Magnetic Ordering of Cr Layers on Fe(100)

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The magnetic ordering at the surface of epitaxial Cr overlayers on Fe(100) is studied by spin-polarized electron-energy-loss spectroscopy. The exchange asymmetry oscillates with the thickness of the Cr overlayer with a period of about two atomic layers, proving directly that the surface Cr layer has a net ferromagnetic moment and that successive layers order antiferromagnetically. The exchange asymmetry is predominantly due to spin-flip scattering. The spin-flip spectrum broadens toward lower energy with increasing thickness, suggesting that Cr may have a surface-enhanced magnetic moment.

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Magnetic superlattices have attracted much attention in recent years. In general, these systems consist of magnetic films interspersed with nonmagnetic films. The magnetic films are aligned ferromagnetically or antiferromagnetically with respect to each other, and oscillate between the two configurations when the spacer-layer thickness is varied. The antiferromagnetically aligned systems result in a giant magnetoresistance effect [1,2] which has important technological applications in magnetic recording. In the majority of these systems it is believed that an RKKY type interaction [3] is responsible for this behavior. Some examples are the Fe/Cu [4], Fe/Mo [5], Co/Ru [6], Co/Cr [6], and Co/Cu [7] systems. These systems generally exhibit a periodicity of about 10 to 15 Å in the spacer-layer thickness.

While the RKKY model predicts shorter periods as well, it is believed that surface roughness can damp out the shorter-period oscillations [8]. Most recently, on carefully prepared systems, short wavelength oscillations have been observed. The wedged Fe/Cr(d)/Fe sandwich system has been studied by Grünberg *et al.* with the surface magneto-optic Kerr effect (SMOKE) [9] and by Unguris, Celotta, and Pierce with scanning electron microscopy with polarization analysis (SEMPA) [10]. Both studies show approximately a 2-monolayer (ML) period, which is the period of antiferromagnetic Cr. A 3-ML period has recently been reported for Fe/Mo/Fe [11] sandwiches. The Fe/Cr/Fe sandwich and the Fe/Cr superlattices have been studied with Brillouin scattering [12], spin-polarized low-energy electron diffraction (SPLEED) [13], magnetoresistance studies [1,6], photoemission studies [14], SEMPA, and SMOKE [9,15].

While the Fe/Cr/Fe sandwiches and superlattices have been studied extensively, very few studies have been performed directly on the magnetic ordering of the Cr overlayer. Bulk Cr forms ferromagnetic (100) planes that couple antiferromagnetically. If this structure carries over to the Cr overlayers, very surface sensitive techniques would be necessary to avoid averaging out the contributions from different layers. Alvarado and Carbone [13] used SPLEED to measure the asymmetry of the Fe/Cr system as both Cr and Fe epitaxial layers were added. They noted the antiferromagnetic Fe-Fe coupling, as well as a monotonically decreasing magnetic signal as Cr was added. Jungblut *et al.* [14], used spin-resolved core-level photoemission spectroscopy to determine that the monolayer Cr couples antiferromagnetically to the Fe, while the spin-polarization signal vanishes for 2 monolayers.

In this paper we present measurements of the magnetic ordering of Cr *overlayers* on Fe(100), providing the first direct evidence of the antiferromagnetic structure of Cr in the Fe/Cr system. We report spin-polarized electron-energy-loss spectroscopy (SPEELS) studies of Cr overlayers on Fe(100). Our data show that low-energy electron techniques are surface sensitive enough to resolve layer-by-layer antiferromagnetic ordering.

The experiments were performed in a UHV system equipped for spin-polarized electron spectroscopies [16]. The Fe(100) substrate was obtained by evaporating about 50 ML of Fe on a Cr(100) crystal (Fig. 1). Clean (100) surfaces were obtained with 2-keV Ne ion sputter-

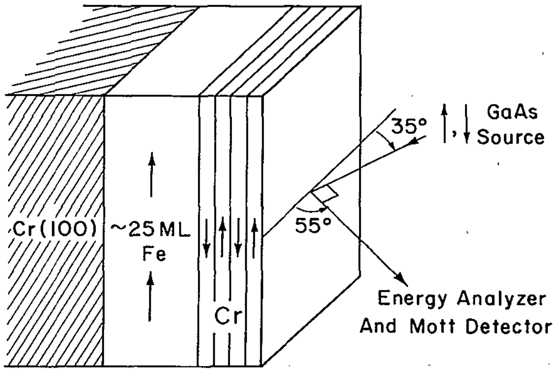

FIG. 1. Schematic of the SPEELS experiment.

ing followed by a 770 °C anneal. The Fe was evaporated at about 2.5 ML/min and a pressure below 1×10^{-9} Torr. The Cr was evaporated at about 0.17 ML/min with a pressure below 5×10^{-10} Torr. Sample quality was monitored with Auger electron spectroscopy and LEED studies. All stages of sample preparation showed a sharp (1×1) LEED pattern. Oxygen could not be monitored accurately with the Auger spectrometer since its peak overlaps a Cr peak; however, one would expect that most of the oxygen contamination would result from CO adsorption. The Auger analysis showed C to be less than 4% of a ML coverage at all stages of the process. Evaporation rates were monitored with a quartz microbalance, as well as the Auger spectrometer. Evaporations were performed at a sample temperature of 200 °C and all data presented were collected between −30 and −60 °C [17]. The Cr crystal served only as a template to grow bcc Fe and does not contribute to the SPEELS signal. The Fe film was pulse magnetized with a few hundred Oe in the plane of the film. All data were taken with remanent magnetizations. The incident polarized electron beam was provided by a GaAs photocathode. The primary electron energy was 31.5 eV. The scattered electrons were collected 20° off specular and passed through a hemispherical energy analyzer and then into a 100-keV Mott detector. The energy resolution was 300 meV.

In a SPEELS experiment one measures the intensity and polarization of the scattered electrons for incident beam polarizations both up and down, relative to the sample majority-spin orientation. The exchange asymmetry is defined as the normalized difference between scattered intensities for up and down incident beam polarizations. Previous SPEELS work on remanently magnetized ferromagnetic systems [16,18] shows that large (30% to 50%) negative asymmetries are obtained for small energy losses. The exchange asymmetries for Fe and several thicknesses of Cr on Fe are shown in Fig. 2 as a function of energy loss. The large asymmetries for the bare Fe film are consistent with previous work on bulk bcc Fe, indicating that the Fe films are remanently magnetized to saturation. Note that throughout this article, for consistency, we define the asymmetry with respect to the Fe majority-spin direction. The 1-ML Cr asymmetry spectrum has a sign opposite to the Fe spectrum, showing that 1-monolayer Cr couples antiferromagnetically to Fe, in agreement with the photoemission studies of Jungblut *et al.* [14]. This spectrum shows a maximum associated with the Cr at about 1.9 eV energy loss. We find that this Cr feature shows a sign opposite to (same as) the Fe asymmetry for an odd (even) number of monolayers of Cr (see Fig. 2). This clearly demonstrates that Cr overlayers on Fe do indeed have antiferromagnetic structure.

Figure 3 shows the asymmetry as a function of Cr thickness. In this study we held the energy loss constant at 1.9 eV and measured the exchange asymmetry. The Cr was deposited in units of 2-min evaporation at a constant rate of $\frac{1}{6}$ ML per minute and the asymmetry measured between each successive deposition, giving a total data cycle time of less than 10 min per data point. Successive thickness studies showed the evaporation rate to be constant to less than 3% variance. The asymmetry shows definite oscillations with Cr thickness. The magnitude of the oscillations shows a decreasing trend with increasing thickness. The measured asymmetry is an average over the area of our electron beam which is several millimeters in diameter, proving the existence of a net macroscopic in-plane oriented surface magnetic moment. For a rough surface, the local film thickness varies. One would expect different local thicknesses to cancel, giving a reduced average macroscopic surface magnetic moment. Thus the reduction in oscillation magnitude may be caused by surface roughness that is less important for

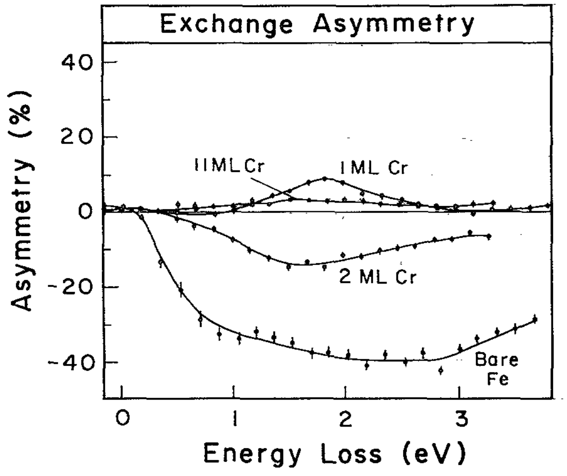

FIG. 2. Exchange asymmetry vs energy loss for various thicknesses of Cr on Fe.

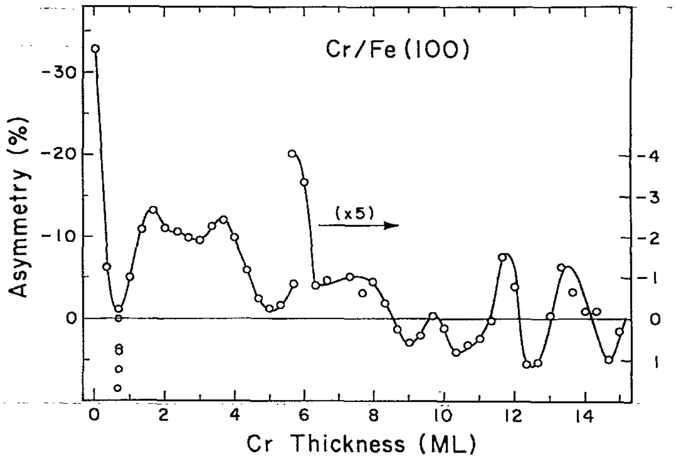

FIG. 3. Exchange asymmetry at 1.9 eV energy loss as a function of Cr thickness. (Note that the asymmetry axis has been inverted with respect to Fig. 2.)

the thinner films. The magnetic ordering of the Cr may be truly planar in the (100) direction, with the reduction in measured oscillation magnitude with Cr thickness being caused by surface roughness. The asymmetry of the submonolayer Cr on Fe is extremely sensitive to surface conditions (structure and cleanliness) and small variations in thickness. A collection of measured asymmetry values for the same nominal $\frac{2}{3}$-ML thickness are shown in Fig. 3. The 1-ML spectrum of Fig. 2 corresponds to the largest asymmetry value observed for this nominal thickness. In different runs we found the oscillation amplitudes to be reproducible within 20% (relative error) for Cr thicknesses greater than 1 ML. The main features of the curve are very reproducible. Asymmetries for films grown at room temperature showed the same period, but with consistently less than half the amplitude, demonstrating greater surface roughness effects.

It is interesting to compare our data on the Cr/Fe system to the Fe/Cr/Fe sandwich system data. In the Fe/Cr/Fe sandwiches, ferromagnetic coupling between the Fe layers has always been found for Cr thicknesses below 4 ML, whereas our Cr/Fe asymmetry data show oscillations as Cr is added, superimposed on a decreasing Fe background signal, even for Cr thicknesses less than 4 ML. The ferromagnetic alignment of the sandwiches for Cr thicknesses less than 4 ML is therefore attributed to a direct Fe-Fe ferromagnetic coupling possibly due to the presence of pin holes in the Cr. Interestingly, the first antiferromagnetic Fe-Fe coupling in the sandwiches is observed for Cr thicknesses between 4 and 5 ML, while our Cr on Fe asymmetry spectra show a sharp dropoff at this Cr thickness. Both results are consistent with the possible closure of Cr pin holes at this thickness. In the thickness range of 7- to 11-ML Cr on Fe we find only small oscillation amplitudes. If one assumes that the Fe-Fe sandwich interactions are mediated by the Cr interlayer, then the reduced Cr oscillations observed in our data could induce a reduced Fe-Fe interaction. We also note that this region corresponds to the first antiferromagnetic region of the long-wavelength period in the Fe/Cr/Fe systems.

Previous work has shown that inelastic scattering asymmetries in a ferromagnetic system are due mainly to an excess of flip-down over flip-up scattering [16,18]. The up and down again refer to incident beam polarization. Figure 4 shows the flip and nonflip scattering rates as a function of energy loss for incident polarization up and down, with respect to the Fe majority spin state. Note that in all cases the nonflip rates are nearly equal, while the flip rates are different and show the Cr feature at about 1.9-eV loss. The Cr feature shows a flip-up (flip-down) excess for an odd (even) number of Cr overlayers. This is again proof of the Cr antiferromagnetic ordering. The SPEELS experiment probes the joint density of states, and the location of the feature at 1.9-eV loss corresponds roughly to the exchange splitting of the system. Both the asymmetry and flip spectra show broadening mainly towards lower energy loss with increased Cr thickness. This broadening may be due to contributions from deeper Cr layers. Since the broadening is towards lower energy, we may infer that these deeper layers have a smaller exchange splitting than the surface layer. Thus this broadening towards lower energy may represent a surface-enhanced Cr moment with moments decreasing as one proceeds into deeper layers. However, we do not have any information on the possible existence of an enhanced Cr magnetic moment at the Fe interface, since our probing depth is only about 1 ML.

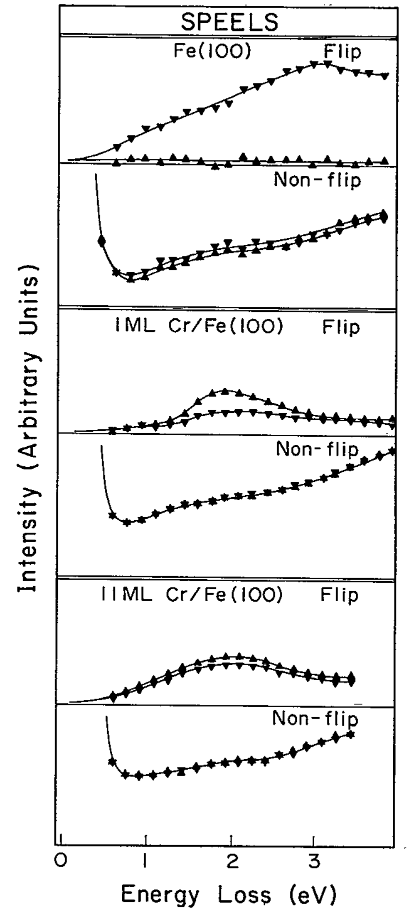

FIG. 4. Flip and nonflip partial scattering rates as a function of energy loss for several thicknesses of Cr on Fe. ▲ and ▼ indicate incident electron spins up and down, respectively, relative to the Fe majority spin direction.

Calculations show a "giant" magnetic moment for the monolayer Cr on Fe(100) as well as the Cr surface layer [19] [$(2.5\text{–}3.1)\mu_B$ vs $0.59\mu_B$ for bulk Cr]. Our results indicate a Cr surface exchange splitting of 1.9 eV, which indicates a strongly enhanced magnetic moment, contrary to the conclusions of Jungblut *et al.* [14].

In addition to the SPEELS study, we have also performed similar studies with spin-polarized secondary electron emission spectroscopy (SPSEES) and SPLEED. Both methods showed oscillations with Cr thickness, however, the SPSEES polarization oscillations were smaller than the SPEELS oscillations by a factor of 3. In electron scattering methods the electron both enters and exits the sample, while in electron emission methods the electron is excited inside the sample and only exits from the crystal. Thus one would expect emission methods to be roughly half as surface sensitive as scattering methods. The reduced surface sensitivity of emission methods means that the signal averages over more layers of the surface, resulting in reduced oscillation amplitudes for the Fe/Cr system. Thus the reduced SPSEES oscillation amplitude is expected. The SPLEED results were similar to the SPEELS results, but SPLEED is very sensitive to changes in angle, energy, and surface structure. The SPEELS experiment proved to be much easier to perform experimentally.

In summary, we have measured the exchange asymmetry as a function of Cr thickness on Fe(100), as well as performing the complete SPEELS experiment for several Cr thicknesses on Fe. We have found an oscillation in the asymmetry with Cr thickness, directly showing the antiferromagnetic order of Cr on an Fe(100) substrate. The magnitude of these oscillations reduces greatly with the first few monolayers, possibly due to surface roughness. Both the asymmetry and the partial flip rates show a feature at about 1.9 eV energy loss, indicating that the surface may have an enhanced magnetic moment. This Cr peak showed broadening with Cr thickness predominantly towards lower energy.

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Note added.—Subsequent to the submission of this manuscript, we learned of SEMPA studies by Unguris, Celotta, and Pierce [20] on a Cr/Fe(100) wedge system, showing similar oscillations with the Cr overlayer thickness.

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