Growth, structure, and magnetism of γ-phase Mn ultrathin films on $Cu_3Au(100)$

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Ultrathin γ-phase (face-center cubic) Mn films were prepared by epitaxial growth on $Cu_3Au(100)$. Kinematic analysis of low energy electron diffraction I/V showed a structure transformation of Mn films from nearly face-center cubic to face-center tetragonal with increasing coverage. No ferromagnetic signal in $Mn/Cu_3Au(100)$ was observed. For 21 ML capping film of Fe on Mn films, the hysteresis loop of Fe was biased. The bias field for 21 ML Fe/15 ML $Mn/Cu_3Au(100)$ was $\sim$200 Oe at 110 K with the blocking temperature $T_b \sim 300$ K. This observation of exchange bias substantiates the theoretical prediction of antiferromagnetism in γ-phase Mn. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1855011]

According to Hund's rules, manganese atom, the transition metal with a half-filled 3*d*-shell, may give rise to magnetic moment as large as 5 μ_B. However, none of the four structures in bulk Mn reveals ferromagnetic behavior. Below 1000 K, α-phase Mn with 58 atoms per cubic cell is stable.[1] Between 1000 and 1368 K, β-Mn exists in two types of atomic sites with 20 atoms per unit cell. γ-phase (face-center cubic: fcc) is stable from 1368 up to 1406 K, and after that, the following δ-phase (body-center cubic: bcc) sustains up to melting point at 1517 K. Antiferromagnetism was observed in α-phase Mn only below 100 K in a noncollinear structure.[1] Other phases are paramagnetic at their existing temperatures, which may be so high as to exceed the Curie or Néel temperature of their possible ferromagnetism (FM) or antiferromagnetism (AF). Therefore, how to prepare stable fcc Mn at lower temperature[2–5] and studying its magnetic behavior[6,7] have become interesting issues and much effort has been made in this topic.

Up to now, there are several methods for preparing γ-phase Mn. One is by rapid quenching of Mn with dilute concentration of Cu,[8] Ni, Pd, or Fe. Mn thus forms a (100) layer–by–layer antiferromagnet with Néel temperature of 540 K. Another method is by epitaxial deposition on suitable substrate. For example, epitaxial Mn layers grow on Fe(100)[4], Ag(100)[5] in the body-center cubic (bct) structure with the same in-plane lattice constant of the substrate. Recently, fct Mn layers are also shown to be stable on fcc substrate, like Cu(100)[3] and $Cu_3Au(100)$[2]. However the detailed crystalline structure and the magnetic property have not been fully resolved. In this work, we studied the fcc-like Mn ultrathin films which were prepared by deposition over $Cu_3Au(100)$ due to the small lattice mismatch between fcc Mn-rich bulk alloy and Cu_3Au. The crystalline structure was characterized as a function of film thickness. Furthermore, Fe overlayer was grown on $Mn/Cu_3Au(100)$, in order to probe the magnetic property of Mn film indirectly.

This experiment was performed in an ultrahigh vacuum (UHV) chamber with base pressure $<2\times10^{-10}$ Torr. The $Cu_3Au(100)$ single crystal with miscut $\leqslant 0.1°$ was cleaned by cycles of 3 keV Ne ion sputtering. After cleaning, the substrate was annealed at 765 K for 5 min and then at 645 K for 30 min to get smooth and well-ordered surface. The $c2\times2$ structure,[9] which indicates the ordered arrangement of Cu and Au atoms, was checked by low energy electron diffraction (LEED). Mn and Fe films were deposited at 300 K. During the deposition, the pressure was better than 4×10^{-10} Torr and the growth condition was monitored by medium energy electron diffraction (MEED). From MEED oscillation, the film thickness was calibrated precisely. Besides, Auger electron spectroscopy (AES) also helped to check the composition of the films. Measurement of LEED and LEED-I/V curves were performed to identify the crystalline structure. From the LEED-I/V curves, the average vertical interlayer distance ($d_\perp$) of the film was determined using the kinematic approximation.[10,11] Magnetic properties of the films were characterized by magneto-optical Kerr effect (MOKE). The MOKE measurement was performed in both longitudinal [along the (001) direction] and polar geometry with modulation and a lock-in technique.

Figure 1 shows the MEED oscillation of Fe and Mn for 8 ML $Mn/Cu_3Au(100)$ and 21ML Fe/8 ML $Mn/Cu_3Au(100)$ at 300 K. The growth of Fe and Mn revealed layer–by–layer growth up to $\sim$12 ML and $\sim$8 ML, respectively. From the LEED images of $Cu_3Au(100)$ and *n* ML $Mn/Cu_3Au(100)$, the coherent growth of Mn films on

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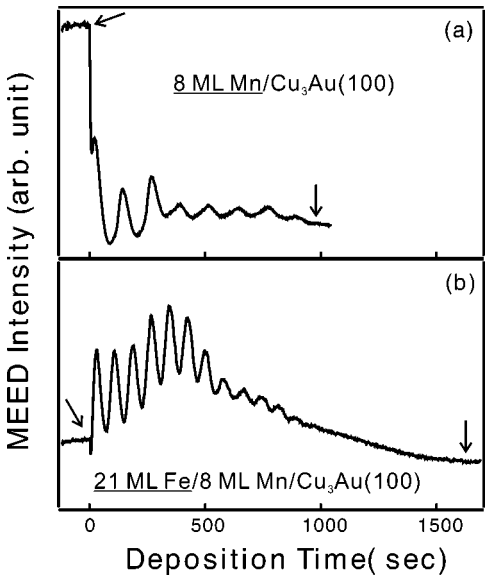

FIG. 1. (a) and (b) are the MEED oscillation curves of 8 ML Mn grown on $Cu_3Au(100)$ and subsequently 21 ML Fe grown on 8 ML $Mn/Cu_3Au(100)$. The arrows indicate the time for shutter opening and closing.

$Cu_3Au(100)$ is concluded and similar result also has been pointed out by Schirmer *et al.*[2] In comparison with $Fe/Cu_3Au(100)$,[9] $Fe/Mn/Cu_3Au(100)$ sustained layer–by–layer growth to higher thickness although $Mn/Cu_3Au(100)$ provides the same in-plane interatomic spacing as $Cu_3Au(100)$. This suggests that the interdiffusion at Fe/Mn interface may play an important role for the growth mode of Fe film.

Figure 2 shows the LEED-I/V curves of the (00) beam for n ML $Mn/Cu_3Au(100)$. The $d_\perp$ was deduced from kinematic fitting[9–11] of theses LEED-I/V curves. The dashed lines and solid lines indicate two series of peaks. At 11.5 and 14.2 ML, the coexistence of these two series was clearly

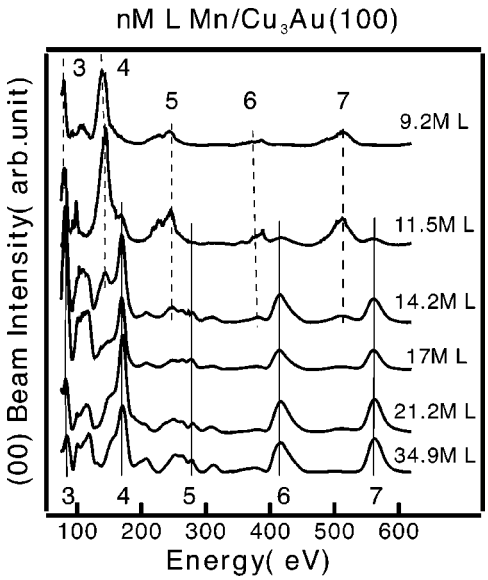

FIG. 2. LEED-I/V curves of n ML $Mn/Cu_3Au(100)$. Solid lines and dashed lines indicate two series of peaks. The numbers are the indices denoting the integer numbers of the single scattering Bragg interference condition (Refs. 9,10).

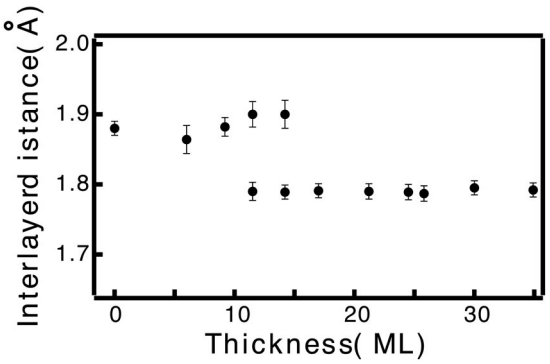

FIG. 3. The interlayer distances $d_\perp$ deduced from the LEED-I/V curves of n ML $Mn/Cu_3Au(100)$.

observed. This indicates that the structural transition started before 11.5 ML and completed after 14.2 ML. After 17 ML, only single phase existed. In Fig. 3, $d_\perp$ of $Cu_3Au(100)$ is 1.88 ± 0.01 Å very close to its bulk value. $d_\perp$ of Mn films changes from ~1.90 Å to ~1.78 Å with increasing thickness. Apparently, the structure of Mn films prefers a stable phase with tetragonal distortion ratio $c/a=1.01$ at lower coverage and another stable phase with $c/a=0.95$ at higher coverage. c and a are the perpendicular and the in-plane lattice constants, respectively. This result is consistent with previous experimental reports[2] and theoretical calculation. In the calculation by Qiu and Marcus,[6] fct Mn is stable with $c/a=1$ in a nonmagnetic phase and $c/a=0.96$ in a AF phase.

Similar to the study[2] of Schirmer *et al.*, $Mn/Cu_3Au(100)$ does not reveal any hysteresis loops in both polar and longitudinal MOKE measurement. In order to detect the possible antiferromagnetism of Mn films, Fe capping layer is added on top of $Mn/Cu_3Au(100)$. After field-cooling from 300 to 110 K under the magnetic field of positive 1000 Oe, the MOKE hysteresis loops were recorded at gradually increasing temperatures with 10 K increment, up to 300 K. With the variation of Mn thickness in 21 ML Fe/n ML Mn/$Cu_3Au(100)$, only above 8–9 ML Mn, we can observe a biased hysteresis loop. Figure 4 gives an example of 21 ML Fe/15 ML $Mn/Cu_3Au(100)$. Exchange bias (H_e) of ~200 Oe is observed at 110 K. In Fig. 5, we also characterize the temperature evolution of H_e and coercivity (H_c) for 14, 21, and 26 ML Fe/15 ML $Mn/Cu_3Au(100)$. All of the H_e gradually approaches zero when near 300 K, indicating that the T_b of FM/AF coupling did not affected by Fe thickness when >14 ML. Both the absolute values of H_e and H_c decrease coherently with the increase of Fe thickness. Since the T_b of FM/AF coupling approaches 300 K, we also have tried undergoing the field-cooling processes after going up to 400 K. The results are also very similar to the case of field cooling from 300 K.

There might be three origins of the AF phase inducing the exchange bias: (1) The first few layers of adjacent fcc Fe are predicted to be AF in some theoretical calculations; (2) the Fe–Mn alloy formed at the interface is found to be AF in certain composition; and (3) the Mn film beneath Fe layer might also induce H_e. To determine the origin of AF, we compare the saturated magnetic signal of n ML Fe/$Cu_3Au(100)$ with n ML Fe/15 ML $Mn/Cu_3Au(100)$ and

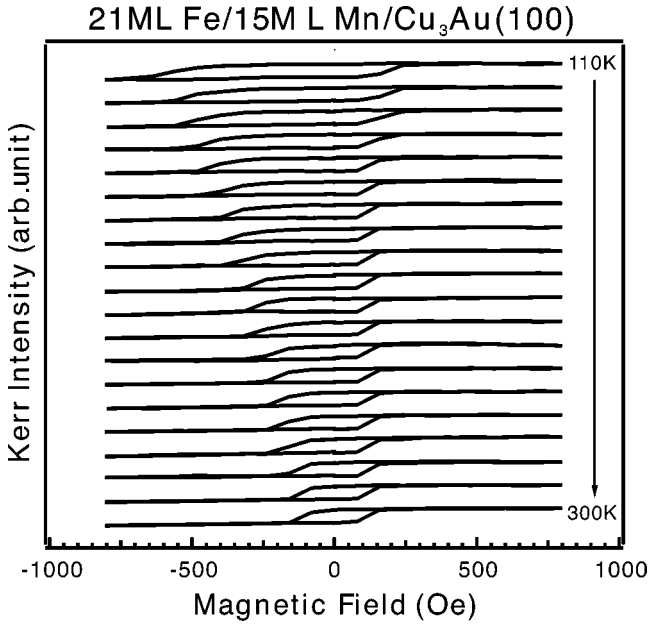

FIG. 4. Longitudinal MOKE hysteresis loops of 21 ML Fe/15 ML $Mn/Cu_3Au(100)$ with increasing temperature from 110 to 300 K ($\Delta T = 10$ K).

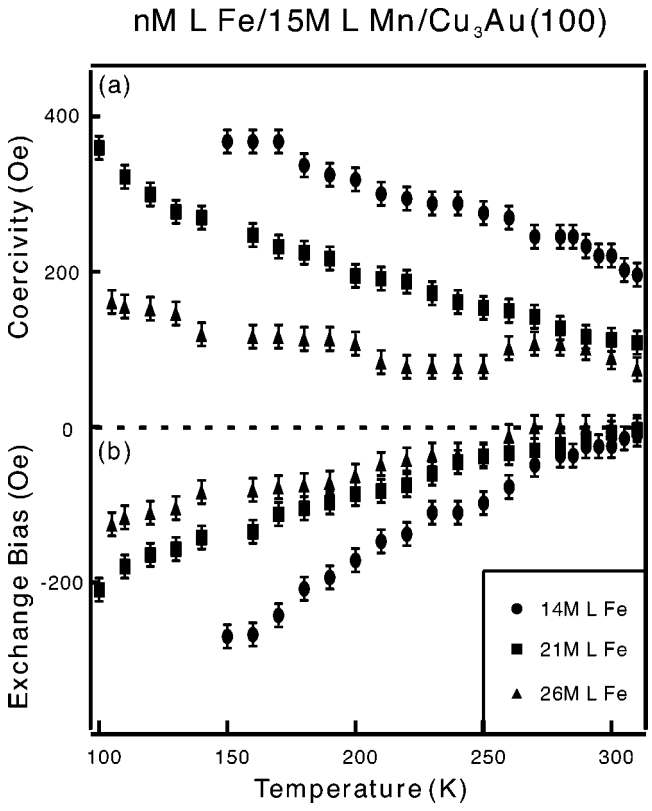

FIG. 5. Variation of (a) H_c and (b) H_e for 14, 21, and 26 ML Fe/15 ML $Mn/Cu_3Au(100)$ with increasing temperature.

conclude that the latter is smaller than the former by ~2 ML of Kerr signal. It means about 2 ML Fe near the Fe/Mn interface is nonferromagnetic. Such thin FeMn alloy or fcc Fe film is excluded from providing the strong FM/AM coupling with $T_b \sim 300$ K. Besides, we also performed an experiment of 21 ML Fe/n ML $Mn/Cu_3Au(100)$. Below 15 ML, the T_b and the H_e became smaller with thinner Mn layer. Since the formation of fcc Fe and thin FeMn alloy are strongly correlated to the condition of Fe/Mn interface, not the total Mn thickness, Mn thickness will not result in such a strong affect on the T_b and the H_e, if the FM/AF coupling comes from the fcc Fe or thin FeMn alloy films. Therefore, the AF is confirmed to originate from Mn film.

In summary, Mn films were coherently grown on $Cu_3Au(100)$ and revealed a structural transition. The crystalline structure changed from nearly fcc to fct with increasing thickness. By capping 21 ML Fe, the antiferromagnetism of this fcc-like Mn films was confirmed through the presence of exchange biased hysteresis loop. For 21 ML Fe/15 ML $Mn/Cu_3Au(100)$, the bias field reached ~200 Oe at 110 K and gradually disappeared while approaching room temperature.

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