Symmetry-Breaking Induced Exchange Bias in Ferromagnetic Ni-Cu-Co and Ni-Fe-Co Sandwiches Grown on a Vicinal Cu(001) Surface

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Ferromagnetic Ni-Cu-Co and Ni-Fe-Co sandwiches were grown epitaxially onto a vicinal Cu(001) substrate and investigated using magneto-optical Kerr effect and x-ray magnetic circular dichroism techniques. We find that the atomic steps of the vicinal surface break the magnetic reversal symmetry to induce an exchange bias in the Ni perpendicular magnetic hysteresis loop. The Ni exchange bias direction can be switched by changing the direction of the in-plane Co magnetization. In addition, the exchange bias can be tailored by changing the Cu or Fe spacer layer thickness.

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Exchange bias [1] refers to a shift of the ferromagnetic hysteresis loop, and is often realized when a ferromagnetic (FM)–antiferromagnetic (AFM) system is cooled down within a magnetic field to below the Néel temperature of the AFM material. Exchange bias has been recognized as one important subject of nanomagnetism research not only because it represents a model system for the study of the FM/AFM interfacial interaction, but also because of its important application in magnetic recording technology. Despite the great progress and many important discoveries [2–7] made in the last decade, a complete understanding of the exchange bias [8–10] remains somewhat controversial. The difficulty lies in the spin compensation of the AFM material which results in a complicated spin structure (or spin frustration) at the FM/AFM interface, making it very difficult to understand and to control the exchange bias. As compared to the complicated FM/AFM interfacial interaction, FM/FM interaction is relatively simple because of the absence of the spin frustration at the FM/FM interface. However, it has been believed for a long time that exchange bias should not exist in a ferromagnetic-only system. The reason for the existence of the exchange bias in a FM/AFM system is that most of the AFM spins are actually frozen within a finite magnetic field so that the AFM material virtually does not respond to the applied magnetic field. The above fact leads to the idea of realizing the exchange bias in a FM-only system by introducing new concepts to freeze some of the FM spins within a finite magnetic field. In fact, in an attempt to open a new direction in the research of the exchange bias, FM-only heterostructures have been applied recently to simulate the exchange bias [11–13]. However, these systems suffer other problems such as the lack of tunability and the nonstability of the exchange bias [14]. Therefore, it is highly demanded to explore new FM-only systems with new concepts that could provide both the magnetic reversal symmetry breaking as well as the tunability and stability of the exchange bias. In this Letter, we report our study on ferromagnetic Ni-Cu-Co and Ni-Fe-Co sandwiches grown on vicinal Cu(1,1,40). Using atomic steps of the vicinal surface, we are able to break the Co magnetic reversal symmetry to generate an exchange bias in the Ni hysteresis loop. Both the direction and the magnitude of the Ni exchange bias can be easily controlled by the Co in-plane magnetization direction and the spacer layer thickness. In addition, the exchange bias is very stable against cycling of the magnetic field.

An electropolished Cu(1,1,40) (2° vicinal angle) single crystal disk was cleaned in an ultrahigh vacuum system by cycles of Ar ion sputtering at ~2 keV and annealing at 600 °C. Ni-Cu-Co and Ni-Fe-Co sandwiches were grown epitaxially onto the Cu(1,1,40) substrate at room temperature, and then capped with 2 nm Cu to prevent contamination. The Co and Ni layers are 8 and 20 ML thick, respectively. Wedge shaped fcc Cu and fcc Fe spacer layer were grown along the $[1\bar{1}0]$ direction to facilitate a continuous change of the oscillatory magnetic interlayer coupling [15]. The samples were characterized by low energy electron diffraction and Auger electron spectroscopy. The result shows that our sample is of contamination-free and high quality single crystalline film, consistent with literature result that Co and Ni grow epitaxially on Cu(001) [16,17]. Magnetic properties of the samples were measured by magneto-optic Kerr effect (MOKE) [18] with the magnetic field either parallel (longitudinal) or perpendicular (polar) to the film. For the purpose of separating the Ni and Co magnetic signals, we grew uniform thickness samples and did element-specific magnetic measurement using x-ray magnetic circular dichroism (XMCD) [19] at beam line 4 of the Advanced Light Source (ALS).

Co/Cu(001) has an in-plane magnetization [20], and Ni/Cu(001) and Ni/Fe/Cu(001) have a perpendicular magnetization above a Ni critical thickness [21,22]. Thus the Ni and Co films should have perpendicular and in-plane magnetizations, respectively, in the Ni-Cu-Co/Cu(001) and Ni-Fe-Co/Cu(001) sandwiches above the Ni critical thickness [23]. Therefore the longitudinal and polar

MOKE signals should mainly come from the in-plane Co and the perpendicular Ni magnetizations, respectively. We first performed the longitudinal MOKE measurement and obtained square shape hysteresis loops for magnetic field parallel (x axis) and perpendicular (y axis) to the atomic steps, showing that the in-plane Co magnetization could be well aligned along the x and y axes even though the vicinal substrate induce a weak in-plane uniaxial magnetic anisotropy [24]. Because of the four stable in-plane Co magnetization axes ($\pm x$ and $\pm y$), we took four polar MOKE measurements after applying an in-plane magnetic field (500 Oe) pulse to align the Co magnetization along the corresponding four in-plane directions. Figure 1 shows polar hysteresis loops from Ni(20 ML)/Fe(8 ML)/Co(8 ML) sample where the 8 ML fcc Fe produces a ferromagnetic interlayer coupling between the Ni and Co magnetizations [23]. For Co magnetization parallel to the atomic steps ($\pm x$ axis), the polar MOKE signal shows identical hysteresis loops for $M_{Co} \| +x$ and $M_{Co} \| -x$ without an exchange bias. This is expected because of the invariance of this system under $+x \rightarrow -x$. For Co magnetization perpendicular to the atomic steps ($\pm y$ axis), we observed a clear exchange bias in the polar hysteresis loops [Fig. 1(b)]. By changing the in-plane Co magnetization from $+y$ to $-y$ directions, the exchange bias switches its sign accordingly. A similar result was also obtained on the Ni-Cu-Co sample so that the observed exchange bias is not related to the AFM nature of the fcc Fe [25]. We here use fcc Fe simply because it provides a stronger interlayer coupling than the Cu spacer layer [26,27].

To further separate the Co and Ni magnetic signals, we measured two samples of Ni(20 ML)/Cu/Co(8 ML)/Cu(1, 1, 40) with $d_{Cu} = 5$ and 7 ML using the XMCD technique which allows the element-specific hysteresis loop measurement [28]. Similar to the MOKE measurement, we magnetized the sample with an in-plane magnetic field (500 Oe) pulse to align the Co magnetization along the $\pm x$ and $\pm y$ axes before taking the polar hysteresis loop measurement. Figure 2 shows the polar XMCD result from $d_{Cu} = 5$ ML sample in which the Ni and Co magnetizations are coupled ferromagnetically. We first took the XMCD measurement for normal incidence of the x-ray which detects only the normal component of the magnetization. For $M_{Co} \| \pm x$, we find no exchange bias in the Ni polar loop. For $M_{Co} \| \pm y$, the Ni polar loop exhibits an exchange bias with its sign depending on the Co in-plane magnetization direction [Fig. 2(a)]. The Co XMCD signal remains ~zero during the sweeping of the perpendicular magnetic field [Fig. 2(b)]. This result confirms that the Ni and Co magnetizations are perpendicular and parallel to the film plane, respectively. To further confirm that the Co in-plane magnetization does not response to the perpendicular magnetic field, we measured the XMCD with the incidence of the x ray at 60 degrees from the surface normal direction [Fig. 2(c) and 2(d)]. Since the XMCD measures the magnetic component parallel to the x-ray beam direction, the off-normal x-ray direction allows us to pick up both the perpendicular and the in-plane magnetic components. Indeed we observed a nonzero Co magnetic XMCD signal for $M_{Co} \| y$ and $M_{Co} \| -y$ [Fig. 2(d)],

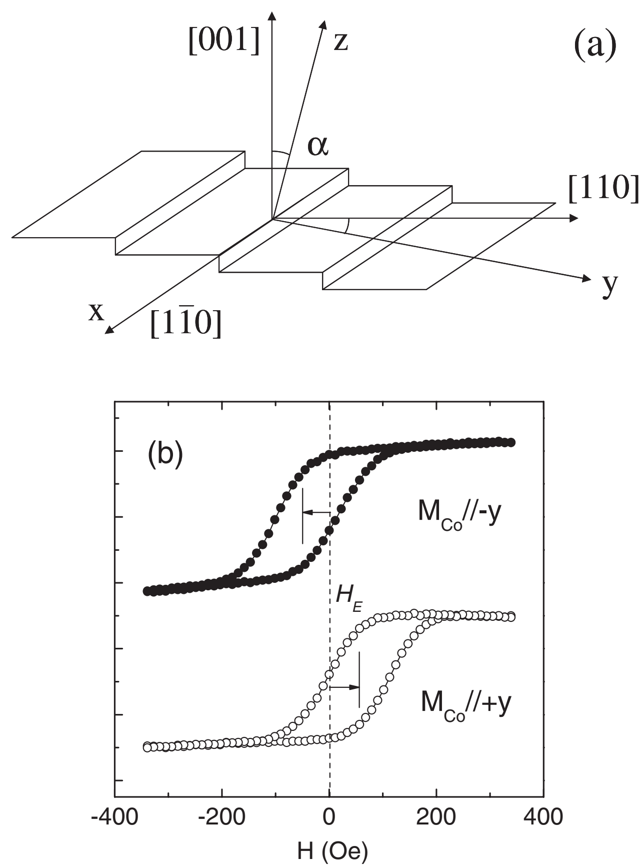

FIG. 1. (a) Schematic drawing of the vicinal surface. (b) The polar MOKE hysteresis loops of Ni(20 ML)/Fe(8 ML)/Co(8 ML)/Cu(1, 1, 40) for $M_{Co} \| -y$ and $M_{Co} \| +y$.

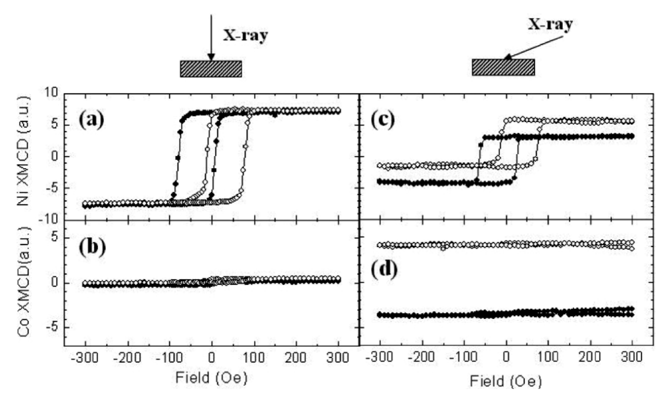

FIG. 2. Hysteresis loops of Ni [(a),(c)] and Co [(b),(d)] of Ni(20 ML)/Cu(5 ML)/Co(8 ML)/Cu(1, 1, 40) measured using XMCD with the applied magnetic field perpendicular to the sample surface. The incident x ray is at normal incidence for (a) and (b), and at 60° incident angle for (c) and (d). Open and solid circles correspond to $M_{Co} \| +y$ and $M_{Co} \| -y$, respectively.

but the Co XMCD signal remains a constant during the sweeping of the perpendicular magnetic field, proving that the Co magnetization does not respond to the perpendicular applied magnetic field. The Ni hysteresis loop again exhibits an exchange bias with its sign depending on the Co in-plane magnetization direction. Note the Ni polar hysteresis loops [Fig. 2(d)] exhibit not only the exchange bias but also different XMCD magnitude for $M_{Co} \| y$ and $M_{Co} \| -y$. The reason will be discussed later. For a 7 ML Cu sample which has an AFM Ni-Co interlayer coupling, the XMCD result confirms that the Ni exchange bias has an opposite sign to the 5 ML Cu sample, showing that the exchange bias comes from the Ni-Co interlayer coupling.

In conventional FM/AFM systems, exchange bias is realized by cooling the sample within a magnetic field to break the magnetic reversal symmetry. Since antiferromagnetism does not play a role in our samples and the Ni exchange bias depends on the Co magnetization direction along the y axis, the exchange bias in our system has to come from the magnetic reversal symmetry breaking due to the atomic steps of the vicinal surface. To understand this symmetry-breaking mechanism more clearly, consider the extreme case that the Ni perpendicular magnetization is parallel to its easy magnetization axis ([001] axis) and the in-plane Co magnetization is in the film plane (xy plane) due to the strong magnetic shape anisotropy. For a flat (001) surface, [001] is the film normal direction so that the Ni and Co magnetizations are exactly perpendicular to each other. Consequently, the magnetic free energy of the Ni-Co system should be invariant under the magnetic reversal of $M_{Ni} \rightarrow -M_{Ni}$ for either $M_{Co} \| \pm x$ or $M_{Co} \| \pm y$. Then there should be no exchange bias in the Ni magnetic hysteresis loop. For a vicinal (001) surface as shown in Fig. 1(a), however, the film normal direction is no longer in the [001] direction but making an angle α to the [001] crystalline axis in the yz plane. Thus the magnetic reversal of $M_{Ni} \rightarrow -M_{Ni}$ is no longer invariant for $M_{Co} \| \pm y$ even though it remains invariant for $M_{Co} \| \pm x$. To be more specific, the Ni-Co interlayer coupling of $-J_{int} M_{Ni} M_{Co}$ should experience a change from $+J_{int} \sin\alpha$ to $-J_{int} \sin\alpha$ for $M_{Co} \| +y$ (or $-J_{int} \sin\alpha$ to $+J_{int} \sin\alpha$ for $M_{Co} \| -y$) under the magnetic reversal of $M_{Ni} \rightarrow -M_{Ni}$. Since the interlayer coupling of $-J_{int} M_{Ni} M_{Co}$ is equivalent to a magnetic field of $J_{int} M_{Co}$ applied to the Ni magnetization [29], the interlayer coupling for $M_{Co} \| \pm y$ is equivalent to creating a shift (or an exchange bias) of $H_{ex} = \pm J_{int} \sin\alpha$ to the Ni polar hysteresis loop. Taking the typical interlayer coupling value of $\sim$1 meV for Cu spacer layer [30] and the fact that the coupling strength decays with square of the interlayer thickness [31], we estimate the exchange bias to be

$$H_E \sim \frac{1 \text{ meV}}{(5 \text{ ML})^2 \mu_{Ni} d_{Ni}(\text{ML})} \sin(2°) \sim 20 \text{ Oe}$$

which has the same order of magnitude as observed in our experiment. Nevertheless, the exchange bias should be proportional to the Ni-Co interlayer coupling. Experimentally, we measured the exchange bias as a function of the space layer thickness for both fcc Cu and Fe. As shown in Fig. 3, the exchange bias exhibits oscillations with the spacer layer thickness in exactly the same manner as the oscillatory magnetic interlayer coupling, supporting the fact that the observed exchange bias is due to the Ni-Co interlayer coupling.

From the above discussion, the Ni magnetization should have a small in-plane magnetic component that is coupled (or locked) to the Co in-plane magnetization during the polar measurement to generate the exchange bias. Note that the XMCD measures the projection of the total magnetization along the x-ray incident direction, the existence of the in-plane component of the Ni magnetization then implies that the Ni XMCD signal for the off-normal x-ray incidence should have a different saturation magnitude for a positive and negative applied magnetic field. This effect was indeed observed in our experiment that the Ni polar hysteresis loop has different saturation XMCD values for positive and negative magnetic fields and that this effect reverses its sign when $M_{Co} \| y$ changes to $M_{Co} \| -y$ [Fig. 2(c)]. From the magnitude difference, we estimate $(M_{Ni})_{\|}/(M_{Ni})_{\perp} \approx 0.2$ which corresponds to a Ni magnetization tilling away from the surface normal direction by $\sim 11°$ towards the Co in-plane magnetization direction. Since [001] is already away from the surface normal direction by 2°, the Ni magnetization is actually tilling away from the [001] direction by $\sim 9°$.

Finally, we tested the stability of the exchange bias in our system. In a conventional AFM/FM system, the exchange bias can be changed by cycling the FM magnetization due to the rearrangement of AFM spins. In our system, the Ni exchange bias is a result of its coupling to the in-plane Co magnetization; thus, it should be retained as long as the Co magnetization holds its direction. Figure 4 shows the stability result from a MOKE measurement on Ni(20 ML)/Fe(8 ML)/Co(8 ML)/Cu(1, 1, 40). A

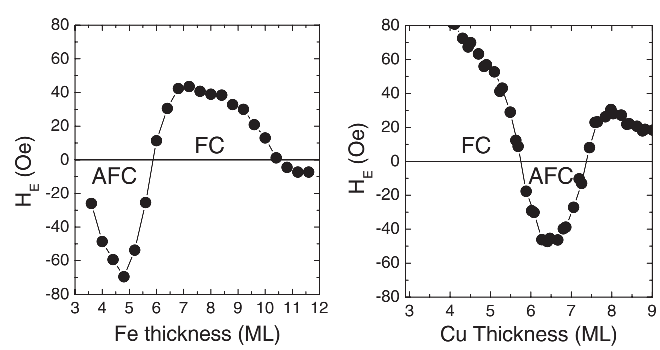

FIG. 3. The exchange bias field measured from the polar MOKE loops as a function of the Fe and Cu spacer layer thickness. FC and AFC denote ferromagnetic and antiferromagnetic interlayer coupling at the corresponding spacer layer thickness.

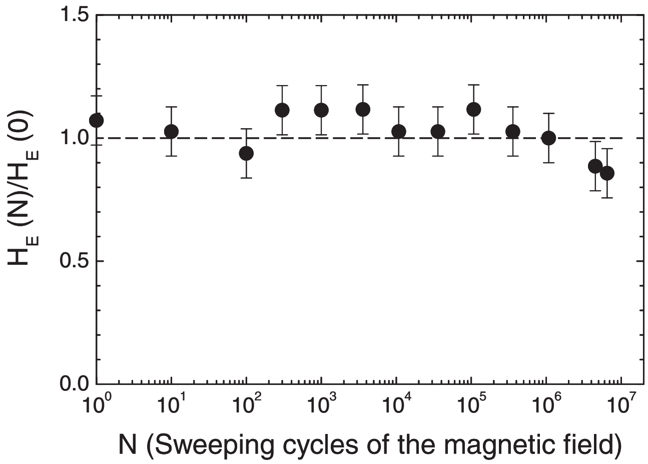

FIG. 4. Exchange bias of Ni(20 ML)/Fe(8 ML)/Co(8 ML)/Cu(1, 1, 40) as a function of the sweeping cycles of the applied perpendicular magnetic field.

perpendicular sweeping magnetic field with an amplitude of 500 Oe was generated by an ac power supply, and the polar MOKE measurement was performed after certain cycles of the magnetic field. The result shows that the exchange bias is very stable up to at least 6×10^6 cycles of the applied magnetic field, which is the maximum number of cycles we applied. In our system, the new concept of creating the exchange bias is (i) we use atomic steps to break the left-right magnetic symmetry of the Co layer to freeze its perpendicular spin component, and (ii) we use indirect interlayer magnetic coupling to create the Ni exchange bias. In this way, no domain wall is involved in creating the exchange bias so that there is no significant training effect as encountered in the spring magnets [14]. In addition, both the direction and the magnitude of the exchange bias in our system can be easily controlled by the in-plane Co magnetization direction and the spacer layer thickness. Therefore the exchange bias mechanism in our system is conceptually different from previously studied systems and thus will open a new direction in the field of exchange bias research. In addition, the easy tunability of the exchange bias in our system is expected to bring more flexibility in future magnetic devices than the traditional FM/AFM system.

In summary, exchange bias is observed in ferromagnetic sandwiches of Ni-Cu-Co and Ni-Fe-Co grown on vicinal Cu(001) surface. For the in-plane Co magnetization perpendicular to the atomic steps, the Ni magnetic hysteresis loop exhibits an exchange bias whose direction and strength can be tuned systematically by the in-plane Co magnetization direction and interlayer magnetic coupling. From the mechanism of the exchange bias in our system, it is safe to predict that our result is a general result for any two coupled FM systems in which the two FM layer magnetizations are almost orthogonal to each other.

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