

Short-period oscillations in photoemission from Cu films on Co(100)

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The electronic structure of Cu films on Co(100) in the k -space region associated with the short-period oscillation of the interlayer coupling in Cu/Co(100) multilayers has been examined by spin- and angle-resolved photoemission and by *ab initio* calculations. The photoemission intensity near the Fermi energy exhibits a rapid oscillatory behavior as a function of film thickness with a period close to that of the interlayer exchange coupling. The photoemission intensity and polarization reflect a modulation of the density of states induced by spin-polarized quantum-well states, in good agreement with *ab initio* calculations of quantum-well states. [S0163-1829(98)50402-X]

The exchange coupling between ferromagnetic layers separated by nonmagnetic materials has attracted much attention recently. There are fundamental problems concerning the coupling mechanism, as two ferromagnetic layers show alternating parallel and antiparallel magnetization as a function of the spacer thickness.¹ Furthermore, if a system with antiparallel alignment is placed in an external field sufficiently large to overcome the interlayer coupling, there is a decrease of the electrical resistance—a “giant magnetoresistance” effect—that can be exploited for sensors.²

Band theory relates the origin of the interlayer coupling to the electronic structure near the Fermi energy. In a Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction picture the oscillatory coupling reflects the topology of the bulk Fermi surface of the spacer material, with periods corresponding to the inverse of the reciprocal-space vectors spanning stationary points.³ More detailed approaches consider how the electronic structure may be modified by the lower dimensionality of thin films. Theoretical investigations⁴ show that changes in the density of states due to quantum well (QW) states modulate the relative energy of the parallel and antiparallel magnetic configurations as the spacer thickness varies. *Ab initio* calculations indicate that the states relevant to the coupling are spin polarized and, as in the RKKY scheme, derive from stationary points of the Fermi surface.

A detailed comparison between experimental and theoretical results requires simple systems with well characterized structures, and epitaxial structures of alternating ferromagnetic Co and nonmagnetic Cu layers provide an ideal test case. The interlayer coupling in Cu/Co(100) multilayers results from the superposition of two oscillatory terms with wavelengths 5.9 ML (Ref. 5) and 2.6 ML (Refs. 6 and 7), respectively. In a RKKY picture the two coupling periods arise from the belly and the neck of Cu Fermi surface cross section in the [100] plane.³ In the QW description the coupling originates from states from $\bar{\Gamma}$ and from the in-plane wave vector $k_{\parallel} = 0.9 \text{ \AA}^{-1}$ along $\bar{\Gamma}\bar{X}$, in the 2D representation of the Brillouin zone. As a function of the Cu thickness the QW states induce a periodic modulation of the density of states at the Fermi energy with the same period as the interlayer exchange coupling. Theoretical calculations demonstrate that this oscillation in the density of states at the Fermi

energy is directly related to the sign reversal of the coupling. The corresponding experimental information on this density of state modulation is provided by photoemission spectroscopy. Direct and inverse photoemission on Cu/Co(100)^{8–10} shows spin-polarized quantum-well states at the $\bar{\Gamma}$ point which modulate the photoemission intensity and the spin polarization at the Fermi energy with a 5.9–6.0 ML periodicity that corresponds to the long coupling period.

On the other hand, no direct observation of a short period oscillation has been obtained in photoemission experiments until now. A recent investigation¹¹ examined the electronic structure of Cu films at $k_{\parallel} = 0.9 \text{ \AA}^{-1}$, and the photoemission spectra show distinct structures due to QW states, with binding energy depending on the film thickness. The binding energy of these states is shown to be consistent with the theoretical expectation, but the relatively coarse (2.8 ML) thickness increments used do not allow the observation of a fast oscillation in the photoemission intensity. The experiment determines an oscillatory behavior of the photoemission intensity at the Fermi energy with an approximate period of 14–19 ML, which was interpreted as the beating frequency resulting from the 2.8 ML sampling frequency of the measurement and a 2.45 ML modulation of the density of states. Weak thickness-dependent structures are also observed in a spectroscopic investigation combining direct and inverse photoemission.¹² The data suggest that quantum-well states approach the Fermi level at 6 and 9.6 ML thickness, which is regarded as consistent with a period of about 3 ML. The spin of the electronic states was not resolved in any of the previous studies.

The present work provides further evidence which links the origin of the shorter period to quantization effects on the Cu band structure. Spin- and angle-resolved photoemission has been used to study the electronic structure of Cu layers on Co(100) as a function of thickness, and we observe directly a short period oscillation of the photoemission intensity near the Fermi energy. Spin polarization measurements show that the oscillations are induced by polarized quantum-well states. In addition, the intensity modulation shows a fine structure reflecting the discreteness on the atomic scale of the incremental film thickness. The results are in excellent agreement with *ab initio* calculations for the short period quantum well states of Cu/Co(100).

Epitaxial structures consisting of fcc Co and Cu films were grown *in situ* on a Cu(100) single crystal. Co films of 15 ML thickness were deposited at room temperature by evaporation on the Cu(100) substrate, and Cu layers grown subsequently on Co. The deposition temperatures were 100 K for the first Cu monolayer and room temperature for the additional layers. The angle-resolved photoemission intensity and spin polarization were measured with a dispersive electron energy analyzer coupled to a Mott spin detector.¹³ In the experimental setup photoemission spectra could be taken during the Cu film growth, so that it was possible to monitor the spectral intensity as a function of film thickness with high sensitivity and accuracy. Changes of the photoemission intensity at selected electron energies and momenta could be followed for incremental thicknesses below 0.07 ML.

The experiments are complemented by *ab initio* calculations for the quantum-well states of Cu/Co/Cu(100) using a Korringa-Kohn-Rostoker Green's function method for layered systems.^{14,15} We consider a Co(100) slab of 5 ML thickness embedded at a variable distance (up to 17 ML) below a Cu(100) surface. The electronic structure and the potentials of the Cu(100) surface and of a Co slab embedded in an infinite Cu host are calculated self-consistently. The resulting potentials for the Co slab and the Cu(100) surface are then used to calculate the density of states of the interacting system. We calculate, in particular, k_{\parallel} -dependent and symmetry-projected density of states of the Cu overlayer as a function of the overlayer thickness. Basically the calculations are similar to the tight binding analysis of Smith *et al.*¹⁶ The essential difference is that all properties are calculated from first principles, in particular the Green's function of bulk Cu, the electronic structure of the Cu(100) surface, as well as the electronic structure of the Co(100) slab embedded into Cu.

Angle-resolved photoemission can selectively probe the electronic states for an in-plane wave vector k_{\parallel} by proper choice of the excitation energy and collection angle. Figure 1 shows photoemission measurements (a) and calculated energy eigenvalues (b) for states with $k_{\parallel}=0.9 \text{ \AA}^{-1}$ along the $\Gamma\bar{X}$ direction of the surface Brillouin zone. The upper curve in Fig. 1(a) represents the photoemission intensity at 0.3 eV binding energy for Cu thicknesses from 4.5 ML to 17 ML. The photoemission intensity was monitored over an energy window of finite width determined by the overall experimental resolution. This is described well by a Gaussian function with 0.3 eV full width at half maximum, as determined from the shape of Fermi cutoff of the electron distribution curves. The photoemission intensity curve [Fig. 1(a)] displays intense and sharp structures as a function of film thickness with maxima at regular intervals superimposed on a decreasing background due to the rapidly attenuated emission from the Co substrate. The separation between the peaks corresponds to a period of 2.3 ± 0.1 ML. A qualitatively similar behavior is displayed by the photoelectron spin polarization in Fig. 2, measured 0.4 eV below the Fermi level as a function of Cu thickness. The spin polarization curve shows two maxima and two minima separated by 2–3 ML thickness.

We show in the following that the structures of the photoemission intensity curve are related to quantum size effects on the Cu electronic structure. In an ideal isolated two-dimensional system the spatial confinement in the direction perpendicular to the plane gives rise to a discrete spectrum of

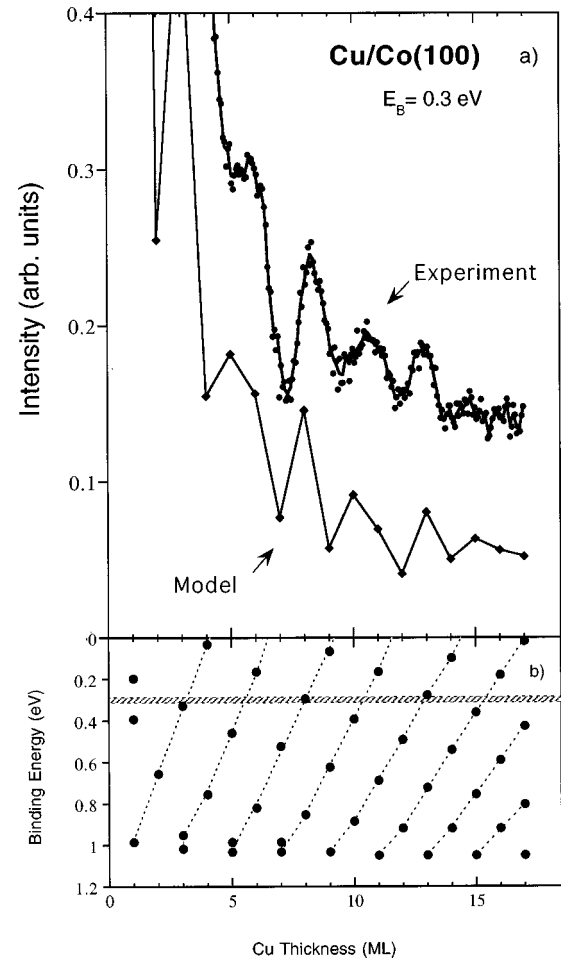


FIG. 1. (a) Circles: Spectral intensity at 0.3 eV binding energy as a function of Cu thickness. The photon energy is 77 eV and the collection angle is 12° from the surface normal [$k_{\parallel}=0.90 \text{ \AA}^{-1}$] along the $\Gamma\bar{X}$ direction of the surface Brillouin zone. Diamonds: Theoretical photoemission intensity calculated with the data of (b) (see text for explanation). (b) Calculated binding energies of minority-spin quantum-well states at $k_{\parallel}=0.94 \text{ \AA}^{-1}$ along the $\Gamma\bar{X}$ direction of the surface Brillouin zone for Cu thicknesses ranging from 1–17 ML. The dashed lines are a guide to the eye showing the quantum-well dispersion with Cu thickness.

n energy levels $E_n=E_n(k_{\parallel})$ for any k_{\parallel} value. In supported film and multilayer systems the interaction at the interfaces with the substrate and the neighbor layers defines the degree of the electron confinement, which in general varies with wave vector, energy, and spin character. For $k_{\parallel}=0.9 \text{ \AA}^{-1}$, along the $\Gamma\bar{X}$ direction, a partial band gap in the Co band structure strongly confines the Cu minority-spin states with binding energy lower than 0.9 eV in the overlayer so that the QW states are well localized in this energy region. Conversely, the hybridization with Co states of corresponding spin and symmetry allows the formation of common delocalized bands for Cu states of majority-spin character. The *ab initio* calculated energy spectrum in Fig. 1(b) shows discrete quantum-well states of minority spin character with a simple dispersion as a function of the film thickness. With increasing thickness branches of states [indicated by the dashed lines in Fig. 1(b)] disperse to lower binding energy and cross the Fermi level with a 2.6 ML interval, which closely corre-

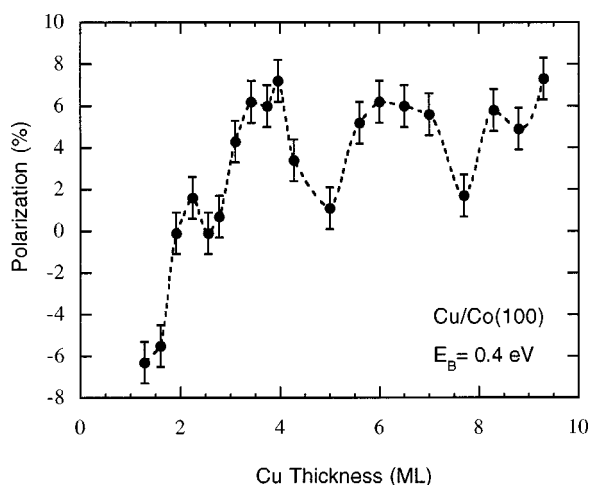


FIG. 2. Electron spin polarization at 0.4 eV binding energy as a function of Cu thickness. The photon energy is 77 eV and the collection angle is 13° from the surface normal [$k_{\parallel}=0.97 \text{ \AA}^{-1}$] along the $\Gamma\bar{X}$ direction of the surface Brillouin zone.

sponds to the experimental short coupling period.^{6,7} The crossing of the quantum-well branches at 0.3 eV binding energy occurs in the calculated spectra with a 2.4 ML period, which is slightly below the period corresponding to the Fermi energy. The experimental period of 2.3 ± 0.1 ML from the photoemission measurements agrees well with the theoretical value. The photoemission intensity curve displays peaks at 6, 8.5, 10.7, 13, and 15 ML Cu thickness, whereas calculated quantum well states are near 0.3 eV for 5–6, 8, 10–11, 13, and 15 ML Cu thickness. The spin polarization measurements in Fig. 2 with minima at about 2.5, 5, and 7.5 ML are also in good agreement with the theoretical results. The calculated minority-spin quantum well branches cross the 0.4 eV binding energy near 3 ML, between 5–6 ML, and between 7–8 ML.

In Fig. 1(a) we compare the experimental data with a curve derived from the theoretical results of Fig. 1(b). In a simple model we assume the single quantum states to contribute to the photoemission spectrum with a finite linewidth corresponding to the experimental energy resolution. The intensity of each quantum-well contribution should be proportional to the inverse of the film thickness, since the spectral weight is expected to decay as the quantum-well wave function becomes delocalized over an increasing number of atomic layers, while the experiment probes the top layers only. We add an exponential background for an escape depth of 2 ML to account for the attenuation of the Co substrate emission. The model curve in Fig. 1(a) agrees well with the experimental one with respect to oscillation period and phase, and both curves show a similar fine structure with peaks of unequal width and noncontinuously decaying strength. The fine structure is due to the discreteness of the thickness incrementing in units of the interlayer spacing. Sharp maxima appear for film thicknesses with quantum-well states in the center of the energy window (e.g., at 8 ML and 13 ML). On the other hand, weaker and broader peaks arise if a quantum-well branch intersects the energy window for a noninteger number of layers (e.g., at 5–6 ML and 10–11 ML).

In order to demonstrate further the relation between the

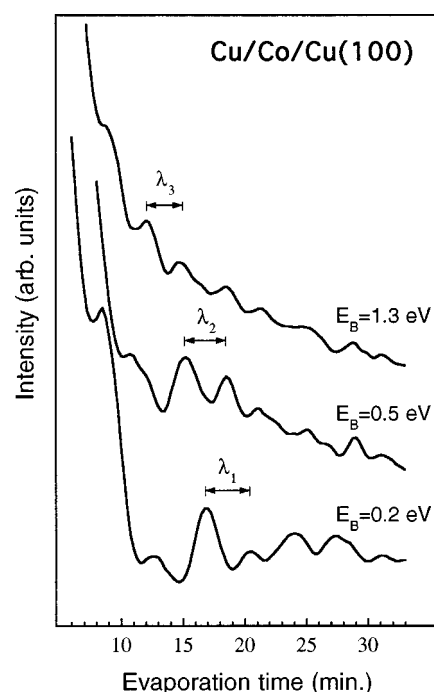


FIG. 3. Spectral intensity at three different binding energies as a function of Cu evaporation time. The experimental geometry is the same as in Fig. 1

structures of the photoemission intensity curves and the quantization effects, we have performed measurements as a function of the initial state energy. Figure 3 presents the photoemission intensity obtained simultaneously for three different binding energies during the growth of a Cu film on Co(100). The period, the phase, and the fine structure of the oscillations clearly display a dependence on the initial state binding energy and the period depends on the binding energy as expected from band structure considerations. We have determined the periods for the three measured intensity curves of Fig. 3 from a Fourier analysis. The period of the three curves in Fig. 3 scales as the ratios 1 : 0.94 : 0.80, at 0.2 eV, 0.5 eV, and 1.3 eV binding energy, respectively. The ratios are in very good agreement with those derived from the Cu band structure calculation, which gives a relationship of 1 : 0.93 : 0.80. Furthermore, the three curves show a fine structure with peaks of various strength and width. The curve for 0.2 eV binding energy shows structures of alternating strength, which is consistent with the theoretical period at 0.2 eV binding energy being close to 2.5 ML, i.e., commensurate to five interlayer spacings. The structures of the intensity curve measured at 1.3 eV are of comparable shape and similar strength, in accordance with the corresponding theoretical period of 2 ML. The 0.5 eV binding energy curve does not show a simple behavior, in qualitative agreement with the calculated period of 2.35 ML, which is incommensurate with simple multiples of the layer spacing.

A fine structure of the photoemission intensity has not been observed previously in photoemission studies of quantum-well states. In fact, photoemission and inverse photoemission results reported regular oscillations with smoothly decaying strength as a function of thickness. The discreteness of the coverage increments, however, gives rise to an intrinsic sampling period that can result in peaks of

varying amplitude and width. Consequently, the occurrence of a fine structure in the oscillatory photoemission behavior should be a general feature that is stronger for fast dispersing states, i.e., for short period oscillations. An obvious requisite for its observation is that the film growth closely approaches a layer-by-layer mode, since roughness quenches the fast oscillations very effectively.

In summary, we have used spin- and angle-resolved photoemission to investigate the electronic structure of Cu films on Co(100) as a function of thickness. The measurements are complemented by *ab initio* calculations for quantum well states. The photoemission intensity oscillates with a short

period corresponding to the one of the interlayer coupling through Cu(100). Spin-resolved measurements show that the oscillations are due to polarized quantum-well states. In addition, the intensity modulation exhibits a previously unobserved fine structure due to the discreteness of the thickness increments corresponding to the spacing of the atomic layers. The experimental results are in good agreement with our *ab initio* calculations and firmly link the origin of the short oscillation period to the existence of polarized quantum well states.

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