Connection between Structure and Electronic Properties in Epitaxial Magnetic Layers

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ABSTRACT

This study explores the consequences of structure on the electronic properties of magnetic multilayers. Epitaxial layers of Co and Cu are grown on Cu(100) in a new deposition system that couples sputter-deposition with MBE and contains a wide range of characterization tools, including RHEED, LEED, and Kerr effect. This system can be coupled *in situ* to spin-polarized, angle-resolved photoemission and to resonant, magnetic X-ray scattering, both employing synchrotron radiation. The interface structure turns out to be critical in determining the coercivity and the presence of quantum well states, which determine oscillatory magnetic coupling.

INTRODUCTION

The emerging field of magnetoelectronics [1,2] calls for magnetic multilayer structures with dimensions in the single digit nanometer regime. Typical structures consist of two ferromagnetic layers separated by a non-magnetic spacer. Metallic spacers produce the effect of giant magnetoresistance (GMR) and insulating spacers are used for magnetic tunneling structures. In both cases the two natural magnetic orientations of the ferromagnetic layers (parallel or antiparallel) have different resistance, an effect that is being used in GMR reading heads for hard disks and for memory cells in magnetic random access memory (MRAM). The combination of Co and Cu that we study is part of commercial hard disk reading heads introduced by IBM. A third phenomenon consists of a periodic switching between the parallel and antiparallel orientation when the spacer thickness increases (oscillatory magnetic coupling).

For layers only a few nanometers thick the interfaces are starting to play a major role in determining the electronic and magnetic properties, such as GMR, magnetic coupling, and

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coercivity. Therefore, a detailed look at the influence of the growth method on the interfaces is warranted. The two primary methods of thin film growth are molecular beam epitaxy (MBE) and sputter-deposition. The first has been utilized primarily in academia, the second in industry. We have constructed a versatile growth chamber that is able to make direct comparisons between the two methods. It contains three electron-beam evaporators and three DC magnetron sputter targets for deposition. The sample is located at the end of a liquid nitrogen dewar for growth at temperatures in a rage from 100 K to room temperature. Sample heating allows deposition at higher temperatures as well. A RHEED electron gun gives real-time information about the growth from any of the deposition sources. The RHEED gun is also equipped with differential pumping, which allows continuous operation even during sputter deposition. Additional, post-growth analysis of structural characteristics is available with a LEED system. Magnetic information can be gathered *in situ* using the magneto-optic Kerr effect (MOKE) with a He-Ne laser.

A relatively clear-cut phenomenon in magnetoelectronics is oscillatory magnetic coupling. It can be understood in a framework where quantum well states in the spacer layer mediate the magnetic coupling [3,4]. They may be viewed as standing electron waves, built up by multiple reflections between the interfaces to the adjacent ferromagnetic layers. Since the interface reflectivity at a ferromagnet is spin-dependent, the quantum well states become spin-polarized [5,6] and carry magnetic information from one magnetic layer to the other, even though the quantum well states themselves reside in a non-magnetic metal, such as Cu. As in an optical interferometer, the roughness of the interfaces has to be small compared to the wavelength of the electrons in order to obtain well-defined standing waves. The relevant wavelength is that of the envelope wave function, which depends on the location of the quantum well states in **k**-space. In Co/Cu/Co(100) structures there exist two oscillation periods of 6 and 2.7 atomic layers, respectively. The long-period oscillations correspond to quantum well states derived from the belly of the Cu Fermi surface, and the short-period oscillations are derived from the neck. Inverse photoemission work on the long-period quantum well states has shown that the density-of-states oscillations that give rise to the long-period oscillations are damped by a factor of 1.8 at a stepped surface with a step spacing of 10 Å [7]. Short-period oscillations have been difficult to obtain reproducibly, because they require interfaces of atomic perfection. Therefore, they are a good criterion for optimizing the growth conditions.

GROWTH CHARACTERIZATION

For characterizing the growth of Co/Cu(100) interfaces we have prepared smooth Cu(100) single crystal surfaces by careful mechanical and electrochemical polishing, followed by a gentle sputter-anneal (500 V Ar^+ at $4 \cdot 10^{-5}$ Torr for 15-30 min). On top of such surfaces we have deposited Co and Cu, using either MBE or DC sputter-deposition. By using the RHEED and MOKE in the growth chamber along with angle-resolved photoemission in an attached chamber, we can compare the differences between the two deposition methods and also comment on the

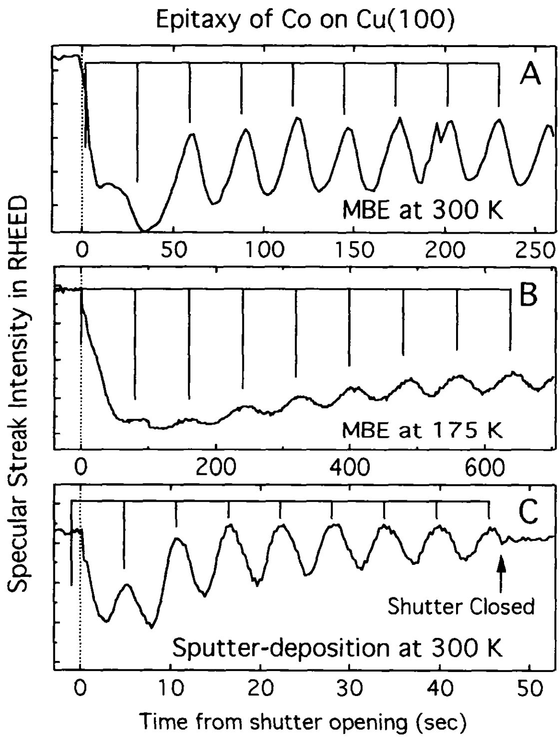

Fig. 1 RHEED oscillations for epitaxial growth of Co on Cu(100).
A) MBE with the substrate at room temperature.
B) MBE with the substrate at 170 K.
C) Sputter deposition with the substrate at room temperature.
The irregularity of the first two oscillations in A) is attributed to intermixing at the interface. It is minimized by deposition onto a cold substrate in B) and by a faster growth rate in C).

changes brought about by variations in other growth parameters such as growth temperature and rate.

The growth, and particularly the formation of the initial interface, is studied by RHEED oscillations at an electron energy of 35 keV and an angle of incidence of 2^0 from grazing along the [011] azimuth. A differentially-pumped RHEED electron gun allows us to record RHEED oscillations even during sputter-deposition. Figure 1 compares the results for different preparation methods. For a room temperature MBE deposition (A) the first two oscillations are quite irregular, leading to a minimum near the location where the first maximum is expected (compare the grid lines). The same effect is observed when reducing the deposition rate to that in (B). This irregularity is attributed to intermixing between the initial Co layer and the Cu substrate, creating a spread-out interface. Co atoms want to exchange positions with Cu surface atoms in order to preserve the low surface energy of Cu. Lowering the temperature of the substrate (B), largely eliminates the irregularities in the first two oscillations, indicating a sharper interface. However, the amplitude of the oscillations is reduced due to imperfect crystallinity. Proper recrystallization requires a subsequent anneal to room temperature. Sputter-deposited films (C) exhibit more regular initial RHEED oscillations, too. This can be attributed to the higher deposition rate. The amplitude of the oscillations remains large because the sample is at room temperature.

MAGNETIC AND ELECTRONIC PROPERTIES

The magnetic properties are characterized by MOKE in the growth chamber. The Co/Cu(100) structures generally exhibit square hysteresis loops with the magnetization along the in-plane [011] easy axis, which are indicative of single-domain switching. The coercivities H_c vary dramatically for different growth methods. Sputter-deposited films generally have the largest coercivities.

For the 8 ML sputtered film in Fig. 1C, a coercivity of H_c = 4.3 Oe is found. Coercive fields were measured over a range of pressures and rates, but no definitive trends were seen. Room temperature MBE growth, like that of Fig. 1A, typically results in coercivities around 2 Oe. Lowering the growth temperature to 170 K, as in Fig. 1B, reduces the coercivity even more, to 1.4 Oe. These values are another indication of the very high film quality, since lower coercivity generally indicates fewer pinning centers for magnetic domains. A low temperature deposition of an additional Cu layer on top of the Co, as done for the creation of the quantum well states in Fig. 3, also has a profound effect on the coercivity measured. For that 3ML Cu / 8ML Co / Cu(100) structure a record low H_c=0.7 Oe is measured. This value is an order of magnitude smaller than typical values found in the literature [8]. The low coercivities we have measured for these metastable fcc Co layers are in contrast to the larger values for the hcp phase of Co, which exhibits a large coercivity due to its anisotropy along the hexagonal axis. In fcc Co, the cubic symmetry allows only a smaller, higher-order bulk anisotropy. The highly-perfect fcc Co films grown here combine both the high magnetization of Co (which gives large GMR signals) with a low switching field, which should make them sensitive magnetic field detectors in GMR structures.

The influence of growth on the electronic structure is demonstrated in Fig. 2. These curves are angle-resolved photoemission spectra of Cu(100) at ϑ = 8° and ϑ = 23° towards the [011] azimuth, using a photon energy hν = 50 eV. A Cu(100) substrate was prepared by polishing and a light sputter-annealing as noted above. The electron distribution curve of this bare substrate is shown with full lines in the two panels. For the dashed curves, Cu was homoepitaxially sputter-deposited onto the substrate at room temperature with an Ar pressure of 2 mTorr during growth (8 Å/min). An Ar pressure of 4 mTorr gave similar results. Compared to the bare substrate, the features of the sputter deposited surface are broadened [9]. The energy

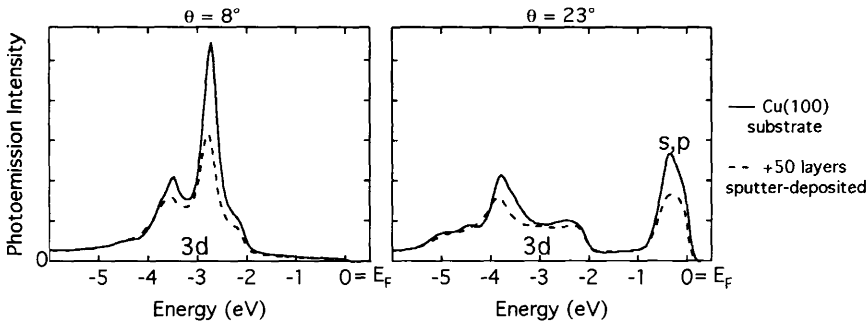

Fig. 2 Influence of growth on the electronic states. Angle-resolved photoemission spectra of a Cu(100) bulk crystal (full lines) are compared to those of an epitaxial film of 50 Cu layers sputter-deposited on top of it (dashed). The peaks are slightly broadened after sputter-deposition, indicating momentum transfer at defects. The photon energy is hν=50 eV. The two emission angles ϑ (from the samle normal) provide a look s,p- and d-bands.

broadening of the peaks is likely due to momentum broadening via scattering at defects. The momentum transfer δk is related to the scattering length l (= average spacing between defects) by the uncertainty relation $\delta k = 1/l$. Using a new generation of photoelectron spectrometers with angular multidetection it has become possible to measure δk accurately enough to derive electron scattering lengths at the Fermi level (see the work in [10]). These spin-dependent scattering lengths are relevant to magnetic transport, such as GMR.

The most sensitive indicator of the interface quality turns out to be spin-polarized photoemission from short-period quantum well states (Fig. 3). Discrete and spin-polarized states are only visible when grown by MBE onto low temperature substrates (170 K) with subsequent anneal to room temperature. The explanation for the spin-polarization of quantum well states is based on a spin-dependent electron reflectivity of the interfaces. In a simple interferometer picture,

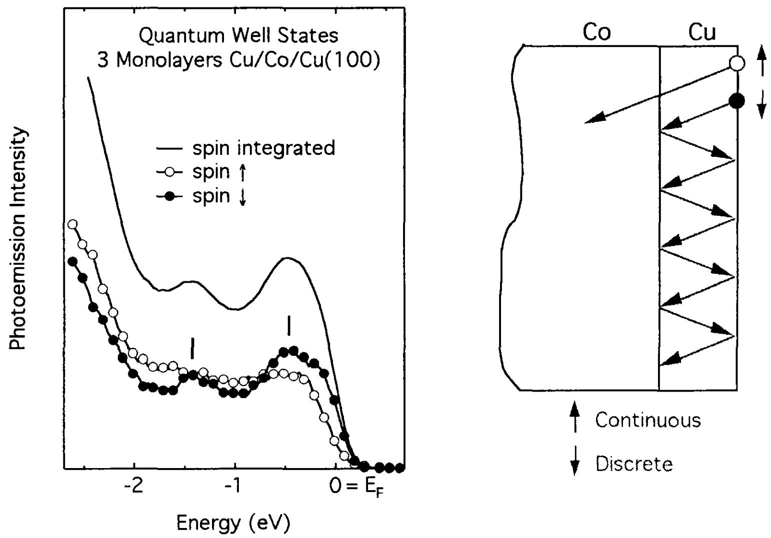

Fig. 3 Spin-resolved photoemission spectra of short-period quantum well states in a Cu/Co/Cu(100) sandwich, grown at 170 K and annealed to room temperature (hν=77 eV, ϑ= 11^0 along [011]). Minority spins are reflected at the interface and confined to the Cu layer, giving rise to peaks for discrete quantum well states (tick marks). Majority spins are transmitted and preserve the continuous, bulk-like spectrum. Such states are not visible with MBE at room temperature or with sputter-deposition, because the larger interface roughness prevents specular reflection of electrons.

quantum well states are standing waves created by reflection of electrons at the interfaces (Fig. 3, right). The reflectivity at a ferromagnetic interface differs between majority and minority spins, because of the magnetic exchange splitting of the bands. For the short-period quantum well states in Co/Cu/Co(100) there is a gap in the minority spin bands of Co. Therefore, minority spins are totally Bragg-reflected at the interface. Majority states, on the other hand, exist on both sides of the interface and are able to propagate. This leads to discrete quantum well states with minority spin character (full symbols and tickmarks in Fig. 3), superimposed on a continuum of bulk-like majority spin states (open symbols).

The GMR effect is based on spin-dependent scattering as well, either in the bulk or at interfaces. The ferromagnetic layers and their interfaces act as spin filters for electrons, based on the different scattering probabilities at the interface and in the bulk: For parallel magnetization of the two ferromagnets we have parallel spin filters with a high transmission, for antiparallel magnetization the spin filters are opposite to each other and block the current. An external magnetic field switches the configuration from antiparallel to parallel, thereby reducing the resistance. There are indications that the interface effect dominates for typical GMR devices. For example, the interface reflectivity can be modified by adding a monolayer of a magnetic material, leading to a substantial increase in the GMR effect [11].

An incisive structural tool that quantifies not only the stochiometric, but also the magnetic roughness of interfaces, is just becoming available at high-powered synchrotron light sources. The technique of resonant X-ray scattering [12] utilizes the anomalous, enhanced scattering amplitude for soft X-rays at the L-edges of transition metal ferromagnets for obtaining both the stochiometric and the magnetic interface roughness. Initial results [12] indicate that the amplitude of the magnetic roughness is smaller and that its correlation length is larger than for the stochiometric roughness. This might be understood qualitatively by the fact that the reduced number of magnetic neighbors at a rough interface causes a reduced Curie temperature for atoms at magnetic interface asperities. That will reduce their magnetic order or destroy it altogether if their Curie temperature drops below room temperature.

SUMMARY

In summary, we compare MBE and sputter-deposition of Cu and Co on Cu(100) with particular emphasis on the quality of the interfaces. The interfaces exhibiting the strongest quantum well states and the lowest coercivity were grown by evaporation at 170 K with subsequent annealing to room temperature. A natural explanation for these sharp interfaces would be that deposition at low temperature suppresses the intermixing reaction of Co atoms with the Cu substrate. After a complete Co film has been formed, annealing to room temperature recrystallizes the Co without causing interdiffusion because Cu and Co are immiscible in the bulk. Judging by the onset of the RHEED oscillations, sputter-deposited films give sharper interfaces than MBE at room temperature, due to their high growth rate, but they exhibit more defects than MBE films grown at low temperature with postanneling.

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