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Letter to the Editor

Direct evidence for complete antiferromagnetic coupling between Co films epitaxially grown on Cu(1 1 1) using Pb as surfactant

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Abstract

Co/Cu films were grown epitaxially on Cu(1 1 1), using Pb as a surfactant. The exchange coupling between Co layers separated by Cu is studied with magnetic circular dichroism in valence band photoemission. Direct evidence for complete antiferromagnetic coupling is observed at 4 monolayer Cu thickness for Co films both with perpendicular and in-plane remanent magnetization from the sign reversal of the dichroic asymmetry.

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Magnetic circular dichroism in ultraviolet photoemission has proven to be extremely useful for the study of the valence band structure of ferromagnetic materials [1]. Magnetic dichroism is the change in photoemission intensity that arises when the magnetization direction or, in circular dichroism, the helicity of the exciting light is reversed. It is thus a differential technique, allowing the

precise identification of magnetic contributions in a spectrum. The low kinetic energy of the photoelectrons (of the order of 10 eV) makes it extremely surface sensitive. Typical attenuation lengths are around 0.5 nm [2]. Because no spin analysis is needed, it is a rather quick method for the investigation of effects related to the spin of valence electrons.

Antiferromagnetic (AF) coupling between adjacent layers of a ferromagnet through a non-magnetic spacer layer may be unambiguously identified taking advantage of magnetic circular dichroism.

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Because of the surface sensitivity in most cases only the signal from the topmost magnetic layer will be detected; a signal arising from the next magnetic layer undergoes a significant attenuation upon passing through both the spacer layer and the top magnetic layer. Depositing an additional magnetic layer onto an already remanently magnetized stack of layers will reveal the direction and the type of magnetization present in the top magnetic layer only, irrespective of the relative thickness. This has the advantage over depth-averaging techniques as magneto-optical Kerr effect (MOKE) that the direction and relative magnitude of the magnetization of the topmost magnetic layer can be probed separately. Magnetic dichroism hence is a way to directly access the type of coupling between a thin magnetic film and a magnetized underlayer. The only prerequisite is that the magnetization of the underlayer remains unaffected by the deposition; for our samples however this is very likely the case, because they are remanently saturated with sufficiently strong coercive fields as judged from the MOKE hysteresis loops.

The interlayer exchange coupling and its oscillation with spacer layer thickness have been very successfully studied in Co/Cu(1 0 0) superlattices and multilayers [3, 4]. In Co/Cu(1 1 1), however, either no AF coupling at all [5] or a significantly reduced and incomplete AF coupling [6, 7] have been reported. This was attributed to the growth mode of Co on Cu(1 1 1) [8]. Deposition of Co without a surfactant results in the three-dimensional growth of a rough film of Co containing stacking defects and a considerable amount of HCP material [9]. Further growth of Cu and Co layers produces superlattices where the AF coupling is strongly suppressed by magnetic bridges across the non-magnetic spacer at low Cu thicknesses [8] or by conformal roughness for thicker Cu films [10].

Predepositing a monolayer of Pb on the Cu(1 1 1) surface prior to the growth of the superlattice effectively eliminates the stacking defects, and reduces the interface roughness [11]. Magneto-optical Kerr-effect (MOKE) measurements of surfactant-grown Co and Cu/Co layers on Cu(1 1 1) revealed that indeed the magnetic properties are controlled by the epitaxial quality of the films [12]. Specifically, surfactant-grown Co/Cu trilayers with

4 monolayers (ML) thickness of the copper spacer exhibited loops that were still unsaturated at magnetic fields of 56 mT, indicating complete AF coupling of the Co layers at that Cu thickness [12].

In this paper we use magnetic circular dichroism to directly demonstrate the complete antiferromagnetic alignment of adjacent Co layers, grown with Pb as a surfactant on Cu(1 1 1), and separated by 4 ML Cu. The antiferromagnetic alignment is shown to be present for both in-plane and perpendicular magnetization.

The experiments were carried out in a UHV-chamber with a base pressure of 1×10^{-8} Pa. The sample was a Cu(1 1 1) crystal with a miscut of less than 0.25° . It was cleaned by Ar bombardment and subsequent annealing to 900 K. Co was evaporated from a high-purity Co rod by electron bombardment, Cu and Pb from crucibles. Typical deposition rates were 0.2 ML/min, while the overall pressure in the chamber did not exceed 2×10^{-8} Pa. No surface contaminations above the Auger electron spectroscopy (AES) detection limit ($\approx 1\%$) were found. The sample temperature was kept at 490 K for Pb deposition, and 300 K for Co and Cu deposition. The quality of the growth was monitored by medium-energy electron diffraction, which was used also for the calibration of the film thicknesses. 1.5 ML of Pb were deposited on the clean Cu(1 1 1) surface prior to the deposition of Co and Cu. It was checked using AES that the complete Pb coverage was floating on the surface after deposition of each Co or Cu layer.

The magnetic characterization was carried out by MOKE. By turning the sample, longitudinal and polar Kerr measurements could be performed.

Photoemission spectra were taken at the 6.5 m normal-incidence monochromator beamline of the Berlin synchrotron radiation facility (BESSY), which offers circularly polarized UV light with a degree of polarization of about 90%. The electron spectrometer is described in detail elsewhere [13]. It was operated at a fixed pass energy of 8 eV, resulting in an overall energy resolution of approximately 200 meV. The spectra were taken in normal electron emission, with the light incident either normally (for perpendicularly magnetized samples), or under an angle of 48° to the surface normal in the $[1 \bar{1} 0]$ azimuth (for in-plane magnetized samples). In the latter case, the magnetization was

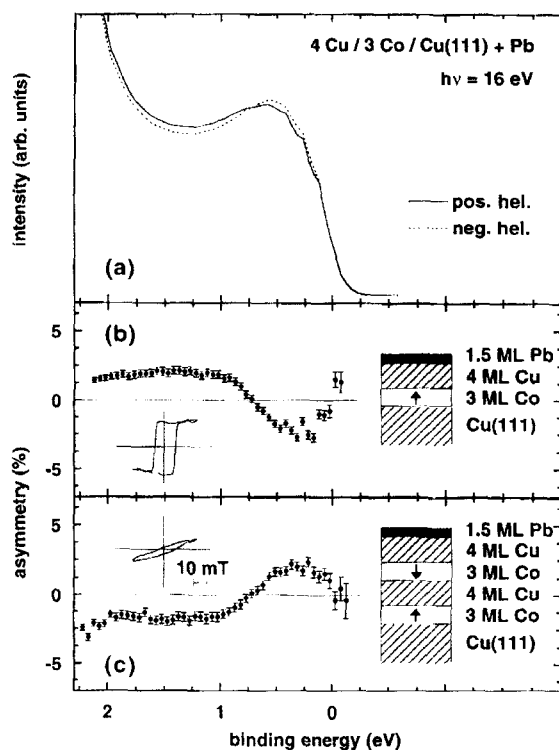


Fig. 1. (a) Photoemission intensity spectra of a 4 ML Cu/3 ML Co/Cu(111) sample, grown with Pb as surfactant, taken at normal emission and normal incidence with $h\nu = 16$ eV. Shown are the spectra for positive (solid line) and negative (dashed line) light helicity. (b) Intensity asymmetry of the spectra of (a), defined as the difference between the two spectra divided by their sum. The inset shows the corresponding MOKE loops. (c) Intensity asymmetry of a 4 ML Cu/3 ML Co/4 ML Cu/3 ML Co/Cu(111) sample, grown with Pb as surfactant, and measured after the deposition of the topmost bilayer onto the remanently magnetized 4 ML Cu/3 ML Co/Cu(111) sample. The inset shows the corresponding MOKE loops.

in the reaction plane defined by the surface normal and the incoming light.

For a Co layer of 3 ML thickness sandwiched between Cu layers and grown using the surfactant, the easy axis of magnetization is perpendicular to the surface. Polar MOKE measurements exhibited rectangular loops, as reproduced in the inset of Fig. 1b. Fig. 1a shows photoemission spectra of a 4 ML Cu/3 ML Co/Cu(111) sample for 16 eV light of positive (solid line) and negative helicity (dashed line). The peak around 0.5 eV binding energy is mainly due to emission from Co d bands. The intensity asymmetry, defined as the difference

between the two spectra of Fig. 1a divided by their sum, is depicted in Fig. 1b. A minus peak at 0.3 eV binding energy with a maximum of -2.5% is followed by a broad plus feature. This asymmetry spectrum is based on the interplay of exchange interaction and spin-orbit coupling, and depends on the sign of the magnetization, and thus the electron spin in the particular electronic states contributing to the photoemission spectra. Upon reversal of the sample magnetization, the asymmetry of Fig. 1b changes sign.

After the sample was remanently saturated, additional Co and Cu layers were deposited to give a 4 ML Cu/3 ML Co/4 ML Cu/3 ML Co/Cu(111) structure. After deposition, photoelectron spectra for both light helicities under the same conditions as before were recorded. The asymmetry of these spectra is given in Fig. 1c. It is obvious that the sign of the dichroic asymmetry has indeed changed sign, proving the antiferromagnetic alignment of the magnetization of the second Co layer with respect to the first. Furthermore, the virtually identical shape (except for the factor -1 , of course) leads us to the conclusion that the electronic structure must be the same as in the first layer. In this case, the first Co layer contributes only about 5% to the dichroism, assuming a mean free electron escape depth of 0.5 nm. This small yet not completely negligible contribution can be the reason for the slightly smaller amplitude of the asymmetry in Fig. 1c compared to Fig. 1b. Considering this, the relative size of the magnetic signal of the top magnetic layer is evaluated to be $97 \pm 7\%$ of that of the bottom layer. In other words, the antiferromagnetic alignment is complete within the experimental error. Subsequent MOKE measurements showed flat hard axis curves which are not saturated at 25 mT, as depicted in the inset of Fig. 1c.

A 6 ML film of Co has its easy axis of magnetization in the plane [12]. MOKE measurements for a single layer of these films also give remanently saturated hysteresis loops (inset of Fig. 2a). Fig. 2a shows the dichroism of a remanently magnetized 6 ML Co/Cu(111) sample. The depicted asymmetry is calculated from the difference of spectra (not shown) taken under 48° incidence with a photon energy of 24 eV. The asymmetry spectrum consists of a plus peak at 0.3 eV binding energy,

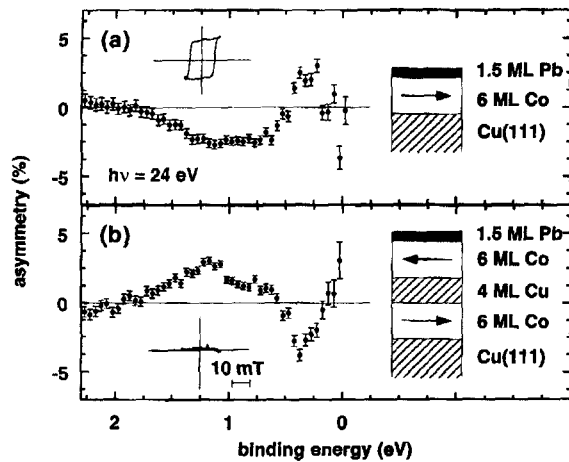


Fig. 2. (a) Intensity asymmetry of a 6 ML Co/Cu(1 1 1) sample, grown with Pb as surfactant, taken at normal emission and 48° light incidence with $h\nu = 24$ eV. The inset shows the corresponding MOKE loops. (b) Intensity asymmetry of a 6 ML Co/4 ML Cu/6 ML Co/Cu(1 1 1) sample, grown with Pb as surfactant, and measured after the deposition of the topmost bilayer onto the remanently magnetized 6 ML Co/Cu(1 1 1) sample. The inset shows the corresponding MOKE loops.

and a broader minus peak reaching from 0.6 to 1.7 eV binding energy. Another bilayer of 4 ML Cu and 6 ML Co is evaporated onto that sample. The asymmetry spectrum after deposition of the second bilayer is depicted in Fig. 2b. Because of the thicker Co layer the first magnetic layer contributes to only 1.5% to the dichroic signal in this case. Again, we observe a complete reversal of sign and an identical shape of both curves. The magnetization of the second Co layer is hence reversed with respect to the first. MOKE measurements of the complete 6 ML Co/4 ML Cu/6 ML Co/Cu(1 1 1) structure are shown as inset in Fig. 2c. Like for the out-of-plane samples, the bilayer loops constituted non-saturated hard axis loops.

Altogether, we have demonstrated that the magnetization in adjacent FCC Co films, grown epitaxially on Cu(1 1 1) and separated by 4 ML Cu, is aligned antiferromagnetically. The as-deposited second bilayer shows qualitatively and quantitatively the same magnetization and electronic structure as the first, with only the sign reversed. The AF alignment is thus complete. The easy axis of magnetization is unchanged by the deposition of an additional bilayer of Co and Cu. With the magnetic

fields accessible here the magnetization of the AF coupled samples could not be fully realigned. The exact strength of the AF coupling therefore remains to be determined.

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