PHYSICAL REVIEW B VOLUME 25, NUMBER 1 1 JANUARY 1982

Magnetic exchange splitting of one layer of cobalt deposited on top of the (111) surface of copper

R. Miranda and Felix Ynduráin
Departamento de Física Fundamental, Universidad Autónoma de Madrid, Madrid, Spain

D. Chandesris and J. Lecante
Laboratoire pour l'Utilisation du Rayonnement Electromagnétique, Orsay and Service de Physique Atomique, Commisariat a l'Energie Atomique, Gif sur Ivette, France

Y. Petroff
Laboratoire pour l'Utilisation du Rayonnement Electromagnétique, Orsay and Laboratoire de Physique des Solides, Université Pierre et Marie Curie, Paris, France

(Received 16 October 1981)

We have measured angle-resolved photoemission spectra for one and two ordered layers of Co deposited on top of a Cu(111) surface using synchrotron radiation ($25 \leq h\nu \leq 80$ eV). The spectrum at normal emission for one monolayer of Co presents the same exchange-splitting-induced three-peak structure that the spectrum of Co(0001). The magnetic-exchange-splitting values at the $\bar{\Gamma}$ and $\bar{K}$ points of the two-dimensional Brillouin zone are 0.7 and 0.5 eV, respectively.

For many years the existence of surface "dead layers," i.e., nonmagnetic surfaces of a magnetic material, was controversial. This idea was supported by the well-known Landau argument[1] showing that at nonzero temperature an infinite two-dimensional system cannot present long-range magnetic order. In the case of an actual crystal, the surface layer of atoms interacts with the rest of the atoms and therefore the Landau idea should not work. Also from the experimental point of view, spin-polarized photoemission experiments[2] show that the surface magnetization of a ferromagnetic transition metal is very similar to the bulk magnetization indicating the absence of a "dead layer." A new approach aimed at studying two-dimensional magnetic order has been focused on the study of thin layers of a magnetic material deposited on top of a nonmagnetic substrate. The main results in this direction are: magnetization measurements of Ni thin films electrodeposited on a Cu substrate from an aqueous nickel salt solution seem to suggest that the two first deposited layers of Ni are nonmagnetic.[3] On the other hand, it has been shown, using spin-polarized photoemission, that ferromagnetism takes place for Ni deposited on Cu at 1.5 monolayers,[4] whereas for Ni condensed on an amorphous substrate ($Pb_{75}B_{25}$) at 10 K, anomalous Hall effect measurements show that ferromagnetism of Ni takes place between two and three layers.[5] Recently ferromagnetic resonance has been used to show that there is not a "dead layer" at the surface of a Ni thin film deposited on top of a quartz substrate.[6] Moreover, amorphous films of Co and Fe condensed at 10 K on $Pb_{75}Bi_{25}$ present a magnetic moment after deposition of the first monolayers.[5] Finally, Mössbauer effect has been used to measure the magnetic moment of two layers of Co deposited on top of Cu and the magnetic moment found is very similar to the Co bulk value.[7]

In a recent work,[8] the deposition of Co on top of a well-ordered Cu surface has been studied using low-energy electron diffraction (LEED) and Auger spectroscopy. Results of this work indicate that Co grows in Cu at room temperature forming well ordered two-dimensional layers. Also, the possibility of one layer of Co on top of Cu being ferromagnetic was advanced based on an electronic structure calculation that gives a high density of states at E_F satisfying the ferromagnetic Stoner condition.[9] This Communication reports the magnetic exchange splitting of one and two ordered layers of cobalt deposited on top of Cu(111) measured using angle-resolved photoemission and synchrotron radiation.

The radiation source was the Orsay 540-MeV storage ring. A 127° cylindrical analyzer was used for energy analysis of the photoemitted electrons, the energy resolution being 150 meV for both monochromator and analyzer. The analyzer can rotate in the plane of incidence of the light to allow angle-resolved photoemission studies with an angular resolution of $\pm 0.5°$. The incident light can be *s* polarized (electric field in the surface plane) or *p* polarized (electric field normal to the surface plane) with a flux of 4×10^{12} photons s^{-1} at 100 eV. For these conditions, typical count rates of 10^4/sec for the *d* band of clean Cu were obtained. The substrate [Cu(111) surface] was carefully cleaned by cycles of argon bombardment (6

$\mu A/cm^2$, 30 min, 500 eV) and annealing (800 K, 5 s). The ordering and cleanliness of the Cu(111) surface, before the Co evaporation, was controlled by LEED and Auger electron spectroscopy (AES). Cobalt was deposited on top of the Cu surface from a thoroughly outgassed high-purity filament. Cobalt does not diffused into the Cu substrate at room temperature and grows layer by layer in registry with the substrate. We have shown previously[8] that the amount of Co deposited can be obtained from the relationship between the peak-to-peak heights of the *MVV* Auger peaks of both substrate and absorbate. Using this calibration we calculate the coverage of the grown films of cobalt to be 0.9 ± 0.2 and 2.1 ± 0.2 monolayers, respectively. The pressure in the chamber was always lower than 1×10^{-10} Torr even when the evaporation of Co took place.

Figure 1 shows two photoemission spectra for electrons with $k_{\parallel}=0$ ($\bar{\Gamma}$ point in the surface Brillouin zone) taken at a photon energy of 25 eV and using *s*-polarized light. The dots indicate the raw experimental data corresponding to one (0.9 ± 0.2) and two (2.1 ± 0.2) monolayers of Co deposited on Cu(111).

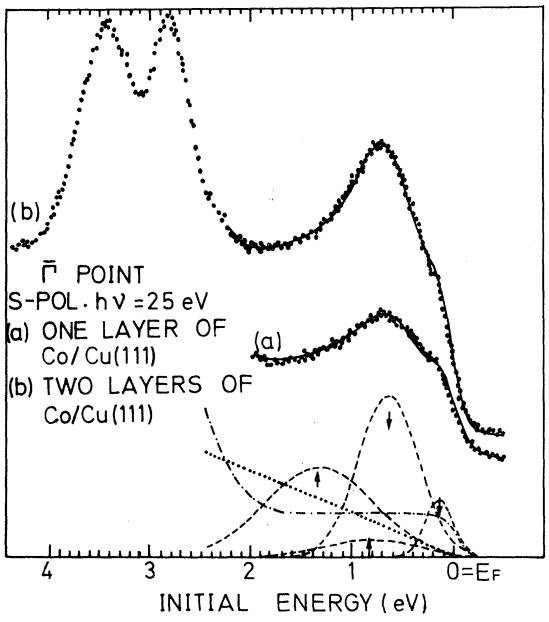

FIG. 1. Angle-resolved energy distribution curves (EDC) of photoelectrons taken at normal emission, using *s*-polarized light. The photon energy is 25 eV. Experimental results for (a) one (0.9 ± 0.2) layer of cobalt on Cu(111) and (b) two (2.1 ± 0.2) layers of cobalt on Cu(111), are shown. The continuous lines show the fit of the experimental data with the following contributions: four Gaussians for the exchange split bands (the lower pair being more intense), the *s*-*p* band of Cu and a secondary electron background. The arrows indicate spin-up (-down) subbands. The same set of Gaussians properly weighted fits the one (0.9) layer and two (2.1) layer spectra.

In Fig. 1 we first identify the peaks at 2.8 and 3.4 eV below the Fermi level as being due to the underneath Cu *d* bands, which are almost identical to those obtained in clean Cu.[10] Notice also that the presence of Co at the Cu surface induces a high density of electronic states near the Fermi level. Although the height of the peaks at -0.2 and -0.7 eV depends indeed on whether we have one or two layers of Co, their position in energy does not change. Moreover, their position, as well as the flat region around -1.8 eV, corresponds to the structures observed in the case of the single crystal Co($\overline{0001}$) surface.[11,12] (It is very important to indicate that the presence of the small peak at -0.2 eV in our spectra is a direct indication that we are dealing with well-ordered Co layers since Heimann *et al.*[12] found that this peak disappears when the Co sample is polycrystalline.) The interpretation of these features is easy since an electronic structure calculation[8] of the Co-Cu(111) system shows that, if Co were nonmagnetic its spectrum should display a two peak structure like Cu *d* bands, but if Co is magnetic, the exchange splitting induces a three peak structure, where the middle peak is higher because the exchange splitting (~ 0.7 eV) is very similar to the separation of the nonmagnetic *d* bands (~ 0.7 eV), making the spin-down low-energy peak overlap with the spin-up high-energy peak. From the above discussion we conclude that *one layer of Co on top of Cu(111) presents a magnetic exchange splitting which is very similar to that of bulk Co.*[11] To get the exchange splitting we have assumed a rigid-band model with nearly the same splitting for up- and down-spin electrons in such a way that the shift at the energy levels due to the magnetism depends only on the local magnetization and on a *k*-averaged electron-electron interaction. The magnetic energy levels are given by[9]

$$E_{\uparrow}=E_0+U\langle n\downarrow\rangle\ ,$$
$$E_{\downarrow}=E_0+U\langle n\uparrow\rangle\ , \tag{1}$$

where E_0 are the nonmagnetic energy levels, U is the intrasite electron-electron Coulomb interaction and $\langle n\uparrow\rangle$ ($\langle n\downarrow\rangle$) indicates the number of electrons with spin up (down). If the local magnetization is nonzero ($m=\langle n\uparrow\rangle-\langle n\downarrow\rangle$) then $E_{\uparrow}\neq E_{\downarrow}$ and the splitting of the bands is $\Delta E=E_{\downarrow}-E_{\uparrow}=Um$.

We have also measured the photoemission spectrum at the corner of the two-dimensional Brillouin zone ($\bar{K}$ point) for one monolayer of Co deposited on Cu(111). Results of our measurements for $h\nu=40$ eV and *s*-polarized light are shown in Fig. 2 in the energy region corresponding to Co states. A theoretical calculation[8] of the electronic structure of the Co-Cu(111) system indicates that if Co were nonmagnetic the spectrum should be a single peak. However, we notice in Fig. 2, in addition to the main peak at -0.3 eV, a shoulder at ~ -1.0 eV. We have fitted

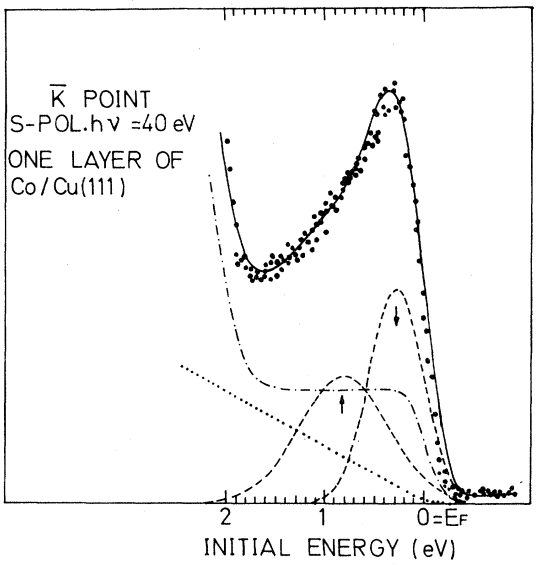

FIG. 2. Energy distribution curve of photoelectons emitted at an angle of $\theta = 30°$ with respect to the normal using s-polarized light and a photon energy of 40 eV. The EDC corresponds to the $\bar{K}$ point in the two-dimensional Brillouin zone. The experimental data corresponding to 0.9 ± 0.2 layers of cobalt on Cu(111) are shown as dots. The continuous line corresponds to the fit of these data with the following contributions: two Gaussians of the same area corresponding to up and down spin electrons of Co (broken lines), the s-p band of underneath Cu (broken-dotted line) and a secondary electron background (dotted line).

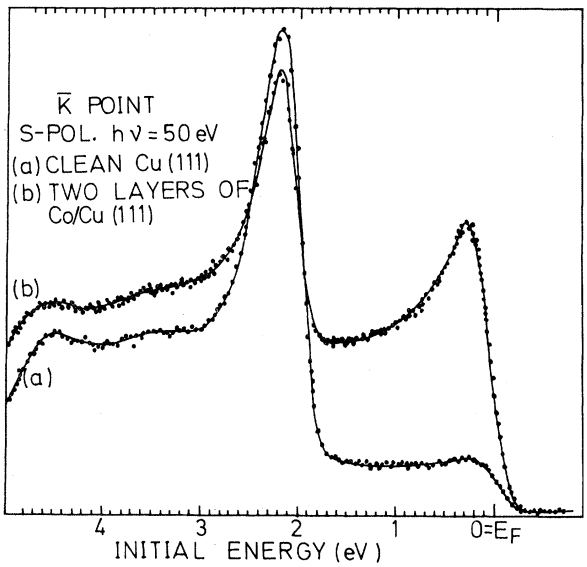

FIG. 3. Energy distribution curves of photoelectrons emitted at an angle of $\theta = 26.5°$ with respect to the normal using s-polarized light and a photon energy of 50 eV. The EDC's correspond to the $\bar{K}$ point in the two-dimensional Brillouin zone for (a) clean Cu(111) surface and (b) two (2.1 ± 0.2) layers of cobalt on Cu(111).

this spectrum with two Gaussians of the same area which would correspond to spin-down and -up electrons, respectively. Since the spin-up band is always much wider than the spin-down band[11,13] due to increased lifetime broadening, its identification is difficult (see also Fig. 1). The exchange splitting in this case, is 0.5 eV instead of being 0.7 eV as at the $\bar{\Gamma}$ point. The change in the value of the exchange splitting as we move along the Brillouin zone emphasizes the fact that Eq. (1) is indeed a first-order approximation.

We mentioned above that the d bands of Cu at the $\bar{\Gamma}$ point are essentially unperturbed by the presence of Co. In a previous paper[8] we concluded that the interaction between d states of Co and Cu is weak. To show this we have heated the Co-Cu(111) system until complete diffusion of Co into bulk Cu takes place (the absence of Co near the Cu surface can be checked by Auger spectroscopy) and we have compared the photoemission spectra obtained for two layers of Co on top of Cu(111) and for a clean Cu(111) surface obtained in this way. Results of our measurements for the $\bar{K}$ point at a photon energy $h\nu = 50$ eV using s-polarized light are shown in Fig. 3. The figure is quite transparent. The single peak of the d bands of Cu at -2.2 eV does not change with the presence of Co indicating a weak interaction between Co and Cu. Another way to see how Co interacts with the Co substrate is to look at the variation in energy of the Co peaks at a given point of the two-dimensional Brillouin zone as a function of the incident photon energy. We have found that the energy position of the prominent Co peak (at both $\bar{\Gamma}$ and $\bar{K}$ points) changes less than 0.3 eV when $h\nu$ varies from 25 to 80 eV. This indicates a small dispersion perpendicular to the surface and therefore a weak interaction with the substrate, since it is found that for a Co single crystal[11] the dispersion of the prominent Co peak at the $\bar{\Gamma}$ point is 0.6 eV when $h\nu$ varies from 9 to 30 eV.

We can summarize our work in the following points:

(i) The deposition and growth of Co on top of Cu(111) can be properly characterized by LEED and Auger spectroscopy indicating no diffusion of Co at room temperature and a well-ordered layer-by-layer growth.

(ii) Angle-resolved photoemission results indicate that one layer of Co at the Cu(111) surface presents magnetic exchange splittings of 0.7 and 0.5 eV at the $\bar{\Gamma}$ and $\bar{K}$ points of the two-dimensional Brillouin zone, respectively.

(iii) The interaction between Co and Cu is rather weak indicating that Co on top of Cu behaves like a quasi-two-dimensional transition metal.

The quasi-two dimensionality and the magnetic exchange splitting of the Co deposited on top of Cu has

important consequences for the Co-Cu(100) system where the observed $c(2\times2)$ reconstruction was interpreted as a magnetic induced charge density wave.[8] Although the magnetism of a (100) layer of Cu is less likely to occur than in a (111) layer, because there are less Co nearest-neighbor atoms, the reported results on the Co-Cu(111) system are very encouraging for performing the same analysis at the Co-Cu(100) system where a ground state with two simultaneous broken symmetries (spin and charge) may exist.[14]

We are indebted to J. M. Rojo for very useful discussions.

[1]L. D. Landau, Phys. Z. Sowjetunion 11, 26 (1937).
[2]H. Alder and M. Campagna, Phys. Rev. B 8, 2075 (1973); M. Landolt and M. Campagna, Phys. Rev. Lett. 38, 663 (1977).
[3]L. Liebermann, J. Clinton, D. M. Edwards, and J. Mathon, Phys. Rev. Lett. 25, 232 (1970).
[4]D. T. Pierce and H. C. Siegman, Phys. Rev. B 9, 4035 (1974).
[5]G. Bergmann, Phys. Rev. Lett. 41, 264 (1978).
[6]W. Göpel, Surf. Sci. 85, 400 (1979).
[7]V. Gradmann, K. Ullrich, J. Pebler, and K. Schmidt, J. Magn. Magn. Mater. 5, 339 (1977).
[8]L. González, R. Miranda, M. Salmerón, J. A. Vergés, and F. Ynduráin, Phys. Rev. B 24, 3245 (1981).
[9]See, for instance, A. Blandin, *Theory of Condensed Matter* (IAEA, Vienna, 1968), p. 691ff.
[10]P. Thiry, Ph.D. thesis (Université Pierre et Marie Curie, 1981) (unpublished).
[11]F. J. Himpsel and D. E. Eastman, Phys. Rev. B 21, 3207 (1980).
[12]P. Heimann, E. Marschall, H. Neddermeyer, M. Pessa, and H. F. Roloff, Phys. Rev. B 16, 2575 (1977).
[13]F. J. Himpsel, J. A. Knapp, and D. E. Eastman, Phys. Rev. B 19, 2919 (1979).
[14]C. A. Balseiro, P. Schlottmann, and F. Ynduráin, Phys. Rev. B 21, 5267 (1980).