Experimental and Model Theoretical Dispersions of Unoccupied Metallic Quantum Well States in the Cu/fccCo/Cu(100) System

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The dispersion with parallel momentum of unoccupied metallic quantum well states in the Cu/fccCo/Cu(100) system has been measured using inverse photoemission and modeled using a phase analysis approach. A flat band near the Fermi level is observed near the neck of the Cu Fermi surface along the $\overline{\Gamma X}$ direction. Appearance of the state coincides with a hybridization gap in the minority spin bands of the underlying Co and its influence on the phase and strength of the short period magnetic coupling in this and similar systems is discussed. [S0031-9007(98)05319-8]

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In many metallic multilayer systems, the magnetic coupling of ferromagnetic (FM) layers oscillates between parallel and antiparallel orientations as a function of nonmagnetic (NM) spacer layer thickness [1–5]. The presence of antiparallel coupling gives rise to the so-called giant magnetoresistance (GMR) effect. The periodicity of the coupling is thought to be determined by the extremal points of the NM layer's Fermi surface in the direction perpendicular to the layers [6–10], and the strength of coupling depends upon how wide a range of states near these critical points can participate. In Cu/fccCo/Cu(100) multilayers, an important model system, there is a long period [5.6 monolayer (ML)] coupling associated with the Fermi surface belly and a short period (2.6 ML) associated with the Fermi surface neck. Magnetic measurements [4,5] show that high quality multilayers exhibit these two periods of oscillation, that coupling is stronger for the short period oscillation, and that the phase (as a function of Cu thickness) and strength of the short period interaction depends on the identity of the FM layer.

The magnetic coupling between FM layers is associated with quantum size effects that modify the electronic structure of the Cu layer. Interface scattering produces metallic quantum well (MQW) states that move to lower binding energy with increasing Cu thickness and modulate the magnetic coupling as they pass through the Fermi level (E_F) [6,9,11–15]. Recently, MQW states have been observed crossing E_F in photoemission and inverse photoemission experiments [11–14,16,17], but these studies have focused either on specific points in the 2-dimensional Brillouin zone (2DBZ) [11–14,16,17], or on the dispersion of MQW states away from E_F [16]. However, the (100) surfaces of the fcc FM 3*d* transition metals have projected band gaps at E_F near the neck of the Cu Fermi surface so that, in the region of the 2DBZ responsible for short period coupling, the dispersion of the Cu MQW states is strongly influenced by the FM layer. These factors will directly affect the strength of magnetic coupling, the phase of magnetic oscillations, and will depend systematically upon the identity of the FM layer.

In this paper we present inverse photoemission (IPE) measurements of the dispersion with parallel momentum ($k_{\|}$) of metallic quantum well states observed in 2, 3, and 4 ML thick Cu overlayers on a thin fccCo film grown on a Cu(100) substrate. We interpret these results using a phase accumulation calculation [18–22]. We observe a Cu MQW state that exhibits a flat dispersion near the neck of the Fermi surface. The flat dispersion means that at thicknesses where the MQW state is near E_F, the film has many states nearby in $k_{\|}$ to contribute to the short period coupling. The model calculation correctly predicts the periodicity of Fermi level crossings, shows that the flat dispersion is caused by the hybridization gap in the minority spin states of the underlying Co, and suggests how location of this gap in the FM layer can affect the strength and phase of the short period magnetic coupling.

The inverse photoemission measurements were performed in the isochromat mode using a spectrometer that has been described in detail elsewhere [23]. An electron gun of the Stoffel-Johnson design [24] directed a well-collimated, monoenergetic electron beam onto the sample, and the incident electron angle was varied along the $\overline{\Gamma X}$ azimuth. Photons of energy 9.5 ± 0.2 eV that were generated by radiative decay of these electrons were collected using a Geiger-Müller tube using a SrF_2 entrance window. Spectra were obtained by monitoring the 9.5 eV emission as a function of incident electron energy, which ranged from $\sim$4–12 eV. The spectrometer also contained low energy electron diffraction (LEED), Auger electron spectroscopy, independent thermal evaporation sources for Cu and Co, a quartz crystal microbalance (QCM), and reflection high energy electron diffraction (RHEED).

The films examined here were prepared *in situ* on a Cu(100) single crystal that was cut and polished to within 0.5° of its high symmetry direction. Atomically clean and well ordered Cu(100) surfaces were obtained by a sequence of Ar^+ ion sputter and 700 K anneal cycles. After $\sim$20 ML of Cu homoepitaxy, a 5 ML fccCo film was deposited on the Cu(100) surface. Finally, Cu films of integer ML thickness were deposited at room

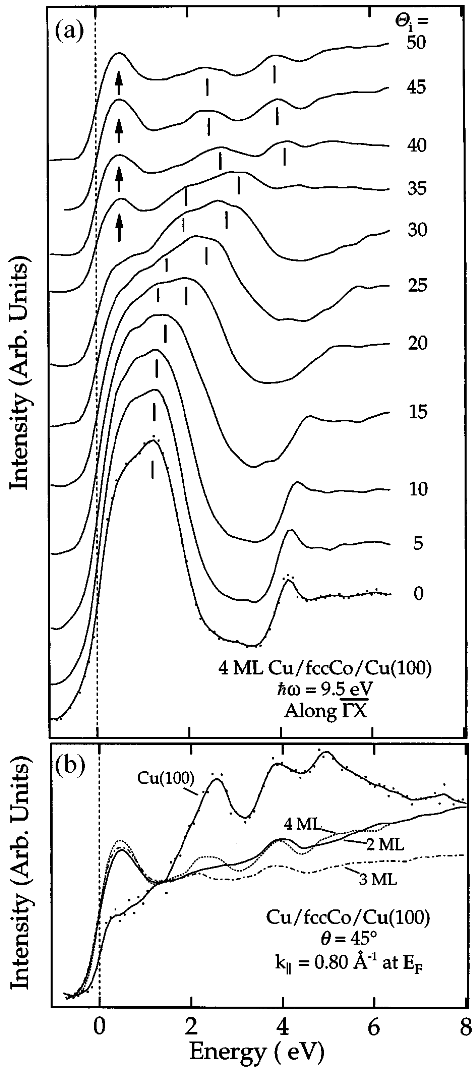

FIG. 1. (a) Inverse photoemission spectra from 4ML Cu/fccCo/Cu(100) as a function of incident angle along the $\overline{\Gamma X}$ direction of the 2D Brillouin zone. The tick marks indicate the MQW levels. The arrows indicate a slowly dispersing feature near E_F that emerges near the neck of the Cu Fermi surface. (b) Inverse photoemission spectra obtained at 45° for different film thicknesses normalized to the intensity minimum at 1.5 eV.

temperature with no further treatment. The QCM, and hence the metal deposition rates, was calibrated to an accuracy of ~10% using Rutherford backscattering. Lack of significant interdiffusion, even for Cu films in the 1–2 ML range, was confirmed by thermal desorption of CO where a single peak at ~170 K was observed with no further desorption at higher temperatures as expected for CO bonded to Co [25]. In addition, the sharp, dispersing image state of the Cu films, as well as the film's LEED and RHEED patterns, were virtually identical to those of Cu(100), indicating flat overlayer growth.

Normal incidence inverse photoemission spectra from a series of Cu thin films of different thickness exhibit [26] MQW levels and Fermi level intensity oscillations in good agreement with earlier measurements [11,12,27]. A series of inverse photoemission spectra obtained as a function of incident angle along the $\overline{\Gamma X}$ azimuth of the 4 ML Cu/fccCo/Cu(100) system is shown in Fig. 1(a). As the polar angle increases, the MQW level initially disperses to higher energy. Near $\theta = 2\theta°$, the MQW feature appears to split, and at larger angles the spectra show an even richer structure. Of particular interest here, however, is the feature near 0.5 eV that first appears near $\theta_i = 35°$. This state remains strong for all larger angles and stays at approximately the same energy as $k_\parallel$ increases. Figure 1(b) shows spectra from the three different Cu film thicknesses for $\theta_i = 45°$, approaching the neck of the Cu Fermi surface. As has been seen in previous photoemission studies [16], the intensity near E_F changes with film thickness. We associate this feature with an MQW state in the Cu film that occurs near the neck of the Fermi surface.

To understand the energy-momentum behavior of these Cu-MQW states, we have modeled the Cu/fccCo(100) system using the phase analysis scheme developed by Smith [18–20,22]. Here we extend the model to finite parallel momentum [19,20] The general idea is that an MQW level is predicted when the total phase accumulated by an electron during a round trip within the Cu film is equal to an integer times 2π. This can be written alternatively as

$$\Delta\phi_B + \Delta\phi_C + 2mk_\perp a = 2\pi n\,, \qquad (1a)$$

$$2mk_\perp a = 2\pi n - \Delta\phi_B - \Delta\phi_C\,, \qquad (1b)$$

where m and n are integers, $k_\perp$ is the perpendicular momentum in the Cu film, a is the Cu monolayer thickness, while $\Delta\phi_B$ and $\Delta\phi_C$ represent the phase shift upon scattering from the surface barrier and Co substrate, respectively. Equation (1b) separates phase accumulation in the film from phase changes upon scattering at the interfaces. In this model, it is convenient to enumerate MQW states by the integer $\nu = m - n$, which relates the nodal structure of the state to the layer thickness. To predict levels away from the center of the SBZ, these quantities must be evaluated at the $k_\parallel$ of interest. The value of $k_\perp$ as a function of energy in the film was calculated using a combined interpolation scheme [28–30] for the bulk Cu band structure.

We model the surface barrier by the image potential for which the scattering properties are well known and, as pointed out in Refs. [18,21] are very similar to those of the empirically determined surface barrier in the energy range of interest. The phase shift from scattering at the surface is therefore given by [19,20]

$$\Delta\phi_B/\pi = \left[\frac{3.4(\mathrm{eV})}{E_\mathrm{V} - \left(E - \hbar^2 k_\parallel^2/2m\right)}\right], \qquad (2)$$

where E_V is the vacuum level. Here, we have included the effect of finite $k_\parallel$ by recognizing that only the part of the energy associated with the momentum perpendicular to the surface is relevant.

The phase shift upon scattering at the Cu/fccCo(100) interface can in principle be modeled accurately with knowledge of the fccCo band structure [10]. In general, however, the phase shift is a slowly varying function of energy, except when a band gap is traversed where it advances by π. In keeping with the spirit of Ref. [22], we assume that the phase shift from the Cu/fccCo interface is constant at energies where the bulk bands are encountered, and is modeled by the functional form

$$\Delta\phi_C = 2\arcsin[(E - E_L)/(E_U - E_L)]^{1/2} - \pi \quad (3)$$

when a band gap is crossed. Here, E_U and E_L represent the upper and lower edges of the Co band gap, respectively. The location of the band gaps in the fccCo band structure, projected into the (100) surface, was determined by a tight binding calculation [31].

In Fig. 2(a) we plot a series of curves showing the energy as a function of accumulated phase at $k_{\|} = 0.98$ Å^{-1}, which is approximately the neck of the Fermi surface. The dashed curves show the phase accumulated in the Cu well for different thickness films, and thin solid curves account for the phase shift from the interfaces. The rapid phase shift between -0.15 and 0.75 eV is the contribution from $\Delta\phi_C$ as the Co band gap is traversed. At the intersections of these two sets of curves, the model predicts an MQW level. Note that levels of successively higher values of ν pass through the Fermi level at regular intervals. At this $k_{\|}$, the model predicts a periodicity of 2.7 ML for the Fermi level crossings of the MQW states, in very good agreement with currently available photoemission data [16], as well as the predicted [7–10] and observed [4,5] short period for magnetic coupling in these systems.

A comparison of the MQW level dispersions predicted by the phase analysis model (lines) to those measured in the IPE experiments (symbols) is shown in Fig. 3. The projected band gaps of Cu(100) and fccCo(100) [31] are also shown. The rapidly dispersing feature at high energy is reasonably well described by the $\nu = 0$ state, although its energy is overestimated by the phase analysis model. A similar overestimate was made in a previous application of this model [22], and is most likely caused its simplicity [32]. The second dispersing level, indicated by the circles in Fig. 3, is most likely related to the $\nu = 1$ state of the 4 ML film. Of primary interest here is the feature near the Fermi level that is observed on all three films for $k_{\|} > 0.6$ Å^{-1}. The level is associated with the $\nu = 1$ MQW state for the 2 and 3 ML films, while for the 4 ML film it is the $\nu = 2$ level that is near the Fermi level. In each case, the phase analysis model predicts that the level has a region of flat dispersion while it is near the neck of the Fermi surface owing to the hybridization gap in the minority Co states. The effect of the Co band gap on the film dispersion can be understood from the phase crossings of Fig. 2(b). The intersection of the solid curves give the predicted location of the $\nu = 1$ level at $k_{\|} = 0.49$ Å^{-1} for 2, 3, and 4 ML Cu thicknesses, while the dashed curves give the same information for $k_{\|} = 0.62$ Å^{-1}. We see that the large phase shift introduced by the band gap results in little energy shift as $k_{\|}$ changes for the 2 and 3 ML films. In fact, the 2 ML film shows a negative dispersion. In contrast, when the state is away from the gap, as in the 4 ML film, the dispersion is very rapid and is similar to that expected for bulk Cu levels.

The phase accumulation calculation shows that the hybridization gap in the minority bands of fccCo causes the flat dispersion of the Cu MQW state near the neck of the Fermi surface. This does not change the periodicity of the Fermi surface crossings and therefore should not affect the length of the short period magnetic coupling. However, the location of band gaps in the FM layer can affect the phase and the strength of the magnetic coupling. Referenced to E_F and compared to fccCo,

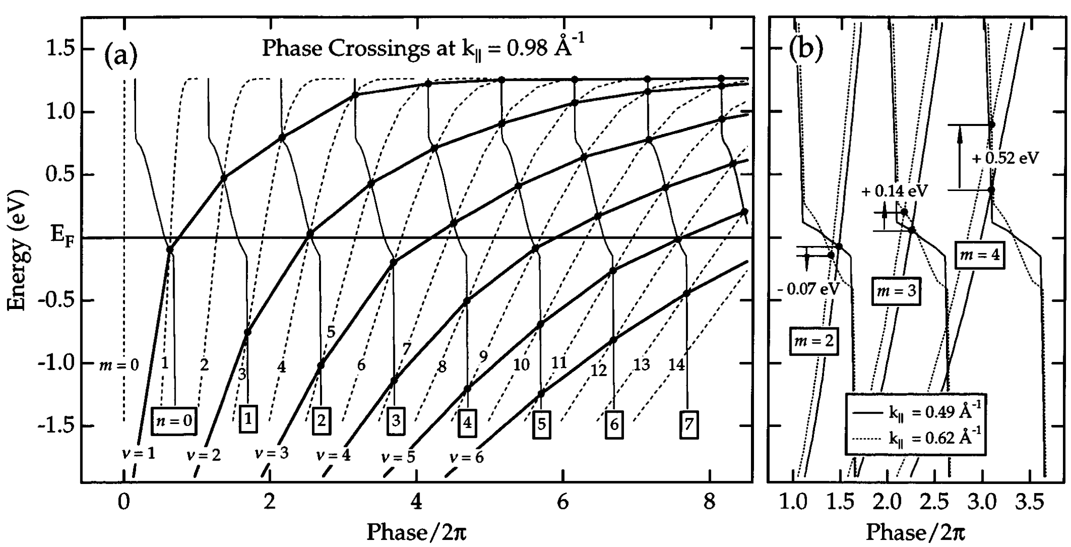

FIG. 2. (a) Phase accumulation at $k = 0.98$ Å^{-1} along $\overline{\Gamma X}$ for a Cu(100) film bounded by fccCo on one side and the image potential on the other. The dashed curves show $2mk_{\perp}a$ and the solid curves are solutions to $2\pi n - \Delta\phi_B - \Delta\phi_C$. (b) Phase accumulation generating the $\nu = 1$ state for $k_{\|} = 0.49$ Å^{-1} (solid curves) and $k_{\|} = 0.62$ Å^{-1} (dotted curves) for different layer thicknesses, m. The rapid phase shift in the Co gap results in a small dispersion for $m = 2$ and 3.

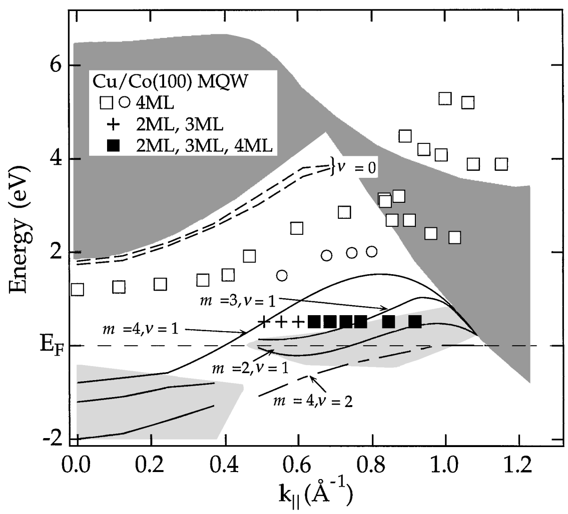

FIG. 3. Measured (symbols) and predicted (lines) dispersions for MQW states in the Cu/fccCo/Cu(100) system. The heavily shaded region is the projected Cu(100) band gap, while the lightly shaded area is the projected band gap in the minority spin states of the underlying fccCo(100).

the hybridization gap near the Cu Fermi surface neck is expected to be at higher energy in fccFe and lower in Ni. Within the phase analysis model, this would imply that the Fermi level crossing of a Cu MQW state of a given ν would be shifted to smaller Cu thickness for fccFe and larger for Ni. This is consistent with the phase shifts seen in the short period magnetic coupling of these systems [5]. Furthermore, as the band gaps at $\overline{\Gamma}$ of Fe, Co, and Ni are well below the Fermi level, only a slight phase is expected from scattering off the bands. This again matches what is observed for the long period magnetic coupling [3,5]. Moreover, the Co band gap by the neck of the Cu Fermi surface ensures that the Cu MQW levels are states rather than resonances, an effect that has been proposed to strengthen the short period coupling [9]. Finally, in models of the magnetic coupling based on the Fermi surface topology of the spacer layer [6–8,10], states in a range of $k_{\|}$ near the neck are expected to contribute to the coupling. For Co, the gap creates a flat MQW band in the Cu film precisely in the energy and momentum region appropriate to contribute to this coupling. Once again, whether fccFe or Ni will induce slowly dispersing MQW states or rapidly dispersing resonances in the Cu depends on the location of its gap.

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