Growth and magnetism of low-temperature deposited Fe/Si(111) films as an intermediate layer for suppression of silicide formation

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Low temperature (LT: 100 K) deposition of Fe on Si(111)7×7 surface effectively reduces Fe-silicide formation at the Fe/Si interface, as compared with conventional room temperature (RT) growth. The interface condition of 5–15 monolayers (ML) LT-Fe/Si(111) remains stable at least up to 350 K. Si segregation was observed after annealing at 400 K. LT-grown Fe films also reveal a relatively flat surface morphology with a roughness of 0.4–0.6 nm. Thus, LT-Fe films were suggested as an intermediate layer for the subsequent RT-growth of Fe. We use a single domain model of magnetic anisotropy to fit the magnetic coercivity evolution of n ML RT-Fe on 5 ML LT-Fe/Si(111). Accordingly, we deduce the surface and volume-contributed magnetic anisotropy for discussion. © 2011 American Institute of Physics. [doi:10.1063/1.3537832]

I. INTRODUCTION

Metallic films on semiconductor surfaces have attracted much attention in recent years. The growth of magnetic materials, like Fe and Co, on semiconductor surfaces, has attracted particular interest and been widely studied because of the potential applications combining magnetism and semiconductor techniques, so-called spintronics. Serious silicide formation at the transition metal/Si interface has long been a key issue. The silicide formation not only randomizes the crystalline structure and electronic structure near the interface but also leads to a thick magnetic dead layer and unstable magnetic anisotropy. Up to now, many buffer layers have been proposed and demonstrated for suppressing silicide formation. In some cases, the buffer layer needs to be thick enough to block the chemical activity of the semiconductor surface. For example, a 6 monolayers (ML) Ag buffer layer was demonstrated to block the alloy formation in Co/Ge(111). In other cases, deposition of submonolayer buffer atoms with high temperature annealing can result in a very stable superstructure on the semiconductor surface, providing a sharp interface for the subsequent deposition of magnetic materials. Nevertheless, inserting an intermediate layer of different materials might increase the complexity when changing the original properties of the semiconducting substrate and magnetic deposit. Therefore, we propose the idea of using a low temperature (LT)-deposited Fe layer, instead of a buffer with different materials, as an intermediate layer for suppressing silicide formation. The Fe/Si interface might be frozen and fixed because of the low mobility and small thermal energy at LT. Such an interface is expected to be stable not only at LT but also after annealing to room temperature (RT), owing to the binding from subsequent deposited layers. With this idea of using LT-Fe as the intermediate layer, not only the planar Si surface, but also many recently developed Si-based nanostructures can be used as suitable templates for magnetic material deposition without introducing new materials in between or destroying the nanopatterning on the substrate. In this study, the Fe/Si interface, thermal stability, and surface morphology of LT:100 K-deposited Fe/Si(111) were investigated. For demonstration, the 5 ML LT-Fe film was used as an intermediate layer for the subsequent RT-growth of Fe. In the coverage-dependent magnetic measurement, the evolution of magnetic anisotropy was also discussed.

II. EXPERIMENT

The entire experimental processes, including sample preparation, transferring, and characterization were performed in an ultrahigh vacuum system with a base pressure better than 3×10−10 torr. The Si(111)7×7 template was prepared by repeated flashing at 1200 °C. For LT and RT-growth, the Fe atoms were deposited while the substrate was at 100 K and 300 K, respectively. The film thickness was calibrated using Auger electron spectroscopy (AES) and scanning tunneling microscope (STM). Both the detections of AES and STM were performed while the sample was at RT. The magnetic behavior was investigated using the magneto-optical Kerr effect (MOKE) in both longitudinal and polar directions with the lock-in technique. The MOKE measurements shown in this report were performed at RT.

III. RESULTS AND DISCUSSION

A. LT-growth of Fe/Si(111)

To investigate the different interface conditions of LT and RT-grown Fe on Si(111), coverage-dependent AES detection was performed. Figure 1 shows the AES signal ratio of Fe$_{2\,\text{eV}}$/Si$_{2\,\text{eV}}$ (left axis) as a function of Fe thickness for RT-Fe films on Si(111) and on 5 ML LT-Fe/Si(111). The AES measurements were carried out at RT. This means for LT-grown films, the sample underwent RT annealing before AES detection. In RT-grown Fe/Si(111) films, the Fe signal becomes observable after 3–4 ML, and then reaches the comparative signal intensity of Si at ∼9 ML. After 10 ML, the Fe signal dominates Si. The question of when the Si really
disappears from the surface can be answered by seeing from the right axis of Fig. 1, the AES ratio of Si$_{92\ eV}$/Fe$_{47\ eV}$. The Si/Fe ratio gradually approaches 0.06 ± 0.03 at 15 ML. This is the minimum ratio of Si/Fe we could observe. The small broad feature of Fe at 87 eV partially overlaps with the possible small Si-92 eV peak. The overlapping actually prevents us from checking the presence of Si more precisely.

Thus, we can conclude, at least to the limit of our AES measurement, the Si(111) substrate is fully covered after deposition of 15 ML RT-grown Fe.

For comparison, the solid squares in Fig. 1 indicate the Fe/Si AES ratio of RT-Fe/5 ML LT-Fe/Si(111). Apparently, the Fe/Si ratio of 5 ML LT-Fe/Si(111) is much larger than 5 ML RT-Fe/Si(111). Sequential deposition on 5 ML LT-Fe/Si(111) gradually increases the Fe/Si ratio in the similar trend of RT-Fe films. The horizontal shift between the two series of data is 3.5 ± 0.5 ML. A recent study of Kataoka et al. showed, for RT-grown Fe/Si(111), metastable FeSi with the CsCl-type structure persists at least to 6 ML of Fe. Afterward, the body-centered-cubic (bcc) Fe(111) film starts to grow. Our AES data supports their conclusion of serious silicide formation in RT-growth. Based on Kataoka et al.’s observation, the equation of Fe/Si AES ratio is written down by assuming the first 6 ML of Fe was mixed with Si, forming 12 ML of FeSi.

$$\frac{Fe}{Si}_{RT} = \frac{I_{Fe}}{I_{Si}} \times \frac{(1 - e^{-(t-6)/\lambda_{Fe}}) + 0.5 \times (e^{-(t-6)/\lambda_{Fe}} - e^{-(t+6)/\lambda_{Fe}})}{(e^{-(t-6)/\lambda_{Si}}) + 0.5 \times (e^{-(t-6)/\lambda_{Si}} - e^{-(t+6)/\lambda_{Si}})} \times (1 - e^{-6/\lambda_{Fe}}),$$

(1)

$$t$$ is the deposited Fe thickness. $\lambda_{Fe}$ and $\lambda_{Si}$ are the mean free path of Fe$_{47\ eV}$ and Si$_{92\ eV}$ Auger electrons. $I_{Fe}/I_{Si}$ is the relative sensitivity between Fe and Si Auger intensity. Note, Eq. (1) is for RT-grown Fe/Si(111) with $t$ ≥ 6 ML. Besides, for RT-Fe/5 ML LT-Fe/Si(111), the Fe/Si AES ratio is written down by assuming no silicide formation at the LT-Fe/Si interface.

$$\frac{Fe}{Si}_{LT} = \frac{I_{Fe}}{I_{Si}} \times \frac{(1 - e^{-6/\lambda_{Fe}})}{(e^{-6/\lambda_{Si}})},$$

(2)

First, the RT-Fe/Si(111) data was fitted by Eq. (1), using the relative sensitivity $I_{Fe}/I_{Si}$=0.66 ± 0.06, which is consistent with the value in the Auger handbook. The fitting curve matches the experimental RT data well, as shown in Fig. 1 with $\lambda_{Fe}=3.6 $ ± 0.3 ML, and $\lambda_{Si}=4.0 $ ± 0.3 ML. With these parameters, the curve of Eq. (2) also matches the experimental data of RT-Fe/5 ML LT-Fe/Si(111). Actually, within the error bars of AES data, we cannot totally exclude the possible presence of 1–2 ML Fe-silicide formation at the interface. However, when considering the detailed chemical composition and AES signal of the Fe-silicide interface, the suppression of silicide formation by LT-Fe is even larger than the shift of 3.5 ± 0.5 ML in Fig. 1. Our experimental and fitting results all indicate the LT-Fe intermediate layer indeed effectively reduces the Fe-silicide thickness by at least 4 ML Fe (~8 ML FeSi) compared with RT-growth. Even after RT-annealing, the LT-grown intermediate layer can sustain the Fe/Si interface and prohibit further silicide formation.

Since LT deposition of Fe on Si(111) can effectively reduce the alloy formation at the interface even after RT annealing, we were curious about the thermal stability of the LT-grown films. Figure 2 shows the AES ratio of Si/Fe as a function of annealing temperature for 5–25 ML LT-Fe films. All the AES detections were performed after keeping the sample at the annealing temperature for 10 min and then cooling it to RT. For 5 and 15 ML LT-Fe, the AES ratio remains invariant during 300–350 K and then we observe the increased Si signal at ~400 K. For 25 ML LT-film, the Si/Fe ratio remains stable up to a higher temperature of 400 K. For a thicker film, like 25 ML LT-Fe, it is hard to judge if the higher stability is truly because of the higher thickness, or
only because the coverage is too thick to detect the slight changes at the Fe/Si interface. Nevertheless, we can still conclude 5–15 ML LT-grown Fe films are stable at least up to 350 K. It also supports our suggestion of using LT-grown layer as an intermediate layer for subsequent RT-growth or processing.

The previous studies of Gallego et al. and Tsushima et al. concluded the Fe(111) films grows epitaxially up to a thick region, maintaining the bcc structure.3,13,25,26 Besides the crystalline structure, the study of surface morphology is still lacking, especially for LT-Fe/Si(111) films. Usually the LT-deposition of thin film is expected to result in statistically flat morphology, due to the low mobility and random landing distribution of deposited atoms. Recent studies of quantum well systems also report the two-step growth, meaning LT-deposition with postannealing may lead to atomically flat surface morphology in some cases of the thin film system. Thus, we try to probe the growth result of LT-deposited Fe surface morphology in some cases of the thin film system.

FIG. 3. (Color online) Surface morphology of (a) 24 and (b) 40 ML LT-grown Fe films, investigated by STM. The figures are all 120×120 nm². The insets (30×30 nm²) reveal the detailed surface structures. The line profiles indicated in the STM images are plotted at the bottom.

In Fig. 5, Hc increases linearly with Fe coverage. The extrapolation of the linear fitting line indicates that the magnetic dead layer could be 3.5±0.5 ML in our RT-MOKE measurement. Thus, we expect, at LT, the magnetic dead layer might be even smaller than 3 ML, which can be due to a slight Fe-silicide formation or the extensive hybridization of Si electronic states into Fe.

In the above sections, compared with RT-grown films, LT-growth can effectively reduce the Fe-silicide formation at the interface, and lead to a flat surface morphology. Thus, for the magnetic study, we used a 5 ML LT-grown Fe film as an intermediate layer for the sequential RT-deposition, since RT-deposition is easier to perform, especially in future applications. The MOKE hysteresis loops for n ML RT-Fe/5 ML LT-Fe/Si(111) measured at RT are shown in Fig. 4. The hysteresis loops are close to the square shape. Both the Kerr saturation signal (Ms) and coercivity field (Hc) increase with Fe coverage but in different ways. The quantitative analysis is shown in Fig. 5. Ms increases linearly with Fe coverage. The extrapolation of the linear fitting line indicates that the magnetic dead layer could be 3.5±0.5 ML in our RT-MOKE measurement. Thus, we expect, at LT, the magnetic dead layer might be even smaller than 3 ML, which can be due to a slight Fe-silicide formation or the extensive hybridization of Si electronic states into Fe.

In Fig. 5, Hc increases monotonically and gradually becomes saturate around 70 Oe. To deduce more detailed insights from the coverage dependent evolution of Hc, we use the Stoner–Wohlfarth single domain model to simulate the
$H_\text{c}$ evolution behavior with Fe film thickness. The energy ($E$) correlated with magnetism in our thin film system can be written as follows: \cite{30,31}

$$E = -H \times M \times \cos(\theta - \phi) + K_{\text{eff}} \times \sin^2 \theta.$$  

(3)

$\theta$ and $\phi$ are the rotation angles of magnetic moment and magnetic field, respectively, relative to the surface normal direction. The first term is Zeeman energy. $M$ is the averaged magnetic moment per unit volume and $H$ is the applied magnetic field. In the longitudinal MOKE measurement, as shown in Fig. 4, we apply the magnetic field in the in-plane directions, and thus $\phi$ only can be $\pm \pi/2$. $K_{\text{eff}}$ in the second term is the uniaxial magnetic anisotropy. Since the easy axis is in the surface plane, one should expect $K_{\text{eff}} < 0$, which we can check later from the fitting results. By minimizing the magnetic energy in Eq. (3), the magnetization direction $\theta$ can be determined as a function of the applied field $H$. Since we have $\phi = \pm \pi/2$, the minima of $E(\theta)$ are always at $\pm \pi/2$. This can be easily checked by solving the equation of $dE(\theta)/d\theta = 0$ with $\phi = \pm \pi/2$. Therefore, in this model, the magnetization only switches between $\theta = \pm \pi/2$, resulting in the square magnetic hysteresis loops, which are actually consistent with our measured loops. Further, from Eq. (3) we can deduce the coercivity field $H_\text{c}$ at which the magnetization direction switches. For example, if the magnetic moment is at $\theta = \pi/2$ initially, we apply the field in the $\phi = -\pi/2$ direction to switch the magnetic moment. The coercivity field can be solved from the following equation:

$$\frac{d^2 E(\theta,\phi = -\pi/2)}{d\theta^2} \bigg|_{\theta = \pi/2} = 0,$$

(4)

$$H_\text{c} = -2 \times K_{\text{eff}}/M.$$  

(5)

In a thin film system, we have both the surface and the volume-contributed magnetic anisotropy. The magnetic anisotropy $K_{\text{eff}}$ can be decomposed as $K_{\text{eff}}=k_v+k_s/t$, \cite{31} where $k_v$ and $k_s$ are volume and surface anisotropy terms, respectively. Since $K_{\text{eff}}$ is the anisotropy energy per volume, the surface contribution $k_s$ will gradually decay with $1/t$, where $t$ is the thickness of thin film. Then we rewrite the $H_\text{c}$ as a function of film thickness $t$

$$H_\text{c}(t) = -\frac{2}{M} \times \left(k_v + \frac{k_s}{t}\right).$$  

(6)

We take the magnetic moment of Fe: $M = 2.5$ $\mu_B$/atom from the literature, \cite{32} and then set $k_v$ and $k_s$ as the free parameters. The solid curve in Fig. 5 is the best fitting by using Eq. (6) with $k_v = -0.5 \pm 0.1$ $\mu$eV/atom and $k_s = 1.9 \pm 0.1$ $\mu$eV/atom. The fitting curve qualitatively reproduces the monotonic increase and saturation of $H_\text{c}$ with the Fe film thickness. With these fitted $k_v$ and $k_s$ values, $K_{\text{eff}} = k_v+k_s/t$ is always negative after 5 ML. This is consistent with the in-plane magnetization of the measured Fe films. Besides, $K_{\text{eff}}$ changes signs from positive to negative at the critical thickness of $t_c = K_v/K_s=4 \pm 1$ ML, indicating the spin reorientation transition (SRT) from surface normal to in-plane. The SRT and $t_c$ is actually consistent with the previous study of Nazir et al. \cite{5} They reported the perpendicular to in-plane SRT at 3.6–5.5 ML for 100 K-grown Fe/Si(111) films.

Compared with other SRT systems, our fitted $K_v$ and $K_s$ are relatively small but actually close to the crystalline anisotropy of bcc Fe, \cite{30,31} One possible reason could be the domain wall motion, which is not considered in the single domain model. Another possibility is the anisotropy model could be interpreted in another way. During the magnetization switching, instead of going through the surface normal direction, the magnetic moment may undergo 180° rotation just on the surface plane. In this case, we can redefine the $\theta$ and $\phi$ relative to another direction, which is perpendicular to the applied magnetic field but on the surface plane. Then we can still keep the same calculation and fitting. The only difference is the magnetic anisotropy is no more the energy difference between surface normal and in-plane directions but between the different directions on the surface plane. Even after modifying the anisotropy from uniaxial to sixfold symmetry, \cite{14} we will still get similar results of $k_v$ and $k_s$. We have to agree, from our experimental data, it is hard to judge which is the real situation in our system. Further work will

FIG. 4. (Color online) Longitudinal MOKE hysteresis loops measured at RT for $n$ ML of RT-grown Fe on an intermediate layer of 5 ML RT-Fe on Si(111).

FIG. 5. Kerr saturation signal ($M_s$: left-axis) and coercivity field ($H_c$: right axis) plotted as function of Fe coverage. The data points were analyzed from the hysteresis loops of $n$ ML RT-Fe/$5$ ML LT-Fe/Si(111) in Fig. 4. The solid line and curve are the fitting results of $M_s$ and $H_c$ using a linear function and Eq. (6), respectively.
be needed for a more advanced conclusion. However, we can at least conclude the volume and surface contributions, $k_v$ and $k_s$, are of opposite signs and compete with each other while the film thickness is increased. Such kind of situation eventually results in an $H_c$ increase and saturation in our measured $n$ ML RT-Fe/5 ML LT-Fe/Si(111) system.

IV. SUMMARY

LT-deposition of Fe/Si(111) films was performed in this experiment. LT-deposition can effectively reduce the Fe-silicide formation at the Fe/Si interface compared with conventional RT growth. The LT-grown Fe films reveal relatively flat surface morphology and its interface condition remains stable around RT. The roughness is within 2–3 atomic ML even for 24–40 ML LT-Fe films. Thus, the LT-Fe films can be used as a suitable intermediate layer for subsequent RT-growth or processing. For demonstration, the coverage-dependent magnetic properties of $n$ ML RT-Fe on 5 ML LT-Fe/Si(111) were investigated. A single domain model of magnetic anisotropy is proposed to fit the magnetic coercivity evolution with film thickness. We deduce the values of surface and volume-contributed magnetic anisotropy, which are inverse signs and their competition is the main origin for the variation in coercivity.

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