Enhanced Fe $3d$ spectral weight near the Fermi level in Fe overlayers on Cr

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Electronic structures of Fe on Cr (Fe/Cr) and Cr/Fe/Cr sandwich films, with an Fe coverage of 1–20 Å, have been investigated by using photoemission spectroscopy (PES). The Fe $3d$ partial spectral weight in Fe/Cr has been extracted as a function of the Fe overlayer thickness. Experimental results are compared with supercell band-structure calculations for a system with monolayer (ML) Fe on each side of five-layer Cr, Fe(1 ML)/Cr(5 ML)/Fe(1 ML), by using the linearized muffin-tin orbital band method. For a very thin Fe overlayer, very interesting features are observed: (i) a sharp emission just at E_F, and (ii) two other structures at higher binding energies which resemble the Cr $3d$ valence bands. The former feature is expected to originate primarily from hybridization between Fe and Cr $3d$ electrons at the Fe/Cr interface, and partially from the Fe $3d$ *surface* states in the Fe overlayer. The latter features also suggest large hybridization between Fe and Cr $3d$ states at the Fe/Cr interface, as confirmed by band-structure calculations. The trends observed in PES measurements are qualitatively consistent with those in band-structure calculations for the Fe(1 ML)/Cr(5 ML)/Fe(1 ML) system. This work suggests the importance of electronic structures, such as Fermi surface effects, in determining the character of the exchange coupling as well as the magnitude of magnetoresistance in the Fe/Cr superlattice.

I. INTRODUCTION

Recently, very large negative magnetoresistance (MR) has been observed in several metallic artificial superlattices, such as Fe/Cr, Co/Cu, and Co/Au,[1–5] which is believed to be associated with antiferromagnetic (AF) interlayer coupling between adjacent ferromagnetic layers across spacer layers.[1,3,6] The existence of the AF coupling was first found in Fe/Cr/Fe sandwiches[7,8] and later in an Fe/Cr superlattice,[9] when the intervening Cr layer thickness is less than about 20 Å. Oscillatory coupling between ferromagnetic layers has been evidenced as a function of the spacer thickness[10–14] and the ferromagnetic layer thickness.[15] The observed oscillation periods are quite large, about $\simeq 10$ Å. In order to account for the novel giant MR effect, some theoretical models have been proposed based on the mechanism of spin-dependent scattering of conduction electrons by magnetic impurities introduced by interface roughness.[16–19] However, the origin of the spin-dependent scattering itself has not been clarified yet. Other models propose the importance of the Fermi surface effect.[20–23] Since conductivity is determined by the Fermi surface geometry, significant differences in the electronic structures for different magnetic configurations will lead to different magnitudes of MR's. Another unresolved problem is the origin of the AF exchange coupling between adjacent magnetic layers or of an oscillatory behavior of the coupling depending on the sublayer thicknesses. A simple Ruderman-Kittel-

Kasuya-Yosida (RKKY) model[24] yields a period of $\lambda_F/2$ (λ_F is the Fermi wavelength of the spacer layer), which is too short to explain experimental results of $\simeq 10$ Å. It is claimed recently that the measured long oscillation periods are obtained by including a realistic description of superlattice one-electron states.[25]

To get further insight into the interlayer coupling of the Fe/Cr superlattice, it is important to investigate its interface electronic structure. There have been several works on oscillatory magnetic coupling using electron spectroscopies, such as photoemission spectroscopy (PES), inverse photoemission spectroscopy (IPES), and spin-polarized photoemission spectroscopy.[26–28] In these works, quantum well states (QWS's) at the Fermi level E_F are involved to correlate with the oscillations of the long-range magnetic coupling in superlattices of Cu/Co(100) and Ag/Fe(100). These QWS's are observed to carry predominantly minority-spin polarization, reflecting a preferential hybridization in the interface. For Fe/Cr(001), Beaurepaire *et al.*[29] performed PES measurements and concluded that an epitaxial thin film of Fe on Cr(001) is ferromagnetic.

In this paper we report synchrotron radiation PES studies of the *in situ*–prepared Fe/Cr bilayer and Cr/Fe/Cr sandwich films in the range of 1–20 Å of Fe coverage. We have extracted Fe $3d$ partial spectral weight (PSW) distributions as a function of the Fe overlayer thickness t_{Fe}. Our results are quite interesting and different from those by Beaurepaire *et al.*[29] in that Fe $3d$

PSW's exhibit new structures for thin Fe coverages, as compared to Fe metal. Experimental results are compared with theoretical density of states (DOS) obtained for a system with monolayer (ML) Fe on each side of five-layer Cr, Fe(1 ML)/Cr(5 ML)/Fe(1 ML), by using the supercell linearized muffin-tin orbital (LMTO) band method.

II. EXPERIMENTAL AND COMPUTATIONAL DETAILS

Photoemission measurements were performed at the Synchrotron Radiation Center (SRC) of the University of Wisconsin-Madison. The base pressure of the chamber was better than 4×10^{-11} Torr. Alternative depositions of Fe ($>$ 99.999%) and Cr ($>$ 99.95%) were done by an evaporation from resistively heated tungsten filaments. The substrate was a Si(001) single crystal with a 400-Å-thick Au film on it. For [001]-oriented growth, there is a good match of the Fe, Cr, and Au lattices.[7,8] Evaporations were done at a low deposition rate and a good vacuum was maintained during deposition: about 0.5 Å/min at the chamber pressure of 2×10^{-10} Torr for Cr and 0.2 Å/min at 7×10^{-11} Torr for Fe, respectively. During vapor depositions, the substrate was kept at room temperature. Thus it is likely that an epitaxial growth was obtained in our experiments, although it was not checked *in situ*. Thicknesses of the deposited films were monitored by quartz crystal sensors, and then corrected by using the measured spectra of Cr 3p and Fe 3p core levels for each evaporation. The cleanliness of the sample surfaces was checked with the valence band spectra obtained at a low photon energy $h\nu$ and also with carbon (C) and oxygen (O) 1s core level spectra. Fe evaporations produced no detectable surface contamination, whereas Cr evaporations caused a little oxygen contamination in our experiments.

A commercial double-pass cylindrical mirror analyzer (CMA) was used to analyze photoelectrons. The overall instrumental resolution was $\sim$ 0.2 eV at $h\nu$ = 70 eV and $\sim$ 0.3 eV at $h\nu$ = 130 eV. The Fermi level of the system was determined from the valence band spectrum of a sputtered Au sample. The spectra were normalized to the incident photon flux.

A supercell of the Fe(1 ML)/Cr(5 ML)/Fe(1 ML) sandwich with three vacuum layers is considered to simulate the experimental system. Electronic band structures and DOS are obtained by employing the self-consistent LMTO band method and the Gaussian broadening method, respectively. For the electron-electron exchange correlation, the von Barth–Hedin form has been utilized in the local spin density functional approximation. Vacuum layers are treated as empty spheres in the LMTO band calculation.

III. RESULTS AND DISCUSSION

We begin with a discussion of the 3p spectra of Cr and Fe core levels.[30] The purpose of presenting the Fe 3p and Cr 3p PES spectra of Fe/Cr is to provide clear evidence of a correlation between the 3p intensities and film thicknesses during the growth of the Fe film.

Figure 1 shows the Cr 3p and Fe 3p core level spectra of a thick Cr film (bottom), Fe/Cr films during Fe growth on Cr (middle five curves), and the Cr/Fe/Cr sandwich (top), all of which were taken at $h\nu$ = 130 eV. The binding energies (BE's) of Cr 3p and Fe 3p core levels, measured at the maxima of the peak intensities, are 42.1 eV and 52.5 eV, respectively. These BE values are similar to those reported for pure Fe metal and Fe/Cr(111).[31] The values of t_{Fe}, labeled in Fig. 1, correspond to the thicknesses that are corrected from those measured by the quartz thickness monitor. In making corrections, relative intensities of Cr 3p and Fe 3p core level data were estimated. Then the values of t_{Fe} were obtained by assuming that the electron emission decays exponentially with the sampling depth,[32] and by using the empirical inelastic mean free paths (IMFP's) for Fe 3p and Cr 3p electrons (5 Å).[33] The Fe thicknesses estimated from our data are in good agreement with those for Fe/Cr(111).[31]

Figure 2 compares valence band energy distribution curves (EDC's) of the Cr/Fe/Cr system, taken at $h\nu$ = 70 eV, as a function of t_{Fe}. All the spectra were measured under the same conditions. At this $h\nu$, Cr and
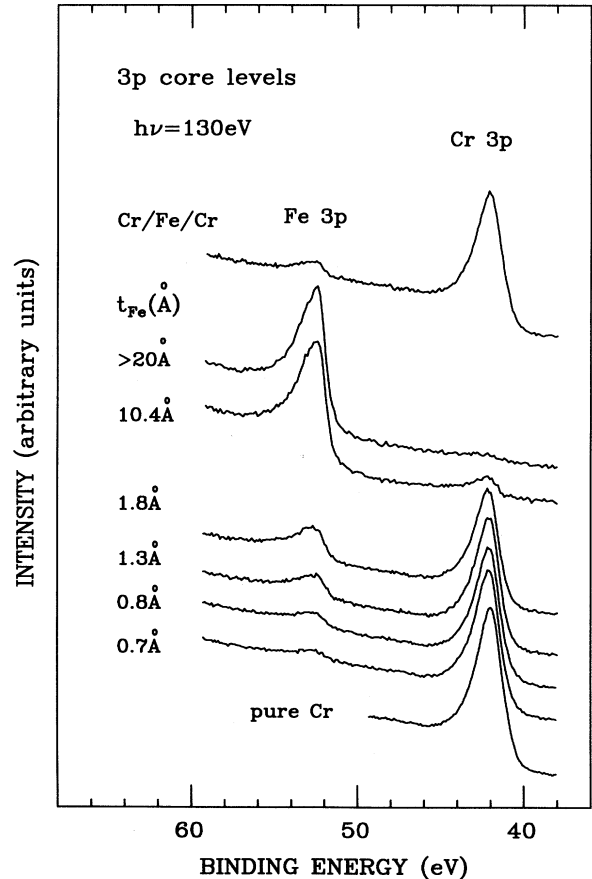

FIG. 1. Cr 3p and Fe 3p core level spectra for Fe/Cr and Cr/Fe/Cr, as a function of the Fe layer thickness t_{Fe}.

Fe $3d$ emissions are dominant over Cr and Fe $4s$ and $4p$ emissions,[34] and so these valence band spectra represent Cr and Fe $3d$ emissions. Except for the weak features at ~ -6 eV, which are due to slight oxygen contamination, the EDC's of thick Cr (bottom) and thick Fe (second from the top) are very similar to those of clean Cr metal and clean Fe metal, respectively.[35,36] Thus the region between E_F and ~ -5 eV in each valence band spectrum in Fig. 2 can be considered to represent intrinsic features of the valence electronic structures of Fe/Cr and Cr/Fe/Cr for different values of t_{Fe}.

As t_{Fe} increases, the spectral intensity near E_F grows progressively, implying an enhancement of the DOS at E_F. This trend arises from the increasing contribution of the Fe $3d$ emission with respect to that of the Cr $3d$ emission with increasing t_{Fe}. When t_{Co} is small, the intensity ratio of the Fe $3d$ emission near E_F to that between -1.5 and -1 eV is larger than the corresponding Cr $3d$ emission, as will be shown in Fig. 3. On the other hand, the position of the main peak (-1.5 eV) remains unchanged up to $t_{Fe} \leq 1.8$ Å, and a shift occurs toward E_F (from ~ -1.5 eV to ~ -0.7 eV) for thicker t_{Fe}. For $t_{Fe} > 10$ Å, the valence band spectrum becomes essentially identical to that of Fe metal.[35] This finding reflects (i) negligible Cr $3d$ emission with respect to Fe $3d$ emis-
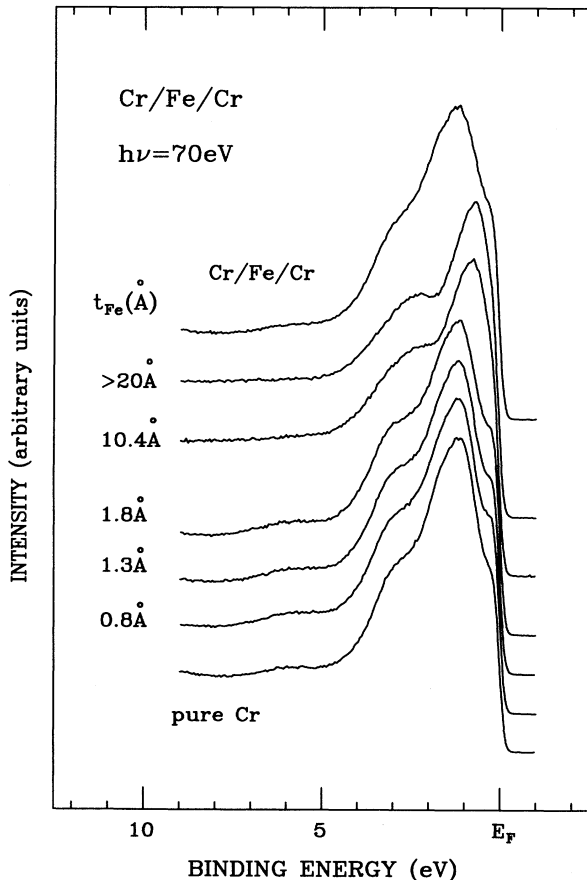
sion due to short IMFP's (~ 5 Å) of Fe and Cr $3d$ electrons at $h\nu = 70$ eV, and (ii) highly effective screening in Fe metal (< 3-4 ML's). According to spin-resolved PES studies of Fe metal[37] as well as band-structure calculations in this work, a main peak at ~ -0.7 eV and a shoulder at ~ -2.5 eV are mainly due to the minority-spin states and majority-spin states, respectively. Thus the separation between the main peak and the shoulder at a higher BE can be a rough measure of the exchange splitting of the Fe $3d$ bands. For the Cr/Fe/Cr sandwich, the line shape of the valence band PES spectrum is similar to that for Fe/Cr with $t_{Fe} \sim 2$ Å.

In order to find changes in the Fe $3d$ valence electronic structures in the Fe/Cr and Cr/Fe/Cr systems, we have attempted to extract the Fe $3d$ partial spectral weight (PSW) as a function of t_{Fe}. We first estimate the fraction of the Fe $3d$ to Cr $3d$ emissions in each valence band spectrum for a given value of t_{Fe}. Then the pure Cr $3d$ spectrum is multiplied by a factor which is equal to the fraction of the Cr $3d$ emission relative to the total $3d$ emission, and subtracted from the measured valence band spectrum. The result is regarded to represent the Fe $3d$ PSW for a given t_{Fe}. In this extraction, it is implicitly assumed that the Cr $3d$ PSW does not change with a thin Fe overlayer on it (for $t_{Fe} < 1.8$ Å).

The extracted Fe $3d$ PSW's in Fe/Cr system are shown in Fig. 3(a) as a function of t_{Fe}. For comparison, the Cr $3d$ and Fe $3d$ spectra of pure Cr and thick Fe are shown at the bottom and the top, respectively. Figure 3(b) shows the calculated Fe $3d$ projected local DOS (PLDOS) in the Fe(1 ML)/Cr(5 ML)/Fe(1 ML) sandwich, which is compared with the calculated $3d$ PLDOS of pure Fe and Cr metals. Note that a DOS peak at E_F exists in the Fe $3d$ PLDOS of Fe(1 ML)/Cr(5 ML)/Fe(1 ML) in Fig. 3(b), which is attributed to minority-spin states of Fe $3d$ electrons. The details of the theoretical results will be published elsewhere.[38] For Cr and Fe metals, the structures in the experimental $3d$ spectra are qualitatively similar to those in the calculated Cr $3d$ PLDOS (bottom) and Fe $3d$ PLDOS (top), respectively. A better agreement between experiment and theory is observed in Fe $3d$ valence bands than in Cr $3d$ bands. However, for both Cr and Fe, peak positions in the calculated $3d$ PLDOS's lie at higher BE's and their full widths at half maximum (FWHM) are larger than in the experimental spectra. Part of such discrepancies may be due to matrix element[39,40] and relaxation effects in the photoemission process, which are neglected in theory curves. Even a larger discrepancy is expected between the theoretical FWHM and the experimental FWHM, if the effect of finite lifetimes of valence holes is included in the calculated Fe $3d$ PLDOS, which is neglected at present.

Extracted Fe $3d$ PSW's in the Fe/Cr system reveal interesting features : (i) a sharp emission just at E_F, (ii) a peak at -1.3 eV (at -2.5 eV in the Fe $3d$ PLDOS), and (iii) a shoulder at ~ -3 eV (at -3.5 eV in the Fe $3d$ PLDOS). A sharp emission at E_F has not been observed in the previous PES studies.[29] It may correspond to *surface* states existing in very thin Fe overlayers on Cr, or may arise from a transfer of Fe $3d$ spectral weight caused by large hybridization with Cr $3d$ states. It will
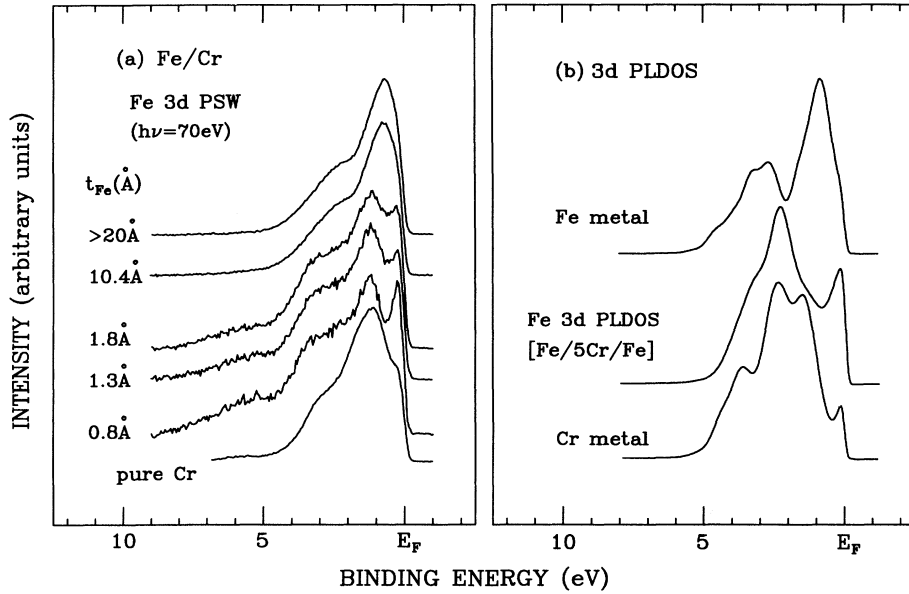

FIG. 2. Comparison of the energy distribution curves (EDC's) for Fe/Cr and Cr/Fe/Cr, taken at $h\nu = 70$ eV.

FIG. 3. (a) Comparison of the extracted Fe $3d$ partial spectral weight (PSW) distribution. (b) Calculated Fe $3d$ PLDOS for Fe metal (top), Fe $3d$ PLDOS for Fe(1 ML)/Cr(5 ML)/Fe(1 ML) (middle), and Cr $3d$ PLDOS for Cr metal, respectively.

be also interesting to find whether these features near E_F in Fe/Cr could be identified as the QWS's, similarly as in other systems.[26–28]

In order to sort out the origin of the structure near E_F, we have performed band-structure calculations and determined the Fe $3d$ PLDOS's for the two model systems, i.e., the 7 ML Fe film with a vacuum on each side and the Fe/Cr superlattice. The former calculation is for identifying the *surface* states in the Fe surface layer, while the latter is for identifying the effect of hybridization between Fe and Cr layers. We have found that both the Fe/Cr superlattice and the 7 ML Fe film possess a peak near E_F in the calculated Fe $3d$ PLDOS, but that the Fe/Cr superlattice exhibits a stronger intensity near E_F than the 7 ML Fe film does. For the 7 ML Fe film, the peak near E_F exists only in the *surface* top layer, reflecting that the peak arises from *surface* states. These theoretical investigations suggest that the observed structure near E_F in the Fe $3d$ PSW's for thin Fe overlayers on Cr originates from (i) the hybridization between Fe and Cr $3d$ electrons at the Fe/Cr interface and (ii) the *surface* states of Fe $3d$ electrons due to a vacuum above the Fe overlayer.

However, the experimental results in Fig. 3(a) seem to support the hybridization-induced mechanism for the observed structure near E_F. It is because a *surface* state should have significant spectral weight even when t_{Fe} is thick, due to short IMFP's of Fe and Cr $3d$ electrons ($\sim$ 5 Å). The latter two structures, at -1.3 eV and ~ -3 eV, resemble the Cr $3d$ valence bands (bottom), suggesting again the existence of a large hybridization between Fe and Cr layers at the interface. Substantially large hybridization between Cr $3d$ and Fe $3d$ electrons is indeed confirmed by the similarity between the calculated PLDOS structures of the Fe layer (shown in this figure) and those of the Cr layer (not presented in this paper) at the interface of the Fe(1 ML)/Cr(5 ML)/Fe(1 ML) sandwich.[38] An important finding in Fig. 3 is that the

trends observed in experiments are consistent with those in band-structure calculations for the Fe(1 ML)/Cr(5 ML)/Fe(1 ML) sandwich.

The effects of hybridization and *surface* states, both of which contribute to the sharp emission at E_F for a very thin Fe overlayer, have opposite effects on magnetism. The magnetic moment of the Fe overlayer on Cr is expected to be enhanced due to *surface* states, compared to that of *bulk* Fe metal, but should be less than the *surface* magnetic moment in the Fe film due to the hybridization effect. This is in agreement with band-structure calculations[41] which predicted a slight enhancement of the magnetic moment for 1 ML Fe on Cr(001), $2.4\mu_B$, as compared to $2.2\mu_B$ in *bulk* Fe, but less than the *surface* magnetic moment of the Fe film, $\sim 3.0\mu_B$.

The comparison for Fe/Cr in Fig. 3 suggests that the electronic structure of the Fe/Cr superlattice will be important in determining its magnetic properties which are responsible for MR effects and the exchange coupling in this system. Therefore it naturally invokes the necessity of further investigation of detailed band structures of the Fe/Cr superlattice, especially the Fermi surface geometry, and finding the correlation between its electronic structures and magnetic properties. To draw a conclusion on the exact role of the electronic structure of the Fe/Cr superlattice in these magnetic properties, further studies are required both theoretically and experimentally. In particular, spin- and angle-resolved PES measurements of the Fe/Cr and Fe/Cr/Fe sandwich films will provide more complete information on their electronic structures.

IV. CONCLUSIONS

The Fe $3d$ PSW's in the Fe/Cr film exhibit very interesting features for about 1 ML thickness of the Fe overlayer, $t_{Fe} < \sim 1.8$ Å. First, a sharp emission just at E_F is observed. This structure seems to originate mainly

from the hybridization between Fe and Cr 3d electrons at the Fe/Cr interface, and partially from the Fe 3d *surface* states of the Fe overlayer. Second, two other structures, which resemble the Cr 3d valence bands, are observed at higher BE's, reflecting the existence of a large hybridization interaction between Fe and Cr layers at the interface. The trends observed in PES measurements for Fe/Cr agree well with those in the PLDOS's obtained from band-structure calculations for the Fe(1 ML)/Cr(5 ML)/Fe(1 ML) sandwich system, which again confirms large hybridization between Fe 3d and Cr 3d electrons at the interface. Our study suggests that the electronic structure of the Fe/Cr superlattice, including its Fermi surface effects, will play an important role in determining the nature of the exchange coupling and the magnitude of MR in this system.

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$$\frac{I(\text{Fe } 3p)}{I(\text{Cr } 3p)} = \frac{\sigma_{\text{Fe}}}{\sigma_{\text{Cr}}} \frac{\lambda_{\text{Fe}}}{\lambda_{\text{Cr}}} \frac{1 - e^{-t_{\text{Fe}}/\lambda_{\text{Fe}}\cos\theta}}{e^{-t_{\text{Fe}}/\lambda_{\text{Cr}}\cos\theta}}.$$

In this equation, λ_i is the IMFP for a given electron and material ($i =$ Fe 3p, Cr 3p), and θ is the analyzer collection angle with respect to the sample surface normal ($\theta = 0°$ for our experimental geometry).

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