Magnetic structure of monolayer-range Cr films deposited on Fe(001)

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Spin polarized photoemission is used to examine the development of the magnetic structure in Cr films deposited on an Fe(001) substrate at 50 °C. The spin polarization of the Cr core levels provides a clear indication that within the initial monolayer the moments are antiferromagnetically aligned to those of the substrate. Through comparison with model tight-binding calculations it is shown that at the monolayer limit the Cr moments are of the order of $1.8\mu_B$ or larger. Subsequent Cr monolayers align antiferromagnetically with respect to the previous layer. The tight-binding calculations further indicate that as the Cr film thickness evolves the moment at the solid-vacuum interface is always enhanced.

INTRODUCTION

There have recently been a number of experimental studies aimed at elucidating the growth mode and magnetic structure of chromium overlayers on an Fe(001) substrate. These studies are partly motivated by the observation that the Fe/Cr(001) multilayer was one of the first systems to show the oscillatory exchange coupling between the adjacent ferromagnetic Fe layers as demonstrated in both light scattering[1] and in magnetoresistance measurements.[2] These observations have been extended with the demonstration that the exchange coupling in the multilayers displays both a long and a short period of oscillation.[3,4]

The Cr/Fe(001) overlayer system is also interesting because whilst Fe and Cr are both bcc structures with an almost identical lattice spacing, the magnetic ground state of Fe is ferromagnetic but that for Cr is antiferromagnetic. Theoretical studies of this system indicate that at the monolayer coverage the Cr aligns ferromagnetically within the layer but antiferromagnetically with respect to the substrate moments.[5,6] Further, these calculations indicate that the moment on the Cr monolayer is $3.6\mu_B$/atom (Ref. 5) and $3.1\mu_B$/atom,[6] i.e., considerably enhanced when compared with the 0.6 μ_B characteristic of bulk Cr. Spin polarized core level photoemission[7] and soft x-ray magnetic circular dichroism[8] have both been used to examine the properties of Cr films grown on Fe(001). These studies concluded that at the monolayer level the Cr was indeed ferromagnetically aligned within the layer but antiferromagnetically aligned to the substrate. The two studies, however, did not find evidence for the large enhancement of the Cr magnetic moment concluding in one case that the moment was $0.6\mu_B$ (Ref. 8) and in the other case $0.5-1.0\mu_B$.[7] However, a more recent *in situ* magnetometry measurement[9] concluded that the Cr monolayer exhibits a large enhancement of the moment to 4 μ_B, i.e., larger than the theoretical predictions. Spin polarized electron-energy-loss spectroscopy (SPEELS) (Ref. 10) and energy-resolved spin polarized secondary electron emission[11] experiments have both concluded that the magnetic moment on the Cr site is $1.8\mu_B$ and that the Cr overlayer is antiferromagnetically coupled to the bulk. Some explanation for this wide range of experimental values for the Cr moment has recently been offered by Vega *et al.*[12] who demonstrate in a theoretical study that the presence of steps in the Fe substrate may lead to strong modifications in the magnetic structure of the Cr overlayer. Unfortunately, complexity is also added to the analysis by recent structural studies which indicate that at room temperature the growth of the Cr on Fe is islandlike rather than layer by layer.[13] Moving to the growth of thicker Cr films both scanning electron microscopy[14] with polarization analysis and SPEELS (Ref. 10) have been used to show that the surface moments change their direction with each new layer.

In this paper we present the results of a spin polarized core level photoemission study of this system. With increased sensitivity we are able to measure the properties at lower coverages than in a previous core level study. Through comparison with model tight-binding calculations we examine the evolution of the magnetic structure in the chromium films as the thickness of the latter increases. Modeling the growth mode on the basis of the island formation found in studies elsewhere we find further evidence that at the monolayer limit the Cr magnetic moments are indeed considerably enhanced.

EXPERIMENT

The spin polarized photoemission studies reported in this paper were carried out with a commercial electron spectrometer backed by a low energy spin polarimeter[15] of the type described by Unguris *et al.*[16] The analyzer collects electrons over a solid angle of $\pm 3°$. The experiment was carried out on beamline X1B at the NSLS.[17] Based on an undulator source this beamline produces an intense flux of photons in the soft x-ray range. The overall energy resolution in the experiment was typically 0.5 eV.

The chromium films were prepared by evaporation onto an Fe(001) substrate that had previously been grown by deposition onto a Ag(001) surface. The initial Fe evaporation was carried out with the silver substrate cooled to 150 °C. The temperature of the Fe film was subsequently raised to

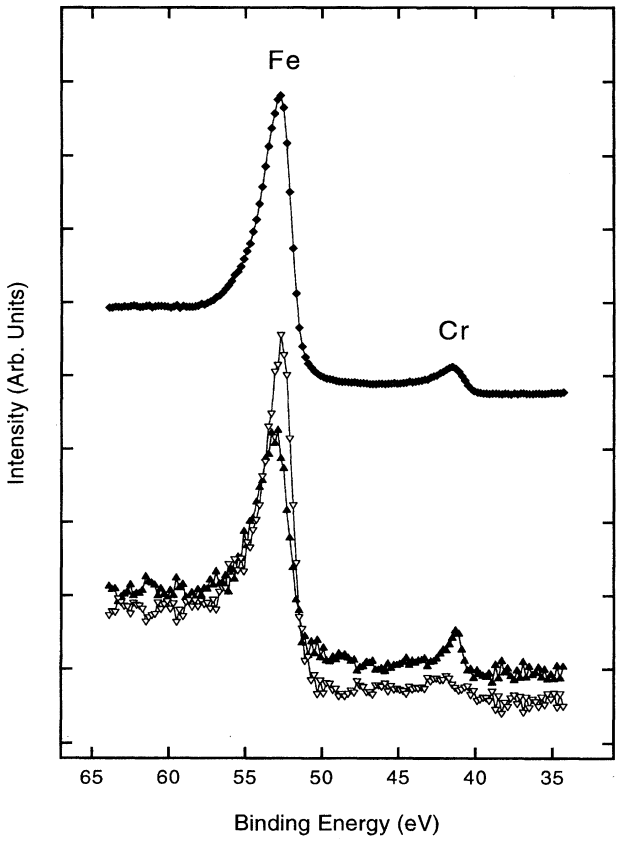

FIG. 1. Spin integrated (upper panel) and spin polarized photoemission spectra (lower panel) showing the Fe $3p$ and Cr $3p$ core levels recorded from 0.6 ML of Cr on Fe(001). In the lower panel the solid up triangles represent the majority spin and the open down triangles represent the minority spin. The incident photon energy is 250 eV.

50 °C for the Cr deposition and then allowed to recool for the photoemission measurement. The different coverages of both the Fe film and the Cr films were monitored through the use of Auger electron spectroscopy. Structural studies performed elsewhere indicate that growing the films at elevated temperatures increases the island or platelet size.[13] However, more recent studies indicate that growth at higher temperatures is more likely to be accompanied by interdiffusion or alloying at the interface.[18]

RESULTS

Figure 1 shows the spin resolved photoemission spectra in the vicinity of the Fe and Cr $3p$ core levels recorded from a Cr coverage corresponding to 0.6 monolayer equivalents (MLE). The incident photon energy is 250 eV. In agreement with previous studies[7] the dominant polarization of the Cr $3p$ core level at a binding energy of 41 eV is majority spin as opposed to the minority spin polarization of the Fe $3p$ core level at a binding energy of 53 eV. The latter spin polarization was found to be -30% in agreement with earlier studies.[19,20] The observation of a spin polarization on the Cr site is an indication that within the Cr layer the moments are

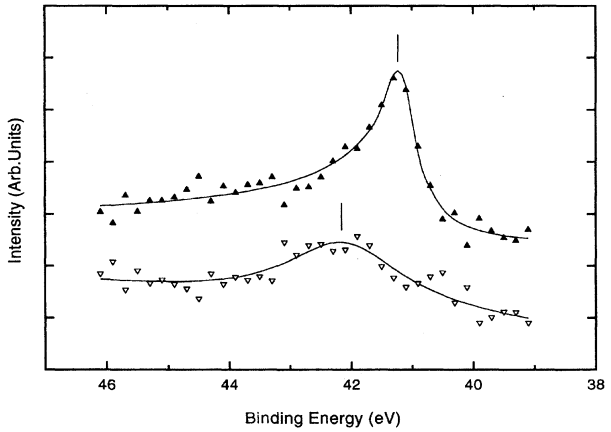

FIG. 2. Expanded photoemission spectra in the vicinity of the Cr $3p$ core levels recorded from 0.6 ML of Cr on Fe(001). The solid up triangles represent the majority spin and the open down triangles represent the minority spin. The incident photon energy is 250 eV.

aligned ferromagnetically. The observation of a dominant majority spin polarization is an indication that the moments on the Cr sites are antiparallel to those of the Fe substrate.

Figure 2 shows an expanded spectrum in the vicinity of the Cr $3p$ core level. The spin polarization and associated splitting between the two spin components is much larger than that observed in a previous study of this system.[7] In the latter study the spectra were recorded from a larger thickness as indicated by the relative intensities of the spin integrated Fe and Cr $3p$ core levels.

Figure 3 shows the spin polarized spectra recorded from a thickness equivalent to four atomic layers. The spectra now show essentially no splitting between the two spin components and an equalization of the intensity in the two components. Although not shown here the net spin polarization of the substrate Fe at this coverage is unchanged from that observed at lower coverages.

Figure 4 shows the spin integrated spectra recorded from the Fe and Cr $3s$ levels at Cr thicknesses corresponding to

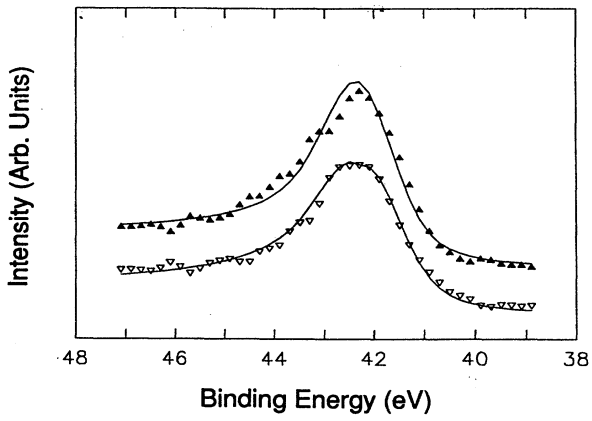

FIG. 3. Expanded photoemission spectra in the vicinity of the Cr $3p$ core levels recorded from 4.0 ML of Cr on Fe(001). The solid up triangles represent the majority spin and the open down triangles represent the minority spin. The incident photon energy is 250 eV.

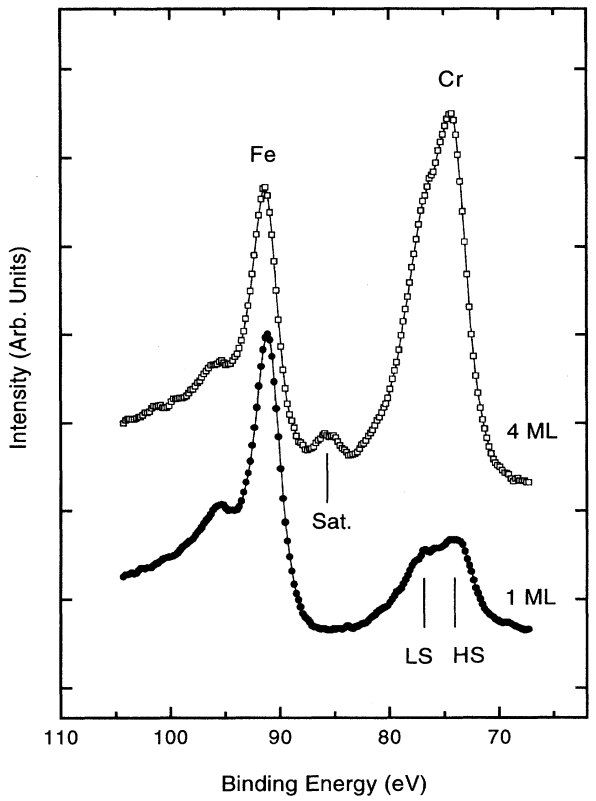

FIG. 4. Spin integrated photoemission spectra showing the Fe $3s$ and Cr $3s$ core levels recorded from 1.0 ML of Cr on Fe(001) (lower spectrum) and 4 ML of Cr on Fe(001) (upper spectrum). The incident photon energy is 250 eV.

one and four monolayers. The Fe $3s$ level is identical to that observed in earlier studies of clean Fe.[21] The Cr $3s$ level on the other hand shows considerably more weight in the satellite or low spin (LS) state, which appears almost equal in intensity to the main high spin (HS) component. The relative intensities of the low and high spin states in the spin integrated or isotropic spectrum will be $S:S+1$ where S is the net spin in the valence bands. Thus the observation of a strong relative intensity in the low spin state as compared to the high spin state is an indication of a large moment on the Cr sites.

In a simple model due to Van Vleck the splitting, ΔE, between the LS and HS states will be given by[22]

$$\Delta E = \frac{2S+1}{2l+1} G^l(3s,3d), \tag{1}$$

where $G^l(3s,3d)$ represents the exchange splitting between the $3s$ and $3d$ valence electrons and S is the net spin in the valence bands. In all studies of $3s$ level photoemission to date the observed splitting is smaller than that predicted by Eq. (1). The reduction has been attributed to interactions with other configurations of the final state ion.[23] In the spectrum corresponding to the thicker Cr film it is possible to observe a small satellite displaced in binding energy by 11.5 eV from the main Cr $3s$ peak. Discussed elsewhere,[24] this latter satellite corresponds to the $3s^2 3p^4 3d^6$ configuration. It is the interaction with this configuration that leads to the reduced splitting between the two components in the main $3s$ emission which represent different total spin states in the $3s3p^6 3d^5$ configuration.

CALCULATIONS AND DISCUSSION

In order to better understand the photoemission spectra presented in the previous section, we have used a spin dependent tight-binding scheme in a slab formulation to examine the evolution of the magnetic structure in the Cr films. We have previously used this approach to examine other thin film systems.[25] The tight-binding calculations were carried out in a slab formulation using an approach based on the use of a Hubbard Hamiltonian

$$H = \sum_k E(k) n_k + \frac{U}{N} \sum_{k,k'} n_{k\downarrow} n_{k'\uparrow}. \tag{2}$$

Here the first term reflects the paramagnetic band structure and the second term represents the modification due to an on-site spin dependent potential, U. This "effective exchange" potential leads to the spin dependent splitting of the electronic states with the resulting formation of local moments.

For the present calculation the parameters associated with a nonorthogonal tight-binding fit to the appropriate paramagnetic band structure were taken from the compilation of Papconstantopoulos.[26] The chromium-iron interaction parameters in the interface were taken as the arithmetic mean of the separate chromium and iron parameters. The two separate bulk lattices have an almost identical lattice parameter so no further scaling was required other than the alignment of the two Fermi levels. To introduce the spin polarization the on-site spin dependent energies for the d blocks were split by an amount Δ with appropriate adjustments to the associated off-diagonal elements. The spin-dependent densities of states obtained from such a method were integrated up to the Fermi level to obtain the resulting layer dependent magnetic moments. Maintaining charge neutrality a self-consistent solution was sought such that for each layer

$$\Delta_L = U_L m_L, \tag{3}$$

where Δ_L is the layer dependent splitting introduced into the d block, U_L the layer dependent Stoner parameter, and m_L the calculated moment for each layer. Values for U_L are taken from local spin density calculations of the susceptibility and related Stoner parameters.[27] The total density of states was calculated by summing over 28 k points evenly distributed throughout the irreducible surface Brillouin zone.

Reported elsewhere, a calculation for an 11 layer Fe(001) slab using this method yields an average internal magnetic moment of $2.2 \pm 0.05 \mu_B$.[28] The same calculation for a 13 layer Cr(001) slab results in an antiferromagnetic alignment of adjacent (001) planes with a bulk moment of $0.6 \mu_B$. As in previous calculations the Cr surface shows an enhanced moment of $2.5 \mu_B$. Interestingly the decay of the absolute value of the moments away from the surface is more gradual in Cr than in Fe where the surface enhancement is apparently screened within one layer.

In Table I we show the calculated magnetic moments for the Fe/Cr system. We start out with the clean iron (001) surface and introduce, layer by layer, five monolayers of Cr.

TABLE I. The calculated magnetic moments for the surface (S) and subsurface ($S1$) layers of Fe and the different layers of Cr as the thickness of the latter is varied from one to five layers.

	Layer dependent moments of Cr/Fe(001) Film Thickness					
Layer	Clean	1Cr	2Cr	3Cr	4Cr	5Cr
$S1$	2.20	2.33	2.31	2.27	2.27	2.26
S	2.96	1.97	1.96	1.92	1.92	1.96
Cr 1		−3.15	−1.74	−0.87	−0.78	−0.76
Cr 2			2.71	1.38	0.80	0.67
Cr 3				−2.56	−1.36	−0.73
Cr 4					2.63	1.35
Cr 5						−2.56

In agreement with earlier tight-binding calculations[5] the initial Cr layer reduces the surface iron moments by 40% from $2.9\mu_B$ to $1.9\mu_B$. The initial Cr layer is antiferromagnetically aligned to the substrate with a large moment of $3.1\ \mu_B$. Reported elsewhere, at this coverage, the same calculation predicts a strong minority spin interface state immediately at the Fermi level at the center of the Brillouin zone. This state has been observed in spin polarized valence band studies.[28]

As indicated in Table I, with the addition of each new Cr layer the internal Cr layers slowly reduce their moments towards the value associated with bulk chromium. However, the newly deposited surface layers always show an enhanced magnetic moment reflecting the presence of the solid-vacuum interface with its reduced number of nearest neighbors. The moment on the interfacial Fe site does not change following the deposition of the first Cr layer. The addition of only a single layer of Fe to these thin Cr layers immediately results in the magnetization profile for the Cr that has previously been found in several earlier multilayer calculations for this system,[29] i.e., the chromium layers adopt moments close to the bulk Cr value.

Both the exchange splitting and the spin polarization of the Cr $3p$ level will reflect in some way the local moment. However, aside from the appropriate exchange integrals the magnitude of the exchange splitting will reflect various configuration interactions[30] as discussed earlier for the $3s$ level. For this study we therefore chose to compare the results of the model calculations with the measured spin polarization which we will assume essentially reflects the polarization characterizing the high spin state of the ion. Unlike the "exchange" splitting between the low and high spin states, the spin polarization in the high spin state will be essentially independent on the configuration interactions and thus may provide a reasonable measure of the net spin in the valence bands. To convert from calculated moment to spin polarization we use a calculation of the spin polarization of the Cr $3p$ core level as a function of the Cr magnetic moment recently reported by Roth *et al.*[7]

In order to model the present experiment we assume that the Cr islands grow in a manner as determined in the scanning tunneling microscopy studies of Pierce *et al.*[13] Thus, at any thickness t as measured by Auger electron spectroscopy, we assume an interface profile of width σ such that the fraction of each layer of thickness nd in the interface is given by

$$P(t,n)=[(2\pi)^{1/2}\sigma(t)]^{-1}\ \exp\{-[(t-nd)/\sigma(t)]^2/2\}, \tag{4}$$

where n is the number of layers and d is the interlayer spacing.

In Fig. 5 we compare the measured spin polarization of the Cr $3p$ level with the modeling described above. The error bars on the x axis reflect the fact that the Auger measurements of the different coverages also have to allow for the island growth. The open squares represent two data points taken from an earlier study.[7] As the growth temperature in that study was at room temperature we have adjusted the thickness for those points to slightly higher values again reflecting the island growth. It is clear that the model appears to provide a reasonable description of the growth. The largest

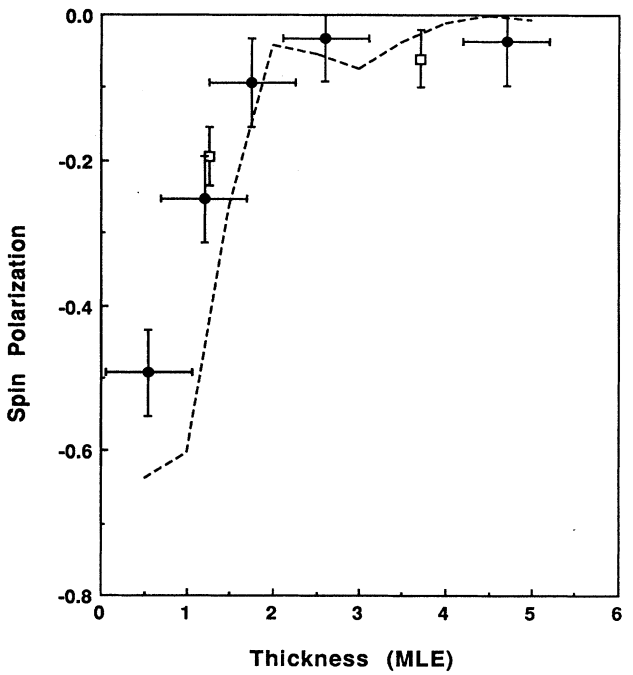

FIG. 5. Comparison of the experimentally observed spin polarization of the Cr $3p$ core level as a function of coverage with model calculations of the expected spin polarization based on the assumption of island growth as described in the text. The open squares represent data points taken from Ref. 7.

spin polarization observed in the present experiment is equivalent to $1.8\mu_B$. However, it must be remembered that the calculations represent the system at a temperature of 0 °C. At higher temperatures we may anticipate a lowering of the magnetization or long range order as T_c is approached. This will effectively lead to a reduction in the observed moment. Some uncertainty in the actual size of the moment also exists because of our method of conversion from magnetic moment to spin polarization.

The rapid drop in the spin polarization between one and two monolayers is an indication that the second Cr monolayer is antiferromagnetically aligned to the initial layer. It does not support the conclusion arrived at in earlier studies[9] that the first two layers are ferromagnetically aligned. The observation that the measured spin polarizations fall below the calculated polarization in the low coverage regime may also reflect two further possibilities: (1) there may be alloying to a limited extent in the interface and (2) the roughness of the Cr film may be less than that used in the model.

SUMMARY

The spin polarized photoemission studies of Cr film growth on the Fe(001) substrate reported here lend support to the prediction of first principle calculations that at the monolayer limit there is a considerable enhancement of the magnetic moment on the Cr site. Further our studies support earlier studies in showing that these Cr moments are ferromagnetically aligned within the layer but antiferromagnetically aligned with respect to the Fe substrate. With increasing coverage, the Cr films develop the antiferromagnetic structure characterizing bulk Cr within the film. However at the solid vacuum interface the films retain a similar magnetization profile to that calculated for a Cr(001) surface.

Clearly the comparison between our modeling and the experiment reflects the method we have used to convert from the calculated moment to a spin polarization. We have chosen to use the results of a calculation reported elsewhere.[7] However, we note that the observed characteristics of both the Cr $3p$ and $3s$ levels at the submonolayer limit qualitatively point to a moment larger than that observed for the substrate Fe.

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