Quantum Well States as Mediators of Magnetic Coupling in Superlattices

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Quantum well states are found at the Fermi level in Cu on Co(100) and Ag on Fe(100) using inverse photoemission. They appear every 5.9 ± 0.5 layers in Cu/Co(100), which agrees with the 5.5- to 6-layer oscillation period of the magnetic coupling in Cu/Co(100) superlattices. For Ag/Fe(100) they connect with minority-spin interface states observed below E_F previously, providing a magnetic coupling channel through the noble metal. These properties are explained in terms of the bulk band structure.

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The electronic structure of metallic superlattices is gaining interest for designing new solids with flexible properties. Recently, an oscillatory magnetic coupling observed in magnetic superlattices [1-9] has created widespread interest, due both to the potential applications in magnetic and magneto-optic storage, and to the unusual oscillation period. The latter is on the order of 10 Å, which is much larger than the Fermi wavelength expected from simple arguments. A variety of theoretical models [10-14] have been proposed to explain this behavior. Our aim is to find the electronic states that mediate the magnetic coupling. We are particularly interested in noble metal spacer layers, because it is not obvious how a noble metal can transmit the magnetic interaction over distances of many atomic layers. In order to narrow the field we notice that states near the Fermi level E_F are expected to contribute the most to magnetic phenomena. After all, the changes in the density of states within kT_C (T_C=Curie temperature) of E_F drive the magnetic phase transition. In momentum space, we will therefore have to consider the whole Fermi surface, but the direction perpendicular to the interfaces of the superlattice will be emphasized by symmetry.

We have investigated such states by inverse photoemission, using Cu(100) films on Co(100) and Ag(100) on Fe(100) as prototypes for one period of the corresponding superlattices. It turns out that there exists a special type of electronic state at the Fermi level that is characteristic of thin films. These are quantum well states, which are created by quantizing the momentum of s,p band states perpendicular to the surface. Similar states have been seen in various metals (see Ref. [15] and references therein). We find that the quantum well states in Cu/Co(100) and Ag/Fe(100) exhibit traits that connect them with oscillatory magnetic coupling. The periodicity of their charge density is equal to the magnetic oscillation period in Cu/Co(100). Its large value of 10.6 Å is explained by the fact that the k vector of the envelope function is not given by the Fermi wave vector k_F, but by the difference between k_F and the wave vector of the Brillouin zone boundary, k_{ZB}. This difference is rather small for the s,p band in most transition and noble metals, explaining the trend towards large periods. Another important ingredient is the spin polarization of the quantum well states, found by interpolation between previous spin-polarized photoemission data and our inverse photoemission results for Ag/Fe(100). It provides a magnetic coupling mechanism through the noble metal. The origin of the spin polarization of these s,p-like states lies in the spin-dependent boundary conditions at the interface with the ferromagnet. For Ag/Fe(100) only minority-spin quantum well states can form at the Fermi level for $k^{\parallel}=0$ since the majority-spin s,p states in Ag(100) can couple to majority-spin states in Fe(100). This is indeed the sign of the observed spin polarization [16].

For obtaining well-defined quantum well states it was critical to produce highly perfect starting surfaces. A fcc Co(100) substrate was produced by depositing about twenty layers of Co onto Cu(100) at room temperature, with the Cu crystal carefully electropolished and sputter annealed at grazing incidence. Fe(100) was cleaned by extensive temperature cycling in 1 atm of H_2 and sputter annealing. Cu and Ag were deposited in a vacuum in the low 10^{-10} Torr range at substrate temperatures between room temperature and 150 °C. The film thickness was monitored with a quartz oscillator and calibrated with medium-energy ion scattering (MEIS).

Results for Cu/Co(100) are shown in Fig. 1. Similar spectra were obtained for Ag/Fe(100) for a smaller range of thicknesses. By changing the initial energy we confirmed that the states in Fig. 1 did not disperse with the momentum perpendicular to the surface $k^{\perp}$ (not shown), and therefore represent two-dimensional states. The strong, nearly periodic dependence of the structures on film thickness is an indicator of quantum well states [15]. They represent standing waves in the noble metal films whose wavelength has to match the film thickness. Every time the film thickness is increased by half a wavelength, the wave function fits in again. Here we are interested in quantum well states at the Fermi level E_F. In order to see clearly where Fermi level crossings occur in Fig. 1, we plot the intensity at E_F versus thickness in Fig. 2. A series of cusps is found, starting at 5.4 Cu layers with a period of 5.9 ± 0.5 layers (1 Cu layer = 1.8 Å). This agrees with the period of 5.5 to 6 layers (Refs. [4] and [3], respectively) observed for the oscillations of the mag-

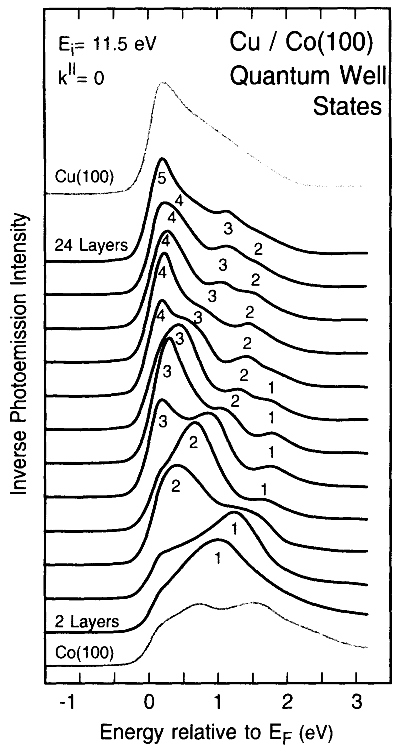

FIG. 1. Inverse photoemission spectra for Cu on Co(100) at normal incidence. The s,p band continuum of bulk Cu(100) (top) is discretized into quantum well states for thin Cu films. For the film thicknesses see the data points in Fig. 2.

netic coupling in Cu/Co(100) superlattices, providing strong circumstantial evidence for a connection. For Ag/Fe(100) the first maximum is found at 4.5 Ag layers, and a weak, second one 5 layers later (1 Ag layer = 2.0 Å). No clear magnetic coupling period has been established for this system [7]. The data for Ag/Fe(100) are summarized in Fig. 3, including previous spin-polarized photoemission results [16] on this system. A minority-spin state with thickness-dependent binding energy was found in this work and interpreted as an interface state. Our inverse photoemission data in Fig. 3 suggest that this state connects with the second member in a series of quantum well states after crossing E_F. This connection establishes that quantum well states in noble metals can be spin polarized, despite their nonmagnetic s,p character, and thus can provide a mechanism for magnetic coupling across noble metal spacer layers. For Cu/Co(100) the occupied counterpart of the quantum well states in Fig. 1 has also been found recently [17].

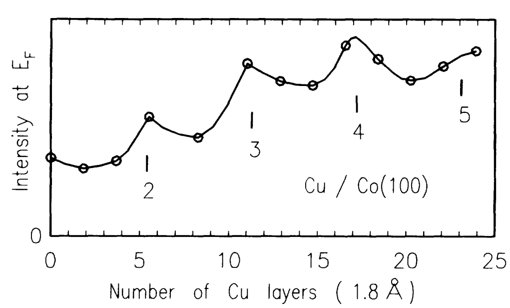

FIG. 2. Inverse photoemission intensity at E_F vs film thickness for the Cu/Co(100) data in Fig. 1. Periodic maxima correspond to quantum well states crossing the Fermi level. The period of 5.9 ± 0.5 monolayers corresponds to the inverse of the distance between the Fermi wave vector k_F and the Brillouin zone boundary k_{ZB}, measured in units of k_{ZB} (compare Fig. 4 for Ag). It also agrees with the period of 5.5 to 6 layers observed in the magnetic coupling for Cu/Co(100) superlattices [3,4].

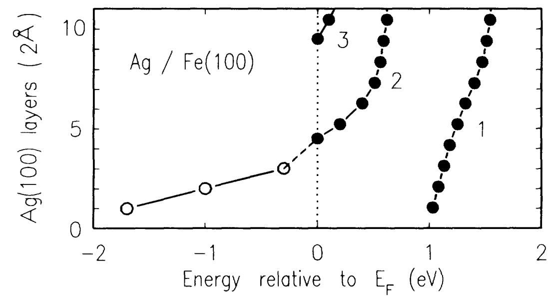

FIG. 3. Energy position of quantum well states for Ag on Fe(100) vs layer thickness. Dots are from inverse photoemission spectra analogous to Figs. 1 and 2; circles represent previous spin-polarized photoemission results [16] for minority-spin interface states. They connect with the second quantum well state.

In order to illuminate the character of the quantum well states observed here and to explain their periodicity and spin polarization we show in Fig. 4 how these states are derived from the bulk s,p band of the noble metal. By borrowing standard techniques from semiconductor

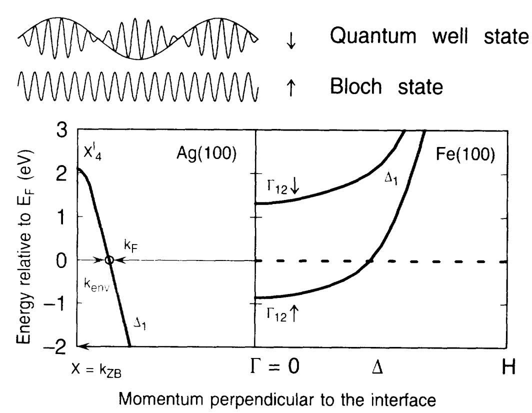

FIG. 4. Character of the wave function for quantum well states in Ag on Fe(100), consisting of a fast-oscillating Bloch wave modulated by an envelope function (top). The corresponding wave vectors (k_{ZB} and k_{env}) are obtained from the bulk band structure (bottom). Only minority-spin states in Ag exhibit quantum well character at the Fermi level since the majority-spin Δ_1 states couple with the corresponding states in Fe.

quantum well calculations [15,18] one finds that the wave function of a quantum well state can be expanded around the Bloch state at the band edge, in our case the X_4' point of the s,p band in Cu and Ag. This fast-oscillating Bloch state is modulated by a slowly varying envelope function, which makes sure that the boundary conditions are met at the two interfaces (Fig. 4, top). The total wave vector of the quantum well state is the sum of the wave vectors of the Bloch state and the envelope function, i.e.,

$$k_{\mathrm{tot}} = k_{\mathrm{ZB}} \pm k_{\mathrm{env}} \quad (1)$$

(see Fig. 4 bottom). This wave vector has to fall onto a bulk band if the film is thick enough to exhibit bulklike bonding [15]. For quantum well states at E_F this means that the total wave vector equals the Fermi wave vector k_F (circle in Fig. 4). With a little arithmetic we arrive at a simple relation between the periodicity in the appearance of quantum well states in Fig. 2 and the Fermi wave vector: The period in monolayers corresponds to the inverse of the distance between the Fermi wave vector k_F and the Brillouin zone boundary k_{ZB}, measured in units of k_{ZB}. To derive this result we note that quantum well states appear with a period of $\lambda_{\mathrm{env}}/2$, since every time the well thickness is increased by half the wavelength λ_{env} of the envelope function the same boundary conditions are satisfied. Thus we have $k_{\mathrm{env}} = 2\pi/\lambda_{\mathrm{env}} = \pi/\text{period}$. Using Eq. (1) with $k_{\mathrm{tot}} = k_F$ and $k_{\mathrm{ZB}} = \pi/d$, where d is the layer spacing, we arrive at the desired relation: $\text{period}/d = (\pi/d)/k_{\mathrm{env}} = k_{\mathrm{ZB}}/k_{\mathrm{env}} = k_{\mathrm{ZB}}/(k_{\mathrm{ZB}} - k_F)$. Indeed, we find that the periods of 5.9 layers for Cu/Co(100) and 5 layers for Ag/Fe(100) correspond to Fermi wave vectors $k_F/k_{\mathrm{ZB}} = (1 - 1/5.9) = 0.83$ and $k_F/k_{\mathrm{ZB}} = (1 - 1/5) = 0.8$, respectively, which are very close to de Haas-van Alphen data (0.827 for Cu and 0.819 for Ag). It is interesting to note that this same periodicity has come up in theoretical treatments of the exchange coupling in superlattices [10-13], where the RKKY coupling is evaluated at discrete layer spacings. By coupling the RKKY wave vector $2k_F$ with a reciprocal-lattice vector $g = \pi/k_{\mathrm{ZB}}$, one obtains $2(k_{\mathrm{ZB}} - k_F) = 2k_{\mathrm{env}} = 2\pi/\text{period}$, which is identical to the result of the quantum well state model.

From the band structures in Fig. 4 we can also explain the spin polarization of quantum well states near E_F in Ag/Fe(100). Only the minority-spin Δ_1 bands exhibit a gap at E_F in Fe(100), allowing the formation of quantum well states in the minority Δ_1 s,p band of Ag(100). The majority states couple with majority states of the same symmetry in Fe(100) and remain a continuum of Bloch states. This agrees with the observation [16] of minority-spin states just below E_F in Ag/Fe(100). When the spin densities of a minority-spin quantum well state and a majority-spin Bloch state are combined, one obtains an oscillatory spin density with a period equal to half the wavelength of the quantum well state, which is the same as the period in the appearance of quantum well states with increasing film thickness. Such an oscillating spin density will surely play an essential role in the oscillatory magnetic coupling through the noble metal.

How do we proceed from the picture of spin-polarized quantum well states to a quantitative description of the oscillatory magnetic coupling in superlattices? It is obvious that such states will mediate magnetic coupling, but for quantitative information we will need the coupling matrix element. In addition, an integration over all momenta parallel to the interface will be required, while we have considered only the critical point $k^{\parallel} = 0$ here. Without getting into these details, we can test some predictions of such a model. For example, the periods of the magnetic coupling in Cu/Fe and Cu/Co superlattices have been found to be equal, but with opposite phases [5,6]. Equal periods are expected if Cu has the same structure in both cases. The opposite phases should be traceable to the different boundary conditions at the interfaces. For example, Fe has mostly majority-spin states at the Fermi level, and may give rise to minority-spin quantum well states in Cu (compare Fig. 4), while Co has only minority-spin states at the Fermi level, thus reversing the spin of the quantum well states.

In summary, we have found quantum well states for Cu/Co(100) and Ag/Fe(100) that exhibit characteristics connecting them with oscillatory magnetic coupling, i.e., their periodicity and their spin polarization. We hope that this input will provide an intuitive understanding of the electronic states that mediate magnetic coupling across noble metals, and that such insight will foster the development of a quantitative theory of magnetic coupling in superlattices.

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