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Rotatable magnetic anisotropy of CoO/Fe/Ag(001) in ultrathin regime of the CoO layer

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Abstract

CoO/Fe thin films were grown epitaxially onto vicinal Ag(001) and investigated using Magneto-Optic Kerr Effect, X-ray Magnetic Circular Dichroism (XMCD), and X-ray Magnetic Linear Dichroism (XMLD) techniques. We show that the CoO film in the ultrathin regime does not induce a uniaxial magnetic anisotropy but a coercivity enhancement. This result provides a mechanism for the microscopic origin of the rotatable magnetic anisotropy. XMLD measurement further reveals that the underlying mechanism is that the CoO spins are totally rotatable in the ultrathin regime to follow the Fe magnetization.

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Understanding the property of ferromagnetic(FM)/antiferromagnetic(AFM) system is one of the most active areas in nanomagnetism research because of its importance to spintronics technology [1]. As a FM/AFM system is cooled down within an external magnetic field to below the Néel temperature (T_N) of the AFM material, the AF will induce an exchange bias and a magnetic anisotropy in the FM layer [2]. While it is well accepted that the exchange bias and the induced magnetic anisotropy are both due to the AFM order of the AFM layer [3,4,5,6], it has been a mystery that the induced anisotropy in the FM layer occurs at a thinner AFM thickness than the exchange bias [1]. For thicker AFM layer, it is clear that the AFM spins after the field cooling induces both a unidirectional anisotropy (exchange bias) and a uniaxial anisotropy in the FM layer [1]. But in the ultrathin regime of the AFM layer, where the exchange bias vanishes, it is not clear whether the induced anisotropy has the same origin or not. Theoretical models have been proposed to attribute the coercivity enhancement into two different origins: one from the induced uniaxial anisotropy and the other from the so-called rotatable anisotropy [7,8]. It was found in experiment that while the uniaxial anisotropy dominates the system in thicker AFM regime as expected, the rotatable anisotropy dominates the system in thinner AFM regime [9,10]. The idea of rotatable anisotropy has been further tested indirectly by studies of the time-dependent dynamics [11,12], temperature dependence of magnetization reversal [13], and the magnetic training effect [14], etc. However, a microscopic origin of the rotatable anisotropy has never been clearly identified. We studied epitaxially grown CoO/Fe/vicinal Ag(001) system in the ultrathin CoO regime where there is no exchange bias but a coercivity enhancement in the Fe film. By studying the effect of the CoO AFM order to the uniaxial anisotropy and the coercivity of the Fe film, we show unambiguously that the CoO AFM order in this regime does not induce a uniaxial anisotropy but only a 4-fold anisotropy to the Fe film. The underlying mechanism is that the CoO spins in this regime are totally rotatable to follow the Fe magnetization switching.

A Ag(001) single crystal substrate was prepared with half of it being flat (001) surface and the other half being a 10° vicinal surface (steps parallel to [110] axis). The substrate is cleaned in an ultra-high vacuum system by cycles of Ar ion sputtering at $\sim 2\text{keV}$ and annealing at 700°C . A 2nm-thick Fe film was grown on top of the Ag(001) substrate, followed by a CoO wedge (0-2nm) growth on top of the Fe film by a reactive deposition of Co under an oxygen pressure of 1×10^{-6} Torr. Low Energy Electron Diffraction (LEED) spots confirm the formation of epitaxial single crystalline CoO film. The crystalline

relationship is that the Ag [110], Fe [100], and CoO [110] axes are parallel to each other which agrees with the literature result [15]. The sample is covered by a 2nm Ag protection layer and then measured by Magneto-Optic Kerr Effect (MOKE), X-ray Magnetic Circular Dichroism (XMCD), and X-ray Magnetic Linear Dichroism (XMLD) at beamlines 4.0.2 of the Advanced Light Source (ALS) of the Lawrence Berkeley National Laboratory.

We first present Fe hysteresis loops taken by MOKE at room temperature which is just below the CoO Néel temperature. The magnetic field is applied in the film plane and is along the Ag [-110] or Fe [100] direction on flat Ag. For CoO/Fe(2nm)/flat Ag(001) (vicinal angle $\alpha=0^\circ$), we observe square shape hysteresis loops with the Fe coercivity increasing with the CoO thickness above $\sim 0.5\text{nm}$ of CoO [Fig. 1(a)]. This is expected because the establishment of CoO AFM order above a critical thickness should enhance the Fe layer coercivity. For CoO/Fe(2nm)/vicinal Ag(001) ($\alpha=10^\circ$), the Fe hysteresis loop displays two split loops with zero magnetic remnance [Fig. 1(a)]. This type of hysteresis loop shows the existence of a step-induced uniaxial anisotropy with the easy magnetization axis parallel to the atomic steps, and with the uniaxial anisotropy strength proportional to the shift field (H_S) of the side loops [16]. As the CoO thickness increases, the Fe coercivity of the CoO/Fe(20Å)/vicinal Ag(001) increases, showing that the CoO AFM order also enhances the Fe coercivity on vicinal surface. But the interesting observation is that the shift field H_S , which is proportional to the step-induced uniaxial anisotropy, does not change as the CoO thickness increases [Fig. 1(b)]. Recall that the Fe film on vicinal Ag(001) should consists of a step-induced uniaxial anisotropy and a crystalline 4-fold anisotropy, the result of Fig. 1 suggests that the CoO AFM order does not induce a uniaxial anisotropy in the Fe film. This effect can be further verified by temperature dependence measurement on CoO(1nm)/Ag(0.5nm)/Fe(2nm)/vicinal Ag(001). The use of 0.5nm Ag is to reduce the CoO/Fe coupling strength so that the effect can be better viewed in a wide temperature range. As the temperature is lowered, the shift field H_S changes little but the coercivity H_C increases dramatically and eventually dominates the H_S to result in a virtual square shape hysteresis loop at low temperature (Fig. 2). This result shows again that the AFM order of the 1nm CoO film does not induce a uniaxial anisotropy to the Fe film. It should be mentioned that once the coercivity dominates the hysteresis loop (e.g., the loops below 150K in Fig. 2), magnetization switching is under a different process and it is unclear that if H_S is still proportional to the uniaxial anisotropy under this situation [17].

Nevertheless, the little change of H_S in the range of $150\text{K} < T < 300\text{K}$ supports the result of Fig. 1 that the CoO AFM order induces little change in the uniaxial anisotropy as compared to the coercivity change.

The next question is if the enhanced Fe coercivity is independent of the Fe uniaxial anisotropy. To answer this question, we measured the CoO(1nm)/Fe(2nm)/vicinal Ag(001) at $T=80\text{K}$ using XMCD at beamline 4 of ALS. Fe hysteresis loops are measured for field applied parallel and perpendicular to the atomic steps (Fig. 3). The Fe coercivity ($H_C=3200$ Oe) is much greater than the step-induced uniaxial anisotropy ($H_S=130$ Oe) after cooling the sample to $T=80\text{K}$, but has exactly the same value for field parallel and perpendicular to the steps. This result shows that the H_C enhancement due to the CoO AFM order at low temperature is independent of the uniaxial anisotropy. It should be mentioned that on flat Ag(001) surface the CoO could develop 2 equivalent in-plane domains along two orthogonal equivalent crystal axes so that local non-zero uniaxial anisotropy could be averaged into a 4-fold anisotropy at macroscopic length scale. On vicinal Ag surface, however, the atomic steps align the Fe magnetization parallel to the steps. Then the interfacial Fe-CoO interaction makes the sample cooling an equivalent field cooling with the “field” being the Fe magnetization (parallel to the steps) so that the CoO-induced uniaxial anisotropy (if it existed) should favor the Fe magnetization parallel to the steps. Therefore the independence of the shift field H_S on CoO AFM order (Fig. 1 and 2) and the independence of the H_C on step orientation (Fig. 3) shows that the CoO AFM order in this regime does not induce a uniaxial anisotropy. By time reversal argument, a magnetic energy term should be expressed by even order terms of the spin components. This leads to the general expression for the uniaxial and 4-fold magnetic anisotropies. As a special case of the magnetic energy, the magnetic energy term in the FM layer due to the AFM/FM interfacial interaction has to take an equivalent form of the uniaxial anisotropy, 4-fold anisotropy, and higher order anisotropy terms (unidirectional or exchange bias is an exception due to time reversal symmetry breaking at low magnetic field). The symmetry in our CoO/Fe/vicinal Ag(001) system requires only a uniaxial anisotropy and a 4-fold anisotropy. Then although the coercivity of a FM layer depends on many factors such as the domain wall pinning [18] and magnetization rotation [19], the facts that the CoO AFM order does not change the uniaxial magnetic anisotropy and that the enhanced H_C is independent of the uniaxial anisotropy lead to the plausible mechanism that the CoO AFM order induces only a 4-fold anisotropy to the CoO/Fe system in this work. However, we

notice that an induced cubic anisotropy is not the only possible mechanism for an enhanced coercivity (e.g., domain pinning), we cannot make a firm conclusion that the CoO AFM order induces a 4-fold anisotropy. This conclusion has to be made by a direct measurement of the magnetic anisotropy in the CoO/Fe/Ag(001) system (e.g., by ferromagnetic resonance or torque magnetometry measurement).

To understand the nature of the possible CoO-induced 4-fold anisotropy, we performed XMLD measurement at the CoO L_3 edge on CoO(1nm)/Fe(2nm)/vicinal Ag(001). X-ray Absorption Spectrum (XAS) was measured at normal x-ray incidence as a function of the x-ray linear polarization angle ϕ (relative to the vicinal step direction). The L_3 ratio (R_{L3}), which is defined as the ratio of the XAS intensity at 778.1 eV and at 778.9 eV (the lower energy intensity divided by the higher energy intensity), is used to quantify the XMLD effect [20]. After field cooling the sample to $T=80\text{K}$, R_{L3} - ϕ relation was measured for applied field parallel and perpendicular to the vicinal steps. First, we find that the measurement result is independent of the field cooling direction (Fig. 4). This result support that the CoO order does not induce a uniaxial magnetic anisotropy. Second, the R_{L3} - ϕ result roughly follows a $R_{L3}=A\cos^2\phi+B$ relation (solid lines in Fig. 4), showing that the Fe magnetization aligns the CoO spin axis. Noting that Fe [100] axis is parallel to CoO [110] axis, the L_3 ratio analysis [17,21,22] then leads to the conclusion that the in-plane CoO AFM spins are 90° -coupled to the Fe spins in the CoO(10Å)/Fe(20Å)/vicinal Ag(001) sample. Finally, after changing the Fe magnetization from parallel to perpendicular to the vicinal steps, the A coefficient of the $R_{L3}=A\cos^2\phi+B$ relation changes its sign (Fig. 4). The same magnitude of the A coefficient after the Fe magnetization switching shows that all CoO spins are also switched by 90 degrees to follow the Fe magnetization switching. This result suggests that the possible mechanism of the CoO-induced 4-fold anisotropy is due to the rotation of the the CoO spins with the Fe magnetization. Although our case is different from the polycrystalline case, the rotation nature of the CoO spins in ultrathin regime agrees with the rotatable anisotropy mechanism in polycrystalline where the rotatable anisotropy involves AF spin rearrangement during the FM magnetization switching.

In summary, using vicinal surface we are able to study the CoO effect on the uniaxial and 4-fold magnetic anisotropies of Fe film in CoO/Fe/vicinal Ag(001). In ultrathin regime of CoO film, we identified that the CoO AFM order does not induce a uniaxial anisotropy but a coercivity enhancement in the Fe film. XMLD measurement

shows that the CoO spins are 90° coupled to the Fe magnetization and switch together with the Fe magnetization. This rotatable nature of CoO spins could explain why CoO order does not induce a uniaxial anisotropy but only a 4-fold anisotropy in the Fe film.

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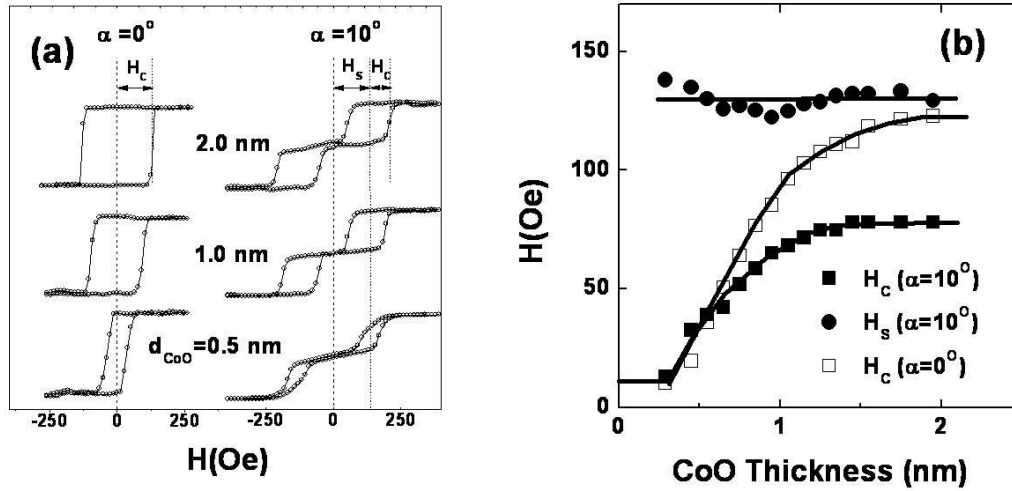


Fig. 1: (a) Hysteresis loops of CoO/Fe(2nm) grown on flat Ag(001) (left) and vicinal Ag(001) (right). α refers to vicinal angle. (b) The coercivity (H_c) and shift field (H_s) versus CoO thickness. While the coercivity increases with the CoO thickness, the shift field is independent of the CoO thickness, showing that the CoO antiferromagnetic order does not enhance the uniaxial magnetic anisotropy.

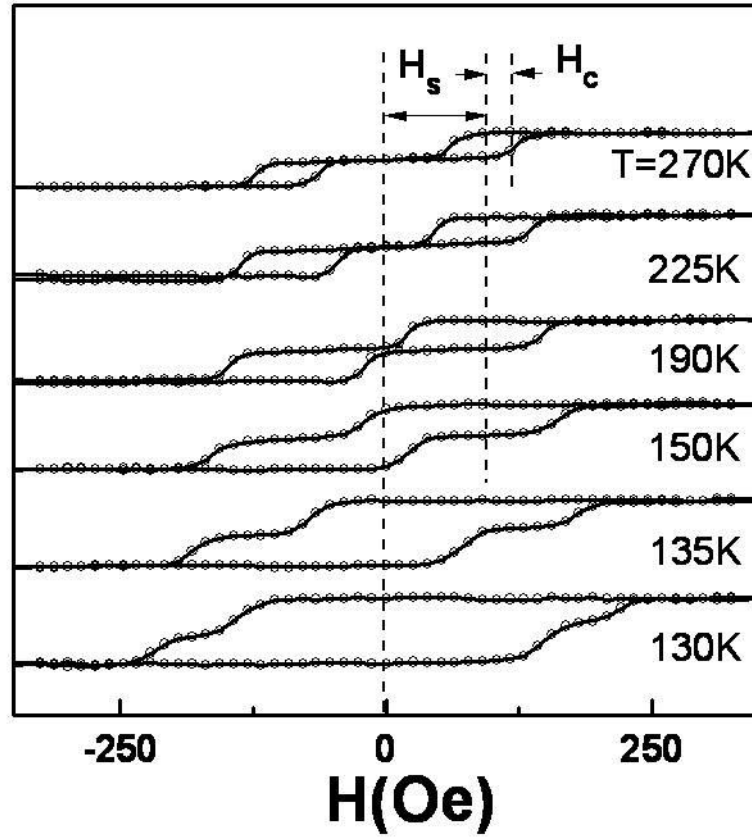


Fig. 2: Hysteresis loops of CoO(1nm)/Ag(0.5nm)/Fe(2nm)/vicinal Ag(001) ($\alpha=10^\circ$). While the coercivity increases with decreasing the temperature, the shift field changes little with the temperature, showing that the CoO antiferromagnetic order does not enhance the uniaxial magnetic anisotropy.

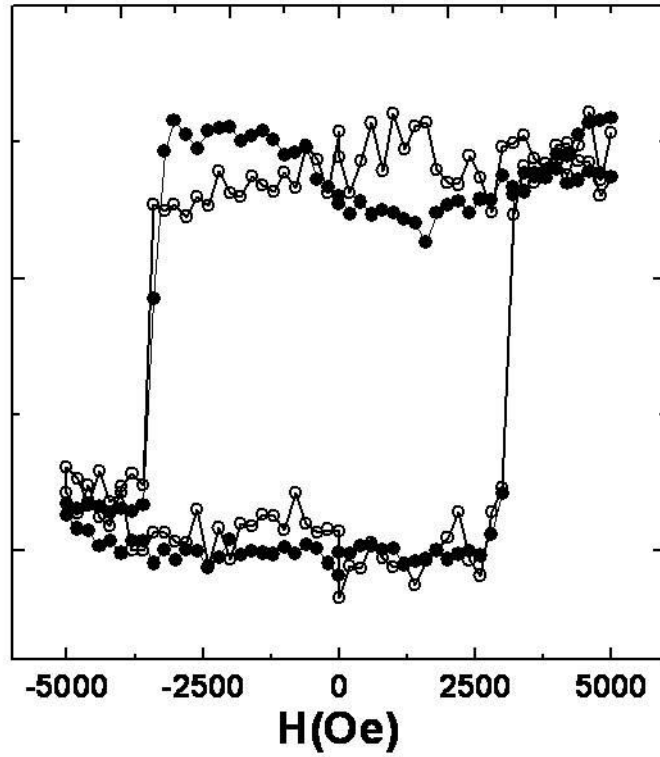


Fig. 3: Hysteresis loops of CoO(1nm)/Ag(0.5nm)/Fe(2nm)/vicinal Ag(001) at $T=80\text{K}$ for magnetic field applied parallel (open circles) and perpendicular (dots) to the atomic steps. The same coercivity in these two cases shows that the coercivity enhancement is independent of the uniaxial anisotropy.

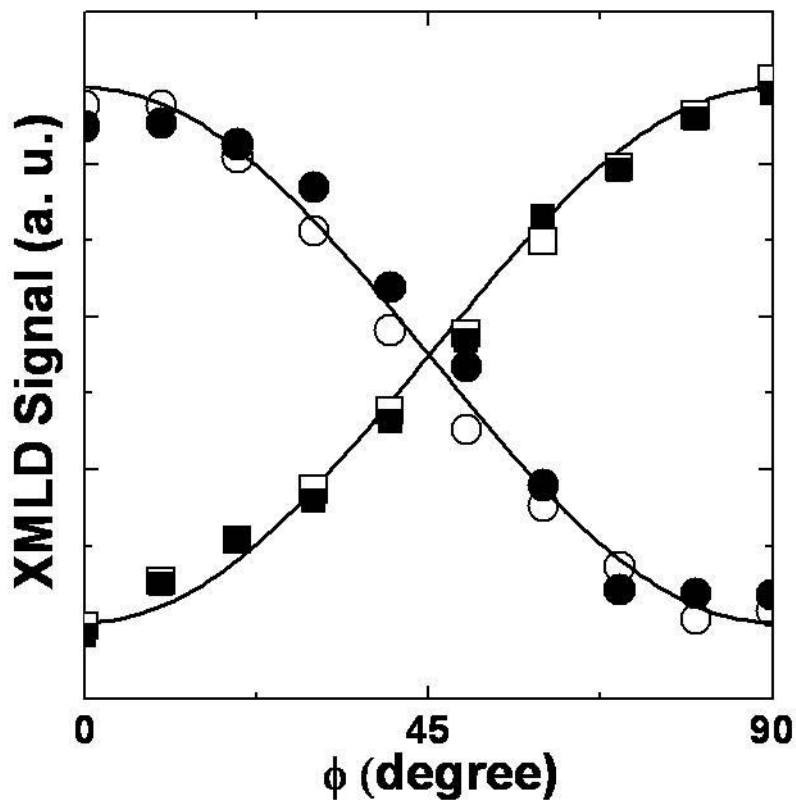


Fig. 4: XMLD of CoO(1nm)/Ag(0.5nm)/Fe(2nm)/vicinal Ag(001) for magnetic field applied parallel (circles) and perpendicular to the atomic steps (squares). Open and closed symbols are for field cooling direction parallel and perpendicular to the atomic steps. The result shows that the CoO is totally rotatable to follow the switching of the Fe magnetization.

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