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# Tailoring epitaxial growth of low-dimensional magnetic structures by using surfactants

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**Abstract**

Ideal artificial materials such as magnetic thin films and superlattices are expected to possess unique properties owing to their reduced symmetry and dimensionality. In real systems, however, the actual behavior is extremely sensitive to the morphological features of the films and interfaces. {Co/Cu} heterostructures on Cu(111) are a prototypical example: their growth is complicated by several difficulties, such as the lack of interlayer diffusion or the appearance of stacking faults. We have been able to overcome these problems by using Pb as a surfactant during growth. In this way, ultrathin Co films and {Co/Cu} superlattices can be grown with custom-chosen properties: we can independently select to have either in-plane or out-of-plane magnetization, and ferro- or antiferro magnetic exchange coupling through the Cu spacer. The surfactant also prevents intermixing at the atomic steps, thus allowing us to grow one-dimensional structures (quantum wires). 

*Keywords:* Epitaxy; Growth; Surface structure and diffusion

## 1. Introduction

The growth of low-dimensional magnetic structures is concentrating very intense efforts, both from theoreticians and experimentalists, owing to the exotic properties expected from these systems. However, in order to realize these expectations the samples must be produced with the finest degree of control at the atomic level. Despite the success obtained in some cases, such as the observation of oscillatory magnetic coupling (OMC) in {Co/Cu} multilayers with (100) orientation [1], further progress in this field has been delayed by the appearance of contradictory results in other systems. The case of {Co/Cu} is particularly interesting; the small lattice mismatch (1.8%) and the immiscibility of these two materials make it an ideal candidate for good-quality heteroepitaxy. In fact, the studies performed on samples with (100) orientation have consistently offered reproducible results corresponding closely to the theoretical predictions [2]. This makes the results obtained for the (111) face especially surprising: superlattices grown by MBE along this direction have repeatedly failed to show complete antiferromagnetic coupling between consecutive magnetic layers [3].

We have performed thorough studies of the growth and crystalline structure of these films, showing that the unsatisfactory magnetic behavior

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was caused by structural imperfections [4]. Several types of defect appear during the epitaxial growth of Co on Cu(111): during the first stages of deposition there is some interdiffusion of the arriving Co atoms into the substrate; this is accompanied by etching of Cu atoms that leave large pools of single-atomic height vacancies. The natural steps of the substrate are decorated by clusters of irregular shape, whose growth stops after reaching a lateral size of ca. 60 Å. Then, double-atomic height islands, apparently made up of a mixture of Co and Cu, nucleate on the terraces. On higher atomic levels, the growth of pure Co continues. However, the structural similarity between the fcc-(111) and the hcp-(0001) faces facilitates the appearance of stacking faults and the adoption by the Co film of its equilibrium hcp structure. This transition is a gradual one, taking place over a thickness range of several monolayers: this means that fractions of fcc and hcp Co coexist laterally within the film [5]. The existence of a lateral displacement of a fraction of the lattice parameter between islands with different stacking sequence necessarily results in discontinuous films. Additionally, atomic steps on the Cu(111) face present large Ehrlich–Schwoebel barriers that practically suppress interlayer diffusion and force multilayer growth. Finally, capping Cu layers grown on top of these poorly ordered Co films present a high degree of twinning due to the loss of stacking coherence across the Co. Given these circumstances, it is not surprising that the {Co/Cu} heterostructures investigated so far have not displayed the expected magnetic properties.

## 2. Results

Since the conflicting magnetic behavior is associated with structural defects, one has to find techniques that allow us to overcome the natural tendencies of the materials involved. We have also demonstrated that using surfactant agents to assist growth can be such a method. In this context, a surfactant is a low-surface-energy element that is introduced in the growth process; during the deposition of the growing material, the surfactant continuously segregates to the film surface and, on doing so, modifies the growth characteristics and the resulting layer structure. In our case, by covering the Cu(111) surface with a full monolayer of Pb before depositing Co, we are not only capable of influencing the diffusivity of the arriving adatoms, but also of preventing them from nucleating at unwanted positions. The presence of Pb greatly increases interlayer diffusion and induces the appearance of layer-by-layer growth, as demonstrated by the He diffraction (TEAS) data presented in Fig. 1. In Fig. 1a, the monotonic decrease of the specularly scattered intensity indicates multilayer growth with a steady accumulation of defects at the surface of the growing film. On the contrary, using Pb results in the appearance of the well-known diffracted intensity oscillations, characteristic of layer-by-layer growth (see Fig. 1b).

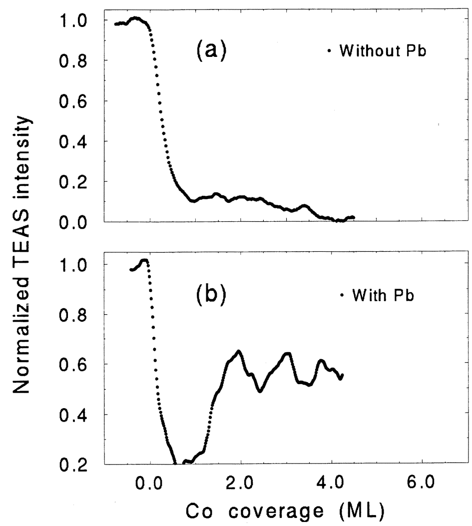


Fig. 1. The growth of Co on Cu(111) observed with He atom diffraction. (a) A Co film grown on clean Cu(111) accumulates roughness due to the lack of interlayer diffusion. (b) The use of Pb as a surfactant reduces the energy barrier at the atomic steps and induces layer-by-layer growth.

The presence of Pb also suppresses the formation of stacking-faults in the Co film, thereby delaying its transition from fcc to hcp [6]. The layers grown using this procedure are flat, continuous and homogeneous; Cu layers deposited on top of them show good-quality fcc structure, without twinning [7,8]. This process can be repeated for a number of periods to construct a crystalline superlattice.

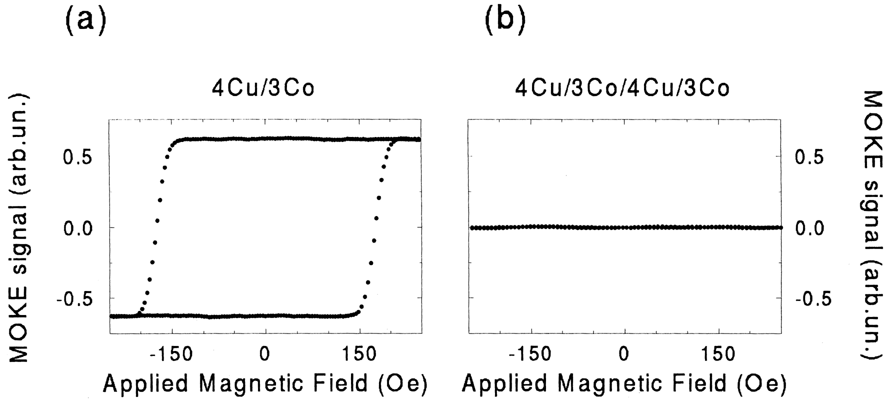


Fig. 2. Polar magneto-optic Kerr effect curves showing the magnetic behavior of a single {Cu/Co} bilayer (a) and a double one (b), grown with the aid of Pb; the absence of magnetization in the second case reveals complete cancellation between both Co layers, which are antiferromagnetically aligned.

The final result is a great overall improvement of the structural quality.

Our own investigations of the magnetic properties of {Co/Cu} heterostructures grown by means of the surfactant method confirm the decisive influence of structural parameters. The high degree of control over the growth process that we can achieve by using the surfactant permits us to choose the desired magnetic behavior. We can select the direction of the easy axis of magnetization (either perpendicular or parallel to the surface) by adjusting the thickness of the Co layers. At the same time the sign of the magnetic coupling transmitted through the Cu spacer layers is determined independently by the thickness of these spacer layers. In this way, we have been able to observe complete AFM coupling for the first time in MBE-grown {Co/Cu} samples of (111) orientation, both with in-plane and out-of-plane magnetization [7,8]. Fig. 2 shows a representative example: Fig. 2a displays the hysteresis curve obtained by means of the polar magneto-optic Kerr effect (MOKE) on a single Co layer covered by Cu; when a second {Cu/Co} bilayer of identical thicknesses is deposited on top of the first one, the MOKE signal disappears, due to the exact cancellation of the magnetization of both Co layers, antiferromagnetically coupled across the Cu spacer.

The surfactant technique also helps us to grow one-dimensional (1D) structures, the so-called "quantum magnetic wires". The objective is to be able to reach the "step-flow" regime, where all the deposited adatoms migrate across the surface and attach to the natural steps of the substrate. The presence of a full monolayer of Pb covering the Cu(111) substrate passivates the atomic steps, where most of the Co–Cu intermixing seems to occur; this has been demonstrated by previous STM experiments, showing that in the presence of Pb the substrate steps are not decorated by clusters as they are when Co is grown on a clean surface [9]. Therefore, another effect of the surfactant agent is to prevent interdiffusion: this allows us to use more elevated substrate temperatures during deposition than when no surfactant is used. Fig. 3 shows the very different behavior displayed by a Co film grown on a 4° miscut Cu(111) crystal, depending on the presence or not of Pb. When no surfactant is used, raising the substrate temperature by a considerable amount increases interdiffusion without much improvement of the film structure: islands continue to be formed on the terraces and disorder is continuously accumulated, as revealed by the steady decrease of the specularly reflected He intensity shown in Fig. 3a; we have found no indications of step-flow growth in the temperature range investigated. On the other hand, by precovering the substrate surface with Pb and carefully adjusting the substrate temperature and deposition rate, we can reach the regime of step-flow growth: the evaporated Co atoms arrive at

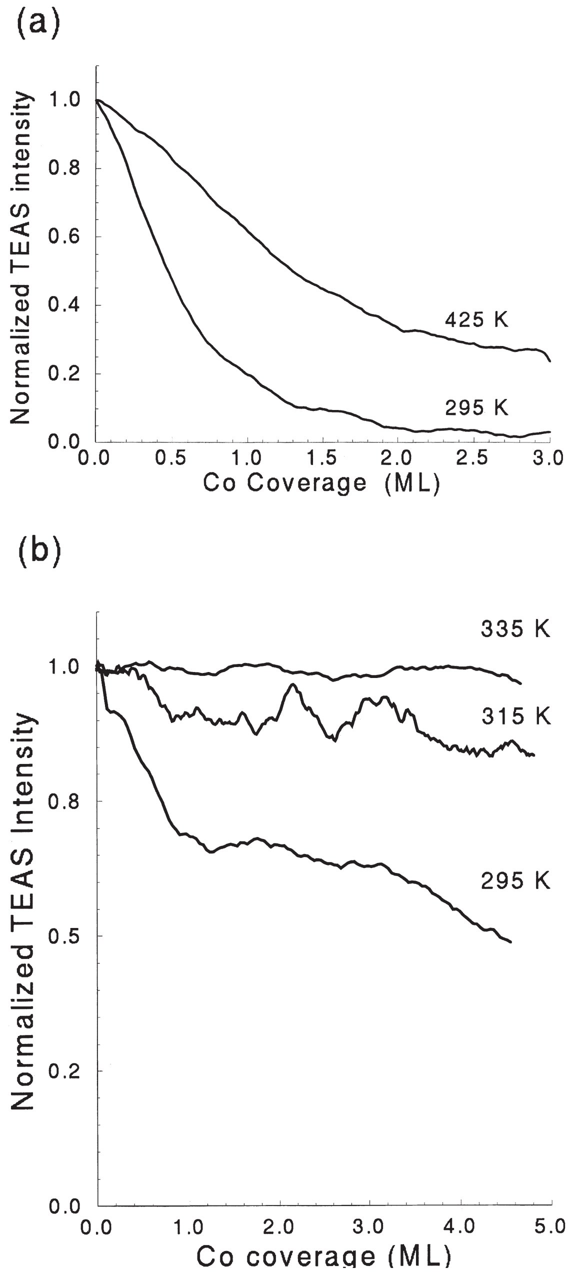


Fig. 3. Specularly reflected He atom intensity measured during the growth of Co on a 4°-miscut Cu(111) substrate. (a) Without surfactant, the existence of interdiffusion prevents us from reaching the step-flow growth mode. (b) When the substrate surface is covered with Pb prior to Co deposition, interdiffusion is suppressed. This allows us to grow at higher temperatures to facilitate the migration of Co atoms to the substrate steps, where they attach to form quasi-1D structures or "quantum wires".

the surface and diffuse until they find a step, where they attach to the last row of substrate atoms. This is demonstrated by the TEAS curves depicted in Fig. 3b: increasing the substrate temperature during Co deposition results in a higher average scattered intensity, indicating a smaller concentration of defects. At 335 K, the specular TEAS intensity remains constant during the whole deposition process: this means that no additional defects are being introduced. Also, $\theta$–$2\theta$ scans (not shown) indicate that the steps present on the surface after growth have single atomic height. Thus, this method allows us to grow Co in the submonolayer range as an array of 1D stripes parallel to the step lines, forming quasi-1D structures.

## 3. Summary

The use of surfactants is an efficient method to control epitaxial growth at the atomic scale. It allows us to overcome the natural limitations of specific systems and to manipulate the growth process in order to produce artificial structures with the desired electronic and magnetic properties.

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