Ni spin switching induced by magnetic frustration in FeMn/Ni/Cu(001)

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FeMn/Ni/Cu(001) bilayer films are grown epitaxially and investigated by photoemission electron microscopy and magneto-optic Kerr effect. We find that as the FeMn overlayer changes from paramagnetic to antiferromagnetic state, it switches the ferromagnetic Ni spin direction from the out-of-plane to an in-plane direction of the film. This phenomenon reveals the mechanism of creating magnetic anisotropy by the out-of-plane spin frustration at the FeMn-Ni interface.

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Controlling the local electron-spin direction in a magnetic nanostructure is a key step toward the spintronics technology.[1] Various methods have been proposed to reach this goal such as the spatial variation in the g factor,[2] tuning of the charge density,[3] spin-torque effect,[4,5] and the voltage-controlled multiferroic antiferromagnet,[6] etc. All these approaches are based to some extent on the spin-charge coupling to modify the electronic states that are coupled to the electron spins. For magnetic materials, such spin-charge coupling often manifests as the spin-orbit coupling which generates the so-called magnetic anisotropy to determine the electron-spin direction. Therefore a control of the electron-spin direction is ultimately related to the manipulation of the magnetic anisotropy.[7–9] Although research on the magnetic anisotropy has been greatly advanced in the last decades, the disadvantage is that once a nanostructure is synthesized the interfacial electronic states are fixed so that it is very difficult to change the magnetic anisotropy anymore. Therefore it has been highly demanded to explore all possible mechanisms to generate the magnetic anisotropy. In this Brief Report, we demonstrate a candidate mechanism to generate the magnetic anisotropy. We show that the spin direction of a Ni thin film in FeMn/Ni/Cu(001) could be switched from out-of-plane to in-plane direction of the film by establishing an antiferromagnetic order of the FeMn film. We attribute this result to the FeMn/Ni interfacial frustration-induced magnetic anisotropy which shifts the Ni-spin-reorientation transition (SRT) thickness[10] by as much as 40%. We choose this system because FeMn/Ni/Cu(001) films can be grown epitaxially and that the FeMn has a well-known 3Q antiferromagnetic spin structure so that well-defined single crystalline ultrathin films can be used for this study with the FeMn Néel temperature easily tuned by changing its film thickness.[11] A 10-mm-diameter Cu(001) single-crystal disk was mechanically polished down to 0.25 μm diamond paste, followed by an electropolish.[12] The substrate was cleaned *in situ* by cycles of Ar^+ sputtering at 2–5 keV and annealing at 600–700 °C. FeMn/Ni/Cu(001) films were grown epitaxially at room temperature with the FeMn and Ni films grown into cross wedges for the purpose of controlling their thicknesses independently. A 10 ML Cu layer was grown on top of the FeMn to protect the sample from contamination. Magnetic properties of the films were measured by magneto-optic Kerr effect (MOKE) and by photoemission electron microscopy (PEEM) at the advanced light source. The magnetic domain images were obtained by taking the ratio of L_3 and L_2 edges utilizing the effect of x-ray magnetic circular dichroism (XMCD).[13] All measurements were made at room temperature.

We first present the Ni domain images (Fig. 1) of FeMn/Ni/Cu(001) at fixed Ni thicknesses of 8.0 ML as a function of the FeMn overlayer thickness. The Ni magnetic domains exhibit two colors below 7.5 ML of FeMn ($d_{FeMn}<7.5$ ML) and multiple colors above 7.5 ML of FeMn. After rotating the sample by 90° with respect to its surface-normal direction, the Ni domain colors remain unchanged for $d_{FeMn}<7.5$ ML but change for $d_{FeMn}>7.5$ ML. Recalling that the Ni domain color is determined by the angle between the incident x ray and the local spin direction, we conclude that the Ni magnetization in Fig. 1 is perpendicular to the film plane for $d_{FeMn}<7.5$ ML and in the film plane for $d_{FeMn}>7.5$ ML, i.e., the FeMn/Ni(8.0 ML)/Cu(001) films undergoes a spin-reorientation transition at 7.5 ML of FeMn thickness. Noticing that the Ni film thickness is fixed at 8.0 ML,

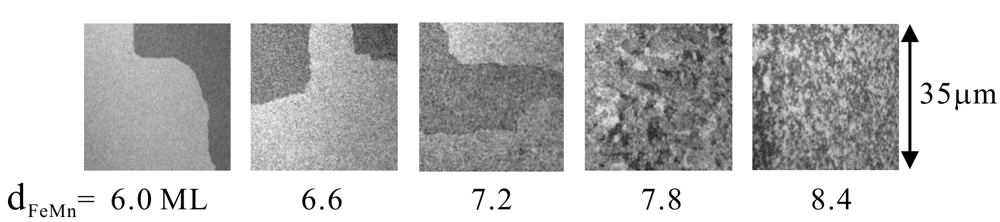

FIG. 1. Ni domain images of FeMn/Ni(8.0ML)/Cu(001) as a function of the FeMn overlayer thickness. The antiferromagnetic order of FeMn overlayer above 7.5 ML switches the Ni spin from out-of-plane to in-plane direction of the film.

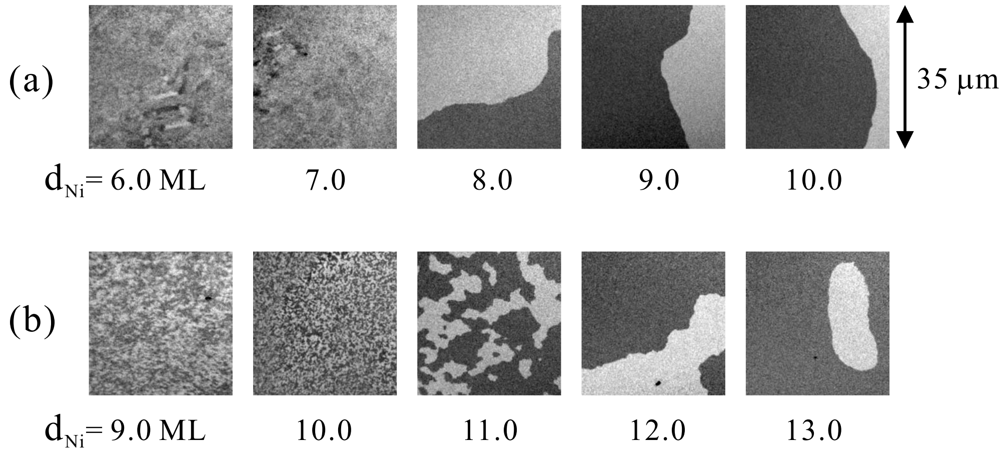

FIG. 2. Ni domain images of FeMn/Ni/Cu(001) as a function the Ni-film thickness. The Ni spin-reorientation transition takes place (a) at d_{SRT}=7.5 ML for paramagnetic FeMn overlayer (d_{FeMn}=6.0 ML) and (b) at d_{SRT}=10.5 ML for antiferromagnetic FeMn overlayer (d_{FeMn}=8.4 ML).

the SRT in Fig. 1 is actually induced by the FeMn overlayer rather than by the Ni film itself as in the conventional SRT in Ni/Cu(001) system.[14] On the other hand, the Ni spin direction should be ultimately determined by its overall magnetic anisotropy. Then the result of Fig. 1 shows that the FeMn film above 7.5 ML thickness must have induced a magnetic anisotropy to the Ni film. This result consequently implies that FeMn film thinner and thicker than 7.5 ML should lead to different Ni SRT as a function of the Ni thickness, respectively. To verify this fact, we show in Fig. 2 the Ni PEEM images as a function of the Ni film thickness at fixed FeMn thicknesses of 6.0 and 8.4 ML, respectively. For each case, the Ni film shows an in-plane to out-of-plane SRT with increasing the Ni thickness. However, the Ni-SRT thickness of d_{SRT}=10.5 ML in the d_{FeMn}=8.4 ML sample is about 40% greater than the d_{SRT}=7.5 ML value in the d_{FeMn}=6.0 ML sample confirming that thicker FeMn film (d_{FeMn}>7.5 ML) induces a magnetic anisotropy which favors an in-plane alignment of the Ni spins. Since both samples have the same FeMn/Ni interface and the interfacial magnetic anisotropy depends very little on the overlayer thickness above 5ML,[15] the results of Figs. 1 and 2 must come from the magnetic state change in the FeMn film. Noticing that the Néel temperature of the FeMn film increases with its film thickness, we attribute the FeMn-induced magnetic anisotropy to the antiferromagnetic order of the FeMn overlayer in FeMn/Ni/Cu(001) film. To support this conclusion, we determined the Ni-SRT thickness d_{SRT} from the PEEM images as a function of the FeMn thickness (Fig. 3). The thickness error from the PEEM image determination is ~0.15 ML. The d_{SRT} remains a constant of 7.5 ML for d_{FeMn}<7 ML, exhibits a sudden increase for 7<d_{FeMn}<8 ML and reaches another constant value of 10.5 ML for d_{FeMn}>8 ML. Then the constant Ni d_{SRT} values for d_{FeMn}<7 ML and d_{FeMn}>8 ML correspond to the paramagnetic and antiferromagnetic states of the FeMn films. The critical thickness value of d_{FeMn}=7.5 ML is similar to the literature value.[16,17]

To further support our conclusion, MOKE measurement was taken at room temperature. Figure 4(a) shows the Ni polar loops, which measure the Ni perpendicular magnetization as a function of the Ni thickness at paramagnetic (d_{FeMn}=4.3 ML) and antiferromagnetic (d_{FeMn}=9.7 ML) state of the FeMn film, respectively. In both cases, the Ni film develops the polar signal above a critical thickness to eventually evolve into a square loop with a full remanence showing the Ni SRT from in-plane to out-of-plane directions with increasing the Ni thickness. However, there are two major differences. First, the Ni-SRT critical thickness is thinner at paramagnetic FeMn [left column in Fig. 4(a)] than at antiferromagnetic FeMn [right column in Fig. 4(a)]. This can be more clearly seen in Fig. 4(b) where the Ni polar remanence ($M_\perp$) is plotted as a function of the Ni thickness for d_{FeMn}=4.3 ML and 9.7 ML, respectively. Second, it is obvious that the Ni coercivity (H_C) at d_{FeMn}=9.7 ML is much greater than at d_{FeMn}=4.3 ML. The H_C at a fixed Ni thickness of 14.5 ML shows that the Ni H_C remains a constant value below 7.5 ML FeMn and then increases rapidly above 7.5 ML FeMn [Fig. 4(c)]. The drastic increase in H_C above an FeMn critical thickness is a signature of the antiferromagnetic order in the FeMn film.[11] Therefore we confirm our conclusion that it is the antiferromagnetic order of the FeMn overlayer above 7.5 ML that induces a magnetic anisotropy to the Ni film.

To understand why the FeMn antiferromagnetic order in-

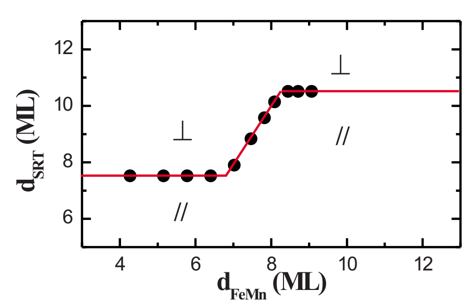

FIG. 3. (Color online) The Ni-SRT critical thickness d_{SRT} as a function of d_{FeMn}. The red solid line is guide to eyes. The antiferromagnetic order of the FeMn film above 7.5 ML generates a magnetic anisotropy to increase the Ni-SRT thickness from 7.5 to 10.5 ML.

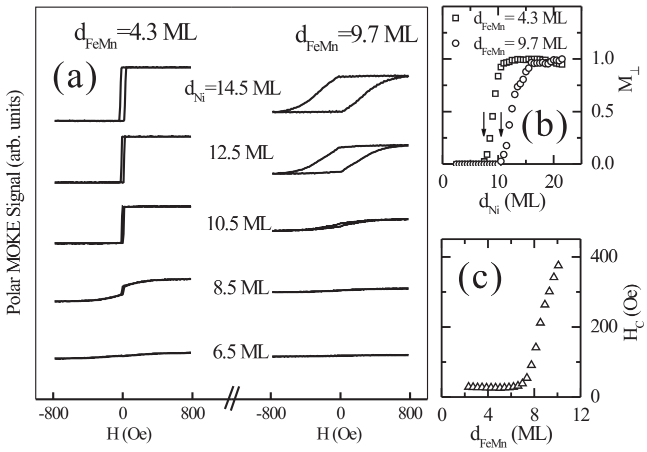

FIG. 4. (a) Polar MOKE hysterisis loops of FeMn/Ni/Cu(001) as a function of Ni thickness for paramagnetic FeMn overlayer (left column, $d_{\mathrm{FeMn}}=4.3$ ML) and antiferromagnetic FeMn overlayer (right column, $d_{\mathrm{FeMn}}=8.4$ ML). (b) The Ni polar remanence as a function of Ni film thickness. Arrows indicate the Ni-SRT thickness. (c) The coercivity of FeMn/Ni(14.5ML)/Cu(001) as a function of the FeMn film thickness.

duces a magnetic anisotropy, we consider the well-known 3Q-like spin structure of the face-centered-cubic (fcc) FeMn lattice [Fig. 5(a)].[18] For FeMn (001) atomic planes, although the in-plane net spin is zero, the out-of-plane net spin is actually nonzero but alternating its direction between neighboring (001) planes. Then at the FeMn/Ni interface with the presence of atomic steps (inevitable in real experimental systems), this kind of 3Q spin structure will give rise to a nonzero perpendicular net spin at each atomic terrace whose direction alternates between neighboring terraces [Fig. 5(b)], as well as an uncompensated in-plane spin only at the [100]-type step edges [Fig. 5(c)] [11]. For the perpendicular FeMn spin component, the FeMn/Ni magnetic coupling will then produce a magnetic frustration due to the atomic terraces.[19] The FeMn-Ni interfacial interaction favors an alternating alignment of the Ni spins between neighboring terraces while the Ni-Ni interaction prefers a parallel alignment of the

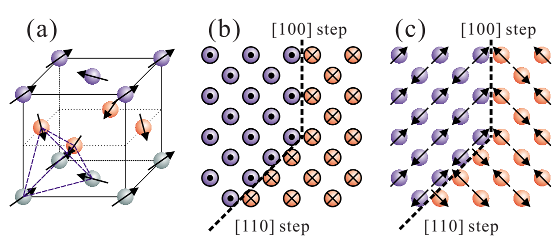

FIG. 5. (Color online) (a) the schematic drawing of 3Q-like FeMn spin structure. Arrows represent the spin orientation. Atoms are painted in three different colors to indicate different (001) planes. The dashed lines in (a) show the tetrahedral unit cell. (b) The out-of-plane, and (c) in-plane FeMn spin components at a (001) island with [100] and [110] steps. The net out-of-plane spin component is nonzero but alternates its direction between neighboring terraces (indicated by dot and cross at the center of atoms). The in-plane spin component has a nonzero net spin only at the [100]-type step edges.

Ni spins. This magnetic frustration is similar to the case of the biquadratic interlayer coupling in magnetic sandwiches[20] and the 90° coupling at the FM/atomic force microscopy (AFM) interfaces[21] where the interlayer/interfacial magnetic coupling competes with the FM intralayer coupling. The competition result is to generate a magnetic anisotropy which favors a perpendicular alignment of the FM spins to the antiferromagnetic spins, similar to the well-known "spin-flop" state in bulk antiferromagnets.[22] Then the FeMn/Ni out-of-plane interfacial magnetic frustration should generate a magnetic anisotropy that favors the Ni spins to be perpendicular to the FeMn out-of-plane spin direction (e.g., in-plane direction for the Ni spins). This explains why the FeMn antiferromagnetic order favors an in-plane alignment of the Ni spins. For the in-plane component of the FeMn spins, the uncompensated spins at $[\pm 1,0,0]$- and $[0,\pm 1,0]$-step edges should create an equivalent fourfold magnetic anisotropy for the in-plane Ni magnetization[11] which could also favor an in-plane alignment of the Ni spins.

To differentiate the above two mechanisms, we performed an experiment using vicinal Cu(001) substrate with the atomic steps parallel to [100] direction. The idea is that the interfacial frustration due to the FeMn out-of-plane spin component should scale linearly with the terrace area so that the magnetic anisotropy (frustration energy per unit area) should be weakly dependent on the step density.[20] On the other hand, the effect due to the in-plane FeMn-uncompensated spin component at the [100]-step edges should obviously scale with the [100]-step density. Therefore a study of the Ni-SRT thickness as a function of the vicinal angle will distinguish these two mechanisms. A curved Cu(001) substrate is used in our experiment to change the vicinal angle (α) continuously.[23] After growing a Ni wedge with its slope along the [100]-step direction and covering the Ni wedge with a uniform FeMn film, MOKE measurement is carried out to determine the Ni-SRT thickness d_{SRT}. It should be mentioned that the roughness of Ni film could smear out the regular step morphology of the vicinal Cu substrate. However, our LEED measurement indicates that double LEED spots persist after the Ni film growth indicating a well-transferred step density from the Cu substrate to the Ni/FeMn interface. Previous study on the step decoration in vicinal Ni/Cu(001) system also indicates that steps from the Cu substrate indeed persist on top of the Ni film.[24] It should also be mentioned that FeMn/Ni/Cu(001) could have a different FeMn/Ni interfacial roughness than Ni/FeMn/Cu(001) thus exhibits a different magnetic behavior.[25] Of course a final answer on the film roughness will depend on an *in situ* surface-morphology measurement using scanning tunneling microscopy. Figure 6 shows the result of d_{SRT} versus the vicinal angle α for paramagnetic ($d_{\mathrm{FeMn}}=5$ ML) and antiferromagnetic ($d_{\mathrm{FeMn}}=17$ ML) FeMn overlayers. The purpose of including the paramagnetic FeMn case is to identify possible effect of the step-induced magnetic anisotropy on the Ni SRT.[9] We find that for paramagnetic FeMn ($d_{\mathrm{FeMn}}=5$ ML), the d_{SRT} value of 7.5ML is independent of α, showing that we can ignore the effect of the step-induced magnetic anisotropy on the Ni SRT. As the FeMn film becomes antiferromagnetic at thicker thickness ($d_{\mathrm{FeMn}}=17$ ML), the Ni d_{SRT} value shifts from 7.5 to 10.5 ML showing the effect of

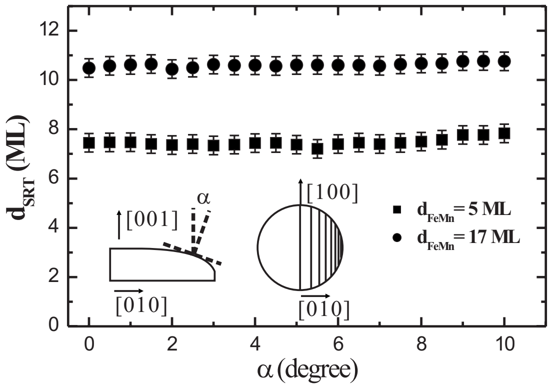

FIG. 6. The Ni-SRT thickness d_{SRT} of FeMn/Ni/Cu(001) as a function of the vicinal angle α for d_{FeMn}=5 ML and 17 ML of FeMn/Ni grown on vicinal Cu(001) with steps parallel to [100].

the FeMn/Ni interfacial frustration on the Ni SRT. More importantly, the d_{SRT} value remains a constant of 10.5 ML rather than increases with the vicinal angle α showing that the FeMn-uncompensated in-plane spins at the [100]-step edges do not have an effect on the Ni SRT. Therefore the result of Fig. 6 favors the conclusion that it is the FeMn out-of-plane spin component that is responsible for the FeMn-induced magnetic anisotropy. Taking the 3 ML Ni-SRT thickness shift and the Ni magnetic anisotropy value in Ni/Cu(001) system [10], we estimate the strength of this frustration-induced magnetic anisotropy to be $\sim$70 μeV/spin, the same order of magnitude as estimated by Koon[21] for an idealized frustrated interface. There sometimes exists induced moment in the antiferromagnetic film at the interface when in contact with a ferromagnetic film. This indeed was observed in FeMn/Co system where both Fe and Mn XMCD signala were detected.[26] However, this induced moment is not responsible for the SRT thickness shift reported in this Brief Report because the induced moment was observed for both paramagnetic and the antiferromagnetic phases of the FeMn film. We would like to point out that the frustration-induced SRT should be a general phenomenon as far as the interfacial crystal plane of the film carries an uncompensated net spin whose direction alternates between neighboring terraces. Finally, another interesting topic for future study could be the exchange bias in this system because the unidirectional and uniaxial magnetic anisotropies due to the interfacial interaction could be separated into two directions in this system.

In summary, we studied the Ni spin-reorientation transition in FeMn/Ni/Cu(001) system and find a 40% Ni-SRT thickness shift as the FeMn overlayer transits from paramagnetic to antiferromagnetic state. We attribute this giant shift to the out-of-plane FeMn-Ni interfacial magnetic frustration which generates a magnetic anisotropy to favor an in-plane alignment of the Ni spins.

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