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Structure and morphology of epitaxial Cu/Co bilayers grown on Cu(111) with Pb as a surfactant

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Abstract

The use of Pb as a surfactant has been shown to improve the magnetic properties of ultrathin Co/Cu heterostructures grown on Cu(111): the thickness range of Co films displaying perpendicular magnetic anisotropy is extended and a complete antiferromagnetic coupling between them is made possible. In this work, we apply low energy electron diffraction (LEED) and scanning tunneling microscopy (STM) to study the growth of epitaxial Cu/Co bilayers on the Cu(111) surface precovered by one monolayer of Pb forming a (4 × 4) superstructure, aiming to understand the origin of their magnetic properties. A quantitative comparison of experimental LEED spectra shows that the Cu layers grow in the twinned fcc structure, the amount of twinning depending on the number of stacking faults in the Co films. STM images of the capping Cu layers show not more than two atomic levels simultaneously exposed. This morphology is similar to that of Co films, which grow in the layer-by-layer mode in the presence of Pb. The result is the formation of sharp interfaces that favour the interface contribution to the magnetic anisotropy and allow the growth of layers with well defined thicknesses. © 2000 Elsevier Science B.V. All rights reserved.

Keywords: Cobalt; Copper; Lead; Low energy electron diffraction (LEED); Magnetic interfaces; Metal–metal magnetic thin film structures; Scanning tunneling microscopy; Single crystal epitaxy

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1. Introduction

Magnetic thin films and superlattices show novel phenomena such as oscillatory magnetic coupling (OMC) across the non magnetic layers and the associated giant magnetoresistance effect (GMR) that allows interesting applications in magnetic reading devices such as spin valves [1]. The Co/Cu system is one of the most extensively studied because of the similarity of the lattice parameters (mismatch is ca. 1.9%) and the possibility of growing fcc Co films by epitaxy, as demonstrated on the Cu(100) surface [2]. The growth of Co on Cu(111) is even more interesting because for thin enough films, perpendicular magnetic anisotropy (PMA) has been reported [3]. However, (111) oriented, molecular beam epitaxy (MBE) grown Co/Cu(111) superlattices show poor magnetic properties: for example, antiferromagnetic (AF) coupling of Co layers is never complete because large parts of the samples remain ferromagnetically (FM) coupled [4].

Recent work has shown that the magnetic properties of Co/Cu heterostructures grown on Cu(111) are significantly improved when the substrate is precovered by 1 monolayer (ML) of Pb,

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which acts as a surfactant. First, the range of thicknesses of Co films displaying PMA is extended [5]. Furthermore, the use of the surfactant reduces the degree of residual FM coupling between Co layers [1] and even allows a complete AF coupling for the appropriate thickness of the Cu spacer layers in MBE-grown samples [5].

It is well known that the structure and morphology affect drastically the magnetic properties in systems of reduced dimensionality. Therefore, a detailed characterization of the films grown with the aid of the surfactant is essential in order to understand the origin of the reported improvements in their magnetic properties. Pb induces the growth of Co [5] and Cu [6] on Cu(111) in the layer-by-layer mode and suppresses the twinning of the capping Cu layers [7], but a systematic characterization of surfactant-grown Cu/Co sandwich layers is still lacking. Having the two different Co/Cu interfaces, these are the simplest structures which contain all the elements present in devices even as complex as superlattices. The aim of the present work is to provide a structural and morphological study of Cu/Co/Cu(111) epitaxial bilayers grown in the presence of Pb. To this end, scanning tunneling microscopy (STM) and low-energy electron diffraction (LEED) are applied. The combination of these two surface-sensitive techniques, which are complementary in several aspects, has been shown to provide a very complete characterization of epitaxial films [8–10].

2. Experimental

Experiments were carried out in a ultrahigh vacuum chamber equipped with a home-made STM unit and a rear-view four-grids LEED optics also suitable for performing Auger electron spectroscopy (AES). The Cu(111) substrate was cleaned by cycles of ion sputtering and annealing. Deposition of Co and Cu was made from reservoirs heated by electron bombardment at rates of ca. 1 ML min^{-1}, while Pb was evaporated by resistively heating a Ta basket containing the Pb. The sample was kept at room temperature. Pb grows in the Stranski–Krastanov mode on Cu(111) [11], so a Pb coverage slightly in excess of 1 ML was deposited on the substrate prior to the Co and Cu depositions. The amount of Pb is easily determined by monitoring the low-energy AES transitions Pb_{94} and Cu_{61}. Deposited coverages of the transition metals were calculated from the ratio of the high energy AES peaks Cu_{920} and Co_{716} following the procedure described in detail in Ref. [9] and were cross-checked with similar relations for the low energy AES transitions and measurements of covered areas in STM images, whenever possible.

STM images were recorded in the constant current mode on the same surfaces subject of the LEED measurements. The latter consisted in the registration of intensity versus energy spectra (*I–V* curves) of the diffraction spots at normal incidence by means of an automated video method[12]. The Pendry *R*-factor R_P was used for quantitative comparison of experimental intensities [13].

3. Structure and morphology of bilayers 3 Cu/ *X* Co/(4 × 4)Pb/Cu(111)

Three different Cu/Co bilayers were grown on the (4 × 4)Pb/Cu(111) surface formed by the deposition of 1 ML Pb on clean Cu(111). The thickness of the Cu layers was the same for all three (3 ML) while it was varied for the Co ones: 1.3, 2.7 and 7 ML, respectively. As previously reported [5,7], Pb diffuses completely to the surface of the growing Cu and Co films. This is shown by the constant intensity of the low-energy Pb_{94} Auger transition, the presence of a (4 × 4) LEED pattern and STM images of atomic resolution. The intensity curves of the (10) and (01) diffracted LEED beams of the three bilayers are shown in Fig. 1, together with two further sets of curves for visual comparison: the curves of the (4 × 4)Pb/Cu(111) structure are displayed at the bottom and the ones for an hypothetical twinned (4 × 4)Pb/Cu(111) crystal at the top. The latter are calculated by the addition of the experimental (10) and (01) curves and simulate a Cu crystal in which both fcc twin-related stacking sequences (…abcabc… and …acbacb…), rotated by 60°, coexist with equal weights on the surface, both covered by Pb forming the (4 × 4) superstructure.

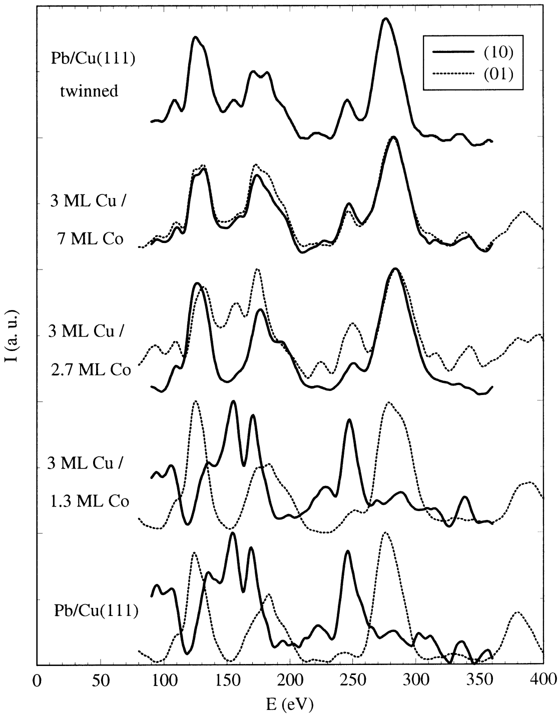


Fig. 1. Experimental LEED *I–V* curves of (10) and (01) beams of three bilayers of different Co thicknesses (Cu thickness is always 3 ML) grown on the (4×4)Pb/Cu(111) surface, whose curves are shown at the bottom of the figure. At the top, the simulated curves for a twinned, Pb covered Cu(111) crystal are displayed for comparison.

Comparing qualitatively the curves in Fig. 1, one concludes that the curves of the bilayer containing the thinnest Co film are very similar to those of the Pb covered substrate, while with increasing Co thickness, the curves resemble more and more to those of a twinned, Pb covered Cu(111) crystal.

In order to quantify the previous observation, we proceeded to fit the intensities of each of the (10) and (01) beams of the bilayers by means of linear combinations of the experimental (10) and (01) beams of the (4×4)Pb/Cu(111) structure, whereby the fraction of the complementary beam was treated as an adjustable parameter. This number also represents the fraction of the twin structure with the stacking sequence complementary to that of the substrate present in the bilayer. This approach can obviously provide only a small fraction of the information available with a dynamical analysis of the LEED intensities, but the relative extensions of the reference structures should be given correctly provided that these are well chosen, that is, are the only ones present in the unknown structure.

Table 1

Fraction of the twin fcc structure with stacking sequence …acbacb… complementary to that of the substrate … abcabca… present in the fits by linear combinations (l.c.) to the structure of 3 ML Cu/*X* ML Co bilayers grown on the (4×4)Pb/Cu(111) surface; the Pendry *R*-factors are given for the ‘best-fit’ structure

3 ML Cu/*X* ML Co/(4×4)Pb/Cu(111)			
X (ML)	1.3	2.7	7
Twin fraction (l.c.) (%)	5^{+15}_{-5}	65 ± 20	55^{+10}_{-20}
R_P (l.c.)	0.25	0.25	0.22

The results of the fits by linear combinations for the three bilayers are shown in Table 1. Error limits have been estimated by the variance of the *R*-factor [13]. The low values for R_P obtained validate the structural model in terms of Cu fcc twin domains. They further confirm that the amount of twinning increases strongly with the Co film thickness, roughly saturating already at ca. 3 ML. The agreement between experimental and calculated curves is shown in Fig. 2 for the bilayer containing 2.7 ML Co.

The morphology of the surfactant-grown bilayers is revealed by STM images like those shown in Fig. 3. They correspond to the heterostructures with 1.3 and 2.7 ML thick Co layers, respectively. The morphology is characterized by a very small roughness, with only one or two atomic levels simultaneously exposed on each single terrace. In Fig. 3a, monoatomic-high Cu islands are visible, which have nucleated on top of the previous atomic plane. In Fig. 3b, growth has proceeded in the step flow mode, that is, the adsorbed atoms have diffused to the lower part of the steps and incorporated there. The difference in the morphologies between the two images is due mainly to the different width of the terraces in the regions observed: in Fig. 3b the mean terrace width is ca. 100 Å, whereas in Fig. 3a it is several times larger. The monoatomic step visible in the centre

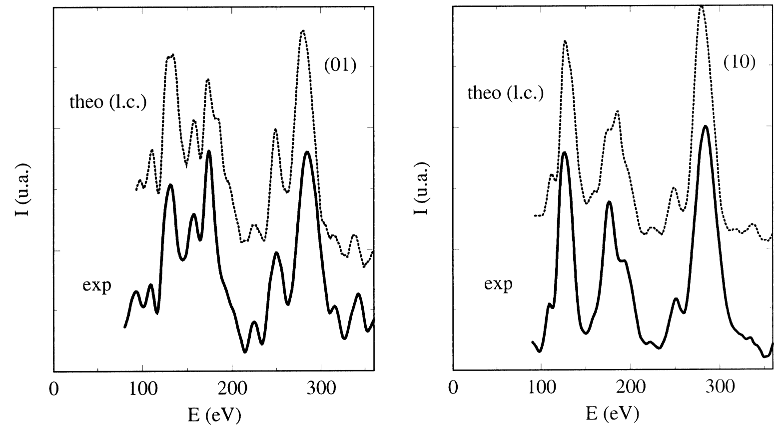
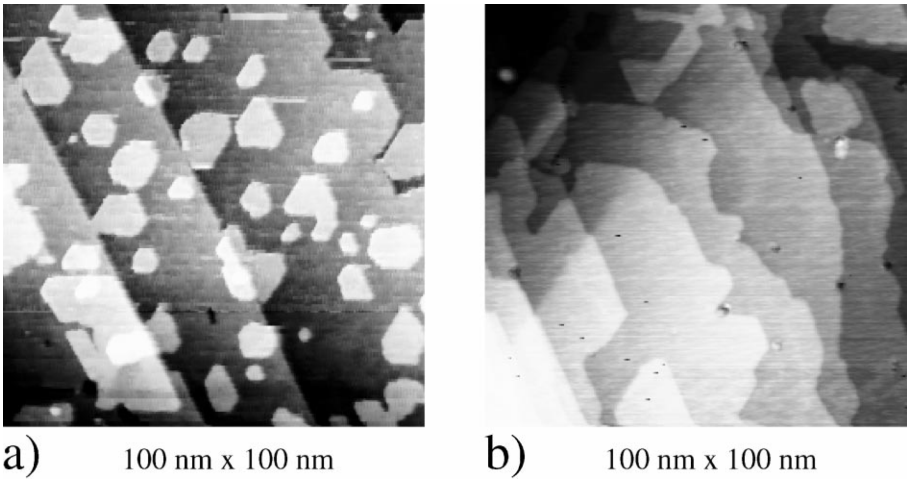


Fig. 2. Comparison of the experimental LEED *I–V* curves of the (10) and (01) beams of the 3 ML Cu/2.7 ML Co/(4 × 4)Pb/Cu(111) bilayer with the result of the best-fit linear combination of the (10) and (01) beams of the Pb covered substrate.



a) 100 nm x 100 nm

b) 100 nm x 100 nm

Fig. 3. STM images (100 nm × 100 nm) of the surfaces of the bilayers formed by deposition of 3 ML of Cu on: (a) 1.3 ML Co and (b) 2.7 ML Co films, previously deposited on the (4 × 4)Pb/Cu(111) surface.

of the image was surely developed after the deposition, probably by a tip impact, because it runs over clearly distinguishable islands and is almost perfectly straight, in contrast to the step at the upper right corner of the image.

4. Discussion

The analysis of the LEED curves points to an fcc stacking sequence at the surface of the bilayers grown with Pb. In Cu layers grown on Co films thinner than 2 ML, the fcc structure with the same stacking sequence as the substrate (say …abcabc…) dominates, while for higher thicknesses, the previous and its twin-related (…acbacb…) coexist with nearly equal weights. This can be rationalized in terms of two facts known from the growth of Co/Cu(111) without surfactant: (a) the first two Co atomic layers grow on Cu(111) mostly with the same fcc stacking sequence as the substrate [8,9]; and (b) Cu atoms deposited on close-packed Co surfaces occupy only the fcc adsorption site and grow following the *local* fcc stacking sequence

[9,10]. So growth of Cu on thick Co films, which are predominantly hcp (...ababa...) stacked [9] and present equal numbers of a- and b-terminated terraces, is expected to produce an equal distribution of the two possible fcc twin orientations.

The result for the bilayer with the intermediate Co thickness is interesting because it gives a higher contribution (65%) of the twin with stacking sequence opposed to that of the substrate itself. With the cautions required by the relatively large error bar of the result (20%), the coincidence of this number with the Co coverage in excess of 2 ML (0.7 ML) points to a third Co layer mostly presenting a stacking fault respect to the substrate's fcc structure. This requires a flat growth on top of the second Cu layer. In fact, it is known from medium energy electron diffraction experiments that the growth of Co on Pb/Cu(111) proceeds in the layer-by-layer mode from the second monolayer on [5]. Our STM images (Fig. 3) prove that the surface smoothness is preserved or even improved by the capping with Cu, with only one or two atomic levels being simultaneously exposed on each original terrace.

Our results show that the structure of the Co/Cu bilayers grown with Pb can be understood in terms of their morphology and the known structure of the bilayers without surfactant, so we conclude that the effect of the latter is mainly on the morphology rather than on the crystallographic structure of the capping Cu layers or, indirectly, of the Co layers underneath.

The net effect of the surfactant is the formation of sharp interfaces between the Co and Cu layers, leading to an increase of the contribution of the interface term in the magnetic anisotropy that favours the orientation of the magnetization perpendicular to the films. This explains the extension of the range of Co thicknesses that show the effect of PMA [5]. On the other hand, the sharp interfaces also explain the observation of complete AF coupling between the Co layers, because they allow the growth of films of well defined thicknesses. This is required for the OMC to be observed, because the oscillation period is of the order of a few monolayers and thus, a structural perfection of the layers down to the atomic level is necessary.

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