Tunable in-plane spin orientation in Fe/Si (557) film by step-induced competing magnetic anisotropies*

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(Received 14 September 2016; revised manuscript received 10 October 2016; published online 15 November 2016)

Although the spin-reorientation transition from out-of-plane to in-plane in Fe/Si film is widely reported, the tuning of in-plane spin orientation is not yet well developed. Here, we report the thickness-, temperature- and Cu-adsorption-induced in-plane spin-reorientation transition processes in Fe/Si (557) film, which can be attributed to the coexistence of two competing step-induced uniaxial magnetic anisotropies, i.e., surface magnetic anisotropy with magnetization easy axis perpendicular to the step and volume magnetic anisotropy with magnetization easy axis parallel to the step. For Fe film thickness smaller than 32 monolayer (ML), the magnitudes of two effects under various temperatures are extracted from the thickness dependence of uniaxial magnetic anisotropy. For Fe film thickness larger than 32 ML, the deviation of experimental results from fitting results is understood by the strain-relief-induced reduction of volume magnetic anisotropy. Additionally, the surface and volume magnetic anisotropies are both greatly reduced after covering Cu capping layer on Fe/Si (557) film while no significant influence of NaCl capping layer on step-induced magnetic anisotropies is observed. The experimental results reported here provide various practical methods for manipulating in-plane spin orientation of Fe/Si films and improve the understanding of step-induced magnetic anisotropies.

Keywords: Fe/Si (557) films, spin-reorientation transition, competing magnetic anisotropies

PACS: 75.70.Ak, 75.30.Gw **DOI:** 10.1088/1674-1056/25/12/127501

1. Introduction

Tailoring the spin orientation of ferromagnetic films was an intensively studied field due to its significant role in developing spintronic device.[1,2] Since the spin orientation in ferromagnetic film is directly determined by the magnetic anisotropy, a better controlling and understanding of magnetic anisotropy becomes one of the challenging topics to engineer the spin order orientation of magnetic material.[1–3] Surface/interface effect has a strong bearing on the magnetic properties of low-dimensional system, and consequently can be taken as an effective method to engineer the magnetic anisotropy of ultrathin film.[4–7] Epitaxial growth of the ferromagnetic film on stepped substrate is one of the most common methods to form a periodical surface morphology of film, resulting in an in-plane uniaxial magnetic anisotropy (UMA) originating from the magnetic shape anisotropy,[8–10] bulk strain,[11,12] and/or Néel pair-bonding of surface atomic.[13] Due to the complex effects induced by stepped substrate, competing magnetic anisotropies are usually employed to modify the spin orientation of ferromagnetic film. For example, in the case of Ag/Fe film deposited on vicinal Ag substrate, the step-induced magnetic anisotropies are the superposition of competing surface and volume magnetic anisotropies.[14,15] Therefore, the spin orientation can be tuned by changing the respective contribution of surface and volume magnetic anisotropy to UMA.[14]

Due to the importance of Fe/Si films in magnetoelectronics and the integration of magnetic devices in silicon technology,[16] the magnetic anisotropy of Fe/Si film has been largely reported.[3,7–10,17–20] In particular, the manipulation of spin-reorientation transition (SRT) processes from out-of-plane to in-plane in Fe/Si film has already been fully understood and studied. For example, for Fe films deposited on flat Si (111) substrate, due to the competition between surface magnetic anisotropy and shape anisotropy, an SRT process from out-of-plane to in-plane with a critical thickness of about 7 monolayer (ML) was observed.[19] Differently, for Fe film grown on vicinal Si (111) substrate, a continuous SRT process in a larger thickness range of 4.5 ML–7.5 ML was obtained.[9] Covering the 4.7-ML Fe film with 80-ML Cu film can also introduce an SRT from out-of-plane to in-plane.[17] However, among the investigations about the magnetic anisotropy of Fe/Si films, tuning of in-plane spin orientation is still not yet developed. Since larger step density may result in larger surface effect, Si (557) with huge step density, i.e., Si (111) 9.45° vicinal substrate,[21] was thus used as the substrate for expecting the emergence of step-induced

*Project supported by the National Basic Research Program of China (Grant Nos. 2015CB921403 and 2016YFA0300701) and the National Natural Sciences Foundation of China (Grant Nos. 51427801, 11374350, 11274360, and 11274361).

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competing magnetic anisotropies. Here, we successfully obtain the competing step-induced UMA in epitaxial Fe/Si (557) film. By modifying the respective contribution of surface and volume magnetic anisotropy to UMA, the in-plane spin orientation in Fe/Si (557) film can be strongly tuned by changing the thickness of Fe film, temperature and Cu capping layer. Our work provides various methods of tuning the spin orientation in Fe/Si (557) film and conduces to the understanding of step-induced magnetic anisotropies.

2. Experiments

The Fe/Si (557) films were prepared in an ultrahigh vacuum molecule beam epitaxial combine system with a base pressure lower than 2×10^{-10} mbar (1 bar $= 10^5$ Pa). The commercial Si (557) substrate was firstly heated at 700 K for over 8 h, and then quickly flashed to 1200 K and quickly cooled down to 400 K, which was repeated over 10 times to form a well defined surface reconstruction, which is confirmed by *in-situ* low energy electron diffraction (LEED) shown in the inset of Fig. 1(a). 1.5-ML Fe was deposited on the Si substrate and then annealed at 600 K for about 4 min to form an $FeSi_2$ buffer layer to prevent the Fe and Si from intermixing.[19] Fe film was again deposited on $FeSi_2$ buffer layer with a deposition rate of $\sim$ 1 ML/min. Cu film with a thickness of 25 ML or NaCl film with a thickness of 14 ML was deposited as the capping layer to prevent the Fe from being oxidized and determine the effect of capping layer on magnetic anisotropy of Fe/Si (557) film. *In-situ* scanning tunneling microscope (STM) was used to obtain the surface morphology of films. *In-situ* MOKE, which supports low temperature measurements, was used to determine sample magnetic anisotropy.

3. Results and discussion

Figures 1(a)–1(d) display the STM images which show the surface morphologies of pure Si (557) substrate, $FeSi_2$ buffer layer, 20-ML Fe film and 25-ML Cu film, respectively. Si (557) substrate has a very narrow terrace with a mean width of $\sim$ 5 nm as shown in Fig. 1(a). Figures 1(c) and 1(d) show the island-like surface morphologies, indicating the Volmer–Weber epitaxial mode for Fe and Cu film. The insets in these

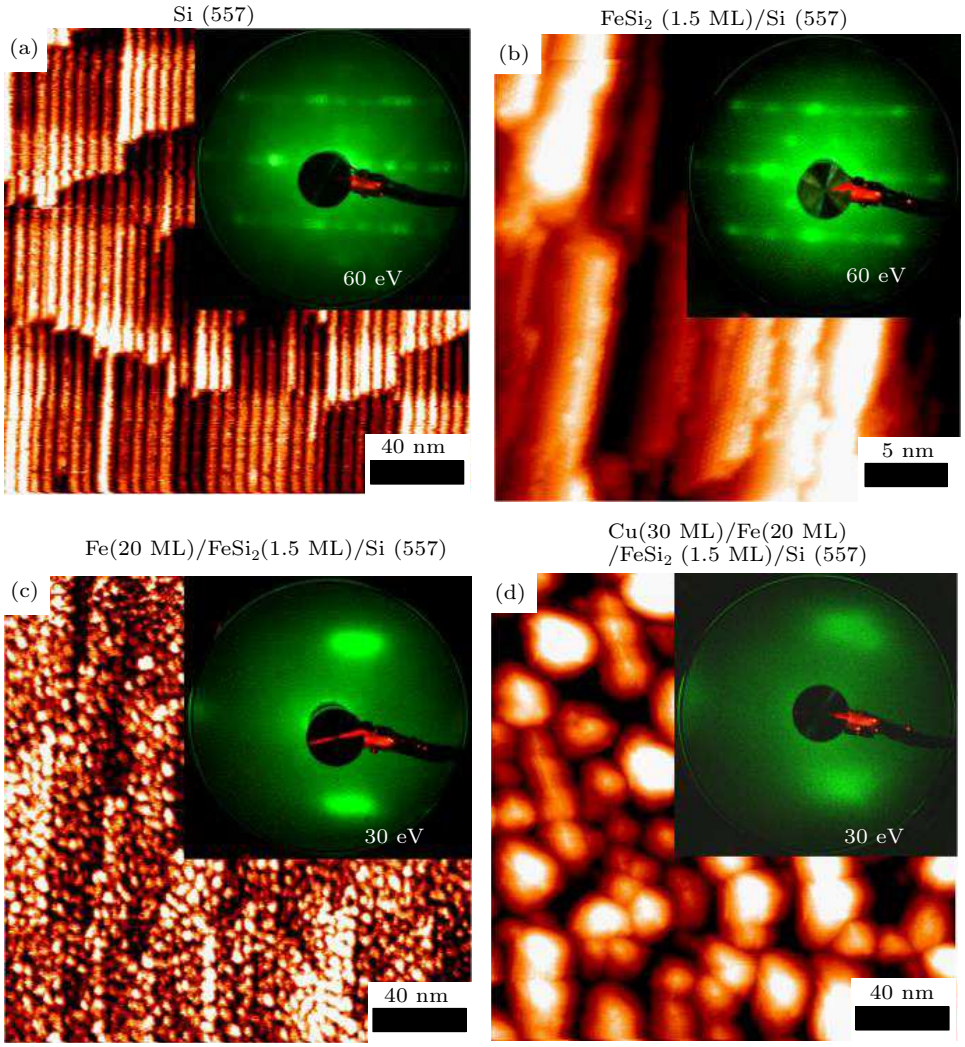


Fig. 1. (color online) STM images for Si (557) substrate (a), $FeSi_2$ buffer layer (b), 20-ML Fe film (c), and 25-ML Cu film (d). Each inset shows the corresponding LEED pattern.

figures show the corresponding LEED patterns. The crystal structure of epitaxial growth of *bcc* α-Fe film exhibits six-fold surface symmetry, which is verified by the LEED pattern (Inset of Fig. 1(c)). Unlike the previous LEED patterns reported in Fe films deposited on Si (111) flat substrate or Si (111) vicinal substrate with small vicinal angle, the elongated shape of the LEED patterns in Fe/Si (557) films can be attributed to the step-induced strong scatter of electrons. However, after growing NaCl capping layer on single crystal Fe film, we observe no distinct LEED pattern (not shown here). The disappearance of LEED pattern indicates that the NaCl capping layer may be amorphous or polycrystalline.

Figure 2 shows the MOKE hysteresis loops parallel and perpendicular to the step in Fe/$FeSi_2$/Si (557) films with different thickness values of Fe film (d_{Fe}) at room temperature. The Fe films deposited on $FeSi_2$ buffer layer were prepared step-by-step. The magnetic hysteresis loops are then obtained by *in-situ* longitudinal MOKE. The asymmetry of magnetization loops shown in Fig. 2 is attributed to the remanence error of magnet, which usually contributes a small shift (about 10 Oe, 1 Oe = 79.5775 A·m^{-1}) of loop.[22] The Kerr hysteresis loops show a split shape with nearly zero remanence for field applied along the step while a square shape for field applied perpendicularly to the step for $d_{Fe} \leq 12$ ML, indicating an in-plane UMA with magnetization easy axis (EA) perpendicular to the step. The split magnetization loop along hard axis (HA) can be characterized by the shift field (H_s) defined by the offset of the minor loop (see the definition shown in Fig. 2(a)), which is typically taken as a measure for the magnitude of UMA. Traditionally, for a perfect flat Fe (111) film, six-fold magnetocrystalline anisotropy is dominated.[8] However, even a small deviation of film plane from (111) plane can change the six-fold magnetic

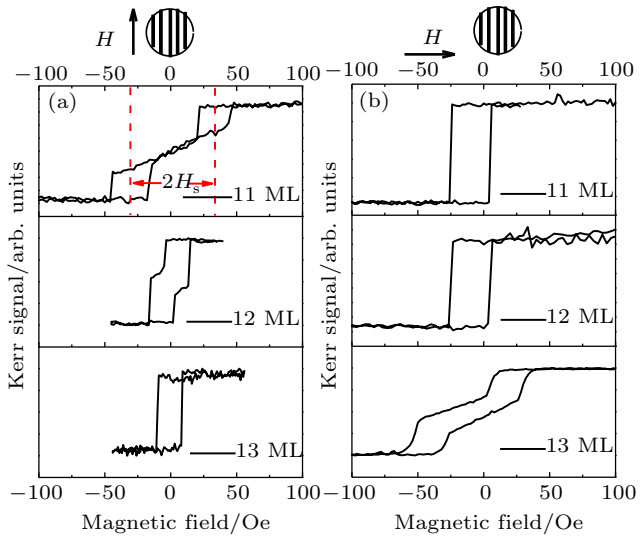


Fig. 2. (color online) *In-situ* longitudinal MOKE hysteresis loops of Fe/Si (557) films in the thickness range of 11 ML–13 ML measured at room temperature for external magnetic field applied parallelly to the step (a) and perpendicular to the step (b).

symmetry into the superposition of a uniaxial and a four-fold contribution.[10] Therefore, although the crystal structure of Fe (557) film is of in-plane six-fold symmetry, the magnetic hysteresis loop of HA reveals a split shape, which is typically obtained in film whose crystal structure is of mainly four-fold symmetry, for example, Fe/CoO/MgO (100) films[23] and Fe/NiO films on Ag (100) vicinal substrate.[24] Interestingly, we observe an in-plane SRT from perpendicular to the step to parallel to the step when further increasing d_{Fe} up to 13 ML. To better clarify the effect of Fe film thickness on magnetic anisotropy of Fe/Si (557) film, the thickness dependence of H_s is obtained.

Figure 3(a) shows the thickness dependence of UMA field in a temperature range of 90 K–300 K. In previous work about the magnetic anisotropy of Fe/Si (111) film, magnetic shape anisotropy plays an important role in determining the magnetic anisotropy due to quasi one-dimensional periodically stepped surface morphology. The magnetic shape anisotropy in Fe/Si (557) film can be accurately obtained from the surface morphology of STM data (see STM image in Fig. 1(c)) by the self-correlation function method developed by Bubendorff *et al.*[25] However, the calculation of magnetic shape anisotsropy yields the UMA field of only about several Oe, which is much smaller than the UMA field ($\sim$ 140 Oe) obtained from experiments for $d_{Fe} = 20$ ML. Therefore, the magnetic shape anisotropy is excluded from further discussion about the origin of magnetic anisotropy. Traditionally, the in-plane UMA is usually divided into two parts, i.e., the surface contribution $H_s^{surface}$ and volume contribution H_s^{volume}. Then the UMA field H_s can be expressed as the following relation:

$$H_s = \frac{H_s^{surface}}{d_{Fe}} + H_s^{volume}. \quad (1)$$

We obtain an excellent fitting quality based on Eq. (1) for $d_{Fe} < 32$ ML (see the solid curves shown in Fig. 3(a)) at various temperatures. Figure 3(b) shows the temperature dependences of $H_s^{surface}$ and H_s^{volume} extracted from fitting results. We define positive value for UMA favoring magnetization axis parallel to the step. It is clearly shown in Fig. 3(b) that $H_s^{surface}$ and H_s^{volume} have negative and positive signs respectively, indicating they support spin orientation perpendicular to the step and parallel to the step, respectively. For $d_{Fe} < 13$ ML, surface magnetic anisotropy is dominated and EA is perpendicular to the step at room temperature. However, the surface contribution to the UMA gradually decreases with further increasing thickness of Fe film and SRT happens when two effects are balanced. The critical thickness in Fe/Si (557) film for in-plane SRT is about 12 ML at room temperature, where the UMA is almost zero and four-fold magnetic anisotropy is dominated. As illustrated in Fig. 2, the values of normalized

remanence of MOKE hysteresis loops parallel and perpendicular to the step are both almost 1 for $d_{Fe} = 12$ ML, indicating two EAs of four-fold magnetic anisotropy in Fe/Si (557) film are parallel and perpendicular to the step, respectively. Besides, the 2-jump loop observed along the step is attributed to two successive 90° domain wall reversals,[26] which is typical for ferromagnetic film deposited on (100) plane of substrate but rarely observed in ferromagnetic film deposited on (111) plane of substrate. The magnetic anisotropy in Fe/Si (557) film can also be modified by the temperature. The surface and volume magnetic anisotropies are both enhanced at low temperature as shown in Fig. 3(b). The critical thickness for the spin-reorientation transition is obtained by setting H_s to be zero based on the anisotropy constants shown in Fig. 3(b). The critical thickness for the spin-reorientation transition changes from ∼ 12.7 ML at 300 K to ∼ 15.2 ML at 90 K and is illustrated in Fig. 3(d), which predicts an in-plane temperature-induced spin-reorientation transition from parallel to the step to perpendicular to the step in a thickness range of ∼ 12.7 ML–15.2 ML when cooling the films down to 90 K. Obviously, in the case of $d_{Fe} = 13.5$ ML, the temperature-driven spin-reorientation transition is verified (see inset of Fig. 3(b)).

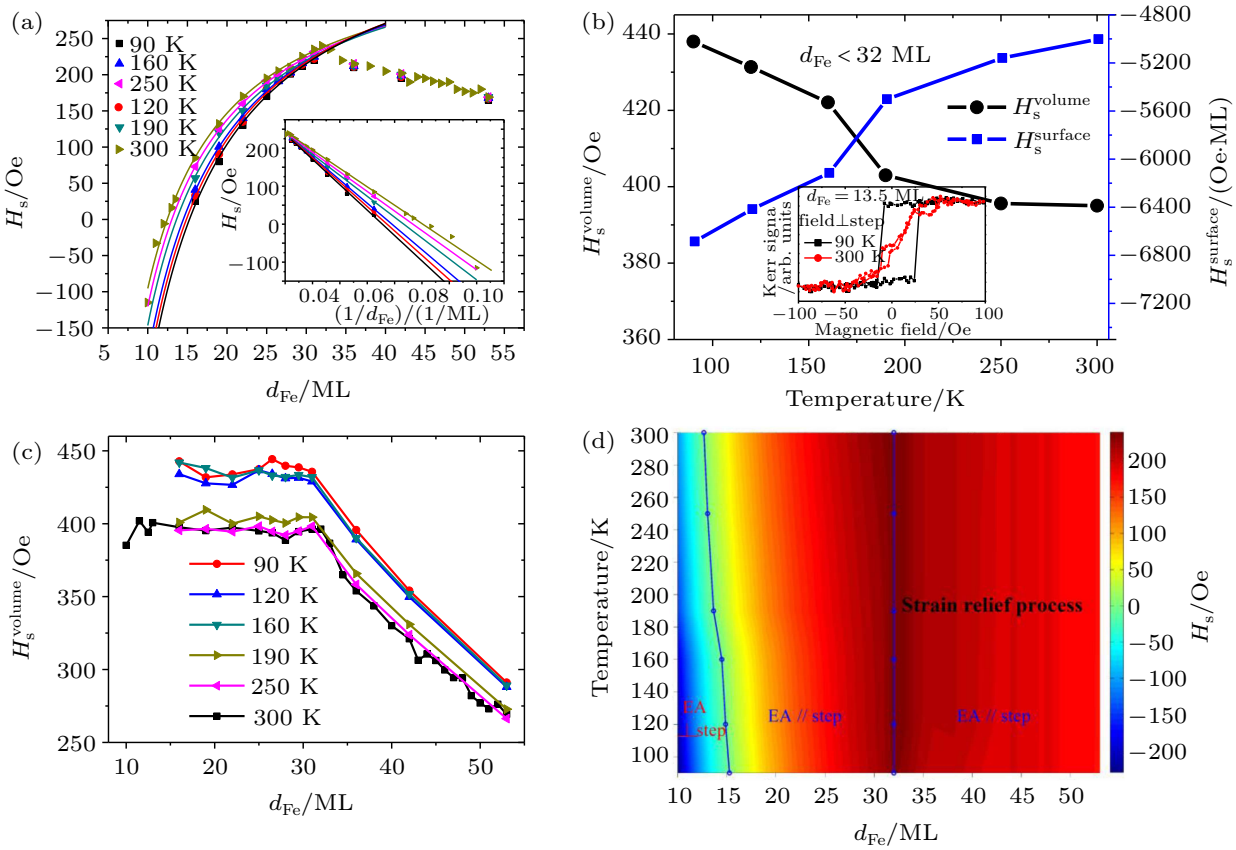


Fig. 3. (color online) (a) Dependences of H_s on thickness of Fe film of Fe/Si (557) film in the temperature range of 90 K–300 K, with the inset showing the plots of H_s versus $1/d_{Fe}$ at different temperatures. The dotted lines and solid lines are the experimental results and fitting results, respectively. (b) Temperature dependences of $H_s^{surface}$ and H_s^{volume} obtained from the fitting results for $d_{Fe} < 32$ ML, with the inset showing the *in-situ* Kerr loops perpendicular to the step for $d_{Fe} = 13.5$ ML for temperatures of 300 K and 90 K. (c) Fe film thickness dependences of H_s^{volume} obtained by setting $H_s^{surface}$ the values from panel (b) in the temperature range of 90 K to 300 K. (d) Thickness and temperature dependences of H_s obtained based on the experimental results and fitted anisotropy fields.

Deviation of experimental data from the fitting line is observed for $d_{Fe} > 32$ ML (see Fig. 3(a)). One of the most possible origins of the volume magnetic anisotropy is the lattice-mismatch-induced volume strain. According to the epitaxial relationships and lattice parameters between Fe and Si substrate (Fe $[1\bar{1}0] \parallel$ p(2×2) iron silicide $\parallel$ Si $[11\bar{2}]$ with $a_{Fe\ [110]} = 2.027$ Å and $a_{Si\ [220]} = 1.92$ Å), compressive stain is induced in Fe film.[17] The volume strain effect cannot persist at large thickness and a strain relief process happens at a certain critical thickness.[11] Figure 3(c) shows the thickness dependences of volume magnetic anisotropy by setting the surface magnetic anisotropy to be a constant obtained from fitting results. The volume magnetic anisotropy keeps almost constant for $d_{Fe} < 32$ ML but decreases quickly with further increasing thickness of Fe film, indicating that the critical thickness for the strain relief process in Fe/Si (557) films is about 32 ML.

Since the step-induced UMA in ferromagnetic layers can be strongly modified by tailoring strain,[17] surface magnetic anisotropy[11,14] or quantum well states[27] when covering the ferromagnetic film with non-magnetic film. Besides, understanding the effects of different capping layers on magnetic

anisotropy is also significant for designing device for spintronic application.[28] Therefore, to better understand the step-induced UMA, NaCl and Cu films are deposited as the capping layer to explore their influence on magnetic anisotropy of Fe/Si (557) film.

Figures 4(a) and 4(b) show the representative effects of NaCl and Cu capping layers on the UMA field of Fe/Si (557) film at room temperature, respectively. The UMA field of Fe/Si (557) film seems to be not affected by the NaCl absorption for $d_{\mathrm{Fe}} = 14$ ML and 50 ML. Differently, the magnetic anisotropy of Fe/Si (557) film can be strongly modified by Cu capping layer. Especially, in the case of $d_{\mathrm{Fe}} < 13$ ML, the spin orientation of Fe/Si (557) film is very sensitive to the Cu adsorption. For example, for $d_{\mathrm{Fe}} = 11$ ML, the magnetization EA switched from perpendicular to the step to parallel to the step after covering only 1-ML Cu capping layer on Fe/Si (557) film as shown in the inset of Fig. 4(b). To better clarify the effects of Cu capping layer on the magnetic anisotropy of Fe/Si (557) film, Cu film thickness (d_{Cu}) dependence of UMA field is obtained. In the case of $d_{\mathrm{Fe}} = 20$ ML, the Cu film thickness dependence of UMA field can be divided into three parts: H_{s} increases rapidly for $d_{\mathrm{Cu}} < 3$ ML (region I), decreases for 3 ML $< d_{\mathrm{Cu}} < 7$ ML (region II), and keeps almost constant for $d_{\mathrm{Cu}} > 7$ ML (region III). The increase of UMA in region I can be attributed to the Fe–Cu pair-bonding-induced reduction of surface magnetic anisotropy.[13] Due to the Volmer–Weber epitaxial growth mode for Cu film (see STM image shown in Fig. 1(d)), Fe film is not fully covered by Cu capping layer until d_{Cu} increases up to 3 ML. In region II, the surface magnetic anisotropy does not change too much. Therefore the reduction of UMA field can be assigned only to the decrease of volume magnetic anisotropy induced by Cu capping layer. The reduction of volume magnetic anisotropy is believed to be directly related to the strain relief process in Fe film. According to the epitaxial relationships and lattice parameters between bulk Fe and Cu capping layer (Cu $[1\bar{1}0] \parallel$ Fe $[1\bar{1}1]$ with $a_{\mathrm{Fe}}\ [1\bar{1}1] = 4.971$ Å and $a_{\mathrm{Cu}\ [1\bar{1}0]} = 5.105$ Å), tensile strain is induced in Cu film.[17] Therefore, considering the intrinsic compressive stain in Fe film, the epitaxial Cu capping layer conduces to the strain relief process in Fe film, and consequently leads to the reduction of volume magnetic anisotropy. In the case of $d_{\mathrm{Fe}} = 50$ ML, the individual contribution of surface magnetic anisotropy to UMA decreases greatly. Therefore, Cu-adsorption-induced strain relief is dominated over the Fe–Cu bond-induced reduction of surface magnetic anisotropy, resulting in only a decreasing trend for UMA field as a function of d_{Cu}. Differently, NaCl capping layer shows polycrystalline or amorphous structure, indicating that non-epitaxial NaCl capping layer cannot bond with Fe film, and consequently neither strain relief nor the reduction of surface magnetic anisotropy occurs.

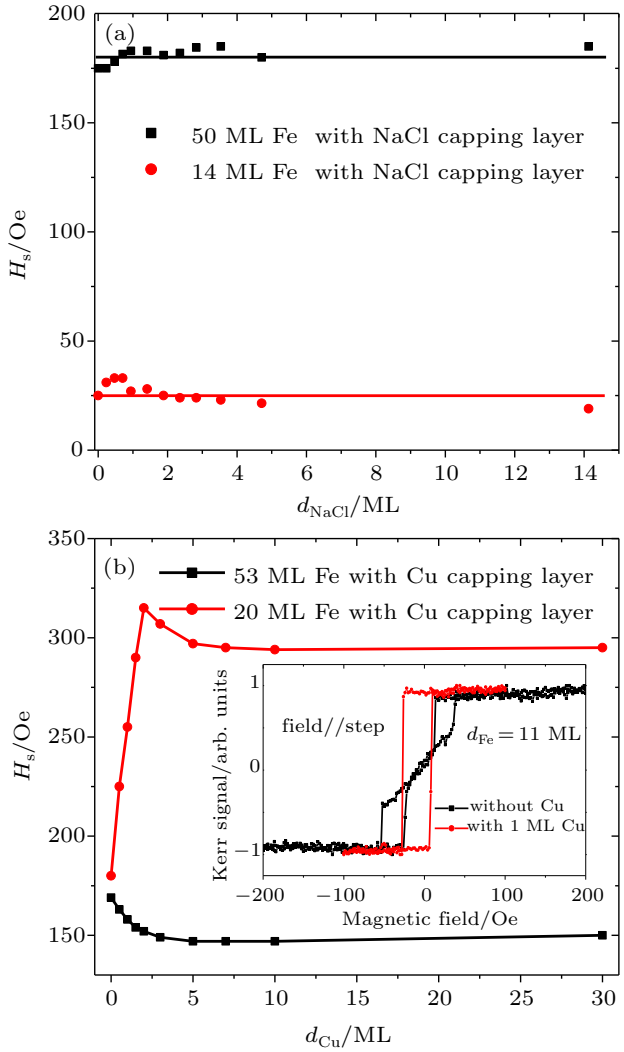


Fig. 4. (color online) Variations of H_{s} with NaCl capping layer thickness (d_{NaCl}) (a) and Cu capping layer thickness (d_{Cu}) (b) for representative Fe film thickness obtained at room temperature. The solid lines are guided by eye. The inset of panel (b) shows the *in-situ* longitudinal Kerr loops for 11-ML Fe with and without 1-ML Cu capping layer along the step edge, measured at room temperature.

4. Conclusions

In summary, the effects of thickness of Fe film, temperature and capping layer on the step-induced UMA of Fe/Si (557) films are systematically investigated. Step-induced competing surface and volume magnetic anisotropies, originating from the Néel pair-bonding of surface atomic and volume strain respectively, are extracted from thickness dependence of UMA field. Therefore, the in-plane spin orientation of Fe/Si (557) film is tunable by modifying the individual contributions of these two competing magnetic anisotropies to the UMA by thickness, temperature and capping layer. Our experimental results provide valuable information for manipulating the in-plane spin orientation and understanding the step-induced UMA in Fe/Si film.

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