Gigantic transverse x-ray magnetic circular dichroism in ultrathin Co in Au/Co/Au(001)

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Abstract. Transverse-geometry x-ray magnetic circular dichroism (TXMCD) measurements on Au/Co-staircase/Au(001) reveal the orbital origin of intrinsic in-plane magnetic anisotropy. A gigantic TXMCD was successfully observed at the Co $L_{3,2}$ edges for Co thickness ($t_{Co}$) in the 2-monolayer regime. A TXMCD-sum-rule analysis shows a remarkable enhancement of an orbital-moment anisotropy ($\Delta m_{orb}$) and of an in-plane magnetic dipole moment ($Tm_{\parallel}$). Both $\Delta m_{orb}$ and $Tm_{\parallel}$ exhibit close similarity in $t_{Co}$ dependence, reflecting the in-plane magnetic anisotropy. These observations evidence that extremely strong, intrinsic, in-plane magnetic anisotropy originates from the anisotropic orbital part of the wave function, dominating the dipole-dipole-interaction-derived, extrinsic, in-plane magnetic anisotropy.

1. Introduction
Magnetic anisotropy (MA) in two-dimensional systems has recently attracted much scientific and technological attention, because MA provides a physically intriguing issue of its intrinsic origin and high potential for spintronic applications. Most of previous studies have been focused on perpendicular magnetic anisotropy (PMA) in magnetic ultrathin films [1], superlattices [2], and nano-clusters [3-5]. This is mainly because PMA could have potential applications for high-density magnetic recording media. PMA is also physically of interest, since it is observed only in systems involving special magnetic interaction that overcomes dipole-dipole interaction which favors in-plane magnetic anisotropy (IMA).

IMA has attracted less interest than PMA, partly due to its little potential for spintronic applications and partly due to its extrinsic origin. Physically, IMA could originate also from the intrinsic, microscopic, magnetic states. But IMA is normally dominated by the extrinsic anisotropy arising from macroscopic dipole-interaction. The intrinsic IMA is thus difficult to separately observe. The intrinsic IMA may be scientifically of interest and of importance, since separately revealing intrinsic IMA provides much information about the microscopic, electronic and magnetic states of low-dimensional systems [6,7]. Observing the intrinsic IMA requires special experimental techniques that allow probing the masked, microscopic electronic states. X-ray magnetic circular dichroism in transverse geometry (TXMCD) has such an ability [8, 9]. Along with the T-geometry sum rules [8], it allows a determination of the orbital-moment anisotropy ($\Delta m_{orb} = m_{orb}^{\parallel} - m_{orb}^{\perp}$) and the anisotropic magnetic dipole moments ($m_{\parallel}^{\parallel}, m_{\perp}^{\parallel}$). Importantly, $\Delta m_{orb}$ is directly related with IMA and $m_{\perp}^{\parallel}$ should be closely connected with $\Delta m_{orb}$.

The origin of intrinsic IMA is not easy to approach by the usual longitudinal XMCD (LXMC) alone, because the spin magnetic moment ($m_{spin}$) dominates LXMC but $m_{spin}$ is essentially isotropic, being not responsible for MA. By contrast, in TXMCD the magnetic field ($B$) is applied to the sample perpendicularly to the photon helicity ($h$) and fully aligns $m_{spin}$ along the field, resulting in $m_{spin}$ perpendic-
ular to \( h \). The contribution of \( m_{\text{spin}} \) to TXMCD is thus effectively “killed.” In contrast, \( m_{\text{orb}} \) and \( m_T \) are not completely perpendicular to \( h \) even in T geometry. Thus, TXMCD allows a determination of the anisotropic \( m_{\text{orb}} \) and \( m_T \), giving information about the intrinsic IMA. Here we report on a TXMCD study of ultrathin Au/Co(\( t_{\text{Co}} \))/Au(001).

2. Experiment
An Au(2 nm)/Co-staircase (2 \( \leq t_{\text{Co}} \leq 9 \text{ ML})/ Au(001)(100 nm) sample was prepared by molecular-beam epitaxy with an MgO(001) substrate. A 100-nm-thick Au(001) seed layer was prepared on the 40-nm Cr buffer layer. The Co thickness (\( t_{\text{Co}} \)) ranged from 2 to 9 monoatomic layers (ML) with each step height of 1 ML. A layer-by-layer growth of Co during deposition was confirmed \textit{in-situ} by reflection high-energy electron diffraction. The Co layer was protected from oxidation with a 2-nm-thick Au capping layer. TXMCD measurements were made at the Co \( L_{3,2} \) edges at room temperature using circularly polarized synchrotron radiation on bending-magnet beam line BL-11A at the Photon Factory. The degree of circular polarization \( (P_C) \) was chosen to be \( P_C = +78\pm3\% \) [3]. Polarization-dependent x-ray absorption spectra (XAS) were taken using the total electron-yield method with \( h \) fixed and \( B \) reversed. The incident-light intensity was monitored with the photocurrent from an Au-coated mirror. The T geometry was set up by rotating the whole superconducting-magnet XMCD apparatus [10] around the vertical axis. The \( \pm5 \)-T field was applied to the sample perpendicular to \( h \) at a light-incident angle of \( \theta = 45^\circ \) with an accuracy of \( \pm0.3^\circ \).

3. Results and discussion
Figure 1 shows the flux-normalized, polarization-dependent Co \( L_{3,2} \)-edge XAS (\( \mu_+ \) and \( \mu_- \)) and TXMCD (\( \Delta \mu = \mu_+ - \mu_- \)) spectra for \( t_{\text{Co}} = 7 \text{ ML} \). Here \( \mu_+ \) and \( \mu_- \) represent the absorption coefficients with their sign defined by the direction of \( B \) with respect to the surface normal (\( n \)), as denoted by an inset of Fig.1. The spectra were taken at room temperature at an incidence angle of \( \theta = 45^\circ \) under a high field of \( B = \pm5 \text{ T} \). Correction for \( P_C \) was made in XAS and TXMCD. The energy integral of the TXMCD spectrum is shown by a red curve in Fig. 1(b). It is crucial to assure no artifact arising from mixing of a strong LXMCD into a weak TXMCD due to possible alignment errors. We measured conventional LXMCD at the Co \( L_{3,2} \) edges on a bcc Co 6-ML film with an MgO(001) substrate. A strong, normal-incidence LXMCD was observed at the Co \( L_{3,2} \) edges but no TXMCD signal was detected on the identical sample. This shows no artifact in the TXMCD.

The TXMCD spectrum is similar in line shape to the separately measured LXMCD. The TXMCD is naturally much weaker in amplitude than the LXMCD. The small TXMCD is reasonable, since \( m_{\text{spin}} \) does not contribute to TXMCD; \( m_{\text{spin}} \) is fully aligned along the high magnetic field (\( B \)) and is perpendicular to \( h \). The observed TXMCD is genuine and indicates the anisotropic orbital moment (\( \Delta m_{\text{orb}} = m_+ - m_- \)) and magnetic dipole moment (\( m_{\text{dip}} = -1/2 m_+ \)). This result leads us to interest in the \( t_{\text{Co}} \) dependence of TXMCD.

Figure 2(a) shows the Co \( L_{3,2} \)-edge TXMCD spectra as functions of \( t_{\text{Co}} \) ranging from 9 to 2 ML. It is evident that the TXMCD rapidly increases in amplitude with decreasing \( t_{\text{Co}} \). Surprisingly, the negative \( L_{3,2} \)-edge TXMCD amplitude amounts to \( \pm15\% \) at the thinnest limit of \( t_{\text{Co}} = 2 \text{ ML} \). This is even comparable...
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increases in amplitude with lowering rules [8, 11, 12]. The T sum rules [8], for the present definition of
Interestingly, the present Co/Au(001) is of strong IMA whereas the previous Co/Au(111) showed strong
of Au/Co/Au(001) at

and out-of-plane orbital (magnetic dipole) moments,
Here
denotes the energy integrals of the TXMCD over the
 comparatively, the TXMCD remarkably increases in amplitude with lowering \( t_{Co} \), showing enhanced \( \Delta m_{orb} \) and \( m^1 \).

To quantify magnetic anisotropy, we determine anisotropic magnetic moments using the TXMCD sum rules [8, 11, 12]. The T sum rules [8], for the present definition of \( \mu^1 \) and \( \mu^2 \), state for \( \theta = 45^\circ \),

\[
\Delta m_{an} = -8/3 \left[ \Delta A^1 + \Delta A^2 \right] \cdot n_h / \left[ A^1 + A^2 \right],
\]

\[
m^1 \perp = -4/21 \left[ \Delta A^1 - 2 \Delta A^2 \right] \cdot n_h / \left[ A^1 + A^2 \right].
\]

Here \( \Delta m_{an} \) is the orbital-moment anisotropy, \( m^1 \perp \) and \( m^1 \parallel \) \( (m^2 \perp \) and \( m^2 \parallel ) \) are the in-plane and out-of-plane orbital (magnetic dipole) moments, \( \Delta A^1 \) and \( \Delta A^2 \) \( (A^1 \) and \( A^2 ) \) are the energy integrals of \( \theta \)-directed \( L^3 \) - and \( L^2 \)-edge TXMCD (XAS), and \( n_h \) is the 3d hole number. There are additional relationships, \( m^1 \perp = -< L^3 \times h > / \hbar \) and \( m^1 \parallel = -< T^3 \times h > / \hbar \) with \( < T^3 > \) being the expectation value of the intra-atomic magnetic dipole operator, \( T = S - 3(r \cdot S)/r^2 \) [12, 13]. Using eqs. (1) and (2) and a theoretical 3d hole number of \( n_h = 2.49 \) for Co [14], we have determined \( \Delta m_{an} = m^1 \perp - m^1 \parallel \) and \( m^1 \).

Figure 3 shows the results for \( \Delta m_{an} \) and \( m^1 \) as functions of \( t_{Co} \). Both \( \Delta m_{an} \) and \( m^1 \) increase steeply with decreasing \( t_{Co} \). Most interestingly, \( \Delta m_{an} \) and \( m^1 \) exhibit almost the same \( t_{Co} \) dependence. Since \( \Delta m_{an} \) is directly related with the IMA energy and \( m^1 \) is closely connected with \( \Delta m_{an} \), our finding indicates that this system is of strong intrinsic IMA with the IMA energy increasing with decreasing \( t_{Co} \). The similar behavior of \( \Delta m_{an} \) and \( m^1 \) with \( t_{Co} \) is reasonable if one takes into account the theoretical prediction that non-zero values of those quantities are the manifestation of the anisotropic orbital part of the electron wave function.

We have obtained for \( t_{Co} = 2 \) ML the numerical results of \( \Delta m_{an} = +0.35 \mu_B \) and \( m^1 = +0.075 \mu_B \). It should be recalled that both \( \Delta m_{an} \) and \( m^1 \) are almost zero under high-symmetry crystal fields in bulk materials. The extremely large \( \Delta m_{an} \) shows an enhanced in-plane orbital moment, indicating strong IMA of Au/Co/Au(001) at \( t_{Co} = 2 \) ML. The present value of \( \Delta m_{an} = +0.35 \mu_B \) sharply contrasts in sign with and is very similar in amplitude to the reported values of \( \Delta m_{an} = -0.12 \mu_B \) for 4-ML Co in Au/Co ultrathin film/Au(111) [1] and \( \Delta m_{an} = -0.11 \mu_B \) for 2-ML-height Co cluster in Au/Co nano-clusters/Au(111) [3]. Interestingly, the present Co/Au(001) is of strong IMA whereas the previous Co/Au(111) showed strong PMA, indicating the strong effect of a substrate orientation on the thin-film magnetic anisotropy.

We estimate an intrinsic IMA energy \( \Delta E_{Co} \) caused by spin-orbit (SO) interaction. We take an approximation incorporating an effective renormalization factor \( \alpha \) [14].
where $\xi_{3d}$ (70 meV) is the 3d SO interaction constant in Co. Using a TXMCD-derived value of $\Delta m_{\text{orb}} = +0.35 \mu_B$ and a reported value of $\alpha \approx 0.2$ for Co [1,14], we obtain an extremely large value of $\Delta E_{\text{SO}} \approx -1.2 \times 10^4$ eV. This is a very large, intrinsic, electronic-state-derived IMA energy. In contrast, a dipole-dipole interaction energy, which favors extrinsic IMA, is estimated to be $-2\pi M_s^2 / 0.93 \approx 0.86 \times 10^4$ eV/Co with a 2-ML-thickness correction in the Co film. We have $|\Delta E_{\text{orb}}| > |2\pi M_s^2 / 0.93|$, which shows that the electronic-state-derived, intrinsic IMA is much stronger than the extrinsic IMA originating from dipole-dipole interaction.

We consider the physical implication of the enhanced $m_{\|}^{\text{orb}}$ with the $x$ axis taken to be in the layer plane. It holds that $m_{\|}^{\text{orb}} (= m_{\|}^{\text{orb}}_i) = -<T_I^x> \mu_B / h$ and $<T_I^x> (=<T_I^y>) = 7 \sum \alpha^i <S^\alpha> \cdot <Q_{\alpha}^i> \approx 7 <S> \cdot <Q_{\alpha}^i>$ [13]. Here $S$ is the spin quantum number of the total system, $i$ denotes the symmetry of the $d$ orbitals ($i = x^2-y^2, 3z^2-r^2, xy, yz, zx$), $<Q_{\alpha}^i> = \sum \alpha <\alpha|Q_{\alpha}^i>| <\alpha|$ represents the $<Q_{\alpha}^i>$ averaged over the occupied five $d$ orbitals with $\alpha$ denoting the occupation probability of orbital $i$. The extremely large $m_{\|}^{\text{orb}}$ indicates a net appearance of $<Q_{\alpha}^i>$ of the total Co 3d states. Using the values of the LXMCD-derived $S = 0.855h (m_{\text{QMS}} = 1.71 \mu_B)$ and the TXMCD-derived $m_{\|}^{\text{orb}} = +0.075 \mu_B$, we obtain a 3d-state-averaged value of $<Q_{\alpha}^i> = 0.0125$. The present non-zero $<Q_{\alpha}^i>$ implies that the four-lobe nature of the out-of-plane orbitals partially manifests itself when observed from the in-plane $x$ axis because of the relative high occupation of the out-of-plane orbitals.

4. Conclusion
We have successfully observed a giant TXMCD at the Co $L_{3,2}$ edges in thin Co layers in Au/Co staircase/Au(001). TXMCD sum-rule analyses have given a remarkably enhanced orbital-moment anisotropy and an enhanced in-plane magnetic dipole moment. Those anisotropic moments were shown to originate from the non-equally populated, anisotropic 3d states and to produce strong intrinsic IMA.

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