Quantum well states in Au/Ru(0001) and their effect on the magnetic properties of a Co overlayer

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Abstract. The magnetic properties of Co/Au/Ru(0001) as a function of thickness-dependent quantum well states (QWS) in the Au spacer layer were investigated using spin-polarized low energy electron microscopy. Au grown epitaxially on Ru(0001) at room temperature was annealed at ~300 °C. Upon annealing, Au forms micron-sized islands across stepped regions of Ru, forming local wedges of different Au thicknesses. Energy scans clearly reveal the existence of QWS in Au/Ru(0001). After depositing Co films on top of the Au, we found that the Curie temperature ($T_c$) and the spin reorientation transition (SRT) of the Co film on the Au islands are different from those of the Co film on the Au wetting layer. However, the QWS of the Au layer have no effect on the $T_c$ and the SRT of the Co film.
Electron confinement in the normal direction of a thin film leads to the formation of quantum well states (QWS). Because QWS modulate the electron population near the Fermi surface, there has been great interest during the last two decades in studying the effect of QWS on the physical properties of thin films. For example, it was found that QWS modulate a film’s stability [1]–[3], electron–phonon interactions [4], superconducting transition temperature [5], surface reactivity [6], surface diffusion [7], etc. In terms of magnetic properties, it was shown that QWS in the spacer layer of a magnetic sandwich [8, 9] are responsible for oscillatory magnetic interlayer coupling [10]. Since many magnetic properties depend sensitively on the density-of-states at the Fermi level, it has been predicted that QWS should modulate many magnetic properties of a magnetic thin film [11]. Although there have been observations of a finite size effect on the Curie temperature ($T_c$) [12] and the magnetic anisotropy [13,14] in magnetic thin films, it is difficult to extend these observations and attribute them to the general effects of QWS. Since it is very difficult to predict the magnitude of QWS effects on magnetic properties, the experimental challenge is to distinguish small variations of measured quantities from true QWS effects. This demands an experimental system that minimizes the experimental error so that a tiny change of the measured physical quantity can be unambiguously identified. For example, in studies of the QWS effects on the $T_c$ of magnetic thin films, the challenge is how to minimize the variations of the film thickness and the sample temperature so that $T_c$ differences in two different samples can be attributed unambiguously to QWS effects. In this paper, we report our study of Co thin films grown on Au/Ru(0001). We first show that Au films on Ru(0001) contain well-defined QWS. Then the $T_c$ and the spin reorientation transition (SRT) of Co films grown on top of Au(wedge)/Ru(0001) were studied using spin-polarized low-energy electron microscopy (SPLEEM). In typical vapor-deposition experiments, it is inevitable to have film thickness variations on a substrate, due to the finite size of the crucible and the finite distance between the crucible and the substrate (the beam spreads over a solid angle). As a result, Co magnetic properties (e.g. $T_c$) might show differences across an Au wedge spacer layer, due to such thickness variations. Such artifacts of subtle thickness variations could be mistaken as effects of Au QWS on the Co magnetic properties. In order to reduce such errors, we fabricated micron-sized Au wedges on step bundles of Ru(0001). On these microscopic Au wedges, deposition-geometry-induced experimental error of the Co thickness across the Au wedge is negligible. By studying $T_c$ and the SRT of Co within the microscopic field of view of the SPLEEM, in real time during the evaporation of Co onto the Au/Ru(0001) wedges, we
essentially eliminate both temperature and thickness inhomogeneity across the measured part of
the sample. We find that although the \( T_c \) and the SRT thickness of the Co film on the Au islands
are different from those of the Co film on the Au wetting layer, the thickness-dependent QWS
of the Au-wedge spacer layer has no observable effect on the \( T_c \) and the SRT thickness of the
Co film at room temperature.

2. Experiment

The experiments were performed in the SPLEEM chamber at the National Center for Electron
Microscopy of the Lawrence Berkeley National Laboratory. The base pressure of the chamber
is \( 2 \times 10^{-11} \) Torr. An Ru(0001) substrate was cleaned \textit{in situ} by repeated cycles of exposure
to oxygen followed by heating to 1800 K [15]. Epitaxial Au films were grown on top of the
Ru(0001) substrate using an electron beam evaporator with the growth pressure remaining
below \( 2 \times 10^{-10} \) Torr. During operation of the SPLEEM, a spin-polarized beam of electrons
is directed to the sample surface at normal incidence, and the reflected specular beam is
magnified in an electron-optical column to form a real-space image of the sample surface.
Both the incident beam and reflected electron beams are guided by electron-optical elements
so that the electron source and the detector can be separated in space [16]. During the Au film
growth, SPLEEM images were recorded while scanning the incident electron energy from 3.6
to 17 eV. The intensity of the elastically scattered electrons from the Au/Ru(0001) sample was
then determined by the laterally averaged and spin-integrated SPLEEM image intensity. This
reflection intensity was recorded as a function of both the electron energy and the Au film
thickness to identify the existence of QWS. For magnetic domain imaging of the Co films
on top of the Au film, a spin-polarized electron beam was directed to the sample surface.
Exploiting the dependence of reflectivity on the relative orientation of film magnetization and
beam polarization, the magnetic domain structure of the magnetic thin film is revealed in
SPLEEM images, obtained by forming pixel-by-pixel difference images from pairs of images
acquired using opposite spin polarization of the incident beam [17].

3. Results and discussion

3.1. QWS of Au films on Ru(0001)

A scanning tunneling microscopy (STM) study showed that Au atoms on Ru(0001) have
high mobility at room temperature and tend to form a pseudomorphic layer [18]. In fact,
Au/Ru(0001) has been one of the model systems for the study of the electronic structure
of Au(111) [19]. In the work reported here, electron reflection intensity was recorded during
Au film growth. Figure 1(a) shows representative energy scans at several selected Au film
thicknesses. The reflection intensity oscillates as a function of the electron energy, showing
the existence of QWS in the Au film. The QWS peak positions are plotted in figure 1(b) to
depict the evolution of QWS with electron energy and Au film thickness. The description of
QWS in metallic thin films has been well established by the so-called phase accumulation
model [20]–[22] with the quantization condition of

\[
2(k_{BZ} - k)d - \Phi = 2\pi \nu. \tag{1}
\]

Here \( d \) is the film thickness, \( k_{BZ} \) is the Brillouin zone vector, \( k \) is the electron momentum, \( \Phi \) is
the total phase gain of the electron at the two interfaces and \( \nu \) is the QWS index. Adopting the

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Figure 1. (a) Electron reflectivity from Au/Ru(0001) as a function of the incident electron energy for Au thicknesses of 2, 3, 5 and 7 ML. The four curves have been offset for clarity. The oscillations of the electron reflectivity show the existence of QWS in the Au film. (b) Black square symbols mark measured energies of the intensity peaks at different Au thicknesses. Lines are computed from equation (1). The standard deviation ($\sigma_E$) between the experimental data and the calculated result is $\sigma_E = 0.23$ eV.

well-determined Au energy band along the (111) direction [23] and assuming a linear energy dependence of $\Phi$ [24], the QWS in the $E\text{--}d$ plane can be fitted using equation (1). The solid lines in figure 1(b) show the excellent agreement between the experimental data and the results calculated using equation (1).

To achieve micron-sized Au wedges across step bundles of Ru(0001), a uniform 5 ML Au film was first deposited on the Ru(0001) substrate at room temperature and then annealed at
Figure 2. LEEM image of Au/Ru(0001) at an incident electron energy of $E = 6.0\,\text{eV}$. The viewing area is $7\,\mu\text{m}$ in diameter. Micron-sized Au islands with flat tops are formed after annealing the RT-grown sample at $\sim 300\,\text{°C}$.

~300 °C for ~20 min. We found that during annealing, the initially uniform Au film breaks into a wetting layer plus many micron-sized islands (figure 2). The islands remain stable when the film is cooled to room temperature. The island formation can be attributed to the fact that the step/terrace structure of the substrate enables a kinetic pathway to the nucleation of new film layers [25, 26]. The islands on top of stepped Ru form local wedges with the thickness difference between neighboring substrate terraces being exactly 1 ML. In fact, the formation of such local wedges on step bundles of the substrate has already been reported in the literature [25, 27, 28]. An interesting result is that the atomic steps from the Ru(0001) substrate are also visible in the Au island with the electron reflectivity varying from terrace to terrace. Figure 3(a) shows an image of a typical Au island on stepped Ru(0001). Dashed lines highlight the step edges of the Ru substrate. It is obvious that the electron reflectivity from different substrate terraces exhibits different magnitudes. This result can be easily understood from the result illustrated in figure 1(b), showing that the existence of QWS in the Au film results in oscillations of the electron reflectivity, both as a function of electron energy at fixed film thickness and as a function of film thickness at fixed electron energy. Thus, as the Au island forms a local wedge on stepped Ru, the electron reflectivity varies from terrace to terrace of the substrate. To further confirm this observation, we imaged the same island at different electron energies. Indeed, as the electron energy varies, electron reflectivity from different terraces within the Au island changes accordingly (figure 3(b)). We measured energy scans on different locations of the Au island as well as on the Au wetting layer. By comparing the local energy scan results with the energy spectra at different Au film thicknesses reproduced in figure 1(b), we can identify the exact local Au film thickness at different substrate terraces within the Au island. Firstly, we find that the Au wetting layer consists of a 2 ML Au film. This result is similar to a number of other thin film systems including Ag/Fe(001), where a 2 ML wetting layer completely covers the substrate [29]. Secondly, the local Au thickness within the island (see figure 3(a)) differs by exactly 1 ML between neighboring substrate terraces. This suggests that the Au island has an atomically flat step-free top facet, with the 1 ML Au thickness difference between neighboring terraces coming only from the Ru atomic steps. In principle, other configurations including atomic steps on the Au top surface might also exist. We believe that the energy penalty associated with the creation of surface steps is the reason why the top facets of the Au
islands tend to be free of atomic steps [30]. However, further studies (e.g. STM) are required for corroborating and understanding in detail the observation of flat top facets on the Au islands.

3.2. Magnetic properties of Co/Au/Ru(0001)

Co films were grown on the Au/Ru(0001) islands in order to study the $T_c$ and the magnetic anisotropy of the Co films. By using micron-sized Au wedges, we minimize measurement errors in temperature and Co-thickness variations across the Au wedge. Earlier studies of ultrathin Co/Au(111) films showed that at room temperature there is a critical thickness where a nonmagnetic-to-ferromagnetic phase transition occurs, and a larger thickness where an SRT from out-of-plane to in-plane magnetization takes place [31, 32]. Since the $T_c$ of an ultrathin film increases with film thickness [33] and the Co SRT thickness is determined by the magnetic anisotropy, an investigation of the Co critical thickness and the Co SRT thickness as a function of...
Figure 4. (a) Energy scan of 1 ML Co on Au. (b) Energy scan of 5 ML Co on Au. The data have been offset for a clearer view.

of the Au film thickness would reveal the effect of the Au QWS on the $T_c$ and the magnetic anisotropy of the Co film, respectively.

Before measuring the magnetic properties of the Co films, a QWS study of the Co film grown on Au was done to investigate the film quality and growth mode of the Co film grown on top of Au islands. It is well known that Co films grow epitaxially on flat Au(111) [29, 30]. Also, earlier studies showed the existence of QWS for Co/Au(111) [34, 35]. However, it is not clear whether QWS exist or what the growth mode is for Co films grown on micron-sized Au islands. We measured the QWS of Co films grown on flat Au and compared them with the QWS of Co films grown on Au islands and on the Au wetting layer. In figure 4, we show representative energy scans for 1 ML Co and 5 ML Co films. First, the existence of QWS indicates epitaxial growth and good crystalline quality of the Co film. For both Co thicknesses, the QWS spectra we found in Co grown on flat Au, on Au islands and on the Au wetting layer agree well with each other. In fact, the QWS for the three cases were identical up to at least $\sim$8 ML Co/Au, which indicates that the Co films grown on flat Au, the Au islands and the Au wetting layer have similar film quality and grow in a layer-by-layer growth mode.

Before the deposition of Co on the Au wedge, an energy scan was performed to confirm the existence of the QWS in the Au wedge. Figure 5(a) shows an image of an Au island on top of Ru step bundles at $E = 5.1$ eV (steps are indicated by the dashed lines). Within the Au
Figure 5. (a) LEEM image (1 × 1.5 µm²) of a micron-sized Au island (dark region) and Au wetting layer (light region) at $E = 5.1$ eV. Dashed lines indicate the step edges of the Ru substrate. Contrast between neighboring steps inside the Au island shows the existence of the QWS in the Au film. Out-of-plane magnetic domain images of Co/Au/Ru(0001) at (b) 3 ML Co and (c) 4 ML Co. The magnetic order of the Co film develops at a thinner thickness on the Au wetting layer than on the Au island, but no QWS effect from the Au film on the magnetic order is observed.

island (dark region), reflectivity contrast is clearly visible between different terraces of the Ru substrate, confirming the existence of QWS in the Au film. Next, we proceeded to study what effect the QWS from the Au underlayer might have on the magnetic properties of Co overlayers deposited on top of the Au. For this purpose, magnetic domain images of the Co film were obtained in real time during the deposition of the Co film. Below 3 ML of Co thickness, the film is paramagnetic and shows no domain contrast on either the Au wedge or on the wetting layer. At $\sim$3 ML thickness, we find the onset of ferromagnetism in the Co film. Imaging a Co film grown on top of the wetting layer with out-of-plane polarized incident electrons reveals out-of-plane magnetized domains (figure 5(b)). At this Co thickness, SPLEEM images obtained with in-plane polarized electrons show no domain contrast, indicating that the Co magnetization is perpendicular to the film plane. In figure 5(b), the black regions have magnetization pointing out of the plane, the white regions have magnetization pointing into the plane and the gray regions are paramagnetic. It can be seen from this figure that the magnetic contrast appears only in the Co film on the wetting layer rather than on the Au island, showing that the $\sim$3 ML Co film on the Au island is still in the paramagnetic state. In fact, on top of the Au islands, magnetic contrast develops only as the Co film reaches $\sim$4 ML thickness (figure 5(c)). Since the critical thickness of the Co film on the Au islands ($\sim$4 ML) is thicker than on the Au wetting layer ($\sim$3 ML), we conclude that the $T_c$ of the Co film on the Au islands is lower than the $T_c$ on the Au wetting layer. However, we did not observe any dependence of the Co critical thickness on the Au film thickness within the Au islands. This result shows that there is no evidence that QWS in the Au film affect the $T_c$ of the Co overlayer.

Below 6 ML thickness, the Co film remains perpendicularly magnetized with no domain contrast visible in the in-plane SPLEEM images. At 6 ML Co thickness, out-of-plane contrast in film regions grown on the wetting layer starts to weaken and, simultaneously, in-plane magnetic
contrast appears (figure 6(a)), showing that the Co film on the wetting layer is undergoing an SRT. At 7 ML, the magnetization of the Co grown on the Au wetting layer is completely in the film plane, but the Co grown on the Au islands remains out-of-plane magnetized (figure 6(b)). On top of the Au islands, at ~8 ML Co thickness, the out-of-plane magnetic contrast starts to become weaker and in-plane magnetic domains appear (figure 6(c)), showing that the Co on the Au islands is undergoing an SRT. Above 8 ML Co, the Co magnetization is completely in the film plane on both the Au wetting layer and the Au islands. Therefore, we conclude that the SRT thickness of the Co film on the Au island (~8 ML) is larger than that on the Au wetting layer (~6 ML). However, for the Co film on top of the Au islands, we observe an identical SRT thickness, regardless of the Au film thickness. The latter fact shows that the Au QWS have no effect on modulation of the Co magnetic anisotropy.

Although we clearly observe QWS within the Au islands, the fact that the Co nonmagnetic-to-ferromagnetic transition thickness and the Co SRT thickness are independent of the Au film thickness within the Au islands shows that the QWS inside the Au layer have no effect on the $T_c$ and the magnetic anisotropy of the Co film. The different Co nonmagnetic-to-ferromagnetic transition thicknesses and the Co SRT thickness on top of the 2 ML Au wetting layer as compared with that on thicker Au films are more likely associated not with the Au QWS but rather with interfacial effects (lattice mismatch or interfacial electronic hybridization), which evolve with the Au film thickness and stabilize above a certain thickness (in our case, above ~3 ML). Although we do not have a detailed characterization of the Au structure, we would...
like to mention an STM study on the similar system of Ag/Ru(0001), where it is shown that the lattice mismatch between Ag and Ru leads to a reconstruction of the first ML of Ag, whereas the second Ag layer resumes its bulk lattice constant [36]. In analogy, it is plausible that the 2 ML Au/Ru(0001) film (wetting layer) has different physical properties when compared with the thicker Au films on Ru(0001). Thus it is not so surprising to observe different magnetic properties for the Co film on top of the 2 ML Au wetting layer as compared with the Co film or thicker Au films. An interesting question is why the Au QWS have no effect on the magnetic properties of the Co overlayer. Although we do not have a definite answer, we would like to discuss the following argument. The importance of QWS for the physical properties is usually associated with the high density-of-states position of $\partial E/\partial k_{\|} = 0$, where $E$ and $k_{\|}$ are the electron energy and the in-plane momentum, respectively. For the Au(111) film, the QWS depend on $k_{\|}$ quadratically so that the $\partial E/\partial k_{\|} = 0$ position is at $k_{\|} = 0$. However, the nesting feature of the Au energy band opens an energy gap at the Fermi level for $k_{\|} = 0$ in the (111) direction so that, as the Au film thickness varies to modulate the QWS energy levels, no QWS with $k_{\|} = 0$ cross the Fermi level [37]. Since most of the physical properties are determined by the electronic states at the Fermi level, we speculate that the absence of QWS with $k_{\|} = 0$ at the Fermi level in the Au(111) system is the reason why the Au QWS have a negligible effect on the $T_c$ and the magnetic anisotropy of the Co film. More rigorous theoretical calculation is needed to confirm our speculation.

4. Summary

In summary, we investigated the QWS of Au/Ru(0001) and the magnetic properties of Co/Au/Ru(0001) bilayers using SPLEEM. By annealing RT-grown Au/Ru(0001) films, we fabricated micron-sized Au wedges across step bundles of Ru(0001), separated by regions covered with a 2 ML Au wetting layer. Energy scans reveal the existence of QWS inside the Au wedges. Magnetic domain images were recorded in real time during the growth of the Co film on the Au wedges. We found that the $T_c$ and the magnetic anisotropy of the Co film are different on the Au wedges and the Au wetting layer. However, the QWS inside the Au wedge have no effect on the $T_c$ and the magnetic anisotropy of the Co film. The latter result shows that the QWS in the Au layer have a negligible effect on the magnetic properties of the Co overlayer.

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