

## Ag/Fe(001) interface

N. B. Brookes,\* Y. Chang, and P. D. Johnson

*Physics Department, Brookhaven National Laboratory, Upton, New York 11973*

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Spin-resolved photoemission studies of the interface formed by depositing silver on an Fe(001) substrate show that the silver  $d$  bands show evidence of hybridization with the substrate  $d$  bands through the presence of a spin polarization for the initial monolayer of silver. Subsequent layers of silver show no evidence of spin polarization in the  $d$  bands. Studies of the dispersion of the interface or quantum-well states associated with these thin films reveal that their effective mass is enhanced due to hybridization with the substrate. This has implications for theoretical models of the exchange coupling in the associated magnetic multilayers, where the bulk band structure is often assumed for the noble-metal intervening layer.

### INTRODUCTION

There is currently considerable interest in the properties of magnetic surfaces, thin films, and multilayers.<sup>1</sup> The technological drive for such studies lies in the possibility of tailoring new materials for both the recording and device industries.<sup>2,3</sup> In particular, transition-metal multilayers are one of the more promising new materials. In these multilayers it is possible to achieve either ferromagnetic or antiferromagnetic coupling of adjacent ferromagnetic layers depending on the thickness of the intervening layer.<sup>4</sup> Example systems include Fe/Cr (Ref. 5) and Cu/Co (Ref. 6) multilayers. The related giant magnetoresistance properties and the large enhancement of the Kerr rotation in these multilayers make them particularly interesting.<sup>7</sup>

Several theories of the oscillatory exchange coupling invoke bulk Fermi-surface spanning vectors that reflect the correct Fermi surface for the intervening layer.<sup>8</sup> Indeed such theories may predict more than one periodicity for the coupling as has been observed experimentally for both the Fe/Cr (Ref. 5) and Cu/Co (Ref. 9) multilayers. While these models are undoubtedly correct in the asymptotic limit of thicker intervening layers, it is unclear that they will necessarily apply to the thinner films where the effects of hybridization in the interface between the magnetic and nonmagnetic layers will be much stronger.

Experimental studies have shown that quantum-well states sample the Fermi surface in finite noble-metal films with a periodicity identical to that observed for the oscillatory exchange coupling in the associated multilayers.<sup>10-14</sup> Elsewhere we have shown that this periodicity for the Fermi-surface crossings directly reflects the discrete periodicity or atomic structure within the thin film.<sup>15</sup> Both the finite quantum-well picture and the bulk electronic band structure lead to the same predicted periodicity for the exchange coupling in the multilayers. However, these two pictures represent models that start from two different extremes, the finite film and the infinite solid. It is therefore of considerable interest to examine further the electronic structure of the thin films.

In this paper we extend our earlier spin-polarized photoemission study of the interfacial properties of thin

silver films deposited on an Fe(001) substrate.<sup>10</sup> We examine the effects of hybridization on the dispersion of the interface or quantum-well states as a function of the silver thickness. Our experimental studies show that in the thinner films hybridization in the interface results in a considerable enhancement of the effective mass of the quantum-well states. We also examine the coverage-dependent changes observed in the vicinity of the silver  $d$  bands where we again demonstrate strong hybridization effects in the ultrathin films.

### EXPERIMENTAL PROCEDURES

The spin-polarized photoemission experiments reported here were carried out on an apparatus that has been described in detail elsewhere.<sup>16</sup> Briefly, the photoemitted electrons are energy and momentum analyzed by a commercial hemispherical analyzer backed by a compact low-energy spin detector.<sup>17</sup> The incident photon flux is provided by the U5 VUV undulator at the National Synchrotron Light Source. The angular resolution of the hemispherical analyzer was  $\pm 1.5^\circ$  and the combined photon and analyzer energy resolution was 0.35 eV or better at the lower photon energies.<sup>16</sup>

One of the principle reasons for choosing to grow silver on the (100) surface of iron is the good lattice match. Indeed the surface unit cell of fcc (001) silver, a  $2.89\text{-}\text{\AA}$  square, represents an excellent match to the  $2.87\text{-}\text{\AA}$  square surface net of bcc Fe(001). A lattice match of less than 1% gives the possibility of good epitaxial growth and indeed successful multilayer growth has been reported.<sup>18</sup>

The Fe(001) crystal was manufactured in the form of a picture frame with each leg cut along a  $\langle 100 \rangle$  direction. A coil for magnetization was wound around one leg. This substrate was cleaned by repeated argon ion bombardment and annealing cycles (750°C). Surface contamination levels were monitored initially using Auger electron spectroscopy and in the final stages using photoelectron spectroscopy. Surface crystallographic order was examined with low-energy electron diffraction (LEED).

The silver films were evaporated from a tungsten basket at room temperature and at a rate of approximately 0.25 ML per minute. The iron and silver Auger ratios

were measured as an estimate of the coverage and the evaporations were monitored using a quadrupole mass spectrometer. LEED measurements showed a good sharp  $p(1 \times 1)$  pattern at all coverages up to approximately 3 ML, after which the patterns became less sharp. As a result, in the following discussion we restrict our observations to the first three monolayers. We found no evidence for intermixing at room temperature.

## RESULTS AND DISCUSSION

### Clean Fe(001)

Since the photoemission results for the clean iron surface are pertinent to the following discussions we briefly review them. The normal emission spin-resolved photoemission spectra recorded from clean Fe(001) with a photon energy of 52 eV and with the light incident at two angles of incidence,  $\vartheta_i = 70^\circ$  and  $35^\circ$ , are shown in Figs. 1(a) and 1(b), respectively. The minority (0.3 eV) and majority (0.7, 2.6 eV) peaks at normal emission are derived from relatively flat bulk bands near  $\Gamma$  in the  $\Gamma$ - $\Delta$ - $H$  direction of the bulk Brillouin zone. At normal emission, selection rules allow only initial states of  $\Delta_1$  and  $\Delta_5$  symmetry;  $\Delta_1$  when the electric vector lies perpendicular to the surface,  $\Delta_5$  when it is parallel. With an angle of incidence,  $\vartheta_i = 70^\circ$ , the  $\Delta_1$  majority peak at a binding energy of 0.7 eV is relatively intense compared to the  $\Delta_5$  exchange split bands at binding energies of 0.3 and 2.6 eV. Shown in Fig. 1(b), these  $\Delta_5$  peaks are found to be stronger at smaller angles of incidence in agreement with previous

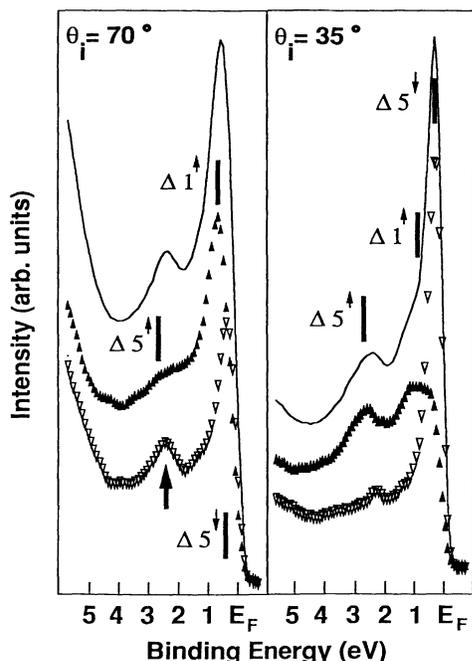


FIG. 1. Spin-integrated and spin-resolved photoemission spectra recorded along the surface normal for photons of incident energy 52 eV. The spectra are shown for two angles of incidence corresponding to  $p$ -polarized light ( $70^\circ$ ) and  $s$ -polarized light ( $35^\circ$ ).

work.<sup>19</sup> The rapidly rising intensity below approximately 5.0 eV binding energy is due to the presence of a relatively intense  $s/p$  band at these energies. At a photon energy  $h\nu = 52$  eV, the  $d$ -band intensity is greatly reduced prior to the  $3p$  threshold. There also appears to be strong coupling to the final state, leading to the intense  $s/p$  band emission.

Quenched by small amounts of oxygen, the peak at 2.4-eV binding energy in the minority-spin spectrum of Fig. 1(a) has previously been assigned to a minority-spin surface resonance<sup>20</sup> with  $\Delta_1$ -type symmetry as indicated by its sensitivity to  $p$ -polarized light. It should be noted that at a photon energy of 52 eV, there is attenuation of the entire spectrum in the presence of oxygen. This suggests a substantial surface sensitivity and, indeed, calculations have indicated the presence of further surface-related features close to the Fermi level. Elsewhere it has been shown experimentally that majority-spin surface state exists near the Fermi level.<sup>21</sup> However, that study was carried out at lower photon energies and with  $s$ -polarized rather than  $p$ -polarized light as used in the present study. The sensitivity to  $s$ -polarized light in the other study<sup>21</sup> indicates that the majority-spin surface state observed close to the Fermi level had  $\Delta_5$  symmetry.

### The Ag/Fe(001) interface

The general phenomena that are observed on depositing silver on Fe(001) have been described previously.<sup>10</sup> In the following we will summarize those results and present our wider findings.

The low-coverage region, illustrated in Fig. 2, shows the attenuation of the minority-spin surface resonance characteristic of the clean surface and the growth of a new feature closer to the Fermi level. With increasing coverage a new feature can be associated with each new monolayer, peaking in intensity at integer monolayer coverage, and attenuating with further deposition. These spectral changes associated with different silver layer thicknesses are illustrated in Fig. 3. Examination of the figure shows that a quantum-well state corresponding to quantum number  $\nu = 1$  moves up to and through the Fermi level at a thickness between 3 and 4 monolayers. Here the quantum number  $\nu$  is related to  $m$ , the number of layers in the well or thin film, and  $n$ , the number of nodes in the state's wave function, by  $\nu = m - n$ .<sup>15</sup> In the highest coverage spectrum of Fig. 3 we are able to identify the quantum-well state corresponding to quantum-well number  $\nu = 2$  at a binding energy of 2.0 eV.

The intensities of the different quantum-well states recorded as a function of coverage provide an indication of the initial growth mode of silver on the Fe(001) substrate. Plotted in Fig. 4, these intensities clearly indicate that the growth proceeds layer by layer, at least for the first few layers. For comparison the dashed lines in the figure show the intensities that would be expected on the basis of island growth with a Poisson-like distribution of island sizes.

The spin-resolved spectra recorded from the 1-ML Ag coverage is shown in Fig. 5. The interface state at 1.7-eV binding energy shows a strong minority-spin polarization.

Reported elsewhere,<sup>10</sup> the subsequent interface states observed with increasing silver deposition show minority-spin polarization. For the monolayer coverage, Fig. 5, a possible majority-spin feature, is also observed as a small peak at  $\sim 3.0$ -eV binding energy. A majority-spin  $\Gamma_{25'}$  bulk transition will also occur at a binding energy close to this. However, the latter peak is sensitive to  $s$ -polarized rather than  $p$ -polarized light. Further, such a feature would be accompanied by the minority-spin component immediately below the Fermi level. The latter is clearly not observable.

The spectra shown above are recorded with an incident photon energy  $h\nu = 52$  eV, an energy at which the surface and interface states are more prominent and the bulk iron  $d$  bands are at a local intensity minimum. If we consider the amplitude of the iron  $d$  bands in Fig. 2 as silver is deposited, we observe that they attenuate more rapidly than would be expected for a 4-Å mean free path, an indication of surface-related emission in the clean surface spectrum. Changing the incident photon energy to 36 eV we obtain an increased substrate sensitivity. Such a spectrum is shown in Fig. 6, again for  $\sim 1$  ML Ag/Fe(001). Now the substrate majority and minority features can be clearly seen at the Fermi level and the silver-induced features are less prominent. The majority-spin peak at

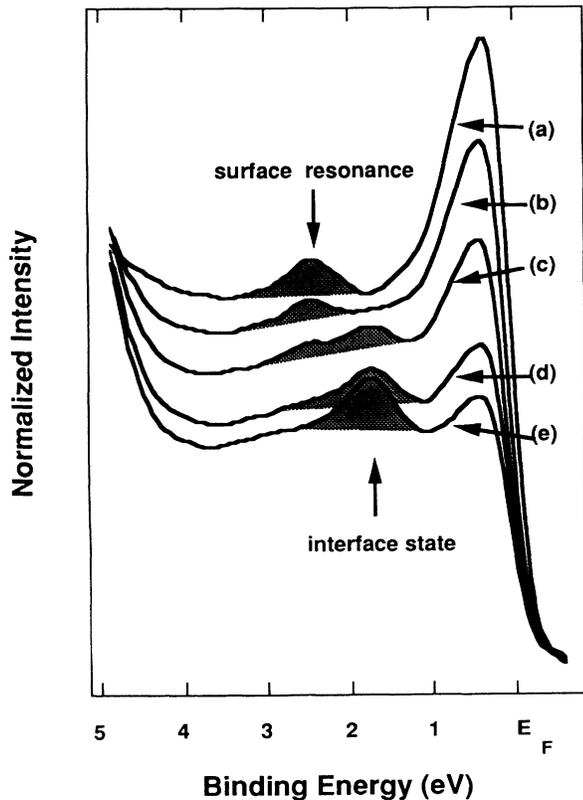


FIG. 2. Spin-integrated photoemission spectra recorded along the surface normal from different submonolayer-coverage silver films deposited on an Fe(001) substrate. The different silver coverages are (a) 0.0 ML, (b) 0.1 ML, (c) 0.2 ML, (d) 0.3 ML, and (e) 0.4 ML. The incident light is  $p$  polarized with an energy of 52 eV.

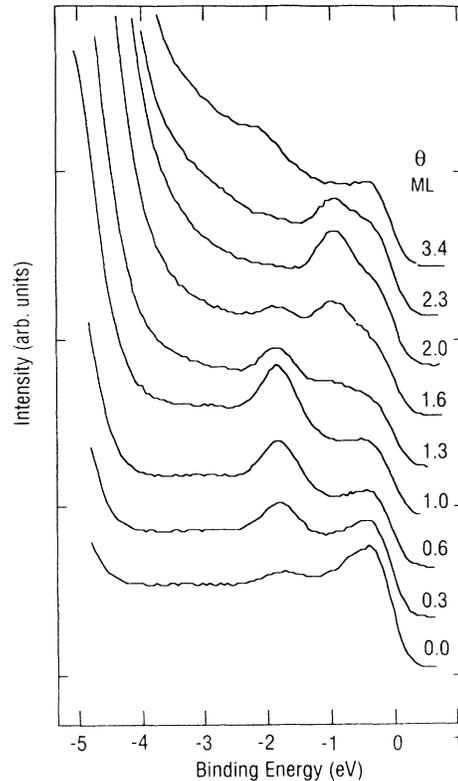


FIG. 3. Spin-integrated photoemission spectra recorded along the surface normal from different thickness silver films deposited on an Fe(001) substrate as indicated. The incident light is  $p$  polarized with an energy of 52 eV.

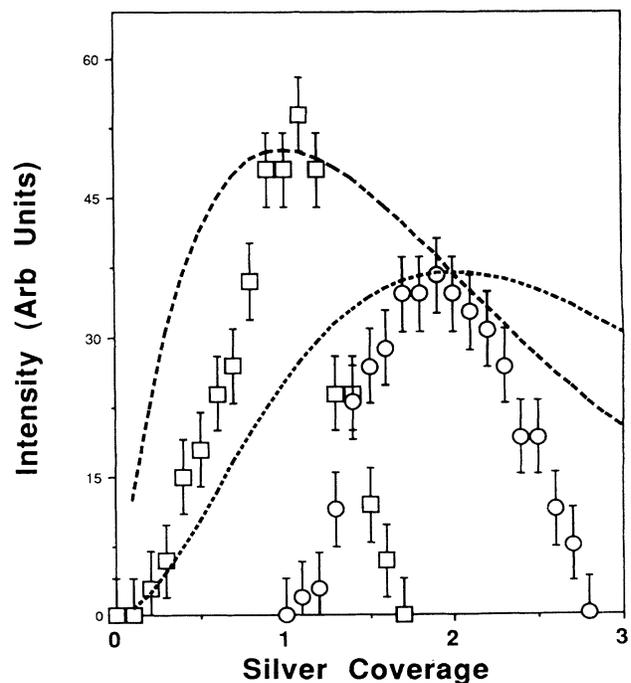


FIG. 4. The measured intensities of the quantum-well states as a function of coverage. The open squares represent the quantum-well state corresponding to one monolayer and the open circles correspond to the second monolayer. The dashed lines represent the intensities that would be expected on the basis of islandlike growth.

3.0-eV binding energy is, however, more prominent. In accord with calculation,<sup>22</sup> earlier experimental studies have previously found that the majority-spin iron bulk  $\Delta_5$  band is observed at a binding energy of 2.4 eV for excitation with photons of energy 35 eV.<sup>20</sup> This is accompanied by a minority-spin transition immediately below the Fermi level. From the present studies it therefore remains unclear whether the peak at 3.0 eV is related to a bulk transition or to a predicted majority interface state. If it is the latter, any difference in intensity for the majority- and minority-spin states on changing photon energy would reflect the relative iron and silver components in the two states with the minority-spin state having more weight in the silver layer and the counterpart majority-spin state having more weight in the iron layers.

Several theories of the oscillatory exchange coupling in the magnetic multilayers involve Fermi-surface spanning vectors at points away from the center of the Brillouin zone. It is therefore of interest to examine the dispersion of the interface or quantum-well states as a function of  $k_{\parallel}$  the momentum parallel to the surface. Figure 7 shows the angular dependence of the 1-ML interface state, which can be easily followed up to  $\sim 8^\circ$  off normal after which it merges with the structure closer to the Fermi level. The zone boundary is at approximately  $16^\circ$  off normal in these spectra. Figure 7 shows this dispersion and the corresponding dispersion for the 2-ML state plotted as a function of  $k_{\parallel}$ . In the latter case the state can be followed across the full zone. Other than at normal emission the 3-ML peak is too weak to observe with any

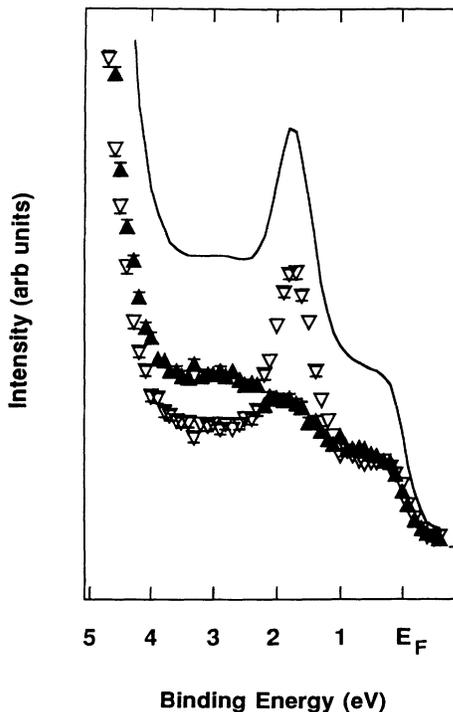


FIG. 5. Spin-resolved spectra recorded from 1 ML of silver deposited on the Fe(001) substrate. The incident light is  $p$  polarized with an energy of 52 eV.

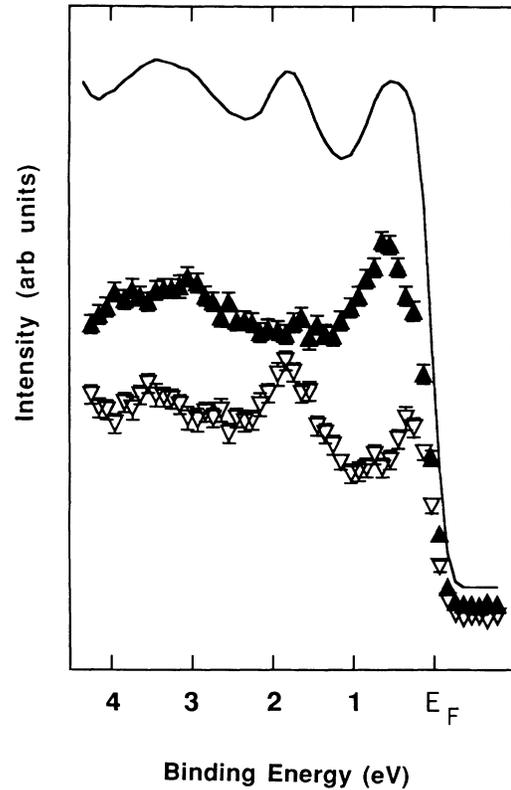


FIG. 6. Spin-integrated and spin-resolved spectra recorded from 1 ML of silver deposited on the Fe(001) substrate. The incident light is  $p$  polarized with an energy of 36 eV.

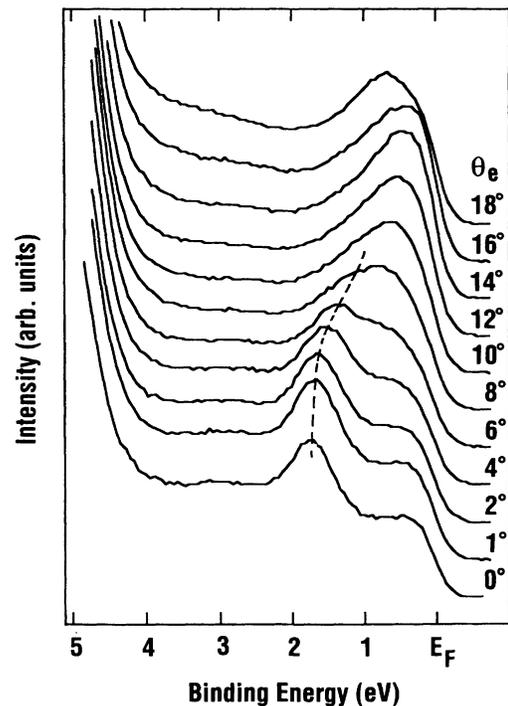


FIG. 7. Spin-integrated spectra showing the angular dependence of the 1-ML Ag interface state. The incident light is  $p$  polarized with an energy of 52 eV.

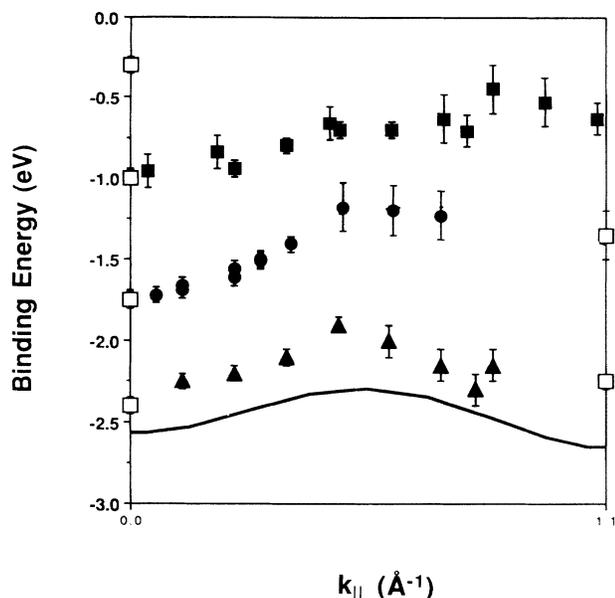


FIG. 8. Observed dispersion as a function of  $k_{\parallel}$  for the clean Fe(001) surface resonance and the interface states corresponding to one 1- and 2-ML coverages of silver. The solid line shows the calculated dispersion of the clean surface state (Ref. 20). The open squares indicate the experimentally observed binding energies at the zone boundaries.

confidence.

We have previously indicated that the measured dispersion of the surface state found on the Fe(001) surface shows good agreement with the calculated dispersion.<sup>20</sup> Indeed this dispersion is largely determined by the dispersion of the bottom of the even symmetry hybridization gap at the center of the surface Brillouin zone. For comparison we again show the experimentally observed and calculated dispersion for this state in Fig. 8. The measured positive dispersion near the center of the zone for this state corresponds to an effective mass of  $1.88m_e$ . The dispersion measured for the 1-ML interface or quantum-well state corresponds to an effective mass of  $2.1m_e$ . At 2 ML of silver the effective mass of the quantum-well state increases to  $2.8m_e$ . We suggest that this increase as the state moves to lower binding energies probably reflects the increased coupling to the substrate minority  $d$  bands as the state moves into resonance. Elsewhere calculations for an isolated Ag(001) (Ref. 23) slab indicate that in this particular azimuth, the quantum-well states may well show a dispersion characterized by an effective mass of the order of  $0.5m_e$ .

#### The Ag $d$ bands

The silver  $4d$  bands also change in a systematic manner. Figure 9 shows the coverage dependence of these bands between submonolayer and 3.5-ML Ag coverages. Obtained with incident  $p$ -polarized light,  $h\nu=60$  eV, the spectra at normal emission are dominated by states of  $\Delta_1$  symmetry. At low coverage, 0.3 ML, we observe a two-peaked structure with the peak at deeper binding energy having the higher intensity. At coverages

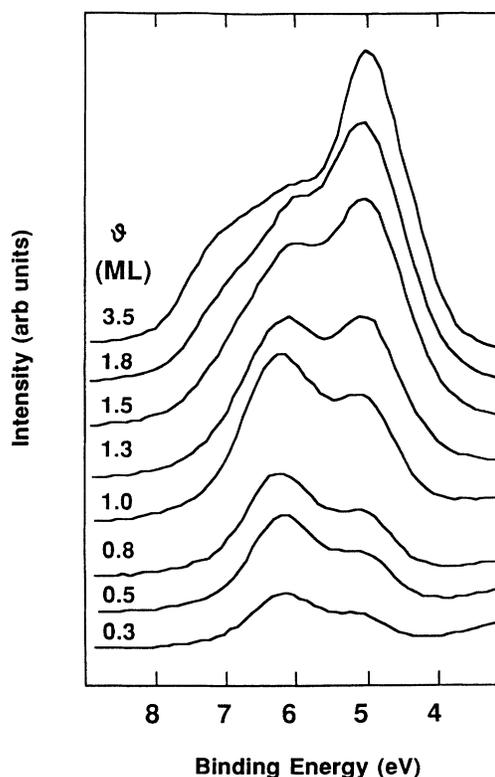


FIG. 9. The coverage dependence of the silver  $d$  bands as a function of silver coverage. The incident light is  $p$  polarized with an energy of 60 eV.

exceeding 1 ML the intensities of the two peaks become equal and by 2 ML the lower binding-energy peak is the more intense. At this coverage an additional shoulder appears at a binding energy of 6.5 eV. The shoulder develops further as 3 ML is approached but beyond this coverage no further changes are observed. The 3-ML spectrum is essentially identical to the spectrum obtained from bulk Ag(001).

With the peaks in Fig. 9 showing different intensities as a function of coverage it is tempting to associate the spectra with a quantization of the  $d$  bands. Indeed such a quantization would be expected and has been discussed elsewhere for other thin-film systems.<sup>24,25</sup> One might associate the peak at a binding energy of 5.0 eV with the first monolayer and the peak at a binding energy of 4.5 eV with the second monolayer. However the photoemission spectra from a 1-ML film look very different as a function of photon energy. Shown in Fig. 9, the two-peaked structure is shown to undergo a complete reversal of intensity between 30- and 60-eV incident photon energies. Such a sensitivity to photon energy may be indicative of silver-iron hybridization. One way of determining the iron contribution to the  $4d$  bands is to change the photon energy to the Cooper minimum for silver, which occurs at  $\sim 120$  eV. At this photon energy the silver  $4d$  cross section is 100 times lower than at 60 eV. Figure 10 shows such a spectrum recorded from a monolayer coverage of Ag at a photon energy of 118 eV. A distinct peak

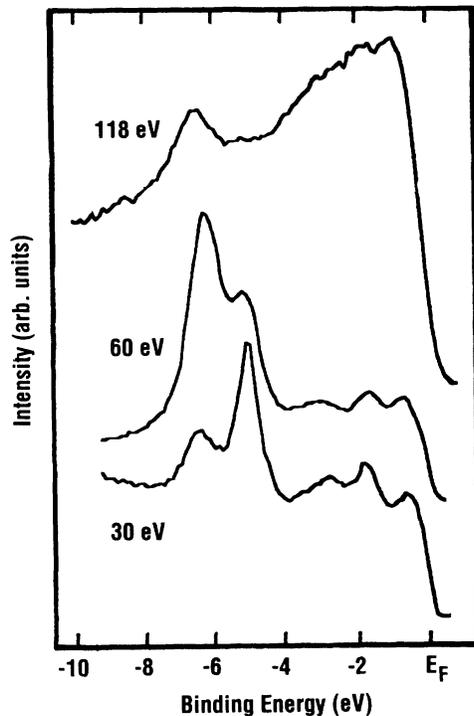


FIG. 10. Spin-integrated spectra recorded from 1 ML of silver deposited on an Fe(001) substrate at photon energies corresponding to 30, 60, and 118 eV.

is seen at 6-eV binding energy and residual intensity at 5 eV. The former peak is an order of magnitude larger than one would expect simply from consideration of the silver cross section when compared with the lower photon energy spectra. However the iron cross section changes only by a factor of 2 over this energy range consistent therefore with the presence of iron intermixing in the silver 4*d* bands at this binding energy.

Figure 11 shows the spin-resolved spectra for the silver 4*d* bands with a silver coverage of  $\sim 0.7$  ML. The intensity difference between the majority and minority spectra for the higher binding-energy peak is further evidence of iron-silver hybridization. The degree of polarization observed in the spectrum was found to be sensitive to surface contamination, and indeed it was reduced after a period of several hours. Similar spin-resolved spectra taken at lower photon energies indicated a similar difference between the minority and majority states although not as large in magnitude. Spin-resolved spectra for a 2-ML film showed no measurable difference between the majority and minority peaks. However, there is a large increase in the silver intensity at 2 ML (three times the single layer) and the shape of the 4*d* bands changes substantially as seen above. It is possible therefore that hybridization effects still exist at the interface but that they are below the present detection limit.

The observation of a spin polarization associated with the *d* bands of the silver at the interface is consistent with the results of a recent magnetic circular dichroism study

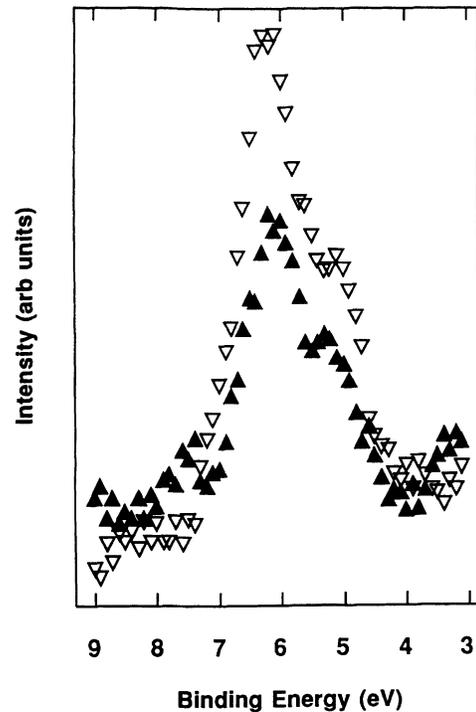


FIG. 11. Spin-resolved spectra recorded from the silver *d* bands at a coverage corresponding to 0.7 ML of silver deposited on the Fe(001) substrate. The incident light is *p* polarized with an energy of 60 eV.

of the related system of copper films deposited on a Co(001) substrate.<sup>26</sup> In that study it was suggested that the large moments observed on the copper atoms were localized at the interface. We should add that while we report in this study the observation of highly spin-polarized electron states associated with the noble metal, it is not possible to quantify the magnitude of magnetic moments from angle-resolved photoemission studies. Theoretical studies reported elsewhere find that a monolayer of silver deposited on an Fe(001) substrate will have a magnetic moment on the silver atoms of the order of  $0.08 \mu_B$ .<sup>27</sup>

## SUMMARY

The experimental observations reported in this paper clearly show the role of hybridization at the interface between two metals, in the present case silver films deposited on an Fe(001) substrate. The hybridization significantly increases the effective masses describing the dispersion of the silver quantum-well states away from the center of the Brillouin zone. Although not observed in the present study these dispersions will presumably become more free-electron-like or even faster as the films become thicker and the effect of the interface is reduced as has been discussed elsewhere in studies of Cu films deposited on Co(001).<sup>28</sup> These observations have important implications for models of the exchange coupling in mag-

netic multilayers invoking Fermi-surface spanning vectors away from the center of the zone. The effect of hybridization on the noble-metal  $d$  bands is also observed. At 1-ML coverage the silver  $d$  bands show evidence of Fe intermixing with associated spin polarization. With increasing coverage this spin polarization disappears suggesting that a strong interaction of the  $d$  bands is confined to the interface.

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\*Present address: ESRF, BP 220, F38043 Grenoble Cedex, France.

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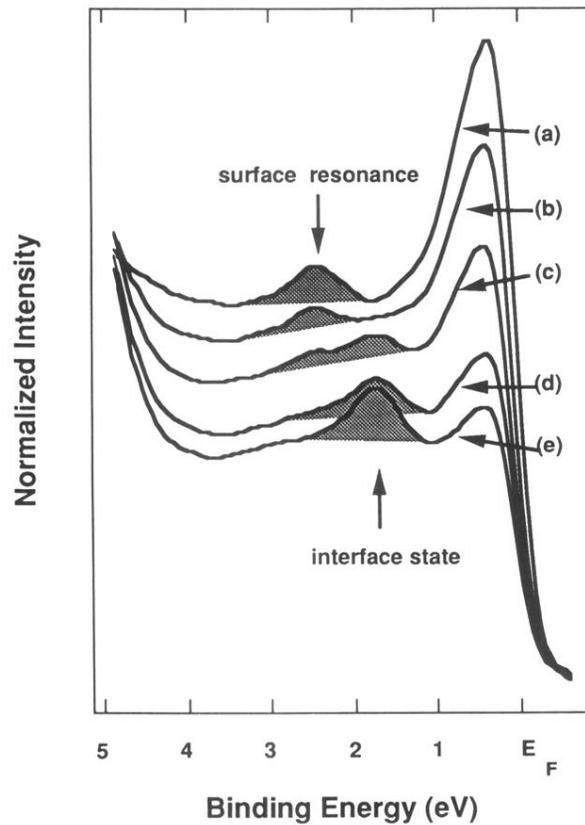


FIG. 2. Spin-integrated photoemission spectra recorded along the surface normal from different submonolayer-coverage silver films deposited on an Fe(001) substrate. The different silver coverages are (a) 0.0 ML, (b) 0.1 ML, (c) 0.2 ML, (d) 0.3 ML, and (e) 0.4 ML. The incident light is  $p$  polarized with an energy of 52 eV.