Oscillatory Magnetic Anisotropy and Quantum Well States in Cu/Co/Cu(100) Films

W. Weber, A. Bischof, and R. Allenspach
IBM Research Division, Zurich Research Laboratory, CH-8803 Rüschlikon, Switzerland

Ch. Würsch, C. H. Back, and D. Pescia
Laboratorium für Festkörperphysik, ETH Hönggerberg, CH-8093 Zürich, Switzerland
(Received 31 July 1995)

The magnetic anisotropy of Co films grown on stepped Cu(100) substrates is found to oscillate with varying Cu overlayer thickness. We relate these oscillations to the occurrence of quantum well states in the Cu overlayer. [S0031-9007(96)00056-7]

PACS numbers: 75.30.Gw, 75.70.Ak

Electronic states in ultrathin films can be confined by the potential barriers at the surfaces or interfaces. This confinement is connected with a quantization of the perpendicular wave vector, giving rise to sharp structures in the electronic density of states, usually called quantum well (QW) states [1]. QW states in metals have been studied with electron spectroscopies in various systems [2–4], and the results suggest that in multilayers such as Co/Cu/Co(100) s, p-like QW states in the nonmagnetic spacer layer transmit the magnetic coupling between adjacent ferromagnetic layers. Likewise, oscillations of the magneto-optical response with varying film thickness have been observed [5–9]. These results were interpreted in terms of quantum size effects. All these observations suggest that as the thickness of the involved layer is increased, the electronic band structure is periodically changed by the occurrence of QW states.

The magnetic anisotropy is caused by the spin-orbit coupling of the electrons and therefore is affected by an altered electronic band structure. Hence one can anticipate that the evolution of QW states with increasing layer thickness also leads to periodic changes in the magnetic anisotropy of a ferromagnetic film. Theoretically a periodic variation of the uniaxial magnetic anisotropy has been predicted [10], indicating that the surface anisotropy is not of a purely local origin in the top atomic layer, but is also influenced—via QW states—by atomic layers that are farther away from the surface. Similar calculations of the fourfold in-plane anisotropy seem yet untractable because of its higher-order symmetry. Up to now, no experimental observation of QW-state-induced anisotropy oscillations has been made.

Here we report on the observation of magnetic anisotropy oscillations. Upon increasing the thickness of a nonmagnetic overlayer beyond the monolayer coverage, the magnetic anisotropy continues to change. For the particular system considered in this Letter—Co films of fixed thickness grown on a stepped Cu(100) substrate—the strength of the magnetic anisotropy is found to oscillate as a function of the Cu overlayer thickness. Moreover, the anisotropy of the bare Co film can be finely tuned by choosing the appropriate film thickness. This leads to the striking observation that the anisotropy not only oscillates in strength but even changes sign: The easy magnetization direction switches repeatedly back and forth with increasing Cu layer thickness. These results show that the magnetic anisotropy of the Co film is influenced by the distant vacuum/Cu interface, which is usually assumed to be magnetically "inactive." As structural changes upon Cu coverage can be excluded, the most likely explanation for this oscillatory behavior is a quantum size effect—the formation of QW states in the Cu overlayer.

The substrates we chose were Cu single crystals oriented 0.1° and 3.4° off the (100) surface, their step edge direction running along the $[1\bar{1}0]$ direction. Prior to film preparation, the Cu crystals were cleaned by sputtering and subsequent annealing not exceeding 800 K. The films were grown by molecular beam epitaxy onto the Cu substrate at ≈315 K with deposition rates of 0.2–0.5 ML/min for Co and 0.05–0.5 ML/min for the Cu overlayer [1 monolayer (ML) = 0.18 nm for both Cu and Co]. The absolute thickness calibration is accurate to within 15% for both Cu and Co. For the magnetic measurements the magneto-optical Kerr effect (light wavelength 633 nm) has been employed [11] to record magnetic hysteresis loops parallel and perpendicular to the preferential Cu substrate step edges during the growth of the film structure.

The hysteresis loops of the uncovered Co film differ markedly for the magnetic field H applied parallel to the step edges (along $[1\bar{1}0]$) and perpendicular to it (along [110]). For H along $[1\bar{1}0]$ a rectangular loop is found, the signature of an easy magnetization axis. Along the [110] direction, on the other hand, an intermediate axis loop consisting of two loops shifted with respect to each other is observed [12]. The preferential step direction of the substrate adds a uniaxial magnetic anisotropy to the fourfold anisotropy of the (100) surface and leads to this completely different loop shape. Such a symmetry breaking has previously been observed on Co films grown on vicinal Cu surfaces [13–15].

 0031-9007/96/76(18)/3424(4)$10.00

We define the shift field H_s as the magnetic field difference between zero field and the center of the single loops (see upper inset in Fig. 1). H_s can be determined with high accuracy and—in the case of a small uniaxial anisotropy contribution—is equal to the uniaxial anisotropy field [16]. Moreover, by determining the linear initial slope s and the saturation Kerr intensity I_s of the intermediate axis hysteresis loops, also the fourfold cubic anisotropy field, being proportional to I_s/s [16], can be evaluated. Since the determination of the slope is not as accurate as that of the shift field, very small cubic anisotropy changes may escape.

A Co film of 5 ML thickness has been grown on the 0.1° off-oriented Cu(100) substrate. During the subsequent growth of the Cu overlayer, hysteresis loops have been measured along the intermediate [110] direction. From each of these loops the shift field H_s has been deduced. The results are given in Fig. 1(a), where H_s as a function of the Cu thickness is shown. At submonolayer coverages of Cu, the Co film shows a drastic decrease of H_s. As discussed elsewhere [12], this is due to a decoration of the Co step atoms by the Cu adsorbate, which changes the electronic band structure at the step atoms.

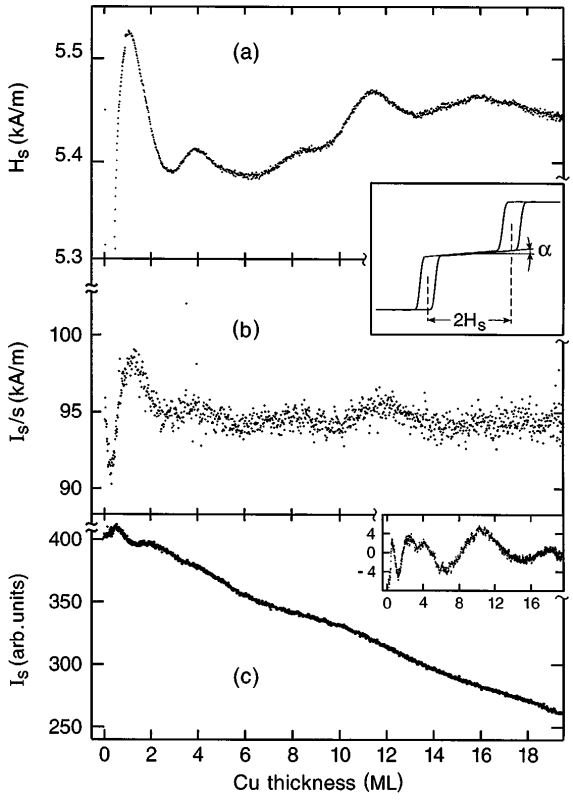

FIG. 1. (a) Shift field H_s, (b) I_s/s, and (c) saturation Kerr intensity I_s of a 5-ML Co film on a 0.1° off-oriented Cu(100) crystal as a function of the Cu coverage. The inset shows a schematic intermediate axis hysteresis loop with the definition of the shift field H_s and the linear initial slope s, with $s = \tan\alpha$. The inset in (c) shows I_s after subtraction of a fitted exponential background.

This anisotropy change appears for different adsorbates such as Cu, Ag, and O [17] and can even be strong enough to change the easy magnetization axis by 90° within the plane. For all adsorbates except Cu, H_s varies monotonously for coverages up to 3 ML.

For Cu, on the other hand, a clear increase of H_s occurs, resulting in a maximum at $d_{\mathrm{Cu}} = 1.2$ ML. Further maxima of H_s can be identified at $d_{\mathrm{Cu}} = 4.1$, 7.7, $\approx$12, and $\approx$16 ML, suggesting an average period of 3–4 ML. This oscillatory behavior was completely missed in Ref. [12], where the Cu overlayer thickness was smaller than 1 ML. Structural changes are excluded as the origin of our observations because the period is much larger than 1 ML [18]. We note that the amplitudes at large Cu coverages are extremely small, being of the order of 1 A/m. The fourfold cubic anisotropy as a function of the Cu coverage is shown in Fig. 1(b). The peaks of I_s/s are found at exactly the same positions as observed for the uniaxial magnetic anisotropy, even though the overall curve is different. These observations show that the addition of Cu, i.e., the displacement of the vacuum/Cu interface from the Co film, influences the magnetic anisotropy of the deeper lying Co film in an oscillatory fashion, even if the Co film is separated by as much as 16 ML from the Cu/Co interface.

The most natural explanation for this behavior is given by the occurrence of QW states in the Cu overlayer, which are known to be very pronounced in the Cu/Co(100) system [2]. We note that the exchange coupling oscillations observed in Co/Cu/Co(100), which are induced by QW states in the Cu spacer layer, exhibit short and long periods of 2.6 and 5.8 ML, respectively [19,20]. These values do not coincide with the period we find for the magnetic anisotropy. The magnetic anisotropy is generally determined by electronic states of the entire Brillouin zone, whereas the exchange coupling is governed only by the electronic states at the Fermi surface. Hence the corresponding QW states can give rise to different periods in experiments on exchange coupling and anisotropy. It is not obvious that a summation over the entire Brillouin zone leads to a period at all. The existence of a period in our experiment thus probably indicates that the anisotropy oscillations are determined by electronic states coming from a restricted area of the Brillouin zone, so that a similar situation as in the exchange coupling case may be realized [21].

Figure 1(c) shows for comparison the saturation Kerr intensity I_s. Clear oscillations in I_s superimposed on an exponential background can be identified. Similar structures in the magneto-optical response have been observed on different nonmagnetic overlayers [7,8] and have been interpreted in terms of spin-polarized QW states. If we compare the magneto-optical response with the anisotropy oscillations we also find no correspondence between the peak separations observed in I_s and the period of the magnetic anisotropy. This indicates that different parts of the Brillouin zone are relevant to the

magneto-optical response and the magnetic anisotropy. Moreover, the peak positions of the magneto-optical response depend strongly on the chosen wavelength [6,7].

The same experiment has also been performed for Cu/Co films on the 3.4° off-oriented Cu(100) crystal. Both I_s and H_s exhibit the qualitatively same oscillatory behavior as in the 0.1° case. In particular, the peak positions are the same. The main difference is that the absolute H_s values are higher by 1 order of magnitude. This is explained by the different miscuts of the Cu substrates, which determine the strength of the uniaxial anisotropy.

If the observed anisotropy oscillations are indeed induced by QW states in Cu, one expects them to be an interface effect. This in particular means that their position should be unaffected by the Co thickness. We therefore repeated the experiment for different Co film thicknesses, i.e., for different initial shift fields and hence different strengths of the uniaxial anisotropy.

Figure 2 summarizes the results for three Co films of different thicknesses d_{Co} and correspondingly different initial shift fields. Hysteresis loops have been measured along both the $[1\bar{1}0]$ and [110] directions for all samples. In Fig. 2, shift fields from loops measured along the $[1\bar{1}0]$ direction are plotted along the negative axis, whereas shift fields measured along the [110] direction are plotted along the positive axis. Thus a sign change indicates a change of the easy magnetization axis by 90°. The curve for d_{Co} is most remarkable: It starts with the easy axis along the $[1\bar{1}0]$ direction, switches to the [110] direction at $d_{\mathrm{Cu}} = 0.05$ ML, and flips back to its original direction at 0.5 ML. Further switches of the easy axis are observed at 2.3, 3.1, and 5.2 ML, and finally back to the [110] direction at 8.0 ML. This means that with increasing Cu overlayer thickness the easy magnetization direction in the Co film switches back and forth repeatedly between the [110] and $[1\bar{1}0]$ directions.

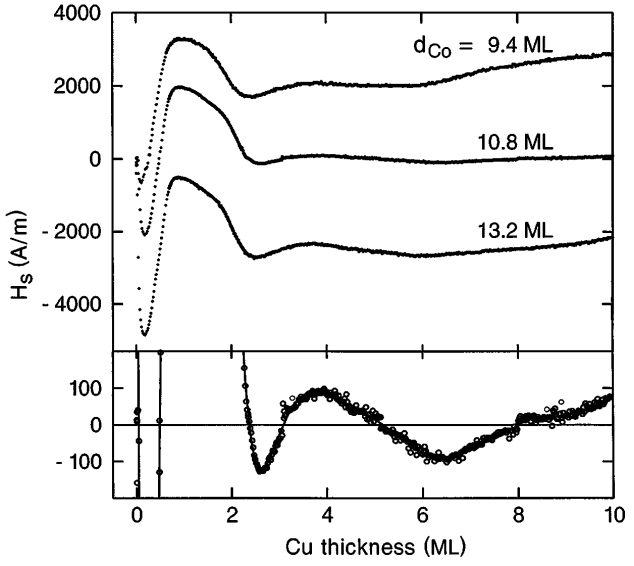

FIG. 2. Shift field H_s vs Cu coverage for three Co films on a 3.4° off-oriented Cu(100) crystal. The Co thicknesses are $d_{\mathrm{Co}} = 9.4$, 10.8, and 13.2 ML. The corresponding initial shift fields at $d_{\mathrm{Cu}} = 0$ L are 12.8, 9.3, and 7.7 kA/m, respectively. Bottom: Enlarged view of the 10.8-ML Co film to highlight the small shift fields at larger Cu coverage and the repeated easy axis switching.

The curves for $d_{\mathrm{Co}} = 9.4$ and 13.2 ML and in Fig. 2 do not show the frequent switching of the easy axis with increasing d_{Cu}. The different initial values of H_s lead to an offset of the uniaxial anisotropy compared to the curve for $d_{\mathrm{Co}} = 10.8$ ML, and hence to oscillations of H_s along the [110] and $[1\bar{1}0]$ axes, respectively. The positions of the maxima and minima, however, agree in all three curves. Thus we conclude that the influence of the Cu overlayer on the Co film is confined to the Co/Cu interface. This further supports our interpretation in terms of QW states. Since QW states are confined to the Cu overlayer they should influence only the Co/Cu interface rather than the entire Co film.

It is known that the Cu QW states have predominantly s, p character. In principle, they should therefore not affect the anisotropy significantly, as the magnetic anisotropy of transition metals such as Co is mainly governed by the d bands. However, it has been shown that the Cu s, p QW states strongly hybridize with more localized d bands [3]. Such a hybridization can thus modify the d band structure at the Co/Cu interface, and hence influence the magnetic anisotropy. As QW states evolve only at specific Cu thicknesses, their influence on the anisotropy is oscillating with thickness.

We note that the concept of QW states loses its strict meaning for very thin layers, for which the notion of interface states is more appropriate. In this context the observation of the anisotropy change or even the switching of the easy axis just above 0.3 ML Cu coverage can be taken as the evolution of an interface state or, in other words, as a precursor of a QW state at very small Cu coverage.

In conclusion, we have observed oscillatory behavior of the magnetic anisotropy by covering a stepped Co(100) film with Cu. This means that the anisotropy of the Co film is influenced by the nonmagnetic vacuum/Cu interface which is displaced by as much as 16 ML from the Co film. We interpret the anisotropy oscillations for Cu coverages above 1 ML as the evolution of QW states in the Cu overlayer, which periodically change the electronic band structure at the interface and hence affect the magnetic anisotropy of the Co film. It is hoped that these experiments will serve as input for more realistic anisotropy calculations, which would contribute to a better understanding of magnetic anisotropies.

We thank L. Bürgi for help with the experiments. Financial support by the KWF, Schweizerischer Nationalfond, and the BBW within the HCM program is gratefully acknowledged.

[1] D. M. Edwards *et al.,* Phys. Rev. Lett. **67**, 493 (1991).
[2] J. E. Ortega *et al.,* Phys. Rev. B **47**, 1540 (1993).
[3] K. Garrison, Y. Chang, and P. D. Johnson, Phys. Rev. Lett. **71**, 2801 (1993).
[4] C. Carbone *et al.,* Phys. Rev. Lett. **71**, 2805 (1993).
[5] Y. Suzuki *et al.,* Phys. Rev. Lett. **68**, 3355 (1992).
[6] W. Geerts *et al.,* Phys. Rev. B **50**, 12 581 (1994).
[7] R. Mégy *et al.,* Phys. Rev. B **51**, 5586 (1995).
[8] A. Carl and D. Weller, Phys. Rev. Lett. **74**, 190 (1995).
[9] H. A. Wierenga *et al.,* Phys. Rev. Lett. **74**, 1462 (1995).
[10] M. Cinal, D. M. Edwards, and J. Mathon, Phys. Rev. B **50**, 3754 (1994); J. Magn. Magn. Mater. **140–144**, 681 (1995).
[11] S. D. Bader, J. Magn. Magn. Mater. **100**, 440 (1991).
[12] W. Weber *et al.,* Nature (London) **374**, 788 (1995).
[13] A. Berger, U. Linke, and H. P. Oepen, Phys. Rev. Lett. **68**, 839 (1992).
[14] P. Krams *et al.,* Phys. Rev. B **49**, 3633 (1994).
[15] W. Wulfhekel *et al.,* Phys. Rev. B **50**, 16 074 (1994).
[16] W. Weber *et al.,* Phys. Rev. Lett. **76**, 1940 (1996).
[17] W. Weber *et al.,* Phys. Rev. B **52**, 14 400 (1995).
[18] A period of 1 ML with increasing Cu thickness—as in Co/Cu(100) upon Co coverage [16]—is not observed, since the growth of the Cu layer varies only the morphology of the Cu top layer. Thus the Cu/Co interface which is responsible for the magnetic surface anisotropy is not affected.
[19] M. T. Johnson *et al.,* Phys. Rev. Lett. **68**, 2688 (1992).
[20] W. Weber, R. Allenspach, and A. Bischof, Europhys. Lett. **31**, 491 (1995); **32**, 379 (1995).
[21] From our data sets we cannot exclude that more than one slightly different periods are present, since the peak positions are not perfectly equidistant.