after that. The origin of this behavior lies in the nature of the Cr SDW.[1] The Cr magnetic moment varies as $\cos(Qz)$ where the SDW wave vector is $Q=(1-\delta)\pi/d$, d is the layer spacing, and δ is a measure of the incommensurability of Q with the lattice wave vector π/d. The amplitude of the Cr moment varies with layer number n as

$$\cos(Qz)=(-1)^n\cos(n\delta\pi). \tag{1}$$

The moment is seen to change sign with each layer and to be modulated by the function $\cos(n\delta\pi)$ which has a period $n=2/\delta$ and a node each δ^{-1} layers. There is an accumulation of phase $\delta\pi$ with each additional Cr layer which gives a phase slip of one layer each 20 layers in the measurement of Fig. 1.

The manifestation of a phase slip in P(Cr) is illustrated schematically in Fig. 3. Between 23 and 24 layers where there is no phase slip, the polarization goes from positive, when the surface is predominantly the twenty third layer, to negative, when it is predominantly the twenty fourth layer, and is zero when equal amounts of layers 23 and 24 are measured. Between 24 and 25 layers there is a phase slip in the oscillations of P(Cr) where the phase abruptly changes by π when there is an additional node in the spin density wave. Just below the phase slip, the 24 layer thick islands give negative P and the 25 layer thick islands give positive P as shown by the arrows in the layers of Fig. 3. Just above the phase slip on the other hand, where there is an additional node in the SDW, 24 layers thick islands give positive P and 25 layer thick islands give negative P. As can be seen from our schematic picture, the moments in the Cr layers are aligned oppositely to each other at the phase slip. Such schematic pictures indicating layer stacking within the film should be viewed with some caution because, while the additional node in the SDW apparently does not occur in the surface layer, we cannot look into the film with the SEMPA measurement to determine just where it does occur. We note also that the thickness where the phase slip occurs is temperature dependent and varies from about 24.5 layers just above T_N to over 38 layers at 1.8 T_N.[2]

Finally, we address the limitations which prevent the extraction of a quantitative measure of the surface magnetic moment from measurements of secondary electron spin polarization. A question, which has been the subject of much debate, is whether or not the surface of an other wise antiferromagnetic Cr crystal is ferromagnetic beyond the trivial ferromagnetic alignment of spins in {100} planes, i.e., does the surface possess unusually large moments that are ferromagnetically coupled to one another? Calculations for the surface of a semi-infinite Cr crystal have predicted an enhanced surface magnetic moment[11–13] and stability of the ferromagnetic surface well above T_N.[14] Thus there is strong motivation to look for evidence of enhanced surface magnetic moments in Cr.

While it may seen appealing to extract a value for the surface magnetic moment from measurement of secondary electron spin polarization, such as our SEMPA measurements of a Cr on Fe(100), there are many difficulties. Ideally one could relate the measured polarization to the average band magnetization using

$$P=\mu/n_v, \tag{2}$$

where n_v is the number of valence electrons per atom and the spin only magnetic moment per atom μ is the net spin density per atom, $n_\uparrow - n_\downarrow$. Equation (2) assumes a uniform representation of the valence electrons in the measured secondary distribution. It is expected to hold best for measurements of the polarization of secondary electrons with kinetic energies in the range of 10–20 eV which is above the energy where spin dependent electron mean free paths cause an enhancement of the polarization.[15] Energies at which there are final state scattering effects[16] that add structure to the polarization measurement must be avoided. Experimental test of Eq. (2) for Fe,[17] Co,[17] Ni,[18,19] and amorphous Fe–Ni alloys[20] show expected trends but lack the precision desired for quantitative evaluation of magnetic moments. In general, P is proportional to μ, but since the constant of proportionality is not known, only relative moments for the same material can be determined. In comparing one material to another, other factors besides differences in μ can influence the spin polarization of the secondary electron cascade such as densities of states, matrix elements for electron excitation, and electron mean free paths.

Even if it is assumed that Eq. (2) can be applied quantitatively for each individual layer of moments in Cr, to obtain a surface magnetic moment one must know or make assumptions about the magnetic moments of the subsurface layers which contribute to the polarization. The secondary electron polarization does not represent either the bulk or the surface but rather some spatial average over the depth probed as determined by l. The total polarization is the sum of the contribution from each layer

$$P=\sum_n P_n I_n \Big/ \sum_n I_n, \tag{3}$$

where $n=0$ is the surface layer. The intensity I_n reflects the fact that the probability of the secondary electrons from layer n reaching the surface and contributing to the measured intensity is proportional to $\exp(-nd/l)$. To extract a value for the surface magnetic moment, one has to compare the measured polarization to a calculation of the polarization using Eqs. (2) and (3) in which the surface magnetic moment is a parameter. The problem is that one does not usually know the subsurface layer moments in order to calculate their contribution to the polarization. Furthermore, all subsurface moments may not have the same value. The SDW structure in the Cr film should be taken into account, but the positions of the nodes of the SDW in the film are not known. Enhanced moments for the first and second layers beneath the surface have been predicted.[12,13]

Clearly, the determination of the surface moment is fraught with difficulties. The constant of proportionality between polarization and magnetic moment is not reliably known. To extract an actual value for the surface magnetic moment requires information about the subsurface contributions to the polarization which also are not known. Although we have discussed the constraints on a quantitative determination of a surface magnetic moment for Cr from a secondary electron spin polarization measurement, many of the same limitations apply to attempts to determine magnetic moments with other spin polarized electron spectroscopies, such as Auger or photoelectron spectroscopy.

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