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Spin polarized photoemission studies of magnetic quantum well states

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Abstract

Quantum well states with discrete binding energies dependent on the thickness of the film are observed in copper films deposited on a Co(001) substrate. They are found to be spin polarized, preferentially with minority spin. These states pass up to and through the Fermi level with a frequency identical to the long period of oscillation in the associated magnetic multilayers. In the 'pre-asymptotic limit' the dispersion of these states away from the center of the zone is described by enhanced effective masses. This has implications for theories of the oscillatory exchange coupling that invoke the bulk Fermi surface.

1. Introduction

There is currently considerable technological interest in the properties of transition metal multilayers [1]. In these multilayers, which include Fe/Cr(001) [2] and Cu/Co(001) [3], it is possible to achieve either ferromagnetic or antiferromagnetic coupling of the adjacent ferromagnetic layers depending on the thickness of the intervening layer [4]. The related giant magnetoresistance properties and the large enhancement of the Kerr rotation in these systems make them particularly interesting [5].

Several theories of the oscillatory exchange coupling invoke bulk Fermi surface spanning vectors that reflect the correct Fermi surface for the intervening layer [6]. Indeed such theories may predict more than one periodicity for the coupling as has been observed experimentally for both the Fe/Cr [2] and Cu/Co [7] multilayers. Whilst 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 non-magnetic layers will be much stronger. The finite thickness of the individual layers within the film will also lead to a discrete quantization of the electronic structure within the layers. In this paper we examine the properties of the 'quantum well' states that reflect this quantization with particular emphasis on Cu thin films deposited on a Co(001) substrate. We show that in the direction along the surface normal these states sample the Fermi surface with a periodicity identical to that observed for the long period oscillatory exchange coupling in the associated Cu/Co(001) multilayers [3,7]. Further we show that these states are highly spin polarized, preferentially with minority spin polarization. Finally, by examining the dispersion of these states as a function of their parallel momentum in the plane of the film we show that the interfacial hybridization leads to a strong modification of the electronic structure and associated Fermi surface of the copper films in the pre-asymptotic limit. This has implications for the theories of the oscillatory exchange coupling in the multilayers that invoke the bulk Fermi surface of the spacer layer.

2. Experimental procedures

The spin-polarized photoemission experiments reported here were carried out on an apparatus that has been described in detail elsewhere [8]. Briefly the photoemitted electrons are energy and momentum analyzed by a commercial hemispherical analyzer backed by a compact low energy spin detector of the type described by Unguris et al. [9]. 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 $\pm 1.5°$ and the combined photon and analyzer energy resolution was 0.35 eV, or better at the lower photon energies [8].

For the experiments described in this paper, Cu films of different thicknesses are deposited onto an fcc Co film that had previously been evaporated onto a Cu(001) substrate. The Co film thickness was typically 20 ML. The deposition rates and hence film thicknesses are monitored directly by a quadrupole mass spectrometer. Low Energy

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Electron Diffraction and Auger electron spectroscopy are used as additional diagnostics.

3. Results and discussion

The quantization of the electronic structure in thin films is shown in Fig. 1 [10]. Here, copper films with different thicknesses are deposited on a Co(001) substrate. The ability to resolve the discrete quantization reflects the energy separation ΔE between the different states. This separation is given by [11]

$$\Delta E = \frac{\partial E}{\partial k}\frac{2\pi}{L}, \tag{1}$$

where $\partial E/\partial k$ reflects the dispersion in the associated bulk bands and L is the thickness of the film. The spectra in the figure clearly show the quantization of the s/p derived bands. With the typical energy resolution available, quantization of the more slowly dispersing d bands is only observed in the thinner films [12,13].

With increasing thickness, the s/p derived quantum well states appear to move up to and through the Fermi level with a periodicity or frequency identical to that observed for the long period oscillation in the exchange coupling in the related multilayers [10,14]. Using phase summation arguments and recognizing that the wave function of the quantum well state acquires another half electron wave length with each layer added to the well [15] it is a simple matter to show that the Fermi surface will be sampled every m layers where

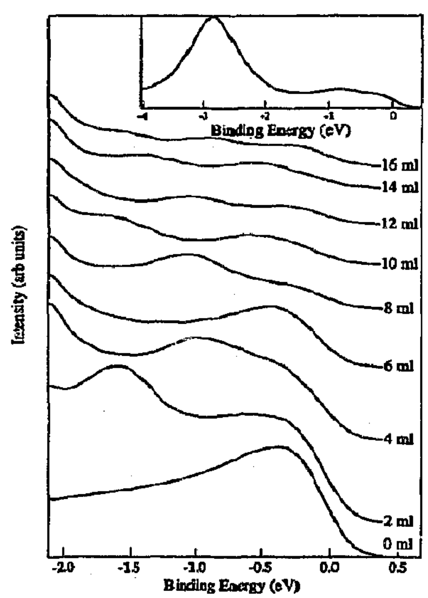

Fig. 1. Photoemission spectra recorded from different thickness copper films deposited on a Co(001) substrate as indicated. The binding energy is referenced with respect to the Fermi level as indicated. The incident photon energy is 24.0 eV and the angle of incidence of the light corresponds to p-polarization. The inset shows a spectrum recorded from a four monolayer thick copper film over a wider energy range.

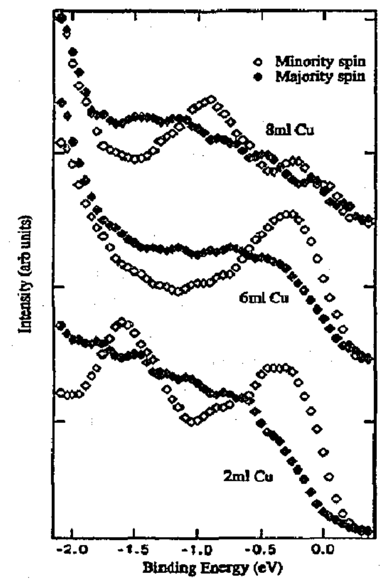

Fig. 2. Spin resolved photoemission spectra recorded from 2, 6 and 8 ML thick copper films. The incident photon energy is 24.0 eV and the angle of incidence of the light corresponds to p-polarization.

$$m = \frac{k_{BZ}}{k_{BZ} - k_F}; \tag{2}$$

k_{BZ} and k_F define the zone boundary and Fermi wave vectors respectively. In the direction of interest, the difference in these wave vectors, $k_{BZ} - k_F$, represents one half the Fermi surface spanning vector.

Fig. 2 shows the spin resolved photoemission spectra recorded from selected copper films [10]. In all cases, the states are preferentially polarized with minority spin even in the thicker films where the photoelectron mean free path will restrict the sampling to the copper layers. Similar results have been obtained both in an earlier study of silver films deposited on an Fe(001) substrate [16] and in another study of the Cu/Co(001) system [17]. The observation that these quantum well states are spin polarized and crossing the Fermi level with the appropriate frequency leads to the suggestion that they are the states that mediate the exchange coupling in the magnetic multilayers [14].

The quantization and spin polarization of these states may be modeled within the tight-binding formalism. Our approach is based on the use of a Hubbard Hamiltonian. Thus

$$H = \sum_k E(k) n_k + \frac{U}{N} \sum_{k',k} n_{k\uparrow} n_{k'\downarrow}, \tag{3}$$

where the first term represents the paramagnetic band structure and the second term represents the modification due to an on-site spin dependent potential U. In the slab formulation, we take the parameters associated with a tight binding fit to the appropriate paramagnetic band structure for each layer in the slab [18], split the on-site spin dependent energies for the d-blocks by an amount Δ_1 and then integrate the spin-dependent densities of states up to the Fermi level to obtain layer dependent moments m_1. A self consistent solution is sought such that for each layer, $\Delta_1 = U_1 m_1$. The 'exchange' potential U_1 is taken as the effective Stoner parameter from LSD calculations of the susceptibilities for the different elements [19].

To simulate the photoemission spectra recorded along the surface normal, we restrict the calculation of the spin dependent densities of states to a narrow region around the center of the surface Brillouin zone. We weight the calculated eigenvectors in each layer of the slab by an escape depth and representative photoionization cross-sections [20]. Fig. 3 shows the results of such a spin-dependent calculation for different thickness copper films deposited on a seven layer Co slab. The minority spin states are observed to move up to and through the Fermi level with the appropriate frequency. States are also observed in the majority spin channel but with considerably less intensity. The spin polarization of these states may be thought of either as a spin dependent asymmetry in the hybridization at the interface as in the present paper or as an asymmetry in the reflectivity of the two spin components from the substrate. Both descriptions are equivalent. Indeed, Bruno has recently examined the reflectivity in the vicinity of the Co substrate spin dependent band gaps and found that at the center of the surface Brillouin zone at energies around the Fermi level, minority spin electrons are preferentially reflected in the Cu/Co(001) system [21].

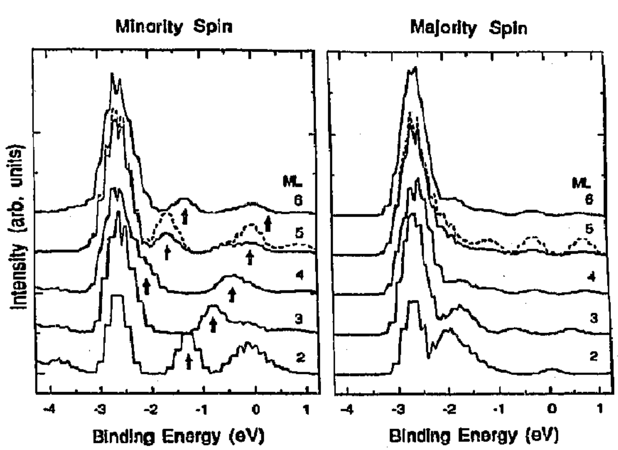

Fig. 3. Calculated spin resolved photoemission 'spectra' from different thickness copper films. The left panel shows minority spin spectra and the right panel shows majority spin spectra. The copper thicknesses range from two through to six monolayers from bottom to top in each panel. The dashed lines indicate spectra calculated for the five monolayer films with equal weight given to s, p and d states. The arrows indicate the relevant quantum well states in the minority spin panel.

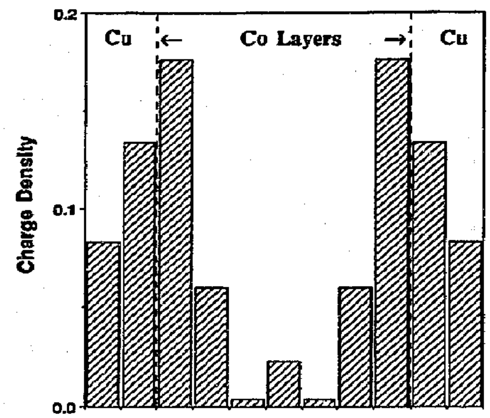

Fig. 4. The calculated layer dependent charge densities for the minority spin quantum well state of a 2 ML Cu film either side of a 7 ML Co(001) slab. The vertical dashed lines indicate the Cu/Co interfaces. In these interfaces the orbital character is predominantly d_{z^2}.

The present calculation highlights other interesting characteristics of these states. Aside from the obvious s/p character, the states also have significant copper d-character. This is shown in the figure for the five monolayer case where we compare the spectra obtained by giving equal weight to the s, p and d electrons with the spectra obtained preferentially from the d-electrons. This reflects the strong $sp_z d_{z^2}$ hybridization that occurs in the vicinity of the copper d bands and explains the observation of a d-moment on the copper site in recent magnetic circular dichroism (MCD) studies of the Cu/Co multilayers [22]. There is also a significant interfacial component in these states as indicated in Fig. 4 for the 2 ML minority spin quantum well state. Whilst we have previously discussed the interfacial characteristics for the ultrathin Ag films deposited on an Fe(001) substrate [16], it should be noted that as a result of the d–d hybridization, the charge density peaks in the interface even for the states associated with thicker Cu films.

Examination of Fig. 4 shows that the quantum well states are characterized by long tails decaying into the underlying Co. These tails arise as a result of hybridization with the evanescent states that exist in the complex momentum plane within the hybridization bandgap of the substrate. Such states have previously been discussed extensively in the literature with particular reference to the formation of surface states in bulk bandgaps [23,24]. The evanescent states have no relevance to the bulk bandstructure but will become important whenever the infinite periodicity is broken at a surface or at an interface. Because of these evanescent states it may be anticipated that there will be a reduction in the 'confinement' of the quantum well states when grown on thinner Co substrates [13], an obser-

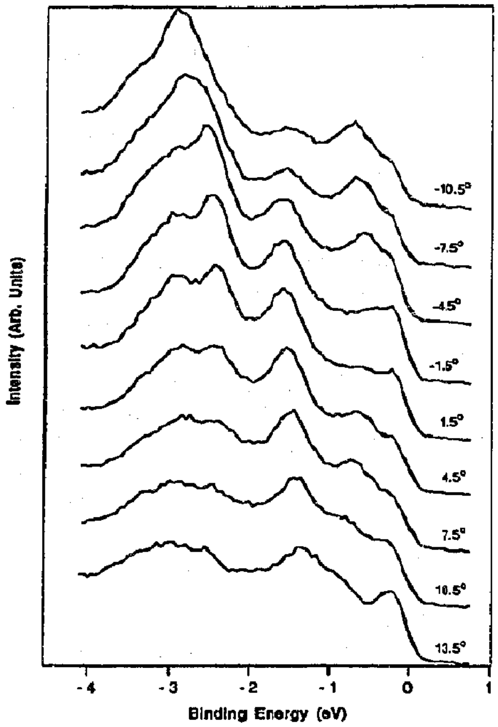

Fig. 5. Photoemission spectra recorded from a 2 ML thick copper film as a function of the angle of emission as indicated. The incident photon energy is 22.7 eV and the angle of incidence of the light corresponds to p-polarization. The quantum well state has a binding energy of 1.6 eV near the center of the zone.

vation that has implications for experiments on magnetic multilayers in which the ferromagnetic layers are replaced by wedges.

As noted earlier, theories invoking the bulk Fermi surface of Cu predict two period lengths for the oscillatory exchange coupling [6]. The long period of oscillation reflecting the spanning vector at the center or belly of the Fermi surface 'dog bone', the short period of oscillation reflecting the spanning vector at the neck of the 'dog bone'. In an isolated copper slab, the latter point on the Fermi surface, approximately $0.75\overline{\Gamma X}$, is sampled by states that have dispersed upwards from the center of the zone with a dispersion in the plane of the film, $E = \hbar^2 k_{\parallel}^2 / 2m_e$, characterized by a free electron like mass m_e [25]. As a function of thickness, we would expect from Eq. (2) the Fermi surface to be sampled with a periodicity inversely related to the bulk Fermi surface spanning vector at this point in the zone. However in the thin films or multilayers this behavior may be modified by the hybridization in the interfaces.

Fig. 5 shows the photoemission spectra recorded from a 2 ML thick copper film deposited on a 20 ML thick Co substrate as a function of the angle of emission in the $\overline{\Gamma X}$ azimuth. At the center of the zone the quantum well state characterizing this thickness is again observed at a binding

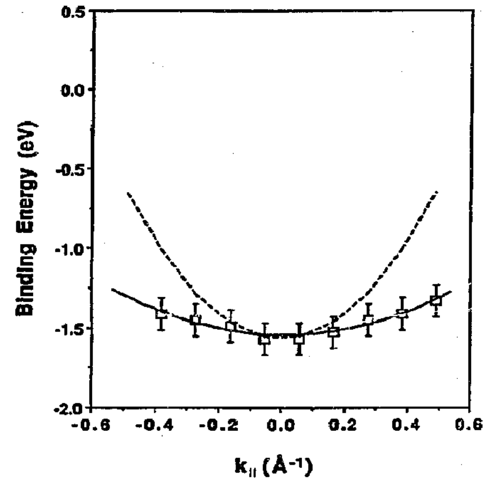

Fig. 6. The dispersion of the quantum well state in Fig. 5 plotted as a function of $k_{\parallel}$. The solid line represents a parabolic fit to the experimental points indicated by the open squares. The dashed line represents the parabolic dispersion that would be characterized by a free electron mass.

energy of 1.6 eV. However away from the center of the zone the dispersion of this state in the plane of the film is clearly not free-electron like but, rather is characterized, as shown in Fig. 6, by an effective mass of approximately $4.0m_e$ where again m_e represents the free electron mass.

Discussed in more detail elsewhere but reproduced in Fig. 7, the modification of the in-plane effective mass extends out to copper film thicknesses of the order of 7–8 ML [26]. This thickness will determine the pre-asymptotic limit below which it will not be valid to describe the properties of the copper layer simply in terms of its bulk

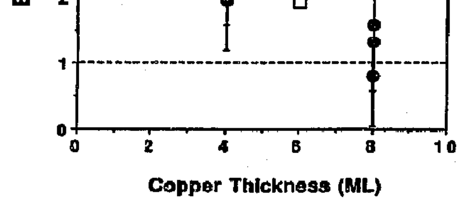

Fig. 7. Plot of the fitted effective masses of the quantum well states in units of free electron mass as a function of the copper film thickness in monolayers (ML). The solid circles indicate the fits to the experimental data; the open squares indicate the results of tight-binding calculations of the effective masses. The dashed line represents the free electron mass.

electronic structure. The effective mass enhancement may be viewed as a competition between on the one hand the properties of the interface and on the other the properties of bulk copper. In the very thin films the interfacial hybridization dominates.

The phenomenon may be reproduced in the tight binding scheme. With some adjustment to the interfacial parameters, it is possible to reproduce the effective mass describing the in-plane dispersion for the 2 ML state. By then fixing these interfacial parameters and increasing the copper film thickness to 4 ML and then 6 ML the calculated effective mass decreases at a rate similar to that of the experiment as shown in Fig. 7. The modification of the effective mass is an indication that the Fermi surface in the copper film will also be modified, an observation that has implications for theories of the oscillatory exchange coupling that invoke the bulk Fermi surface [6]. Indeed in the pre-asymptotic regime it is likely that the calculated coupling periods involving spanning vectors away from the center of the zone will be modified as a result of this interfacial hybridization.

4. Summary

Our studies have highlighted the importance of the interaction in the interface for determining the coupling in the magnetic multilayers. An asymmetry in the spin dependent interaction at the interface produces the spin polarization in the quantum well states. Further the hybridization in the interface modifies the in-plane effective mass, a phenomenon which cannot be ignored in descriptions of the pre-asymptotic regime. It is therefore clear that further studies of the interfacial interactions will shed new light on the oscillatory exchange coupling in the magnetic multilayers.

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