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d-like quantum-well states in (111)-oriented metallic overlayers on Fe and Co

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(111)-oriented metallic overlayers of Cu, Au, Pd, and Pt with thicknesses up to 3 atomic layers (AL) on Fe(110) or Co(0001) show evidence for *d*-like quantum-well (QW) states in spin-resolved photoemission spectroscopy. The noble-metal *d*-like QW states are not spin polarized and are well below the Fermi level. In contrast, the *d*-like QW states of Pd and Pt approach the Fermi level for about 3 AL, but are spin polarized only for the first AL. Thus both types of QW states do not mediate the antiferromagnetic interlayer exchange coupling in corresponding (111)-oriented magnetic multilayers.

During the last years enormous interest has been paid to the study of magnetic multilayers consisting of alternating paramagnetic and ferromagnetic layers. This interest has been spurred by the multitude of new magnetic phenomena such as perpendicular magnetic anisotropy,[1,2] induced magnetic polarization,[3–5] and oscillatory interlayer exchange coupling.[6] Recently, it has been pointed out[7,8] that spin-polarized quantum-well (QW) states in noble-metal overlayers on Fe(100) or Co(100), which are derived from *sp* electrons of the noble metal, mediate this oscillatory interlayer exchange coupling in corresponding (100)-oriented magnetic multilayers. However, the interlayer exchange coupling in (111)-oriented magnetic multilayers such as molecular-beam-epitaxy-grown Cu/Co(111) multilayers is not yet well understood. A well-defined oscillatory coupling as in (100)-oriented Cu/Co multilayers has not been found.[9] There is evidence for an oscillatory coupling of only a few percent of the sample from spin-polarized neutron reflection.[10]

Prompted by these observations we have studied the electronic structure of (111)-oriented overlayers of Cu, Au, Pd, and Pt on Fe(110) and Co(0001) substrate films by spin- and angle-resolved photoemission. Our attention has been focused on the existence of QW states in such (111)-oriented overlayers. We have found in normal emission evidence for QW states derived from *d* electrons of the overlayer. Moreover, the spin polarization and the thickness-dependent binding energy of these *d*-like QW states have been examined in order to clarify whether they can mediate the antiferromagnetic interlayer exchange coupling in corresponding magnetic multilayers.

The experiments were performed with two different experimental setups. The apparatus used for spin-resolved photoemission is described elsewhere.[11] Unpolarized vacuum ultraviolet light of a noble-gas resonance lamp with photon energies of 21.2, 16.85, and 11.83 eV is used. The energy and angular resolution of the system are 200 meV and ±3°, respectively. Spin analysis of the photoelectrons is done with a 100-keV Mott detector. The base pressure of the chamber (1×10^{-10} mbar) rose to 5×10^{-10} mbar during electron-beam evaporation. The film quality and the growth mode was examined by low-energy electron diffraction (LEED) and Auger electron spectroscopy (AES). Additional angle-resolved photoemission experiments without spin analysis were performed at the TGM3 beamline at BESSY, Berlin.[12]

Following previous work,[13,14] thick Fe or Co films were evaporated onto a W(110) single crystal held at an elevated temperature (T=450 K for Fe and T=400 K for Co) with deposition rates of 2 Å/min. The film thickness was measured with a calibrated quartz microbalance. Fe and Co films grow epitaxially with bcc(110) and hcp(0001) orientation, respectively. Onto these substrates films (in the following denoted as "ferromagnetic substrates") the paramagnetic overlayers (Cu,Au,Pd,Pt) were evaporated at a rate of 0.5 Å/min, with the substrate held at room temperature to avoid interdiffusion with the ferromagnetic substrate.

The LEED patterns of the overlayers on the ferromagnetic substrate indicate the fcc(111) orientation for all studied systems. Detailed LEED and AES studies of Pd(111)/Fe(110), Pd(111)/Co(0001), and Pt(111)/Co(0001) are reported in Refs. 3 and 12. Cu(111) grows pseudomorphically on Co(0001) (1.8% misfit) as indicated by the sharp $p(1\times1)$ LEED pattern with a low background. In contrast, the LEED pattern of 1 atomic layer (AL) Au on Co(0001) displays a linear superposition of the LEED pattern of the Co(0001) and the Au(111) surface nets. The in-plane lattice constant of 1 AL Au is slightly smaller ($\approx$2%) than the corresponding bulk lattice spacing, which is attained at 8 AL. Au grows epitaxially on bcc-Fe(110) in the "Nishiyama-Wasserman" mode, which is suggested by the (1×8) superstructure visible in the LEED pattern of 1 AL Au on Fe(110).

The growth mode of Cu on Co(0001) has been studied by AES. Unfortunately, the low-energy Auger lines of Co (53 eV) and of Cu (60 eV) interfere. Thus we measured the 716-eV Co Auger line. It exhibits an exponential decrease [corresponding to $\exp(-d/\Lambda)$] with a de-

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cay length of $\Lambda = 12.4$ Å, when Co is covered by Cu of thickness d. Since this decay length has approximately the same value as the inelastic mean free path of electrons of 700-eV kinetic energy,[15] the observed exponential decrease of the Co Auger lines is at least consistent with a layer-by-layer growth. Analogously the growth mode of Au on Fe or Co has been confirmed to be consistent with a layer-by-layer growth.

In the following the photoemission spectra of the overlayer systems Pd/Fe(110) and Au/Co(0001) will be discussed, which are representative of all studied systems. Figure 1 shows the photoemission spectra of Fe(110) ($x = 0.0$ AL), which is successively covered by up to 7 AL Pd. The spectra were taken in normal emission for a photon energy $h\nu = 16.85$ eV. Moreover, all spectra were taken under identical conditions to get comparable absolute intensities. At 0.5 AL Pd coverage a first peak evolves at -1.5 eV binding energy and saturates in intensity at 1 AL Pd. With further coverage (1.5 AL) a second peak is seen at -0.7 eV, which saturates in intensity at 2 AL PD. With coverages more than 4 AL Pd the spectra resemble the spectrum of a pure Pd(111) surface.[16] Analogously the photoemission spectra of the overlayer system Au/Co(0001) are shown in Fig. 2. A first peak evolves for 0.5 AL Au coverage at -3.1 eV binding energy and saturates at 1.5 AL. At 1.5 AL Au coverage a second peak emerges at -2.8 eV binding energy and saturates at 3 AL Au. At 5 AL Au the spectrum resembles that of a Au(111) surface.[17]

All studied systems show similar behavior. Especially, a first and a second additional peak appear in the coverage-dependent spectra, which saturate in intensity at 1 to 1.5 AL and 2 to 3 AL, respectively. However, only one additional peak at 1 AL Cu is observed for Cu overlayers on Co(0001). Table I lists the binding energies of all observed peaks.

On varying the photon energy the binding energy of these peaks remains constant. Thus they do not disperse with $k_\perp$, i.e., the wave-vector component of the photoelectron perpendicular to the surface. Thus the two additional peaks of each overlayer system correspond to *two-dimensional* (2D) *states.*

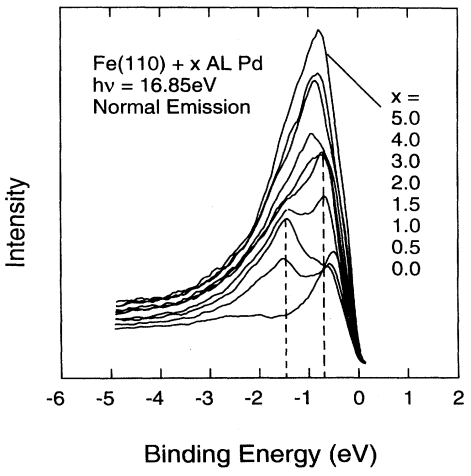

FIG. 1. Spin-integrated photoemission energy distribution curves for various Pd coverages (in atomic layers=AL) on Fe(110) plotted on the same absolute intensity scale.

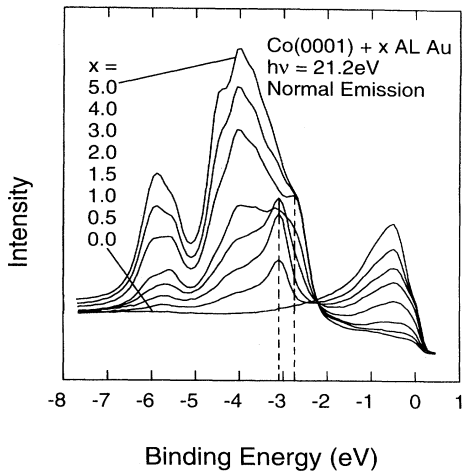

FIG. 2. Spin-integrated photoemission energy distribution curves for various Au coverages (in atomic layers=AL) on Co(0001) plotted on the same absolute intensity scale.

Moreover, the electronic character of these 2D states has been analyzed. The 2D states of the Pd and Pt overlayers have been shown to exhibit a minimum of the photoemission intensity at photon energies of about 110 and about 180 eV, respectively. This minimum coincides with the Cooper minimum of the atomic photoionization cross section for Pd-4d and Pt-5d subshells. Thus the 2D states of the Pd and Pt overlayers possess mainly Pd-4d and Pt-5d character, respectively.[12] In the case of noble-metal overlayers the big difference between the atomic photoionization cross section of s and d electrons [$\sigma_{Au}(5d)/\sigma_{Au}(6s) = 1365$; $\sigma_{Cu}(3d)/\sigma_{Cu}(4s) = 208$ at $h\nu = 21.2$ eV (Ref. 18)] suggests that the noble-metal 2D states are derived from noble-metal d electrons. Therefore all states listed in Table I possess *d-electron character.*

In order to further characterize the nature of these d-like 2D states, it is interesting to inspect the bulk band structure of both the substrates and overlayers, since there are not yet band-structure calculations available for these overlayer systems. A textbook-type example is given for Au(111) on Co(0001). According to the electronic band structure of Co(0001),[19] the 2D states of the Au overlayer lie in an energy gap of Co from -3.8 to

TABLE I. Binding energies of the quantum-well states formed by the various paramagnetic overlayers on Fe(110) or Co(0001).

Substrate film	Paramagnetic overlayer	Binding energy of the QW state for overlayer thicknesses 1–1.5 AL (eV)	2–3 AL (eV)
Co(0001)	Pd(111)	−1.3	−0.6
	Pt(111)	−1.3	−0.6
	Cu(111)	−2.5	...
	Au(111)	−3.1	−2.8
Fe(110)	Pd(111)	−1.5	−0.7
	Au(111)	−3.3	−2.7

−2.7 eV along the [0001] direction of the first Brillouin zone. Thus the Au-5*d* electrons of 1 AL Au on Co are bound in a quantum well, which is formed by the energy gap at the Au/Co interface on the one hand and by the potential step of the vacuum/Au interface on the other hand. Therefore the 2D state of 1 AL Au is most likely a *d-like quantum-well state.* Increasing the overlayer thickness the binding energy of this QW state at −3.1 eV is shifted to −2.8 eV. Such a behavior has been observed before in the case of *sp*-like QW states.[7,8] This binding energy versus thickness relationship of a QW state was shown to map in first approximation the bulk band structure of *sp* electrons, with the wave vector approaching that of the zone boundary with increasing overlayer thickness. If in first-order approximation this model is adopted for our *d*-like QW states, the QW state will follow the Au *d* band, which disperses upwards from the Γ point to the zone boundary.[17] Thus the binding energy of the quantum-well state shifts upwards from −3.1 to −2.8 eV in going from 1.5 to 3.0 AL Au. At higher Au coverages another binding energy of the QW state would be expected near −2.5 eV. But this binding energy lies outside the energy gap. Therefore at coverages more than 4 AL the quantum-well state disappears and the electronic structure of the Au overlayer converges to that of the bulk.

The 2D states of the other overlayer systems do not lie in an energy gap of the Fe or Co substrate. In spite of this different Hartree-Fock potentials of the overlayer and the substrate can cause a potential step at the overlayer/substrate interface, which together with the potential step at the vacuum/overlayer interface form a quantum well. Moreover, different symmetries of the 2D states and the electronic states of the substrate could establish a potential barrier. Such a "symmetry gap" prevents electrons of the overlayer from propagating into the substrate. In general, the confinement of the overlayer electrons will depend on the reflection coefficients at each interface, which may even be spin dependent. From this point of view, all 2D states observed in the different overlayer systems can be termed QW states.

Recently, the spin polarization of *sp*-like QW states of Ag and Cu overlayers on Fe(100) and Co(100) substrates, respectively, has been examined using spin-resolved photoemission.[8] These QW states are negatively polarized, i.e., show a confinement of mainly the spin-down component. Moreover, they cross the Fermi level with increasing overlayer thickness. Because of this they induce a negatively polarized spin density of electrons at the Fermi level through which the antiferromagnetic interlayer exchange coupling is mediated. In the case of *d*-like QW states of noble-metal overlayers the situation is quite different. First, they do not cross the Fermi level. Second, they are not spin polarized. The latter is proven by the spin-resolved photoemission spectrum of 1 AL Au on Co in Fig. 3. Neither a spin splitting nor a spin polarization is measurable, not even after subtraction of the positively polarized Co background. The same holds for all *d*-like QW states of the noble-metal overlayers. Therefore these QW states cannot mediate the antiferromagnetic interlayer exchange coupling in (111)-oriented magnetic multilayers with noble-metal spacers. Instead, it should be looked for *sp*-like QW states with wave vectors near the "stationary points of the Fermi surface."[20] Recent angle-resolved off-normal photoemission measurements on (111)-oriented overlayers of Au on Fe(110) or Co(0001) have indeed shown evidence for *sp*-like QW states.[21]

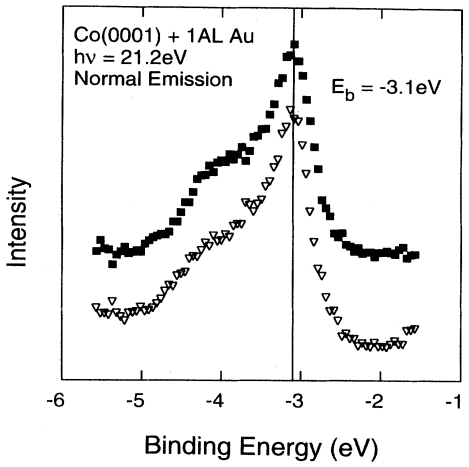

FIG. 3. Spin-resolved photoemission energy distribution curves of 1 AL Au on Co(0001). Solid symbols correspond to the majority-spin direction and open symbols to the minority-spin direction.

The QW state of 1 AL Pd or Pt is spin split, but the QW state of 2 AL Pd or Pt exhibits neither a spin splitting nor a spin polarization.[22] This hints at an induced magnetic moment of the Pd or Pt overlayer which is mainly confined to the first atomic layer.

In conclusion, in normal emission there is a difference between (100)- and (111)-oriented noble-metal overlayers on ferromagnets in that we do not observe any *sp*-like QW states crossing the Fermi level with increasing overlayer thickness in (111)-oriented systems. Instead we observe *d*-like QW states, which are located far below the Fermi level in the case of noble metals, but which cross the Fermi level in the case of Pd or Pt. In any case, there is no spin polarization for the noble-metal *d*-like QW states and only spin polarization for the first AL in the case of the strong paramagnets Pd or Pt. Thus these *d*-like QW states do not mediate the antiferromagnetic interlayer exchange coupling in corresponding magnetic multilayers.

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