

Unoccupied electronic states of ultrathin films of Co/Cu(111) and Fe/Cu(111)

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Angle-resolved inverse photoemission has been used to study changes in the unoccupied electronic states for ultrathin films of Co and Fe epitaxially deposited on Cu(111). Combining inverse photoemission and photoemission measurements of the occupied valence states reveals changes in the exchange splitting of the $3d$ band. The exchange splitting in a 15-Å-thick Co film is $\Delta E_{\text{ex}} = 1.05 \pm 0.1$ eV, a value close to that measured for bulk Co(0001), $\Delta E_{\text{ex}} = 1.1 \pm 0.1$ eV. Similar electronic structure is observed in thinner epitaxial Co films, 15 Å down to 2 Å thick, despite the fact that films cease to be continuous below 4 Å (i.e., 2 layers). The inverse photoemission spectra for the Fe/Cu(111) films show a more complicated evolution with increasing film thickness. Above 10 Å, the Fe films undergo a structural transition to the bcc(110) phase showing a 0.5-eV shift higher in energy of the uppermost minority-spin $3d$ band, indicative of a transition to the higher-spin-state bcc phase with an exchange splitting of 1.9 ± 0.2 eV. The lower energy of the uppermost minority-spin $3d$ band for the thinner films possibly indicates reduced exchange splitting in the low-spin phase of fcc Fe(111).

I. INTRODUCTION

The properties of ultrathin epitaxial ferromagnetic films of transition metals on nonmagnetic substrates is a subject of considerable interest.¹ Interesting magnetic properties of these systems include thickness-dependent Curie temperatures,² critical exponents,² magnetic anisotropies,⁴ and micromagnetic properties.⁵ However, little is known about the electronic structure of these ultrathin films. In particular, the study of the unoccupied electronic states in thin magnetic films has only just begun. In this paper, we report inverse photoemission and photoemission measurements of the development of the electronic states as a function of film thickness for two thin-film systems: Co and Fe layers on Cu(111).

The magnetic moment per atom is given by the difference in occupation of the majority and minority $3d$ states. It has also been correlated to the exchange splitting of the $3d$ band by the empirical relation: exchange splitting in eV \approx magnetization per atom in Bohr magnetons.⁶ As the film thickness approaches a single layer, the magnetic moment per atom has been predicted to remain nearly constant for the Co/Cu(111) system⁷ [$1.56\mu_b$ for bulk hcp Co versus $1.63\mu_b$ for the Co/Cu(111) monolayer]. Photoemission measurements of 1 and 2 monolayer Co/Cu(111) films showed no enhancement in the exchange splitting.^{8,9} However, the majority and minority bands are not clearly resolved in photoemission, due to overlapping transitions from bands of different symmetry. Recent magnetic measurements¹⁰ of a Co/Cu(111) monolayer have shown the Co magnetic moment is roughly bulklike for a monolayer. For the Fe/Cu(111) system, magnetic measurements¹¹ have shown the low-spin phase of fcc Fe can be stabilized on Cu(111) with a magnetic moment per atom of $0.5\mu_b$ compared to the magnetic

moment of $2.2\mu_b$ for bulk bcc Fe, with no dependence of the magnetic moment per atom on film thickness. However, these films were grown at 400°C and are representative of intermixed Fe-Cu films on Cu(111).

The perpendicular dispersion of the unoccupied part of the $3d$ band can be determined with inverse photoemission by varying the incident electron energy and measuring the energy distribution of emitted photons at each incident energy. It should be quantized for ultrathin films, becoming dispersionless for a two-dimensional monolayer film. Changes due to band narrowing and variations in the exchange splitting have been studied by measuring the unoccupied minority-spin $3d$ states, which can be isolated rather well with inverse photoemission.

II. EXPERIMENT

Inverse photoemission measurements were performed using a tunable instrument described elsewhere.¹² The energy resolution of this instrument is better than 0.26 eV (photons plus electrons) at low (10–20 eV) incident electron energies. Since the energy spectrum of the emitted photons is measured for constant initial electron energy, each spectrum measures the distribution of unoccupied electronic states for a single value of the crystal momentum. Tuning the initial electron energy through the 10–20-eV range for normal incidence electrons, the intensity for $3d$ states increases relative to the intensity for s,p -like states. This allows the orbital character of the electronic states observed in the spectra to be determined. With enhanced resolution, small differences in the energy and intensity of image potential surface states can also be monitored for use as an analytical tool. The binding energy of the image states is referenced to the vacuum level, so changes in the work function are reflected in their po-

sition.¹³ The width and intensity of the image states reflect changes in the surface potential barrier, which is sensitive to changes at the surface. For example, Fe/Ag(001) and Co/Ag(001) films, which exhibit poor low-energy electron diffraction (LEED) patterns, do not show well-defined image potential states, whereas more ordered films do. Complimentary photoemission measurements of the occupied states were performed in a separate experimental apparatus using a conventional spectrometer and discharge light source.¹⁴

The Cu(111) sample was mechanically polished to 1 μm and then electropolished. Repeated cycles of glancing incidence Ar^+ ion bombardment at 500 eV and annealing to 650 K produced a clean, well-ordered surface as determined by LEED. The substrate surface quality was further confirmed by the intensity of the $n=1$ image state, which has a greater intensity than the $n=0$, s,p surface state for high-quality surfaces. Co and Fe thicknesses were determined with a calibrated quartz-crystal microbalance. Film structure and purity was checked with LEED and from the position and intensity of the image states observed in inverse photoemission. The Co films were observed to grow epitaxially with a different stacking arrangement than the fcc Cu(111) substrate. The LEED pattern for films thicker than 4 \AA (2 layers) displayed a sixfold pattern similar to bulk Co(0001) rather than the threefold pattern of the Cu(111) substrate. The LEED patterns for Fe films thicker than 10 \AA (5 layers) exhibited streaks indicative of the formation of oriented domains with the bcc(110) symmetry of the close-packed Fe(110) surface. This structural transition has also recently been reported in a LEED study of this particular epitaxial system¹⁵ and Auger electron-diffraction measurements.¹⁶

III. BULK ANALOG

A comparison of a 15- \AA (7.5-layer) Co film and a 17- \AA (8.5-layer) Fe film (1 layer=2 \AA) is shown in Fig. 1. These films exhibit similar electronic structure to that of their bulk counterparts:^{17,18} hcp Co(0001) and bcc Fe(110). The band filling of Fe is less than that for Co, so there are more unoccupied electronic states for Fe. For Co, only emission from the uppermost minority-spin $3d$ band is evident. For Fe, some emission from the majority-spin $3d$ band is seen as a peak intersected by the Fermi level. The peak associated with the minority-spin $3d$ band is much broader than the one associated with majority-spin $3d$ band because it has a greater lifetime broadening. The lifetime broadening, which is the imaginary part of the self-energy, increases proportional to the peak energy relative to the Fermi energy¹⁸ E_F so bulk transitions farther from E_F will have greater widths. The image potential surface states, which probe the work function, are the small peaks near the vacuum level, about 4.5 eV above the Fermi energy. Observations of the dependence of the electronic structure on film thickness are discussed below.

hcp Co(0001)

In order to gain some insight for interpretation of the thin-film inverse photoemission data, measurements for

bulk hcp Co(0001) will be presented first. All data were taken with normal incident electrons ($k''=0$). This simplifies the interpretation of inverse photoemission spectra since only a single $3d$ band is observed above E_F . It has minority spin. The surface mesh of hcp Co(0001) has the same symmetry as fcc Cu(111) and a direct comparison can be made of energy bands observed along the surface normal.^{19,20}

Figure 2 shows a set of spectra from Co(0001) taken with varying incident electron energy. The main peak in the spectra is assigned to the uppermost minority-spin $3d$ band.¹⁷ For decreasing initial electron energy, the emission from this $3d$ band decreases because its cross section is reduced due to the dipole selection rule, which forbids the transition at the zone center for the hcp(0001) surface and the zone boundary for the fcc(111) surface. The low-energy spectra also show another feature emerging at the Fermi level, which is the high-energy tail of a surface state of s,p origin that has been previously reported in photoemission.¹⁹ The tick marks show the dispersion of the uppermost minority band to lower energy as the initial energy is changed. The dispersion of the uppermost minority $3d$ band together with corresponding angle-resolved photoemission data²⁰ for the majority $3d$ band is plotted in Fig. 3. The energies of the initial-state upper

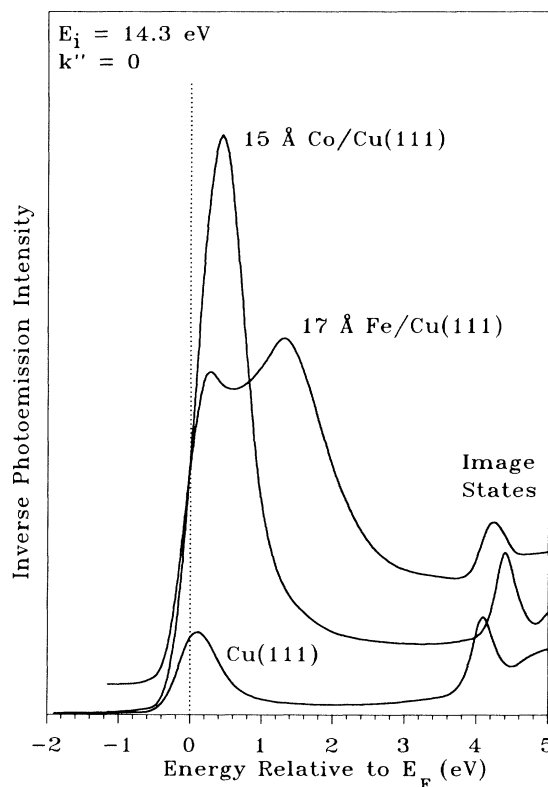


FIG. 1. Inverse photoemission spectra for 15 \AA of Co and 17 \AA of Fe on Cu(111). The prominent peak in each case corresponds to the uppermost minority-spin $3d$ band. This simplifies interpretation of inverse photoemission spectra. The majority band in Fe is clipped by the Fermi level, so the spectrum is indicative of an exchange splitting greater than 1.2 eV.

bound, indicated along the top of Fig. 3, were used to determine the value of crystal momentum. This figure reveals an exchange splitting of 1.1 ± 0.1 eV for the bulk hcp $3d$ band. The corresponding fcc(111) symmetry points are also labeled.

IV. THIN FILMS

For comparison of thin-film fcc Co(111) and fcc Fe(111) data, incident energies of 14.3 and 10.8 eV were chosen. At 14.3 eV the emission from the $3d$ states of the Co (or Fe) is stronger than the s,p -like surface state near the Fermi energy of the Cu substrate.²¹ Although the surface state of the Cu substrate is occupied at $k''=0$, it rapidly disperses²² above E_F for small k'' and is detected due to the finite angular resolution of the spectrometer and momentum broadening by surface imperfections. For 14.3 eV initial energy, the upper band is halfway to the Brillouin-zone boundary, so any shift in energy of the $3d$ band corresponding to a change in the exchange splitting or average band energy should be evident; however, this measurement is relatively insensitive to changes in the bandwidth. For 10.8 eV, perpendicular momentum nearer the fcc zone boundary is probed, so measurements at this initial energy should reveal any band narrowing due to the reduced dimensionality for thinner films.

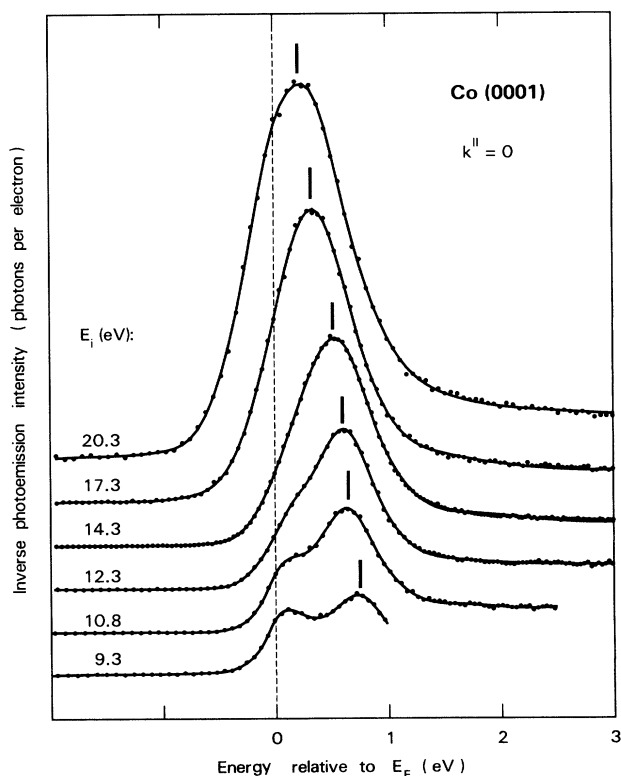


FIG. 2. Energy-dependent inverse photoemission spectra for bulk hcp Co(0001), showing the dispersion of the uppermost minority band along the surface normal. E_i is the incident electron energy. The intensity of the $3d$ band decreases at lower incident energy due to a dipole selection rule that forbids the transition at the hcp Brillouin-zone center.

Difference spectra are also presented with emission from the s,p surface state of the Cu substrate subtracted from the thin-film spectra assuming an exponential attenuation of the incident low-energy electrons in Co and Fe with a mean free path of 5 Å estimated from photoemission results.²³

A. Co films on Cu(111)

Inverse photoemission spectra from Co films as a function of film thickness for an initial electron energy of 14.3 eV are shown in Fig. 4. The energy of the minority $3d$ band of the Co appears shifted down in energy for the thin films. An increase in energy is expected if the monolayer (2 Å) film has an increased magnetic moment. The apparent decrease with decreasing film thickness is due to the overlapping emission from the Cu substrate s,p surface state. The difference spectra (dashed line) reveal the energy of this band to be nearly constant at 0.1 eV below that of the bulk hcp Co(0001). This downward shift is probably due to the small ($\sim 2\%$) expansion of the close-packed surface net. The 2-Å Co film difference spectrum appears to have a slightly lower band energy. However, this could be due to incomplete wetting of the substrate that is not accounted for with a simple exponential attenuation of the substrate s,p surface state emission and/or an underestimated mean free path.^{24,25} Incomplete wetting of the substrate is evident of the image state measurements (Fig. 4 inset). The image state energy for the 2-Å Co film is midway between the Cu(111) and Co(0001) values, which suggests the substrate is not completely covered. The linear increase with initial coverage

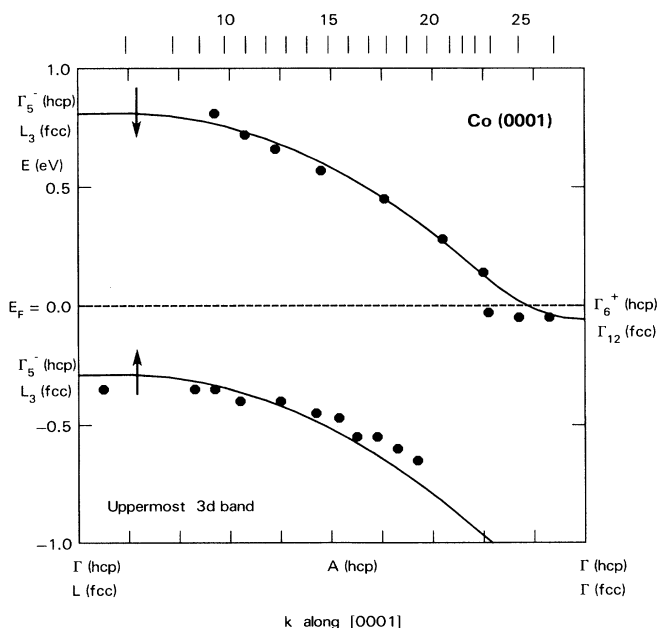


FIG. 3. Band dispersion for the uppermost $3d$ band of bulk hcp Co(0001), including photoemission data from Ref. 19. Note the bulk exchange splitting of 1.1 eV. The corresponding fcc symmetry points are also labeled. The energies of the initial upper-state band are marked along the top, in units of eV.

that the film is complete at 4 \AA (2 monolayers) coverage.

For the 10.8-eV data presented in Fig. 5, the intensity of the emission from the s,p state of the substrate is enhanced relative to the $3d$ states of the film. Both the 2- and 4- \AA films show the relative intensity at the Fermi level is higher than the corresponding 14.3-eV spectra. This is due to the enhanced emission from the s,p surface state of the Cu substrate. The difference spectra that account for this enhanced emission (dashed lines) show the energy of the minority $3d$ band is again 0.1 eV lower than the bulk Cu(0001) $3d$ band. These measurements show ultrathin films of Co on Cu (111) have a bulklike electronic structure with no evidence for any significant band narrowing or a change in the exchange splitting as the film thickness is reduced to a single monolayer that agglomerates to form bilayers on the surface.

A combination of HeI photoemission for a 16- \AA Co film and inverse photoemission measurements for a 15- \AA Co film is shown in Fig. 6. The incident electron energy for the inverse photoemission spectrum is 20.3 eV. This is high enough to minimize any interference from the substrate. For HeI incident light, the photon energy of

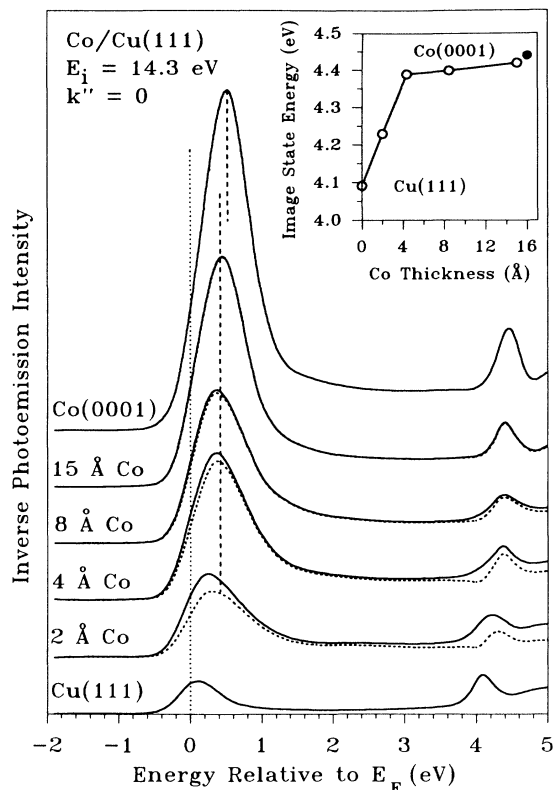


FIG. 4. Thickness-dependent inverse photoemission spectra for Co/Cu(111) for an incident electron energy of 14.3 eV. Difference spectra, produced by subtracting the Cu(111) spectrum multiplied by an exponential attenuation factor with a mean free path for the incident electrons of 5 Å, are shown as dashed lines. A downward shift of the $3d$ band of 0.1 eV is indicated. The inset shows the position of the image potential surface states vs thickness, which indicates an exposure of greater than 2 layers (4 Å) is required to fully cover the surface.

21.2 eV matches the incident electron energy for the inverse photoemission so the same band at nearly the same value of crystal momentum is measured. An exchange splitting of 1.05 ± 0.1 eV is measured, which is slightly lower than the measured bulk Co(0001) value of 1.1 ± 0.1 eV in Fig. 3.

B. Fe films on Cu(111): Transition from fcc(111) to bcc (110)

Inverse photoemission spectra at an incident electron energy of 14.3 eV for Fe films as a function of film thickness is shown in Fig. 7. The energy of the uppermost $3d$ band of the Fe abruptly changes from roughly 0.8 to 1.3 eV as the film thickness is increased, which reflects the change in exchange splitting of low-spin fcc Fe(111) films evolving into a bulklike bcc Fe(110) phase. The arrow indicates the energy of the bcc Fe(110) majority band taken from photoemission results,²⁶ which determines the bulk Fe(110) exchange splitting to be 1.9 ± 0.2 eV. The image state energies are also sensitive to this structural transition (Fig. 7 inset). However, unlike the Co films (Fig. 4), the image state energies of the Fe films evolve more slowly over the first 10 Å but then fall short of the bulk Fe(110) value. This signifies incomplete coverage of the

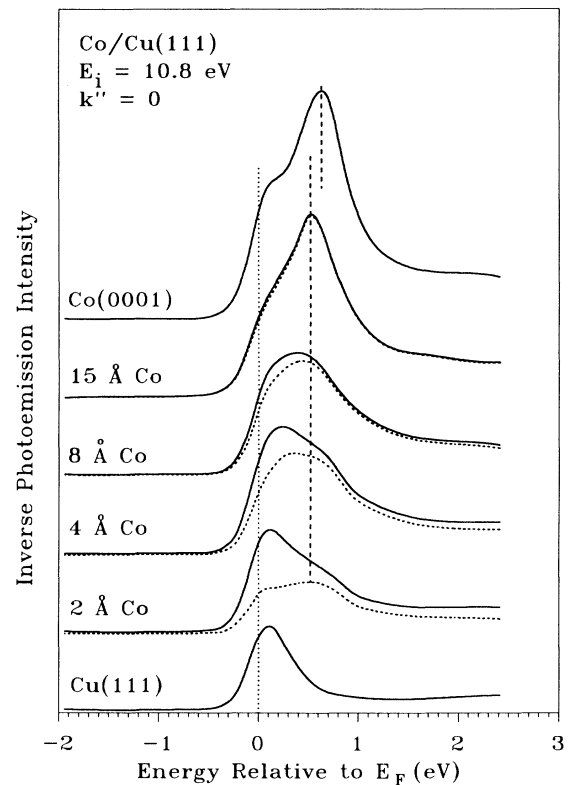


FIG. 5. Thickness-dependent inverse photoemission spectra for Co/Cu(111) for an incident electron energy of 10.8 eV. For thinner films, the peak at the Fermi level is due to enhanced emission from the s,p surface state derived from the Cu(111) substrate. The difference spectra (dashed lines) indicate the $3d$ band is shifted downward in energy by 0.1 eV as in Fig. 4.

surface up to 5 monolayers and/or magnetoelastic strain effects due to the changing magnitude of the spin moment.²⁷ The structural transition could relieve strain in the film by forming facets of bcc Fe(110) with different orientations¹⁵ and associated exposed areas of the substrate, lowering the effective work function of the film.

The 10.8-eV incident energy inverse photoemission spectra for the Fe films are shown in Fig. 8. The energy of the uppermost minority-spin 3*d* band shifts from 0.8 to 1.45 eV, which further illustrates the change in electronic structure associated with the structural transition. The 10-Å film has an electronic structure midway between the two extremes with the minority-spin 3*d* band at about 1.1 eV. The downward shift of the uppermost 3*d* band for thinner films is indicative of a reduced exchange splitting of low-spin fcc Fe films²⁷ on Cu, which is in agreement with recent spin-polarized neutron-scattering measurements.²⁸ The 10-Å Fe film is an intermediate phase, probably a mixture of low-spin fcc (111) domains and bulklike bcc(110) domains.

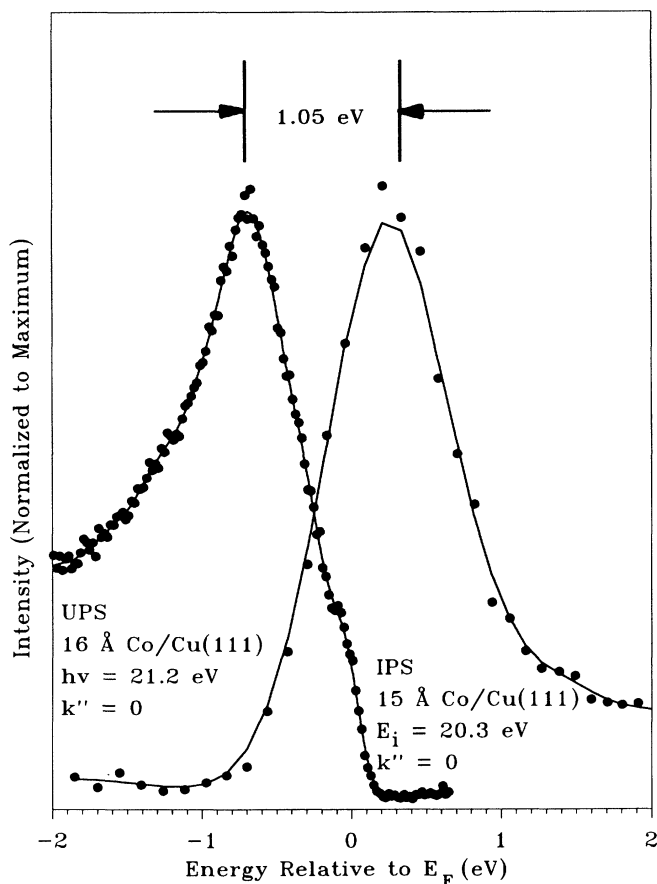


FIG. 6. Photoemission and inverse photoemission spectra for thick Co/Cu(111) films at $k''=0$. The incident electron energy for inverse photoemission of $E_i=20.3$ eV was chosen to probe the same value of perpendicular momentum of the photoemission experiment with $h\nu=21.2$ eV. The measured exchange splitting of 1.05 ± 0.1 eV is shown.

C. Annealing behavior

Surface segregation can be monitored using inverse photoemission by following the energy of the $n=1$ image state, which is sensitive to changes in the work function. For Co deposition on the Cu(111), the image state shifts upward in energy, which reflects the work function increase for Co(0001) as opposed to Cu(111). No change in the image state energy for the 15-Å film upon annealing to 150°C is observed, which shows Cu surface segregation had not occurred. A sixfold LEED pattern with spots as sharp as the substrate LEED pattern and a low background intensity was obtained for the annealed 15-Å film, indicating that annealing improves the long-range order of the film. This behavior is supported by Auger electron spectroscopy measurements,²⁹ which set a lower limit on the segregation temperature of 250°C.

For Fe on Cu(111), as shown in Fig. 9, annealing a 17-Å film to 200°C has a noticeable effect on the inverse photoemission spectrum. Cu segregation is more pronounced for this system since the Fe agglomerates on the surface to form facets¹⁵ with associated exposed areas of the substrate. For the annealed film, the intensity at the Fermi energy is significantly enhanced and the image

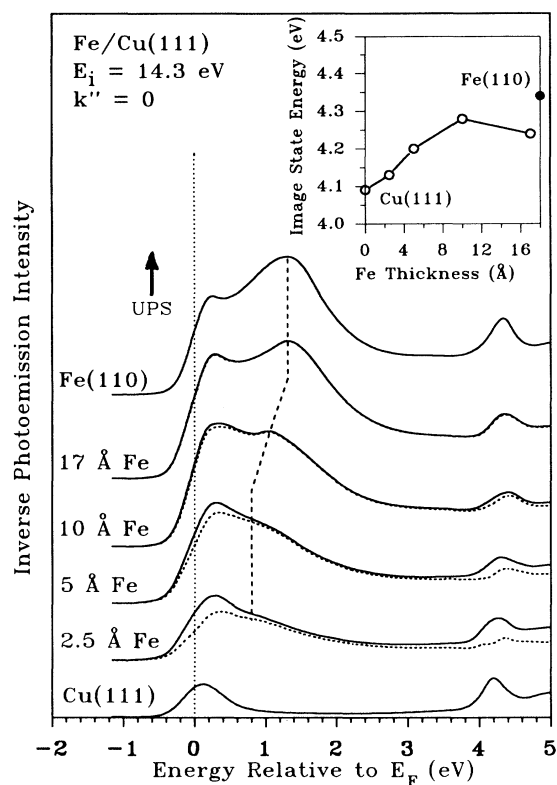


FIG. 7. Thickness-dependent inverse photoemission spectra for Fe/Cu(111) at an incident electron energy of 14.3 eV. The position of the bulk bcc Fe(110) majority band taken from Ref. 26 is indicated by the spin-up arrow. The difference spectra, produced as in Fig. 4, reveal the change in electronic structure associated with the structural transition at 10-Å film thickness. The inset shows the energies of the image states have a complicated behavior due to the structural transition.

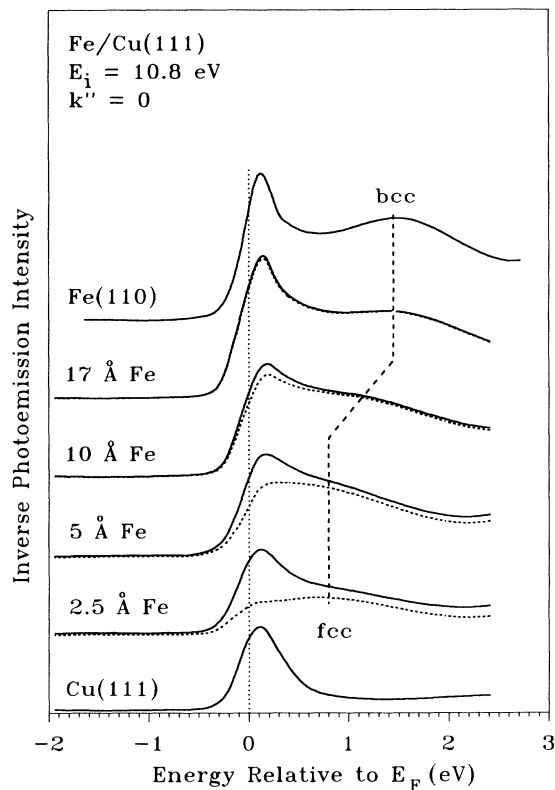


FIG. 8. Thickness-dependent inverse photoemission spectra for Fe/Cu(111) at an incident electron energy of 10.8 eV. The difference spectra (dashed lines) reveal the thin films of fcc Fe(111) are in the low-spin ferromagnetic phase with an associated downward shift of approximately 0.6 eV for the uppermost minority-spin 3d band.

state energy is the same at the Cu substrate. This indicates a layer of Cu segregates to the surface upon annealing. The sharpness of the transition at the Fermi energy is strongly suggestive of a Cu(111) monolayer *s,p* surface state. This monolayer state is possibly related to the bulk Cu(111) *s,p* surface state, which could be shifted up in energy by a corresponding difference in the inner potential of the underlying Fe.

V. SUMMARY

The growth and evolution with thickness of the uppermost unoccupied minority-spin 3d state above the Fermi energy of Co and Fe layers deposited on Cu(111) surfaces were observed with inverse photoemission. Band narrowing or enhanced exchange splitting in the monolayer limit for Co films were not observed, which is supported by theoretical predictions⁷ of no enhancement in the ex-

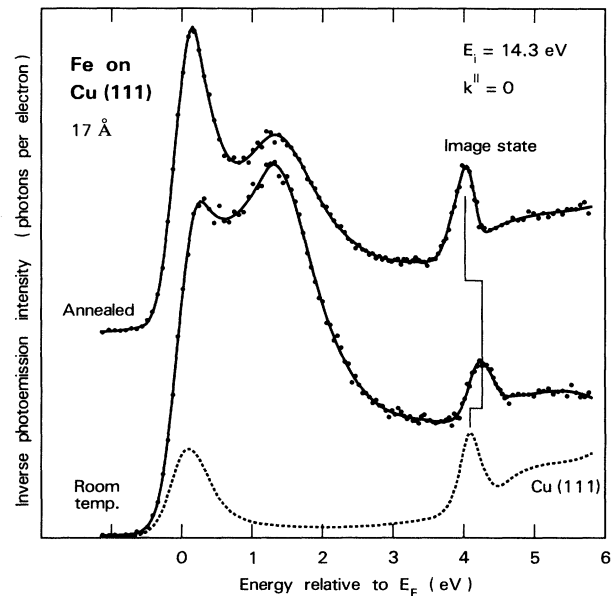


FIG. 9. Effect of annealing a 17-Å Fe film on Cu(111) to 200°C. The annealed film exhibits a sharp *s,p*-like surface state near the Fermi energy associated with Cu surface segregation. The image potential surface state shifts in energy back to the original Cu(111) energy, confirming the presence of a Cu overlayer on the surface.

change splitting for a monolayer of Co on Cu(111). However, the absence of an enhancement could be due to incomplete wetting of the substrate for the Co film. Combining inverse photoemission and photoemission measurements reveal an exchange splitting in Co/Cu(111) films of $\Delta E_{\text{ex}} = 1.05 \pm 0.1$ eV, similar to the measured exchange splitting in bulk hcp Co(0001) of $\Delta E_{\text{ex}} = 1.1 \pm 0.1$ eV. For Fe on Cu(111), a corresponding change in the electronic structure was measured for the low-spin fcc(111) to bulklike bcc(110) structural transition at 5 layers, with a decrease in the energy of the uppermost minority-spin 3d band for thinner fcc Fe(111) films. In addition, inverse photoemission has been shown to be a sensitive probe of the morphological composition of the surfaces of these films by observing changes in the energies of the image potential surface states upon annealing these metastable structures.

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