Ferromagnetism of Thin Epitaxial fcc Cobalt Films on Cu(001) Observed by Spin-Polarized Photoemission

D. Pescia,[a] G. Zampieri,[b] M. Stampanoni, G. L. Bona, R. F. Willis,[a] and F. Meier
Laboratorium für Festkörperphysik, Eidgenössische Technische Hochschule Zürich—Hönggerberg, CH-8093 Zürich, Switzerland
(Received 29 September 1986)

The $p(1\times1)$ Co monolayer epitaxially grown on a Cu(001) substrate is ferromagnetic. The remanence magnetization of 1-monolayer and thicker films is zero, although a uniaxial anisotropy perpendicular to the plane of the films exists. Up to 400 K no variation of the magnetization as a function of temperature is observed.

PACS numbers: 75.50.Cc, 75.70.Ak, 79.60.Cn

Recent improvements[1-4] in the preparation of well-characterized epitaxial thin films have opened a new perspective in the field of magnetism in two dimensions. On the theoretical side, increasingly sophisticated methods have been devised to investigate the magnetic ground state of, e.g., a single layer of a 3*d* transition metal on a nonmagnetic substrate.[5] Experimentally, many results —some of them rather spectacular—have been reported on a variety of thin magnetic films.[6] Much of the work suffered from incomplete characterization or nonoptimal quality of the films. In fact, only a few systems are known, so far, where truly epitaxial growth of a magnetic material occurs, among them $p(1\times1)$ cobalt on copper (001). In this paper we report on the magnetic properties of perfectly epitaxially grown single-layer and multilayer films of fcc Co on Cu(001). For this system we found that the monolayer film is already ferromagnetically ordered, the magnetization curves for films up to 9 monolayers have the same shape, there is no remanence perpendicular to the film plane, and the Curie point even for the monolayer film must be much higher than 400 K, the highest measuring temperature. Briefly, the fcc Co/Cu(001) system behaves rather identically for all film thicknesses studied and shows within the resolution of the experiment no specific features of two dimensionality such as magnetic dead layers, thickness-dependent magnetic anisotropy, or strongly thickness-dependent variations of the Curie temperature. For a theoretical discussion of such effects see, e.g., Ref. 6.

The experimental technique used to study the magnetic properties of the thin cobalt films is spin-polarized photoemission. Details of the equipment are described by Campagna *et al.*[7] The magnetic field to saturate the sample is applied perpendicular to the film plane. As has been pointed out before, this field geometry is unfavorable for electron-optical reasons[8] and prevents angle-resolved spin-polarized photoemission. On the other hand, the applied field is externally variable and the sample can be forced into saturation, quite in contrast to the closed-flux geometry.[8] In addition, the measurement of the polarization perpendicular to the film plane is advantageous in view of the recently postulated strong perpendicular anisotropies predicting a remanence magnetization normal to the film plane.[9] An experiment measuring the in-plane magnetization cannot distinguish between the unmagnetized state and perpendicular remanence, a problem which has been encountered recently.[10]

Before attempting the spin-polarized photoemission experiment a complete low-energy electron diffraction and Auger spectroscopical study has been performed of the epitaxial growth of Co on Cu(001).[11] Epitaxial growth without any island formation is recognized by the fact that for a sufficiently thick Co layer the Auger spectrum consists only of the Co lines whereas the LEED pattern is indistinguishable from that of the copper substrate. The thickness calibration is done by comparison of the peak ratios of the Co and Cu lines as a function of the evaporation dose; see Fig. 1. The measured points are well fitted by a series of straight segments by use of standard reliability-test procedures.[11] Each segment represents the growth of a single monolayer. It must be concluded that the film is growing layer by layer as is already recognized in the pioneering work of Gonzalez *et al.*[12] In fact our independent calibration of the film thickness is in excellent agreement with Fig. 1 of Ref. 12. The deposition of the Co occurs by evaporation from a resistively heated Co wire. No traces of any contaminant—including carbon—were recorded in the Auger spectra and the perfectly sharp LEED pattern showed no extra spots at all beside the $p(1\times1)$ geometry.

The spin-polarized photoemission experiments were performed *in situ* at a fixed photon energy of 7.9 eV. The photothreshold of all samples was 5.0 ± 0.2 eV. With the use of a Mott detector, all electrons emitted from the sample were analyzed for their spin polarization, $P=(N_\uparrow-N_\downarrow)/(N_\uparrow+N_\downarrow)$, along the normal to the sample. $N_\uparrow$ ($N_\downarrow$) is the number of electrons with spin magnetic moment parallel (antiparallel) to the applied magnetic field. The emission from Cu is less, by a factor of 2, compared with that from Co, which has an appre-

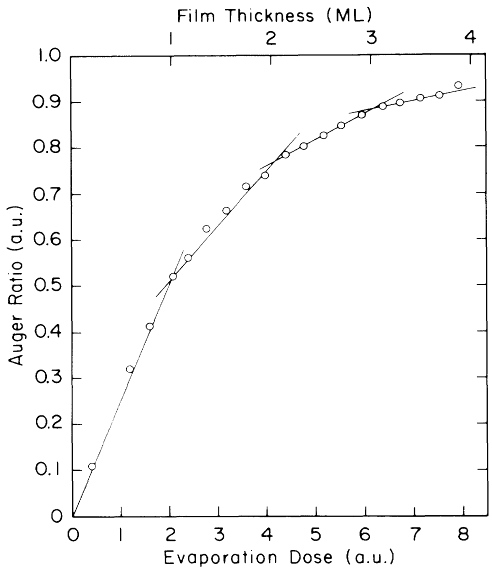

FIG. 1. Auger peak ratio $(Co_1+Co_2)/(Co_1+Co_2+Cu_4+Cu_5)$ vs evaporation dose for epitaxially grown Co on Cu(001). Co_1 and Co_2 are the amplitudes of the Auger transition of cobalt at 656 and 716 eV, respectively. Cu_4 and Cu_5 are the amplitudes of the Auger transition of copper at 845 and 920 eV, respectively. For a detailed discussion of this curve see Refs. 11 and 12. A change of slope corresponds to the onset of a new monolayer. Accordingly, a calibration of the film thickness is given in the upper horizontal scale. With use of this calibration curve the uncertainty in the thickness of the films of Fig. 2 is ± 0.2 monolayer.

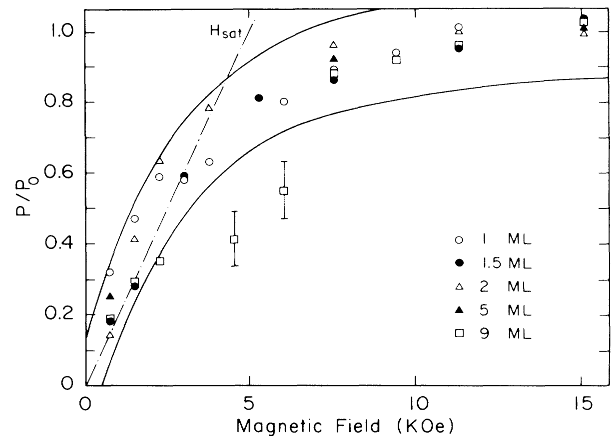

FIG. 2. Normalized polarization P/P_0 as a function of the externally applied magnetic field perpendicular to the film plane. Data are given for five film thicknesses from 1 to 9 monolayers (ML) for $T=300$ K. The saturation polarization P_0 for the various films is P_0(1 ML)=9%, P_0(1.5 ML)=9.5%, P_0(2 ML)=9.5%, P_0(5 ML)=15%, P_0(9 ML)=20%. The width of the outlined area corresponds to the average statistical uncertainty of the individual measurements. The straight line through the origin gives the initial slope of P/P_0. Its intercept with the saturation line $P/P_0=1$ occurs at $H_{sat}=5\pm 2$ kOe.

ciable density of states near E_F because of the unfilled $3d$ shell. The saturation polarization P_0 of the 1-monolayer film was 9%, of the 9-monolayer film 20%. With the assumption of a mean free path of 8 Å in cobalt, which is a reasonable value for d-electron materials at these excitation energies,[13] the reduction of P_0 with decreasing film thickness is entirely consistent with the additional unpolarized-electron emission from the Cu substrate.

The dependence of the normalized polarization P/P_0 versus applied magnetic field is shown in Fig. 2 for five different film thicknesses between 1 and 9 monolayers. The saturation polarizations P_0 of all films are given in the caption of Fig. 2. P is positive for all films at the photon energy used. Although no band calculation of fcc Co exists for the copper lattice spacing of $a=3.61$ Å, the work of Moruzzi, Janak, and Williams[14] predicts for $a=3.46$ Å negative spin polarization at photothreshold, typical of a strong ferromagnet with filled majority-spin d bands. Note, that earlier work on hcp Co gave $P=+20\%$ near threshold.[15]

The width of the outlined region in Fig. 2 is the average statistical error of the individual measurements at fixed H. The two stray points at 4.5 and 6 kOe of the 9-monolayer film are not taken into account, but their uncertainty is given, too. Except for these two points all other measurements fall inside the error region showing that the $P(H)$ dependence is the same for all film thicknesses within the accuracy of the experiment. The remanence of all films has been checked separately by sweeping the magnetic field through a complete hysteresis loop and measuring the polarization for H fields close to zero. Then, the high counting rate provides high statistical accuracy to the measurement. Any remanence polarization—if present at all—must be smaller than 1%.

Recently, it has been established by spin-polarization neutron-reflection measurements[16] that Co films thicker than 4 monolayers are magnetized in plane. Because of the fact that the field dependence of the spin polarization is the same for all films, see Fig. 2, we conclude that even the 1-monolayer film has in-plane magnetization at $H=0$. Note that the absence of remanence does not necessarily imply that the film is in-plane magnetized. Zero remanence could also occur as a consequence of stripe domain formation where the magnetization of "up" domains—with magnetization perpendicular to the film plane—is compensated by "down" domains. However, it is a well-known result of the theory of domains in perpendicularly magnetized films[17] that the films become single domain for vanishing thickness. Again, this argument clearly points to the fact that the films investigated in this work have their magnetization in plane.

For a film with only shape anisotropy the magnetization lies in plane.[18] If a perpendicular magnetic field is applied, the magnetization along **H** occurs by coherent rotation of the spins out of the film plane—without any domain motion—and it increases linearly with H. With the magnetization denoted by M_0, saturation is reached at $4\pi M_0$ which is 18 kOe for fcc Co.[19] Clearly, no such behavior is evident from Fig. 2. Bearing in mind that $P(H)$ is proportional to the normal component of $M(H)$, we point out two conspicuous features of Fig. 2. (1) The initial slope of $P(H)$ is much steeper than the shape-anisotropy-only value of $1/4\pi$ since the extrapolation of the $P(H)/P_0$ line at $H=0$ reaches 1 not at 18 kOe but already around 5 kOe. This is shown in Fig. 2 by the dash-dotted line. (2) The actual $P(H)$ dependence is not linear but rounded and approaches saturation only asymptotically.

The steep rise of $P(H)$ at $H=0$ is due to a strong anisotropy component perpendicular to the plane of the film. However, it is just not sufficient to bring about a nonzero remanence requiring a perpendicular uniaxial anisotropy constant $K > 2\pi M_0^2$. The rounding of the $P(H)$ curve arises naturally if the easy axis of the film—or the direction of uniaxial anisotropy—does not lie exactly along the surface normal but takes on a nonzero angle relative to it.[18] Then saturation is reached only asymptotically, i.e., for infinitely large H fields.

A rough estimate of the component of the uniaxial anisotropy perpendicular to the plane of the film can be given, with the assumption that the angle between the easy axis and the surface normal is negligibly small.[20] Then, the initial slope of P/P_0 is $(4\pi M_0 - 2K/M_0)^{-1}$. The straight line having this slope intercepts the saturation line $P/P_0 = 1$ at $H_{sat} = 5$ kOe; see Fig. 2. The uncertainty of H_{sat} is estimated to be ± 2 kOe. Then, with $M_0 = 18/4\pi = 1.4$ kOe, K becomes 9.3 ± 1.4 kOe.[2] The anisotropy field $H_A = 2K/M_0$ amounts to 13 ± 2 kOe giving rise to an anisotropy barrier $U_A = \frac{1}{2} H_A M_0$ of 0.064 ± 0.010 meV/atom. This value is comparable with the one calculated by Gay and Richter[9] for a monolayer of V at the lattice constant of silver and approximately 5 times smaller than that for Fe at the same geometry. Surprisingly, the $P(H)$ behavior—and therefore also the anisotropy—is independent of film thickness.

With regard to the temperature dependence of the polarization shown in Fig. 3 for a single-monolayer film, it is evident that P is constant up to the highest measuring temperatures. Indicated in the figure is the statistical error of each measurement. For experiments on magnetic samples, small differences in the alignment of the electron beam upon reversal of the **H** field can hardly be avoided, thus producing errors generally slightly larger than expected from counting statistics only.[21] As Fig. 3 shows, no decrease of P is observed with increasing T, pointing to a Curie point much higher than 400 K. An exact determination is not possible because of instrumental limitations—the sample is held close to the helium bath containing the superconducting coil used to generate the external magnetic field—and because around

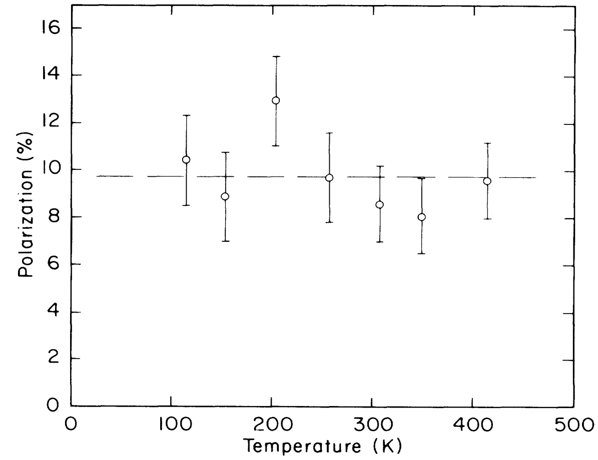

FIG. 3. Temperature dependence of the spin polarization for a 1-monolayer film measured in saturation. Applied field: 15 kOe. Indicated is the statistical error of each measurement.

600 K diffusion of Co into the Cu substrate occurs within minutes. However, the data show that the Curie point for a monolayer fcc Co/Cu(001) film is certainly far above room temperature, contrary to findings reported for other, similar systems.[6]

In conclusion, we have established that 1 monolayer of Co on Cu(001) is ferromagnetic. The polarization as function of an externally applied perpendicular magnetic field shows no remanence and is consistent with the magnetization being in plane. The Curie point even of the monolayer film is far above room temperature. The cobalt films studied present a truly epitaxial system. With the use of spin-polarized photoemission as a very direct probe of magnetism, a most surprising insensitivity of the magnetic properties with respect to the film thickness has been found even on a monolayer scale.

We thank H. C. Siegmann and M. Aeschlimann for stimulating discussions. The expert technical assistance of K. Brunner was a decisive help in carrying out this experiment. Two of us (D.P.) and (R.F.W.) acknowledge the hospitality of the Laboratorium für Festkörperphysik. The curve of Fig. 1 is published with the permission of A. Clark of Cavendish Laboratory, Cambridge, United Kingdom. The financial support of the Schweizerische Nationalfonds and the Science and Engineering Research Council (United Kingdom) is gratefully acknowledged.

[a]Permanent address: Cavendish Laboratory, University of Cambridge, Cambridge CB3 0HE, United Kingdom.

[b]Permanent address: Centro Atomico Bariloche, 8400 San Carlos de Bariloche, Río Negro, Argentina.

[1]M. Thompson and J. L. Erskine, Phys. Rev. B **31**, 6832 (1985).

[2]M. F. Onellion, C. L. Fu, M. A. Thompson, J. L. Erskine, and A. J. Freeman, Phys. Rev. B **33**, 7322 (1986).

[3]A. Amiri, G. Jennings, D. Pescia, R. F. Willis, K. Prince, M. Surman, and A. Bradshaw, Solid State Commun. **57**, 329 (1986).

[4]R. Miranda, D. Chandesris, and J. Lecante, Surf. Sci. **130**, 269 (1983).

[5]A. J. Freeman, C. L. Fu, S. Ohnishi, and M. Weinert, in *Polarized Electrons in Surface Physics,* edited by R. Feder (World Scientific, Singapore, 1985).

[6]U. Gradmann, Thin Solid Films **126**, 107 (1985); *Dynamical Phenomena at Surfaces, Interfaces, and Superlattices,* edited by F. Nizzoli, K.-H. Rieder, and R. F. Willis (Springer-Verlag, Berlin, 1985), Vol. 3.

[7]M. Campagna, D. T. Pierce, F. Meier, K. Sattler, and H. C. Siegmann, Adv. Electron. Electron Phys. **41**, 113 (1976).

[8]E. Kisker, R. Clauberg, and W. Gudat, Rev. Sci. Instrum. **53**, 1137 (1982).

[9]J. G. Gay and R. Richter, Phys. Rev. Lett. **56**, 2728 (1986).

[10]B. T. Jonker, K. H. Walker, E. Kisker, G. A. Prinz, and C. Carbone, Phys. Rev. Lett. **57**, 142 (1986).

[11]A. Clark, D. Pescia, and R. F. Willis, to be published.

[12]L. Gonzalez, R. Miranda, M. Salmeron, J. A. Verges, and F. Yndurain, Phys. Rev. B **24**, 3245 (1981).

[13]D. T. Pierce and H. C. Siegmann, Phys. Rev. B **9**, 4035 (1974). Note that the escape depths determined by the overlayer technique tend to become shorter with increasing perfection of the overlayers. For epitaxial Co, the electronic mean free path at 716 eV is 3.0 Å; see Ref. 12. For expitaxial iron electrons excited slightly above the vacuum level have a mean free path of 8 Å; see G. Fernando, Y. C. Lee, P. A. Montano, B. R. Cooper, E. R. Maag, H. M. Naik, and S. D. Bader, J. Vac. Sci. Technol. (to be published).

[14]V. L. Moruzzi, J. F. Janak, and A. R. Williams, *Calculated Electronic Properties of Metals* (Pergamon, New York, 1978).

[15]G. Busch, M. Campagna, and H. C. Siegmann, Phys. Rev. B **4**, 746 (1971).

[16]J. A. C. Bland, D. Pescia, and R. F. Willis, Phys. Rev. Lett. (to be published).

[17]C. Kooy and U. Enz, Philips Res. Rep. **15**, 7 (1960), p. 22 and Fig. 12 in particular.

[18]E. C. Stoner and E. P. Wohlfarth, Philos. Trans. Roy. Soc. London, Ser. A **240**, 599 (1948).

[19]It has been found by neutron-scattering methods that the magnetic moment per atom of hcp and fcc cobalt is identical; see Ref. 15.

[20]Denoting this angle by θ, the exact expression for the initial slope of P/P_0 is $[4\pi M_0 - (2K/M_0)\cos 2\theta]^{-1}$; see Ref. 17.

[21]G. Busch, M. Campagna, and H. C. Siegmann, J. Appl. Phys. **41**, 1044 (1970).