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### Title

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# Element-specific study of epitaxial NiO/Ag/CoO/Fe films grown on vicinal Ag(001) using photoemission electron microscopy

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NiO/Ag/CoO/Fe single crystalline films are grown epitaxially on a vicinal Ag(001) substrate using molecular beam epitaxy and investigated by photoemission electron microscopy. We find that after zero-field cooling, the in-plane Fe magnetization switches from parallel to perpendicular direction of the atomic steps of the vicinal surface at thinner CoO thickness but remains in its original direction parallel to the steps at thicker CoO thickness. CoO and NiO domain imaging result shows that both CoO/Fe and NiO/CoO spins are perpendicularly coupled, suggesting that the Fe magnetization switching may be associated with the rotatable-frozen spin transition of the CoO film.

Magnetic interfacial interaction in antiferromagnetic/ferromagnetic (AFM/FM) systems generates many fascinating properties such as the exchange bias effect that as a AFM/FM system is cooled down within an external magnetic field to below the Néel temperature ( $T_N$ ) of the AFM layer, the FM layer hysteresis loop shifts in magnetic field.<sup>1,2</sup> Although originated from the AFM order,<sup>3,4</sup> the exchange bias depends sensitively on the detailed magnetic states of the AFM layer such as the cooling field orientation<sup>2</sup> and the magnetic domain states of the AFM layer,<sup>5</sup> suggesting that the AFM/FM interfacial interaction could lead to many metastable spin configurations. In an effort to provide a deep understanding, epitaxial single crystalline NiO/Fe<sup>6</sup> and CoO/Fe<sup>7,8</sup> systems were recently fabricated from which the AFM states of the single crystalline NiO and CoO layers can be probed directly using x-ray magnetic linear dichroism (XMLD).<sup>9</sup> With these advances it was identified that the NiO/Fe(001) and CoO/Fe(001) process a perpendicular interfacial couplings between the AFM and the FM spins above an AFM layer critical thickness<sup>6,10</sup> while the Fe/CoO(001) processes a collinear interfacial couplings only.<sup>11</sup> These different behaviors in AFM/FM and FM/AFM interfacial couplings well explain the mysterious perpendicular interlayer coupling between two FM layers across an AFM spacer layer.<sup>12,13</sup> Recently, the identification of the AFM rotatable spins in CoO/Fe(001) system<sup>10</sup> also reveals the mechanism of the rotatable magnetic anisotropy in the ultrathin regime of the AFM layer.<sup>14,15</sup> Despite the above progress, it remains unclear at the microscopic level on how the AFM/FM coupling leads to the different metastable spin configurations in an AFM/FM system. We studied single crystalline CoO/Fe and NiO/Ag/CoO/Fe films grown epitaxially on a vicinal Ag(001) substrate. By aligning the Fe magnetization parallel to the Ag[110] atomic steps of the vicinal surface at room temperature, we show that the AFM order of the CoO layer at low temperature switches the Fe magnetization to the per-

pendicular direction of the atomic steps below a critical CoO thickness.

A vicinal Ag(001) single crystal ( $\sim 2^\circ$  vicinal angle) with atomic steps parallel to the Ag [110] crystal axis was prepared by Ar ion sputtering at 2 keV and annealing at  $\sim 500$ – $600$  °C in an ultrahigh vacuum system.<sup>16</sup> A 30 monolayer (ML) Fe film was deposited on top of the Ag substrate followed by a CoO wedge, 2 ML Ag, and a NiO wedge to form the sample of NiO(wedge)/Ag(2 ML)/CoO(wedge)/Fe(30 ML)/Ag(001). The NiO and CoO films were grown by a reactive deposition of Ni and Co under an oxygen pressure of  $1 \times 10^{-6}$  Torr,<sup>10</sup> and the two wedges are orthogonal to each other so that their thicknesses can be changed independently. The sample was covered by a 2 nm Ag protection layer. Low energy electron diffraction measurement confirms the formation of single crystalline film with the bcc Fe [100] axis parallel to the fcc Ag [110] axis, and with the fcc CoO and NiO [110] axis parallel to the Fe [100] axis.<sup>17,18</sup> The sample was taken to the photoemission electron microscopy (PEEM3) beamline at the Advanced Light Source of Lawrence Berkeley National Laboratory. Element-specific domain imaging was performed for Fe using the effect of x-ray magnetic circular dichroism (XMCD), and for CoO and NiO using the effect of XMLD. Methods of taking domain images and assigning spin directions were reported in an earlier paper.<sup>19</sup>

We first present the result of CoO/Fe film. After magnetizing the film with an external magnetic field parallel to the atomic steps of the vicinal surface, the sample was transferred to the PEEM stage. Fe domain image confirms the formation of Fe single domain state with the magnetization parallel to the atomic steps (Fig. 1). No CoO XMLD signal was detected, showing that the CoO film is above its Néel temperature at room temperature. After cooling the sample to 100 K within zero magnetic field to below the  $T_N$  of the CoO film, it is surprising to find that the Fe magnetization switches by  $90^\circ$  to the perpendicular direction of the atomic steps below 11 ML CoO thickness ( $d_{\text{CoO}} < 11$  ML) but re-

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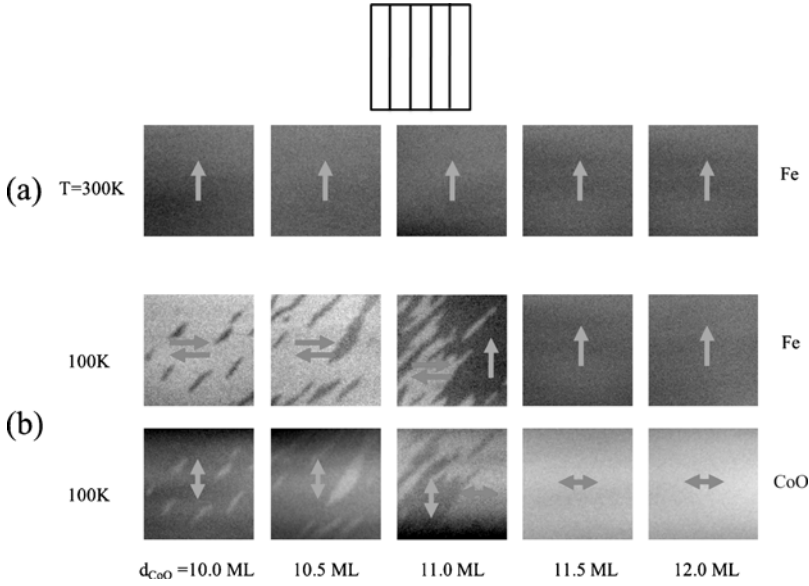


FIG. 1. (Color online) Fe and CoO magnetic domains of CoO/Fe/vicinal Ag(001) at (a)  $T=300$  K (above the CoO Néel temperature), and (b)  $T=100$  K (below the CoO Néel temperature). Arrows indicate the Fe and CoO spin directions, and the atomic steps of the vicinal surface are parallel to the vertical axis of each image. The CoO/Fe perpendicular magnetic coupling switches the Fe magnetization from parallel to perpendicular direction of the atomic steps of the vicinal surface for  $d_{\text{CoO}} < 11$  ML.

mains its original direction parallel to the atomic steps for  $d_{\text{CoO}} > 11$  ML (Fig. 1). XMCD imaging of the CoO film shows that the CoO spins are always coupled to the Fe spins perpendicularly (Fig. 1), both above and below  $d_{\text{CoO}} = 11$  ML, in agreement with our earlier result for CoO/Fe grown on flat Ag(001).<sup>10</sup> The  $90^\circ$ -coupling at the AFM/FM interface is due to the “spin-flop” coupling mechanism<sup>20</sup> which gives raises a magnetic anisotropy ( $K_u$ ) to the FM film whose strength is  $K_u = J_{FA}^2/J_A$ , where  $J_{FA}$  is the AFM/FM interfacial coupling,  $J_A \sim 100$  erg/cm<sup>2</sup> is the AFM (CoO) exchange interaction. In theory,  $K_u$  is estimated to be  $\sim 0.8$  erg/cm<sup>2</sup> for a perfect compensated interface.<sup>21</sup> In experiment,  $K_u$  can be estimated from the saturation field of the hard axis loop  $K_u = H_S M_F d_F$ . With the typical values of  $H_S \sim 3000$  Oe,  $M_F \sim 1700$  Oe, and  $d_F \sim 10$  ML for Fe/CoO layer, the CoO/Fe coupling induced anisotropy is estimated to be  $K_u \sim H_S M_F d_F \sim 0.7$  erg/cm<sup>2</sup> which is roughly the same order of magnitude as the theoretical estimation. From here we can also estimate the CoO/Fe interfacial coupling to be  $J_{FA} = \sqrt{J_A K_u} \sim 8$  erg/cm<sup>2</sup>.

It is well known that vicinal Ag(001) surface with atomic steps parallel to Ag[110] axis induces an in-plane uniaxial magnetic anisotropy to the Fe film ( $K_{\text{Step}} \sim 0.1-0.2$  erg/cm<sup>2</sup>) that favors a parallel alignment of the Fe magnetization to the atomic steps at all temperatures.<sup>22,23</sup> Note that the CoO/Fe interfacial coupling favors a perpendicular alignment between the CoO and Fe spins, thus the existence of the Fe  $90^\circ$  switching implies that the vicinal surface also favors CoO spins to be parallel to the atomic steps so that the CoO/Fe perpendicular interfacial coupling would have to switch the Fe spins to the perpendicular direction of the steps by sacrificing the Fe or Co step-induced magnetic anisotropy. Then the interesting question is why this occurs only at  $d_{\text{CoO}} < 11$  ML? While we could not give a firm answer, we offer two possible explanations. One is that the CoO step-induced anisotropy accidentally changes its sign at  $d_{\text{CoO}} = 11$  ML, favoring the CoO spins to be parallel to the atomic steps for  $d_{\text{CoO}} < 11$  ML and perpendicular to the atomic steps at  $d_{\text{CoO}} > 11$  ML. This mechanism is unlikely although we cannot rule it out completely. The other and plausible mechanism is that the CoO step-induced anisotropy does not change its sign. As the sample is cooled down

to establish the CoO AFM order, the CoO/Fe  $90^\circ$ -coupling should initially align the CoO spins perpendicular to the vicinal steps in order to keep the CoO/Fe  $90^\circ$  coupling. Then after the CoO spins are well ordered at low temperature, the CoO spins prefer to go to its easy axis to be parallel to the steps by switching the Fe spins to the perpendicular direction of the steps. But this can only occur when the CoO spins are rotatable. That is why we observe a parallel alignment of the CoO spins to the steps only at  $d_{\text{CoO}} < 11$  ML because our previous work shows that CoO spins are rotatable at  $d_{\text{CoO}} < 11$  ML but frozen at  $d_{\text{CoO}} > 11$  ML.<sup>10</sup> The second explanation also requires that the Co/Fe  $90^\circ$ -coupling and the induced anisotropy are stronger than the Fe step-induced anisotropy so that as the CoO spins are parallel to the steps at  $d_{\text{CoO}} < 11$  ML, the Fe spin switch to its hard axis (perpendicular to the steps) in order to maintain the  $90^\circ$  alignment between and CoO spins. This is justified from our estimation of  $J_{FA} \sim 8$  erg/cm<sup>2</sup>  $>$   $K_u \sim 0.7$  erg/cm<sup>2</sup>  $>$   $K_{\text{Step}} = 0.1-0.2$  erg/cm<sup>2</sup>. It should also be mentioned that the CoO has a very strong anisotropy ( $K_{\text{CoO}} \sim 2.7 \times 10^8$  erg/cm<sup>3</sup>).<sup>2</sup> That is why once the CoO spins are frozen at  $d_{\text{CoO}} > 11$  ML, they are no longer rotatable anymore.

We further studied NiO/Ag(2 ML)/CoO/Fe film on vicinal Ag(001) in which the NiO and CoO are magnetically coupled across the 2 ML Ag layer. Figure 2 shows the magnetic domains of Fe, CoO, and NiO layers at  $d_{\text{NiO}} = 3$  ML. First, we find that the NiO and CoO spins are always coupled perpendicular to each other throughout the thickness range studied. This result shows indirectly the high quality of our sample because if large pinholes existed in the 2 ML Ag spacer layer, we would expect a collinear coupling between the CoO and NiO spins due to their direct contact. The  $90^\circ$ -coupling between two FM layer across a spacer layer is usually attributed to the spin frustration<sup>24</sup> but this mechanism cannot be applied directly to the interlayer coupling between two AFM layers across a spacer layer. Future theoretical study is needed to explain our observation. Second, we find that the Fe  $90^\circ$  switching occurs at a thinner CoO critical thickness than the CoO/Fe case. The CoO critical thickness is determined as a function of the NiO thickness. The result shows that the CoO critical thickness decreases monotonically with increasing the NiO thickness (Fig. 3). The result of

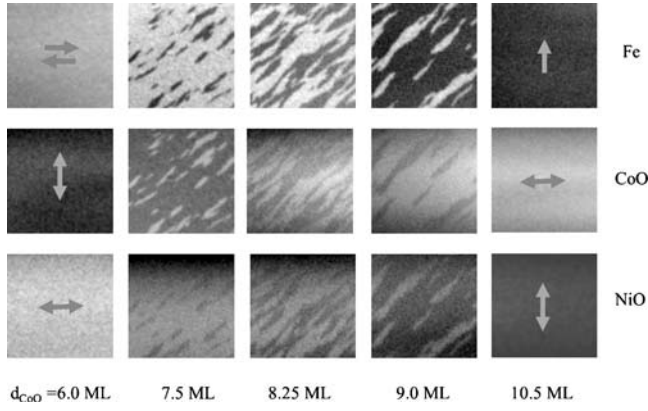


FIG. 2. (Color online) Fe, CoO, and NiO magnetic domains of NiO(3 ML)/Ag(2 ML)/CoO/Fe/vicinal Ag(001) at  $T=100$  K. Fe  $90^\circ$  magnetization switching persists but at a thinner CoO thickness than the CoO/Fe bilayer.

Fig. 3 favors the frozen spin mechanism over the sign change in the step-induced CoO anisotropy because the step-induced CoO anisotropy is unlikely to change its sign by adding the NiO/Ag overlayer. On the other hand, the addition of a NiO film which is magnetically coupled to the CoO layer is equivalent to increasing the magnitude of the fourfold CoO anisotropy so that frozen spins should occur at a thinner CoO thickness than that without the NiO film. Although this could explain why the Fe  $90^\circ$  switching occurs at a thinner CoO thickness by adding the NiO/Ag overlayer, a firm determination of the underlying mechanism requires more experiments in the future.

In summary, single crystalline Co/Fe and NiO/Ag/CoO/Fe films were grown epitaxially on vicinal Ag(001) substrate. The Fe magnetization was first magnetized parallel to atomic steps of the vicinal surface at room temperature and then was cooled down to 100 K in zero magnetic field.

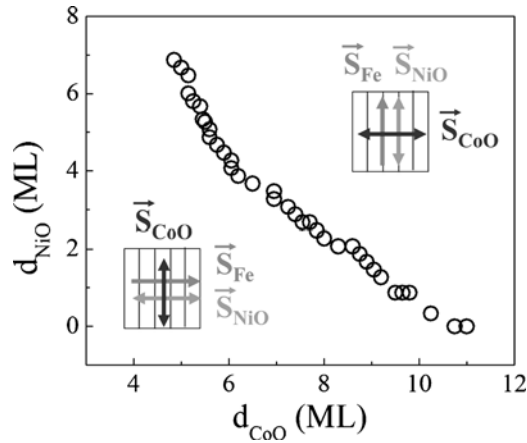


FIG. 3. (Color online) The Fe  $90^\circ$  magnetization switching point in the CoO–NiO thickness plane at  $T=100$  K. Schematic drawings show the spin directions of Fe, CoO, and NiO on the vicinal surface.

Element-specific domain imaging shows that the Fe spins in the Fe/CoO film are switched to perpendicular direction of the atomic steps at low temperature for  $d_{\text{CoO}} < 11$  ML and remains its direction parallel to the atomic steps at  $d_{\text{CoO}} > 11$  ML. The CoO spins are coupled perpendicularly to the Fe spins. For NiO/Ag/CoO/Fe film, we find that the NiO and CoO spins are also coupled perpendicularly, and that the Fe spin  $90^\circ$  switching occurs at thinner CoO thickness with increasing the NiO thickness.

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