Quantum-well states and the short period of oscillation in Cu/Co(001) multilayers

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We report a combined photoemission and inverse-photoemission study of the quantum-well states formed in copper thin films deposited on a fcc Co(001) substrate. In particular, we examine the k point that would correspond to the Fermi-surface crossing that would allow the quantum-well states to mediate the short period of exchange coupling in the associated magnetic multilayers. The states do not display a strong intensity in photoemission, an effect that we attribute to interfacial roughness. [S0163-1829(96)11348-5]

There is currently considerable interest in the properties of magnetic multilayers. These systems show an oscillatory exchange coupling between adjacent ferromagnetic layers[1] and a giant magnetoresistance dependent on the thickness of the intervening nonmagnetic layer.[2] In the case of noble metal films deposited on ferromagnetic substrates, it has been shown that ''quantum-well'' (QW) states reflecting the finite layer thickness exist within the film and that these states are highly spin polarized.[3–5] The latter spin polarization arises because of the marked asymmetry in the spin-dependent reflectivities of the states at the interface with the ferromagnetic layer.[6] As the thickness of the noble metal film increases, the QW states sample the Fermi surface with a frequency identical to that observed for the exchange coupling in the associated multilayers.[4,5,7,8] These observations taken together, the spin polarization and the frequency of Fermi surface crossings, lend support to the idea that it is the quantum-well states that mediate the oscillatory coupling in the magnetic multilayers[8,9] and indeed theoretical studies indicate that the relative strength of the oscillatory coupling may be directly related to the reflectivity of the quantum-well states at the interface.[6,10]

In the case of Cu/Co(100) multilayers, several theories predict more than one period of oscillation for coupling in the ⟨001⟩ direction.[11] These two periods, corresponding to spanning vectors at the belly and neck of the copper Fermi surface ''dog bone,'' have been observed experimentally in several studies[12] although in general the long period of oscillation has proven easier to observe.[13] Contrary to such experimental observations, recent theoretical studies suggest that the short period oscillation should in fact dominate the coupling.[14,15] This prediction reflects the observation that away from the center of the Brillouin zone at the point corresponding to the neck of the Cu Fermi surface dog bone, the minority spin hybridization band gap in the Co substrate spans the Fermi surface. This differs from the center of the zone where the top of the minority spin gap lies 0.5 eV below the Fermi level. It is the reflectivity in the vicinity of these gaps that determines the degree of confinement of the quantum-well states. We may therefore anticipate that out in the zone the minority spin quantum-well states will be more ''confined'' within the copper layer and hence the related short period oscillations should be stronger. A recent experimental study has, in fact, shown that in certain samples the short oscillation period does indeed dominate the coupling.[16]

In the present paper we report the results of a search for the quantum-well states that should lead to the short oscillation period. Using photoemission and inverse photoemission, we examine the development of quantum-well states in copper films deposited on a Co(001) substrate at the k point corresponding to $k_{\parallel}=0.765\overline{\Gamma}\,\overline{X}$. The states display two-dimensional character in that they do not disperse with $k_{\perp}$. However, when compared with the states at the center of the zone, they do not in fact show the dramatic enhancement in their intensity that we might expect on the basis of increased confinement. We associate the lack of intensity with interfacial roughness.[15]

The angle-resolved photoemission studies were carried out on the U5 undulator beam line at the National Synchrotron Light Source.[17] The photoelectrons are analyzed in energy and momentum with a commercial hemispherical analyzer. The total energy resolution (spectrometer and monochromator) and the angular resolution are ~300 meV and ±1°, respectively. The momentum-resolved inverse photoemission studies were carried out using an ultraviolet spectrometer and an electron source that have been described in detail elsewhere.[18] This instrument has the capability of examining the unoccupied electronic structure at points far removed from the center of the zone. It is therefore ideal for the present experiment. The energy and angular resolution of the instrument are of the order of 300 meV and ±2°, respectively.

Cu films of various thickness were deposited onto a fcc Co(001) film at room temperature in a base pressure of 3×10^{-10} Torr. The fcc Co film was prepared by epitaxially growing Co onto a freshly cleaned Cu(001) substrate to a thickness of approximately 20 monolayers (ML's). Elsewhere we have shown that certainly for the states at the center of the zone, a Co thickness of three to four layers is required to completely confine the sp-derived quantum well states.[19] In the inverse photoemission experiment the cleanliness and the surface order of all films were monitored using Auger electron emission (AES) and low-energy electron diffraction, respectively. For the photoemission studies the pho-

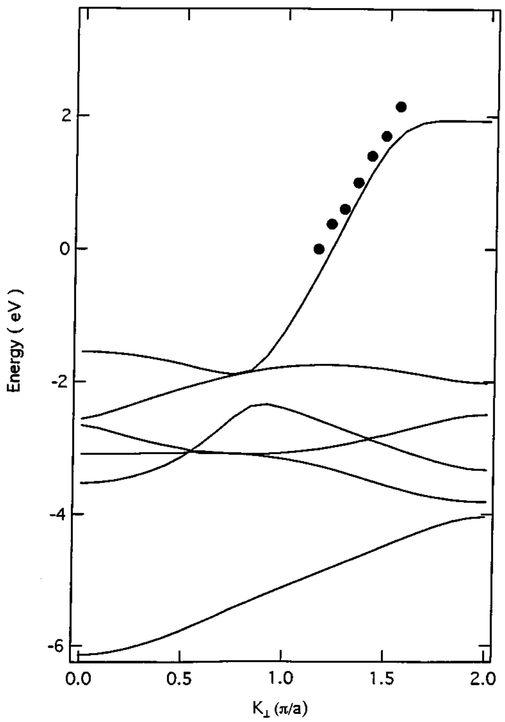

FIG. 1. Tight-binding calculation of the copper band structure as a function of $k_\perp$ for $k_\parallel=0.765\ \overline{\Gamma}\,\overline{X}$. The filled circles indicate the experimentally determined dispersion of band 6 as derived from the spectra of Fig. 3(a).

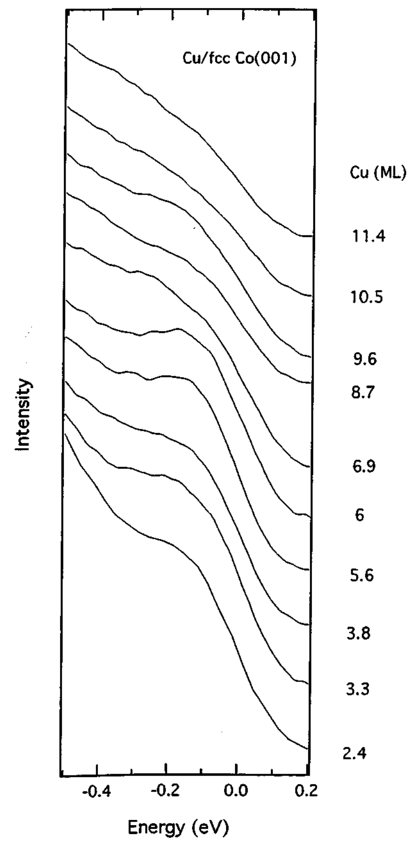

FIG. 2. Photoemission spectra recorded from copper films as a function of thickness. The photon energy is 21 eV and the angle of emission corresponds to a $k_\parallel=0.765\overline{\Gamma}\,\overline{X}$ for the Fermi level.

toemission itself was used as a monitor of cleanliness. The relative thicknesses were monitored with a quartz crystal and a quadrupole mass spectrometer. The absolute thickness was calibrated via the attenuation of the photoelectron and Auger electron intensities.

We first search for quantization effects in the Cu thin films by examining the occupied states immediately below the Fermi level with photoemission. In Fig. 1 we show the calculated electronic structure for bulk copper as a function of k perpendicular at a point corresponding to k parallel equal to $0.765\ \overline{\Gamma}\ \overline{X}$. These calculated bands are derived from a tight-binding fit to a first-principles calculated band structure.[20] At k perpendicular corresponding to approximately $1.2\pi/a$, the calculation indicates an s-p band crossing the Fermi level. A Fermi surface crossing at this point will correspond to a spanning vector q of length $1.6\pi/a$ and therefore an oscillation period for the related multilayers of 2.5 ML's. As noted earlier, a substrate minority spin gap exists in the vicinity of the Fermi level at this point in the Brillouin zone and there should therefore be strong confinement of any QW states derived from the Cu s-p band.

In Fig. 2 we show a series of photoemission spectra recorded from different thickness Cu films deposited on the Co substrate as indicated. The spectra are recorded at an emission angle such that the Fermi edge corresponds to a $k_\parallel=0.765\overline{\Gamma}\ \overline{X}$. It is evident from the figure that a small peak appears in the vicinity of the Fermi level at a thickness of approximately six monolayers. A much weaker peak is also observed at the Fermi level for a thickness of 9.6 ML's. These observations are consistent with the predicted short period of oscillation of approximately three layers. However, we note that the intensity of the state is not particularly strong even though the states fall within a substrate band gap, and furthermore the photon energy has been selected to maximize the cross section.

It should be remembered that in an off-normal photoemission spectrum, such as those shown in Fig. 2, only the emission immediately at the Fermi level corresponds to the chosen $k_\parallel$. We therefore continue our study by examining the properties of the quantum-well states in the unoccupied region. In Fig. 3(a) we show a series of inverse photoemission spectra recorded from a clean Cu(001) sample and in Fig. 3(b) a second series of spectra recorded from a 10-ML Cu film deposited on an fcc Co(001) substrate as a function of the incident electron kinetic energy. For each spectrum the incident angle of the electrons was selected to maintain $k_\parallel=0.765\ \overline{\Gamma}\,\overline{X}$ in the initial state. Because $k_\parallel$ is determined by the initial state, all binding energies in each spectrum now correspond to the same $k_\parallel$.

Figure 3(a) clearly shows a state dispersing down to and through the Fermi level as the electron energy increases from 14 to 22 eV. In Fig. 1 we compare these experimental observations with the calculated electronic structure. The $k_\perp$ for each experimental point is determined from the equality

$$k_\perp=\left[\frac{2m}{\hbar^2}(E_{\rm KE}-V_0)-k_\parallel^2\right]^{1/2}, \tag{1}$$

where $E_{\rm KE}$ is the kinetic energy of the incident electrons relative to the Fermi level and V_0 represents the inner poten-

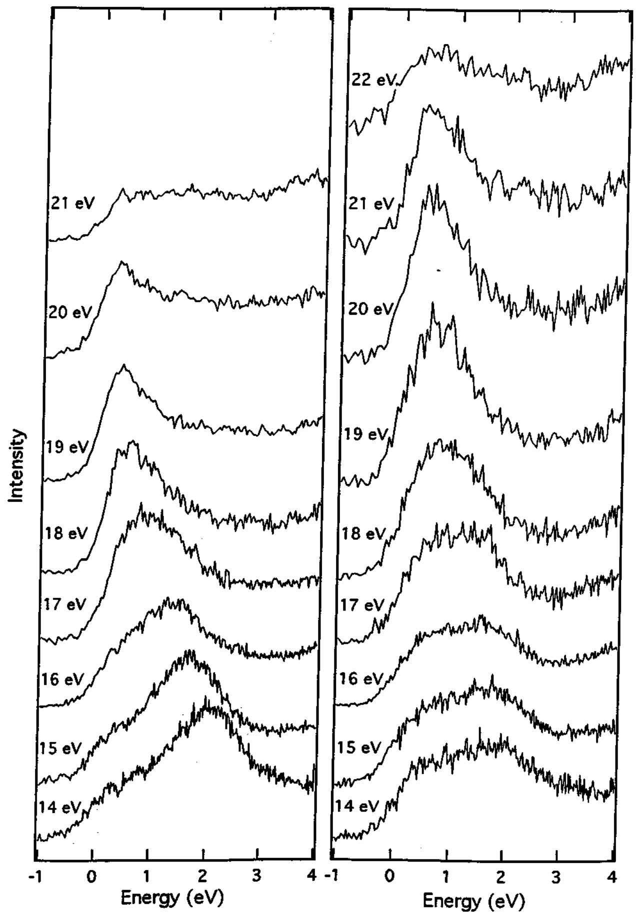

FIG. 3. (a) Inverse-photoemission spectra recorded from a Cu(001) crystal as a function of the incident electron beam energy as indicated. The incident angle is adjusted for each spectra to maintain $k_{\|}=0.765\ \overline{\Gamma}\ \overline{X}$. (b) As in (a), but now for a copper film of thickness 10 ML as determined by Auger electron spectroscopy.

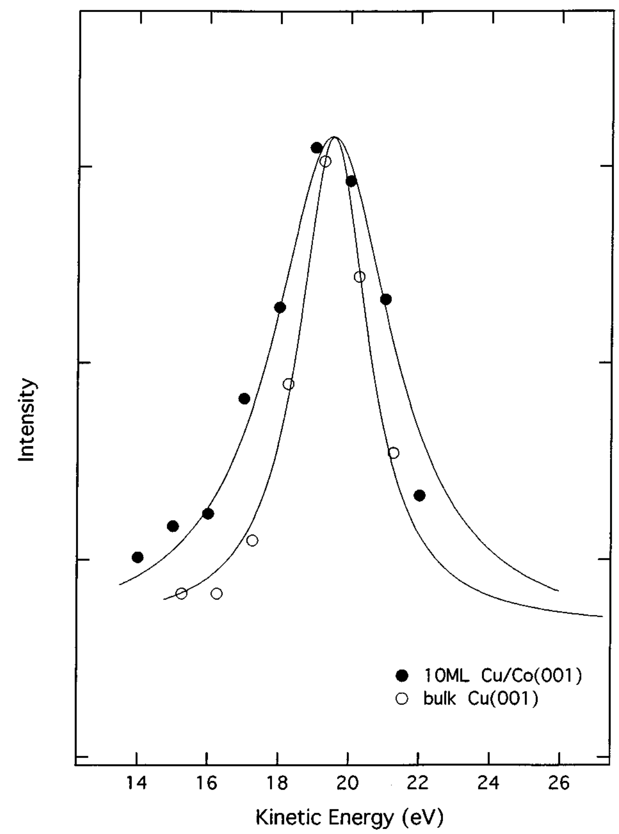

FIG. 4. The intensities as a function of incident beam energy measured 0.6 eV above the Fermi level for the bulk spectra, Fig. 3(a) and the copper film, Fig. 3(b). The solid lines represent Lorentzian fits to the experimental data.

tial. From the comparison between experiment and calculation it is clear that we are examining the electronic structure at the appropriate point in the Brillouin zone.

The quantization effects in the thin film are evident in Fig. 3(b). Unlike the spectra in Fig. 3(a), the spectra in Fig. 3(b) now show a number of peaks which do not disperse as the photon energy is changed. These peaks are at binding energies of 0.6, 1.5, and 1.9 eV above the Fermi level with the nondispersion most evident for the state immediately above the Fermi level. Indeed examination of Fig. 3(b) shows that this latter state persists in the spectra even as the incident electron energy is increased to the higher energies for which the bulk transition would no longer exist. The lack of dispersion in the binding energy is consistent with localized two-dimensional states and, hence, the quantization of the associated bulk bands. Our tight-binding analysis places the top of the minority spin band gap in the Co substrate approximately 1.0 eV above the Fermi level. Consequently, because the quantum-well states at 1.5 and 1.9 eV above the Fermi level no longer fall within this gap they are less confined and never gain the same intensity as the state immediately above the Fermi level.

In Fig. 4 we compare the intensity of emission as a function of incident electron beam energy for the quantum-well state 0.6 eV above the Fermi level in Fig. 3(b) with the intensity of the bulk Cu spectra in Fig. 3(a) at a point 0.6 eV above the Fermi level. In each case a representative background has been subtracted. In the thin film the wave function of the QW state retains many of the characteristics of the parent bulk band. The intensity therefore peaks at an energy close to that of the appropriate direct transition in bulk copper. Such effects are well known in studies of photoexcitation from surface states.[21]

The quantum-well state represents a constant final state (CFS) as the initial state is varied by changing the incident electron beam energy. Smith *et al.*[22] have considered in considerable detail the linewidths that we might anticipate in such studies. In particular, they examine the linewidths of the constant initial state (CIS) mode in photoemission for normal and off-normal emission. The CFS mode in inverse photoemission is simply related to the CIS of photoemission through time reversal. Smith *et al.* restrict their analysis to the case of emission at constant polar angle. However in the present studies the polar angle is varied to maintain constant $k_{\|}$ as the incident electron beam energy is varied. It is a trivial extension of their analysis to show that the measured linewidth Γ_m in an inverse photoemission CFS study at constant $k_{\|}$ is given by

$$\frac{\Gamma_m}{|\nu_i|}=\frac{\Gamma_i}{|\nu_i|}+\frac{\Gamma_f}{|\nu_f|}. \tag{2}$$

Γ_i and Γ_f represent the inverse lifetime of the initial and final states, respectively, and ν_i and ν_f are the perpendicular group velocities in the related bands. A more complete analysis will include an angular-dependent term that accounts for the finite angular spread of the incident beam.[23]

We first consider the intensity variation or cross-section scan obtained in the bulk case. Eastman *et al.*[24] have previously shown that such a method provides a good measurement of the lifetime in the final state of photoemission or in the present case the initial state in inverse photoemission. Because of the proximity of the final state to the Fermi level we follow Eastman *et al.* and approximate Γ_f by zero. Then from Eq. (2) the lifetime in the initial state is obtained from $\Gamma_i = \Gamma_m^{\text{bulk}}$. Fitting the intensities associated with the bulk data in Fig. 3 with a Lorenztian leads to a full width at half maximum (FWHM) or inverse lifetime in excellent agreement with data for noble metals published elsewhere.[25] In fact, the earlier data were recorded in a photoemission study[25] with almost identical angular acceptance to the present study. Turning to the intensity variation for the QW state we first note that the width of the thin film is sufficiently large that we may consider the initial state to be essentially identical for both the bulk studies and the thin-film data. If we make the simple assumption that the initial state is represented by a plane wave then we may assume that the intensity I_0 of the QW state will reflect the width of the well W such that[26]

$$I_0 \propto \left| \frac{\sin(k-k')W/2}{(k-k')W/2} \right|^2, \tag{3}$$

where k and k' represent wave vectors for the initial and final states. Thus the width of the intensity peak is inversely related to the width of the well and in the limit that the well becomes infinitely thick (the bulk case), I_0 reduces to a δ function $\delta(k-k')$. In fact, in the present case the inverse of the difference in the Lorentzian FWHM for the bulk and QW state cross sections is close to the width of the well as determined by AES. However such a correlation needs to be examined further in more detailed studies.

We now consider the width of the QW state itself in Fig. 3(b). Examining the spectra at the higher incident energies where there is no interference from adjacent QW states we find that the width of the QW state at 0.6 eV above the Fermi level is approximately 1.0 eV. Interestingly we note that because we are using a spectrometer the individual spectra in Fig. 3(b) represent CIS spectra in that the initial state is constant and each point in the spectrum represents a different photon energy. Smith *et al.*[22] have again considered the widths that would be anticipated in such a spectrum. If we assume no dispersion of the QW state with k perpendicular as evidenced by the lack of dispersion in Fig. 3(b) then we can show that the measured linewidth of the QW state Γ_m is given by

$$\Gamma_m = \Gamma_f^{\text{QW}} + 2\hbar \nu_{\parallel} \delta k_{\parallel} \tag{4}$$

where Γ_f^{QW} represents the inverse lifetime of the QW state, $\nu_{\parallel}$ represents the group velocity parallel to the surface, and $\delta k_{\parallel}$ reflects the finite angular resolution.

In a recent theoretical study of copper films deposited on Co(001), Gelderen *et al.*[27] have calculated the widths of the QW states found in the copper films at the center of the zone. They find that the states have an infinitesimal width when they fall within the substrate band gap but that they are extremely broad when they move out of the gap to become resonances. Thus in our study, if the QW state is within the gap, Γ_f^{QW} is effectively zero again and Γ_m will be determined entirely by the energy and angular resolution of our instrument. From the width of the Fermi edge we estimate that at the higher incident energies the energy resolution is approximately 0.5 eV. The angular term in Eq. (4) reflects the group velocity of the state parallel to the surface. Comparison with calculations for bulk copper suggest that at this particular point in the zone the rate of dispersion of the state is approximately 0.7 eV Å.[28] We are therefore still left with a broadening of the order of 0.2 eV. We suggest that this additional broadening reflects scattering parallel to the interface. Momentum broadening perpendicular to the interface results in an even larger width because the rate of dispersion of the associated Cu bulk band perpendicular to the interface is an order of magnitude larger than that parallel to the interface.[28] Further, momentum broadening perpendicular to the interface would also broaden QW states at the center of the zone. However all studies of the latter states[4,5,19] appear to indicate that their experimental widths are limited solely by the experimental resolution.

A broadening of 0.2 eV represents a mean scattering length of 3.5 Å parallel to the film. This appears anomolously small and may indicate interface mixing. However we note that if we have underestimated the broadening due to finite angular resolution the mean scattering length that we would obtain would be larger. A detailed study by Meinel and co-workers[29] has also examined the influence of interface and surface roughness on the intensity of quantum-well states observed in thin films. They found that for states at the center of the zone roughness at either interface can easily lead to reduction in the peak intensity by a factor of 40% or more. Momentum broadening as a result of impurities or step imperfections has also been studied extensively in the case of surface states.[30]

In summary, we have demonstrated the existence of quantum-well states in copper at the Brillouin-zone point corresponding to the neck of the Fermi surface dog bone. We have shown that the intensity of the QW states reflects the direct transitions observed for the parent bulk bands. However in our photoemission study the states never show the intensity that we might anticipate for good confinement in the vicinity of a substrate band gap. We suggest that this reflects the interfacial roughness but note that scattering at any impurity center will couple the quantum-well states to other momenta which will allow reduced confinement. An understanding of such effects would benefit from further study with higher energy and more importantly higher momentum resolution. Indeed studies of the linewidths of the QW states may offer an interesting probe of the roughness of buried interfaces.

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