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VALENCE BAND PHOTOEMISSION STUDY OF Fe OVERLAYERS ON Cr

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Electronic structures of Fe overlayers on Cr (Fe/Cr) films, with an Fe coverage of $1 - 20\AA$, have been investigated by using photoemission spectroscopy. Experimental results are compared with supercell band structure calculations for a system with monolayer (ML) Fe on each side of five layer Cr, Fe(1ML)/Cr(5ML)/Fe(1ML). The extracted Fe $3d$ partial spectral weight in Fe/Cr exhibits very interesting features for very thin Fe overlayers. First, a sharp emission near the Fermi energy is observed, which 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. Second, other structures are observed at higher binding energies which resemble the Cr $3d$ valence bands, also suggesting large hybridization between Fe and Cr $3d$ states at the Fe/Cr interface. These conjectures are confirmed by band structure calculations for Fe(1ML)/Cr(5ML)/Fe(1ML).

I. INTRODUCTION

Fe/Cr multilayers have attracted much attention since the giant magnetoresistance (GMR) effect and antiferromagnetic (AF) interlayer coupling between ferromagnetic Fe layers were observed [1–3]. Similar phenomena have been observed in other ferromagnetic/nonmagnetic multilayers [4–7]. It is believed that GMR is associated with antiferromagnetic (AF) interlayer coupling between adjacent ferromagnetic layers across spacer layers [2,3,5,8,9]. Oscillatory coupling between ferromagnetic layers has been evidenced as the spacer thickness [10–14] or the ferromagnetic layer thickness varies [15]. The observed oscillation periods are quite large, about $\simeq 10\AA$. In spite of extensive work on these multilayers, the origins of GMR effect, of the AF exchange coupling between adjacent ferromagnetic layers, and of an oscillatory behavior of the coupling depending on the sublayer thicknesses have not been understood yet.

Some theoretical models have been proposed to account for the novel GMR effect, based on the mechanism of spin-dependent scattering of conduction electrons by magnetic impurities introduced by interface roughness [16–18]. These models have been substantiated by recent experimental results [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], which does not require the existence of interface roughness. 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. As to the AF exchange coupling and an oscillatory behavior of the coupling between ferromagnetic layers, a simple Ruderman-Kittel-Kasuya-Yosida (RKKY) model [24] yields a period of $\lambda_F/2$ (λ_F: Fermi wavelength of the spacer layer). However, this is too short to explain experimental results of $\simeq 10\AA$. It is recently claimed that the measured long oscillation periods can be obtained by including a realistic description of superlattice one-electron states [25].

It is important to investigate the interface electronic structure [26,27] to get further insight into the interlayer coupling of these multilayers. Several electron spectroscopy studies have been reported on oscillatory magnetic coupling, such as photoemission spectroscopy (PES), inverse photoemission spectroscopy (IPES), and spin polarized photoemission spectroscopy studies [28–30]. In these works, the quantum well states

(QWS's) at the Fermi level E_F are observed, which are considered to correlate with the oscillations of the long range magnetic coupling in Cu/Co(100) and Ag/Fe(100) multilayers. These QWS's carry predominantly minority spin polarization, reflecting a preferential hybridization in the interface.

In this paper we report synchrotron radiation PES studies of the *in-situ* prepared Fe/Cr bilayer films in the range of $1-20\AA$ of Fe coverage. We have extracted Fe $3d$ partial spectral weight (PSW) distributions as a function of the Fe overlayer thickness t_{Fe}. It is found that Fe $3d$ PSW's exhibit new structures for thin Fe coverages, as compared to Fe metal. We have also compared experimental results with theoretical density of states (DOS) obtained for a system with monolayer (ML) Fe on each side of five layer Cr, Fe(1ML)/Cr(5ML)/Fe(1ML), 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 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 $400\AA$ thick Au film on it. Evaporations were done at a low deposition rate and a good vacuum was maintained during deposition : about $0.5\AA$/min at the chamber pressure of 2×10^{-10} Torr for Cr, and $0.2\AA$/min at 7×10^{-11} Torr for Fe, respectively. The substrate was kept at room temperature during evaporation. Deposited film thicknesses were monitored by quartz crystal sensors. The cleanliness of the sample surfaces was checked with the valence band spectra, taken 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 ~ 0.2 eV at $h\nu = 70$ eV and ~ 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. All the spectra were normalized to the incident photon flux.

A supercell of the Fe(1ML)/Cr(5ML)/Fe(1ML) 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. For comparison, we have also calculated electronic structures of an Fe film, with seven Fe layers and three vacuum layers on each side, and an Fe/Cr superlattice.

III. RESULTS AND DISCUSSION

Figure 1(a) shows the extracted Fe $3d$ partial spectral weights (PSW's) in Fe/Cr as a function of the Fe layer thickness 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. The extraction procedure is as follows. First, the fraction of the Fe $3d$ to Cr $3d$ emissions in each valence band spectrum is estimated for a given value of t_{Fe}. Here we used valence band PES spectra, taken at $h\nu = 70$ eV, where Cr and Fe $3d$ emissions are dominant over Cr and Fe $4s$ and $4p$ emissions [32]. Then the pure Cr $3d$ spectrum is multiplied by the factor which is equal to the fraction of the Cr $3d$ emission relative to the total $3d$ emission, and then 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 $t_{Fe} < 1.8\AA$).

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, and (iii) a shoulder at ~ -3 eV. A sharp emission at E_F has not been observed in previous

PES studies [26]. It may correspond to *surface* states existing in very thin Fe overlayers on Cr, or may arise from a transfer of Fe 3*d* spectral weight caused by large hybridization with Cr 3*d* states. For $t_{Fe} > 10$Å, the valence band spectrum becomes essentially identical to that of Fe metal [33,34]. For Fe 3*d* PSW, 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, according to spin-resolved PES studies of Fe metal [35] as well as band structure calculations in this work.

Figure 1(b) shows the calculated Fe 3*d* projected local DOS (PLDOS) in Fe(1ML)/Cr(5ML)/Fe(1ML) sandwich, which is compared with the calculated 3*d* PLDOS of pure Fe and Cr metals. Note that a DOS peak at E_F exists in the Fe 3*d* PLDOS of Fe(1ML)/Cr(5ML)/Fe(1ML) in Fig. 1(b), which is attributed to minority-spin states of Fe 3*d* electrons. There is also a peak at -2.5 eV with a shoulder at ~ -3.5 eV. For Cr and Fe metals, the structures in the experimental 3*d* spectra are qualitatively similar to those in the calculated Cr 3*d* PLDOS (bottom) and Fe 3*d* PLDOS (top), respectively. A better agreement between experiment and theory is observed in Fe 3*d* valence bands than in Cr 3*d* bands. However, for both Cr and Fe, peak positions in the calculated 3*d* PLDOS's lie at higher BE's and their FWHM's are larger than in the experimental spectra. Part of such discrepancies may be due to matrix element and relaxation effects in photoemission process, which are neglected in theory curves.

Figure 2 compares the calculated Fe 3*d* PLDOS's for two model systems, i.e., the 7ML Fe film (thin solid line) with a vacuum on each side and the Fe/Cr superlattice (thick solid line). The Fe 3*d* PLDOS of Fe metal (dashed line) is also provided for comparison. The purpose of these theoretical investigations is to sort out the origin of the near E_F structure in Fe overlayers on Cr. The former calculation is for identifying the *surface* states

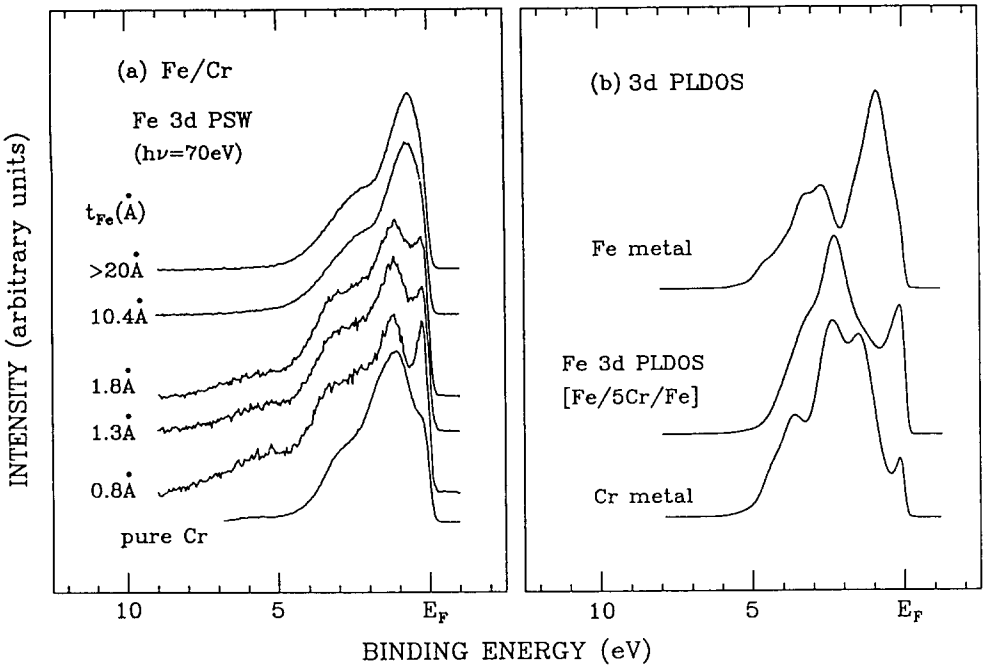

FIG. 1 (a) Comparison of the extracted Fe 3*d* partial spectral weight (PSW) distribution. (b) Calculated Fe 3*d* PLDOS for Fe metal (top), Fe 3*d* PLDOS for Fe(1ML)/Cr(5ML)/Fe(1ML) (middle), and Cr 3*d* PLDOS for Cr metal, respectively.

in the Fe *surface* layer, while the latter is for identifying the effect of hybridization between Fe and Cr layers. This figure shows that both the Fe/Cr superlattice and the 7ML Fe film possess a peak near E_F in the calculated Fe 3*d* PLDOS, and that the Fe/Cr superlattice exhibits a stronger intensity near E_F than the 7ML Fe film does. For the 7ML Fe film, the peak near E_F exists only in the *surface* top layer, reflecting that the peak arises from *surface* states. This theoretical comparison suggests that the observed structure near E_F in the Fe 3*d* PSW's for thin Fe overlayers on Cr originates from: (i) the hybridization between Fe and Cr 3*d* electrons at the Fe/Cr interface, and (ii) the *surface* states of Fe 3*d* electrons due to a vacuum above the Fe overlayer.

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 [36] which predicted a slight enhancement of the magnetic moment for 1ML 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, $\sim 3.0\mu_B$.

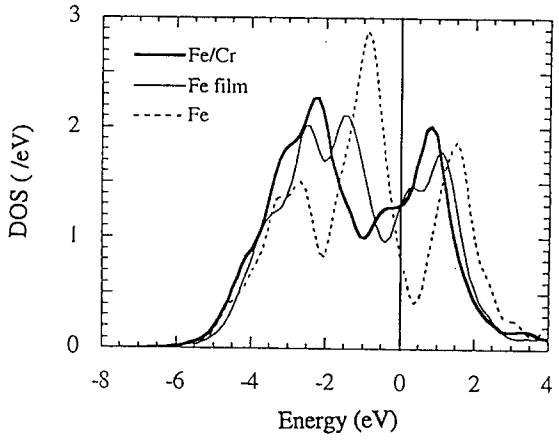

FIG. 2 Comparison of the calculated Fe 3*d* PLDOS's for an Fe/Cr superlattice (thick solid lines), for a top surface Fe layer in a 7 ML Fe film (thin solid lines), and for Fe metal (dashed lines), respectively.

The experimental results in Fig. 1(a) seem to support the hybridization-induced mechanism for the observed structure near E_F, because a *surface* state should have significant spectral weight even when t_{Fe} is thick, due to short electron mean free paths of Fe and Cr 3*d* electrons (~ 5Å) [37]. The latter two structures, at -1.3 eV and ~ -3 eV, resemble the Cr 3*d* valence bands (bottom), suggesting again the existence of a large hybridization between Fe and Cr layers at the interface. An important finding in Fig. 1 is that the trends observed in experiments are consistent with those in band structure calculations for the Fe(1ML)/Cr(5ML)/Fe(1ML) sandwich. Therefore our finding suggests that the electronic structure of the Fe/Cr multilayer might be important in determining its magnetic properties, such as GMR effects and the oscillatory exchange coupling.

IV. CONCLUSIONS

The Fe 3*d* PSW's are extracted for the Fe/Cr film as a function of an Fe coverage. It is found that the Fe 3*d* PSW's exhibit very interesting features for $t_{Fe} <\sim 1.8$Å, such as a sharp emission just at E_F and other structures, similar to those of bulk Cr 3*d* spectrum. A sharp emission just at E_F seems to originate mainly from the hybridization between Fe and Cr 3*d* electrons at the Fe/Cr interface, and partially from the Fe 3*d* *surface* states of the Fe overlayer. In addition, two other structures are observed at higher BE's. These latter features resemble those of bulk Cr 3*d* spectrum, and so reflect the existence of a large hybridization interaction between Fe and Cr layers at the interface. The trends observed in valence band spectra in Fe/Cr agree well with those in the PLDOS's obtained from band structure calculations for the Fe(1ML)/Cr(5ML)/Fe(1ML) sandwich system. This finding also confirms large hybridization between Fe 3*d* and Cr 3*d* electrons at the interface. Our studies suggest that the details of the electronic structure of the Fe/Cr multilayer are important in determining its oscillatory AF coupling and GMR effect.

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