RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 48, NUMBER 5 1 AUGUST 1993-I

Magnetism of Mn layers on Fe(100)

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(Received 24 March 1993)

The magnetic state of epitaxial overlayers of Mn grown on Fe(100) is studied using spin-polarized electron energy loss spectroscopy. Nonzero exchange asymmetries are found, demonstrating that the surface layer of the Mn overlayers has a net magnetic moment. The exchange asymmetry oscillates with a period of about two atomic layers as the Mn overlayer thickness is varied, proving that the Mn forms ferromagnetic (100) sheets and that the sheets align antiferromagnetically. The average Mn exchange splitting is found to be 2.9 eV, indicating a magnetic moment of the order $3\mu_B$.

The coupling between ferromagnetic layers separated by spacer layers has attracted much attention recently. Many systems have been found in which the coupling oscillates between ferromagnetic and antiferromagnetic alignment of the magnetic layers with increasing spacer layer thickness.[1] For paramagnetic spacers the coupling can be described by an RKKY-type interaction.[2] In carefully prepared samples more than one oscillation period is observed due to several critical points on the Fermi surface. A different situation arises when the spacer layer is itself a material with magnetic ordering. In the Fe/Cr/Fe(100) system it was found that the coupling oscillates with a two-monolayer period,[3] suggesting that the Cr spacer layer exhibits antiferromagnetic ordering that resembles the antiferromagnetic structure (or more precisely the spin density wave state) of bulk chromium. Very recently it was shown that the surface of ultrathin epitaxial layers of Cr on Fe(100) has magnetic order below[4] and above[5] the bulk Cr Néel temperature, directly showing the magnetic order in the Cr layers induced by the Fe substrate. So far the Cr/Fe(100) interface is the only $3d$-$3d$ transition-metal interface system that has been studied in detail. The Cr monolayer has been shown to couple antiferromagnetically to Fe(100) by spin-polarized core level spectroscopy.[6] Several rare-earth elements were also shown to couple antiferromagnetically to an Fe(100) substrate by spin-polarized photoemission[7] and spin-polarized Auger spectroscopy.[8]

In this paper we report on the magnetic order of epitaxial Mn overlayers on Fe(100). Bulk Mn is a very complicated system which can exist in various structures. Mn has a tendency towards antiferromagnetic order and we find that, indeed, magnetic order can be induced in ultrathin Mn layers grown on ferromagnetic Fe(100) substrates.

Spin-polarized electron energy loss spectroscopy (SPEELS) studies were used to probe the magnetic state and structure of the Mn overlayers. The experimental arrangement[9] is identical to that of the Cr/Fe(100) studies reported previously.[4] The Fe substrate was obtained by epitaxial growth of 60–70 Å of Fe on a clean Cr(100) crystal. The base pressure of the chamber was in the low 10^{-11} Torr range. During Fe evaporation, the chamber pressure was below 5×10^{-10} Torr, with an evaporation rate of 4–6 Å/min. Auger studies showed that Fe/Cr intermixing begins at temperatures of 320 °C to 350 °C, well above the Cr substrate temperature of 200 °C used during Fe growth. The subsequent Mn evaporations were performed from an Al_2O_3 crucible at sample temperatures between 90 °C and 145 °C, pressures of about 1×10^{-9} Torr, and rates of 0.2–2 Å/min. All thickness measurements were obtained by comparing Auger peak intensities, with calibration performed using a quartz microbalance thickness monitor. Both low-energy election diffraction (LEED) and Auger electron spectroscopy studies were used to monitor sample quality. At all stages of the sample preparation a good (1×1) LEED pattern was obtained, although the Mn overlayers gave rise to more diffuse LEED patterns. Auger studies showed carbon to be less than 4% of a monolayer at all times, while the oxygen peak overlapped with a Mn peak and thus was difficult to monitor quantitatively.

The SPEELS experiment performed consists of scattering incident spin-polarized electrons off the sample and collecting scattered electrons in the off-specular (20°) geometry. The scattered electrons are then energy and spin analyzed using a 180° hemispherical energy analyzer with an energy resolution of 300 meV and a 100 keV Mott detector, respectively. Primary electrons of 31.5 eV generated in a GaAs photocathode were used. Both the intensity and polarization were measured for scattered electrons from incident beam polarizations both up and down relative to the remanent magnetic state of the Fe substrate. One defines the exchange asymmetry as the normalized difference between scattering intensities for incident up and incident down electron spins.

The exchange asymmetry of the Mn overlayers was found to be critically dependent on the method of sample preparation. In order to find the optimal growth conditions, we performed anneal studies of the Mn overlayers. The top panel of Fig. 1 shows the exchange asymmetry of 3 ML Mn grown at −100 °C, as a function of the maximum anneal temperature. The data plotted are for asymmetries measured after the sample was cooled back down to a temperature of about −100 °C. The lower panel of Fig. 1 shows the ratio of the Fe (703 eV) Auger peak to the Mn (542 eV) Auger peak as a function of maximum anneal temperature. It is evident that at temperatures above 50 °C the film undergoes some structural changes that slightly lower the Fe/Mn Auger ratio and

dramatically increase the measured asymmetry. These observations are consistent with an atomic smoothing of the sample surface. At temperatures above 150 °C intermixing begins. All subsequent studies were performed on Mn overlayers grown at substrate temperatures of 90 °C to 145 °C. The asymmetries were found to be temperature dependent and larger at low temperatures. All SPEELS data were taken between −70 °C and −120 °C.

The exchange asymmetry spectra for the bare Fe film as well as 3 ML Mn/Fe are shown in Fig. 2(a). The large negative values of the Fe asymmetry are typical of a ferromagnet magnetized to saturation.[9,10] Since all spin polarizations are defined with respect to the Fe majority-spin direction, the positive values of the Mn overlayer asymmetry indicates that the Mn surface layer is aligned antiferromagnetically to the Fe substrate. Also shown are the spin-flip and nonflip (spin conserving) partial scattering rates for incident up and down electrons. Spin-flip scattering consists of an incident electron filling a vacant state and scattering out an electron from a filled state of opposite spin. Thus the flip scattering cross section probes a convolution of empty states of one spin with filled states of the opposite spin. Generally, the energy loss of the spin-flip scattering maximum corresponds to the average exchange splitting,[9,10] which in turn correlates with the magnetic moment with a relationship of roughly 1 eV/1μ_B. Thus the location of the spin-flip scattering maximum at 2.9 eV loss indicates that the surface of the Mn overlayer on Fe(100) has an average exchange splitting of 2.9 eV, and this corresponds to a magnetic moment of the order of 2.9μ_B. While Mn films of different thicknesses showed different values of the asymmetries, all Mn films measured showed spin-flip scattering maxima at 2.9 eV loss.

The magnetism of Mn is complicated. As the structure of Mn is varied, the magnetic structure predicted from calculations runs through paramagnetic, ferromagnetic, and antiferromagnetic states. Additionally, the crystal structure of Mn is by itself a very complicated system. Room-temperature bulk Mn forms a complex structure with a unit cell of 58 atoms.[11] Epitaxial Mn layers on Fe(100) (Ref. 12) as well as Ag(100),[13,14] Cu(100),[13] and Pd(100) (Ref. 15) grow in a distorted face-centered-tetragonal (fct) structure. These epitaxial Mn layers take on the in-plane spacing of the square surface net of the substrate and distort out of plane, yielding the fct structure. In the case of Mn on Fe(100) the in-plane spacing is a=2.87 Å and the out-of-plane spacing is about 3.3 Å, giving a layer spacing of 1.65 Å.[12] This structure can be regarded as either a body-centered tetragonal (bct) with c/a=1.15 or a fct with c/a=0.81.

While many calculations have been performed for various bulk structures of Mn, none have been performed for the particular structure of Mn overlayers on Fe(100). Calculations for the correct atomic volume but with a bcc structure predict a moment of 2.6μ_B,[16] while calculations for the fct structure but with slightly smaller atomic volume predict moments of 2.3μ_B–2.5μ_B for various c/a ratios, including 0.81.[17] Additionally, the spin-dependent

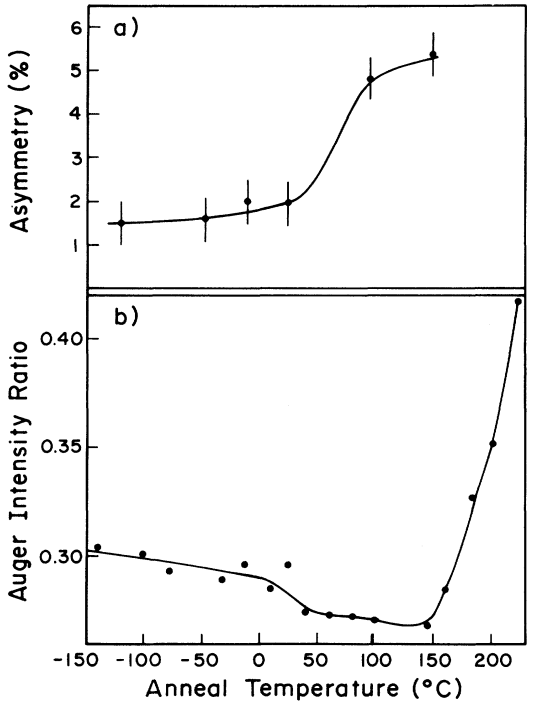

FIG. 1. (a) The exchange asymmetry of 2 ML Mn/Fe(100) vs maximum anneal temperature. The sample was grown with a substrate temperature of −100 °C and measured at −100 °C. (b) The ratio of the Fe (703 eV) Auger peak intensity to the Mn (542 eV) Auger peak intensity of 1.2 ML Mn/Fe(100) vs anneal temperature. A primary energy of 3 keV was used.

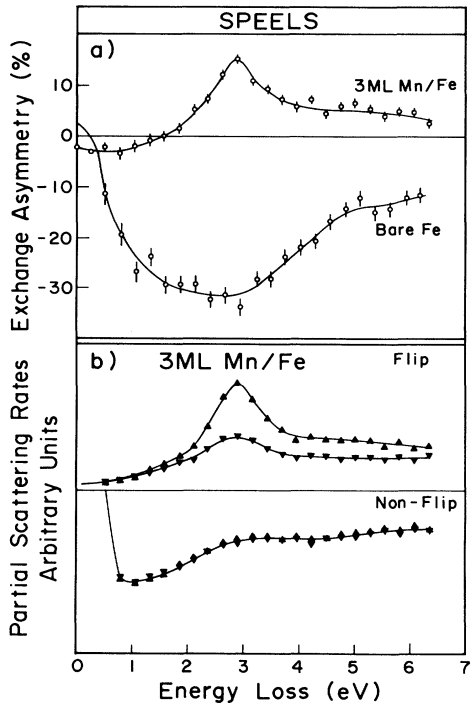

FIG. 2. (a) The exchange asymmetry vs electron energy loss for 3 ML Mn/Fe(100) and for Fe(100). (b) The flip and nonflip partial scattering rates for 3 ML Mn/Fe(100) for incident electrons of spin parallel (▲) and antiparallel (▼) to the Fe substrate majority spin direction.

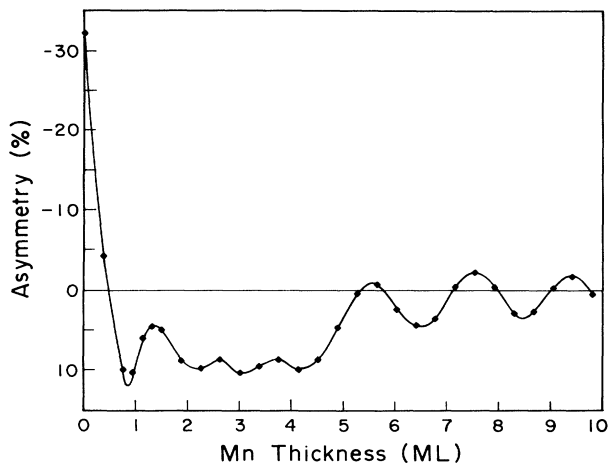

FIG. 3. The exchange asymmetry at 2.9 eV energy loss vs Mn overlayer thickness. Statistical errors are contained within the symbol size.

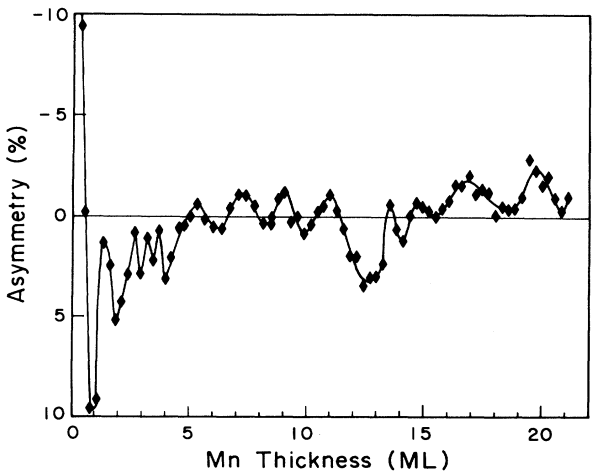

FIG. 4. The exchange asymmetry at 2.9 eV energy loss vs Mn overlayer thickness. Note the scale change from Fig. 3. Statistical errors are contained within the symbol size.

density-of-state curves shown with these calculations show apparent average exchange splittings of 2.9–3.3 eV.[16–18] These values are remarkably close to our measured value of 2.9 eV.

In order to probe the magnetic structure of these Mn overlayers, thickness-dependent data were taken with the energy loss held constant at the Mn asymmetry peak of 2.9 eV (Figs. 3 and 4). Since the probing depth of our experiment is only about a monolayer, the measured asymmetry represents the magnetic state of the surface layer for the given thickness.[4] The thickness sweeps are very similar to the Cr/Fe(100) SPEELS studies reported earlier, with two-monolayer oscillations providing strong evidence for layer-by-layer antiferromagnetic ordering of the Mn overlayers. Although substrate and sample quality did affect the measurements, all samples showed the same features. Fe substrates showing asymmetries between -25% and -35% were grown and used. Measurements were taken between each successive Mn evaporation, giving a cycle time of 10 to 13 min per data point. Figure 3 shows near-perfect 2 ML period oscillations as large as 10% in the last two periods, while the higher data point density of the data shown in Fig. 4 increased the sample exposure to residual gas per monolayer growth and thus Fig. 4 shows reduced oscillation amplitudes (2%). In addition to the two-monolayer period, Fig. 4 also shows longer period contributions as well. The oscillations were present even at our maximum measured thickness of 21 ML.

It is interesting to compare our measurements with previous results for the Cr/Fe,[4,5] Fe/Cr/Fe,[3] and Fe/Mn/Fe[12] systems. The primary difference between the Mn/Fe data reported here and the previously reported Cr/Fe (Ref. 4) studies takes place in the thickness range of 2–5 ML. In this thickness region, the Mn magnetic surfaces align *anti*ferromagnetically with the Fe, while the Cr surfaces aligned ferromagnetically with the Fe substrate. Although a proper theoretical treatment of these overlayer systems would require first-principle electronic structure calculations for films as thin as 5 ML or less, the Cr/Fe(100) RKKY calculations of Shi, Levy, and Fry[19] seem to be in qualitative agreement with the observed Cr overlayer data,[4,5] even for the thin films. For films thicker than 5 ML we find that the Mn overlayers form ferromagnetic (100) sheets aligned antiferromagnetically with respect to each other in the (100) direction, as the Cr overlayers do. The coupling between Fe layers in an Fe/Cr/Fe sandwich system was found to oscillate between ferromagnetic and antiferromagnetic with a 2 ML period for thicknesses greater than 5 ML.[3] In conjunction with the Cr overlayer studies, this result implies that the Fe coupling is mediated by the magnetic state of the Cr spacer layer. Since the Mn overlayers on Fe form a layer-by-layer antiferromagnet very similar to the Cr overlayers, one would expect that the Fe/Mn/Fe system would oscillate just as in the Fe/Cr/Fe system. However, this is in contradiction with the recent studies of Purcell *et al.*, who observed only antiferromagnetic couplings for Mn thicknesses greater than 4–7 ML, with oscillatory modulations in the coupling strength.[12] We do not understand this apparent discrepancy at present.

In conclusion, we have measured the exchange asymmetry on the surface of Mn overlayers on Fe(100) and found evidence for a layer-by-layer antiferromagnetic structure coupled to the Fe substrate, with films less than 5 ML thick showing a predominantly antiferromagnetic alignment. We have found the average exchange splitting of the surface Mn to be 2.9 eV. The measured value of the exchange splitting corresponds to a Mn overlayer surface magnetic moment of the order of $2.9\mu_B$, comparing well with calculated values.

This project was supported by the NSF through Grant No. DMR 9119815.

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