281. WE-Heraeus Seminar
"Spin-Orbit Interaction and Local Structure
in Magnetic Systems with Reduced Dimensions"
June 12-15, 2002, Wandlitz, Germany

X-ray magnetic circular dichroism study on spin reorientation transitions of magnetic thin films induced by surface chemisorption

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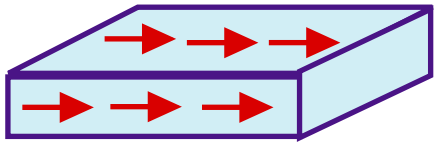
All the works presented today were done in Ohta Lab, my previous affiliation.

Magnetic anisotropy of ultrathin films

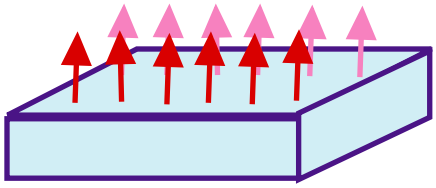
Perpendicular magnetic anisotropy (PMA)



easy axis surface parallel

shape anisotropy $-2\pi M_S^2$

magnetic dipole-dipole interaction



easy axis surface normal

some other origins

PMA new recording media with higher density

Phenomelogical description of anisotropic energy

$$\Delta E = -2\pi M_s^2 + K_{2v} + (K_{2s} + K_{2i}) / d$$

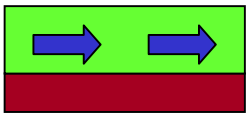
Shape anisotropy

Bulk magnetoelastic anisotropy

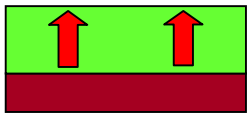
Surface / Interface magnetocrystalline anisotropy

thickness

When $\Delta E < 0$



When $\Delta E > 0$



Co/Pd(111), Co/Pt(111), Co/Au(111) *etc.*

$K_{2v} < 0$, $K_{2s} < 0$, $K_{2i} > 0$ Interface anisotropy stabilizes PMA

Ni/Cu(001)

$K_{2v} > 0$, $K_{2s} < 0$, $K_{2i} < 0$ Bulk magnetoelastic anisotropy stabilizes PMA

Note: Surface anisotropy contributes to the stabilization of the in-plane magnetization in both cases.

Microscopic origin of magnetic anisotropy

Spin-orbit interaction anisotropic

cf. exchange (spin-spin) interaction not anisotropic

$\mathbf{S}_i \cdot \mathbf{S}_j$ scalar product

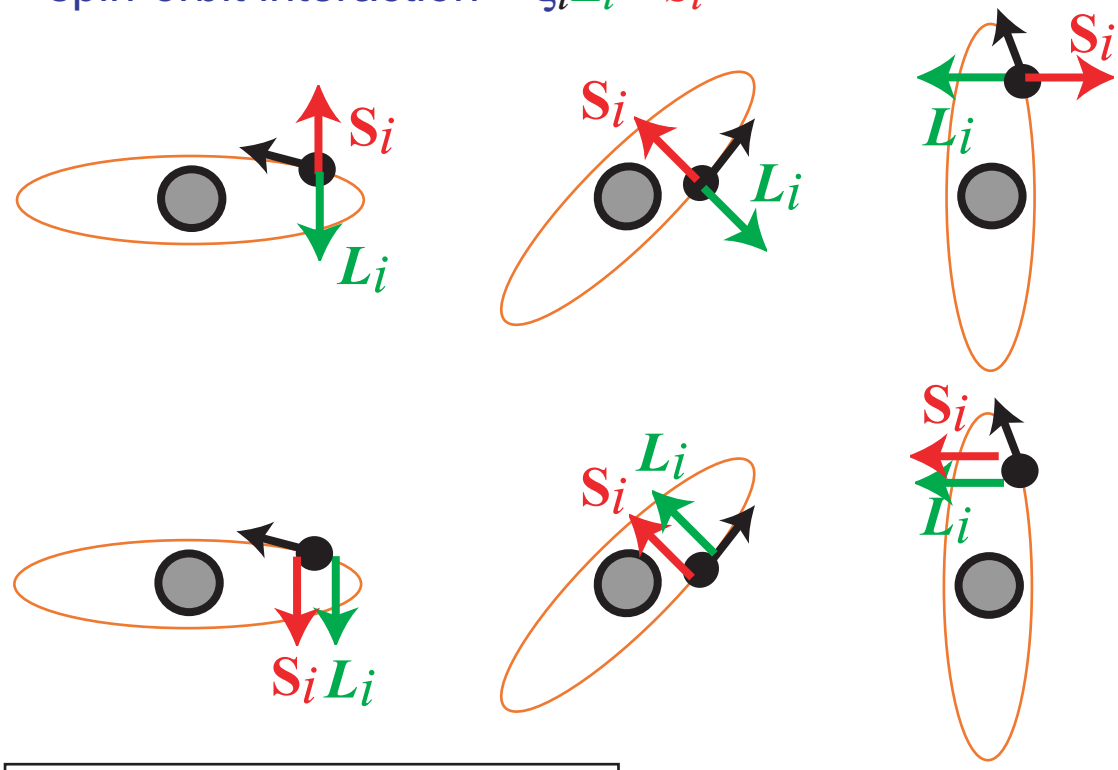
same energies without spin-orbit interaction

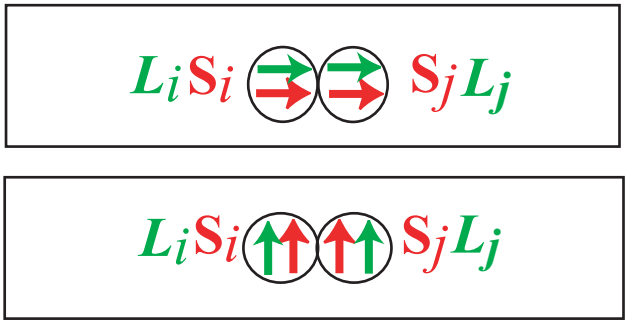
Orbital angular momentum $\boldsymbol{L}_i$

anisotropic in ultrathin films

or more generally, except for spherical symmetry

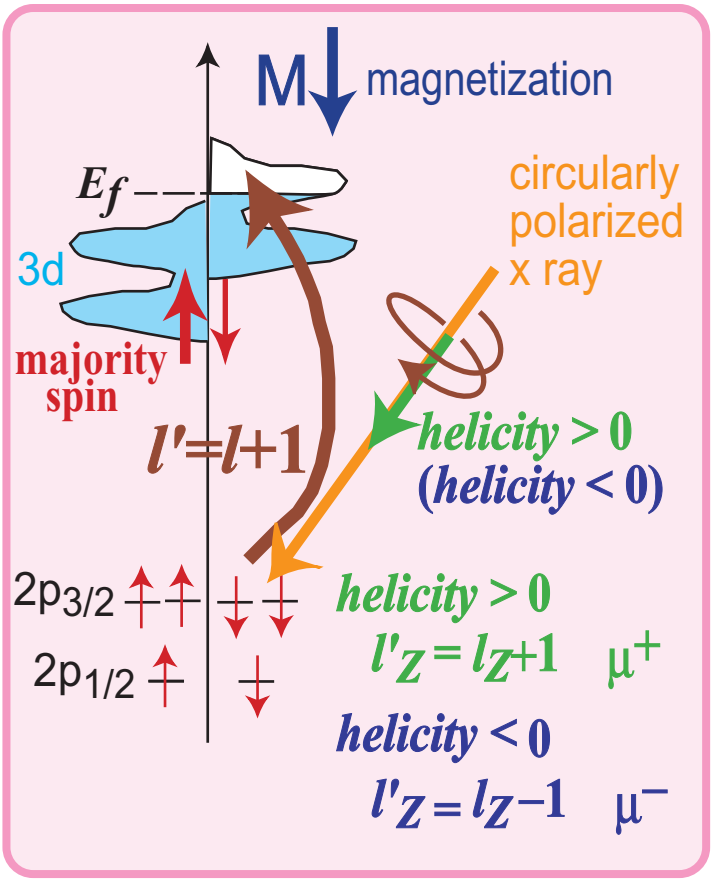
spin-orbit interaction $\xi_i \mathbf{L}_i \cdot \mathbf{S}_i$

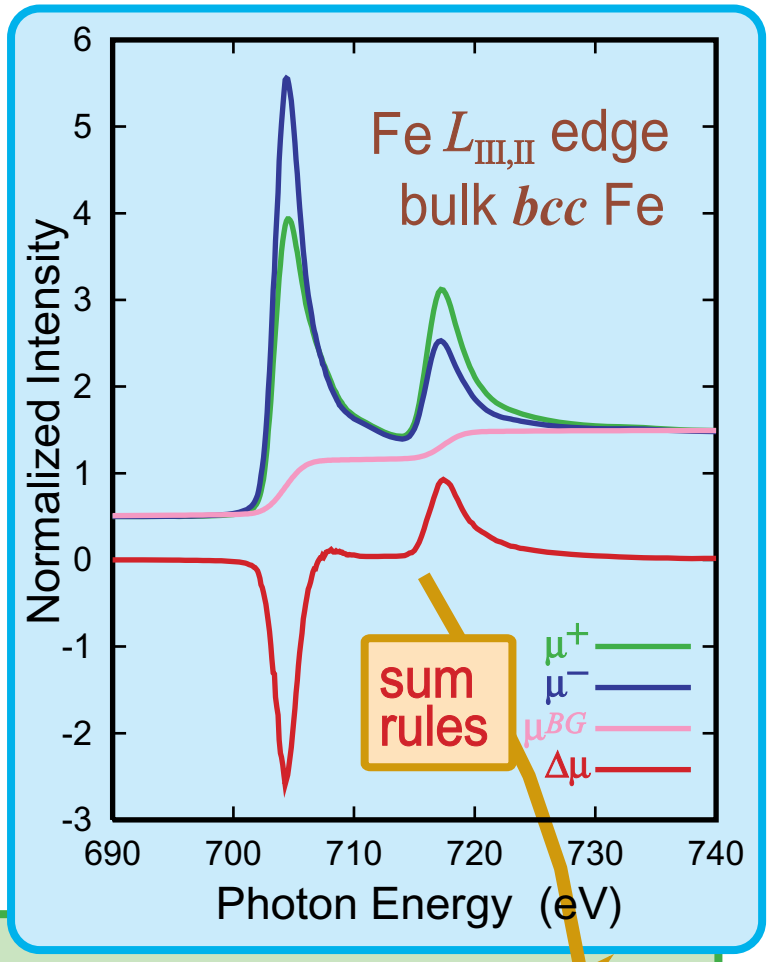




different energies with spin-orbit interaction

X-ray Magnetic Circular Dichroism





Fe $m_l = 0.071$
$m_s = 2.36$

$L_{III,II}$-edge sum rules

Orbital $$m_l = -\frac{4}{3}\frac{n_h \mu_B}{P_c \cos\theta}\frac{\int_{L_{III}+L_{II}} \Delta\mu \, dE}{\int_{L_{III}+L_{II}} (\mu^+ + \mu^- - 2\mu^{BG}) dE}$$

Spin $$m_s = -\frac{2 n_h \mu_B}{P_c \cos\theta}\frac{\int_{L_{III}} \Delta\mu \, dE - 2\int_{L_{II}} \Delta\mu \, dE}{\int_{L_{III}+L_{II}} (\mu^+ + \mu^- - 2\mu^{BG}) dE} + 7 \langle T_z \rangle \mu_B$$

n_h hole number

$\langle T_z \rangle$ magnetic dipole moment

μ_B Bohr magneton

P_c circular polarization factor

θ angle between k and M

μ^+ ↑↑ spectrum

μ^- ↑↓ spectrum

μ^{BG} continuum step

$$\Delta\mu = \frac{\mu^+ - \mu^-}{P_c \cos\theta}$$

Element-specific magnetization

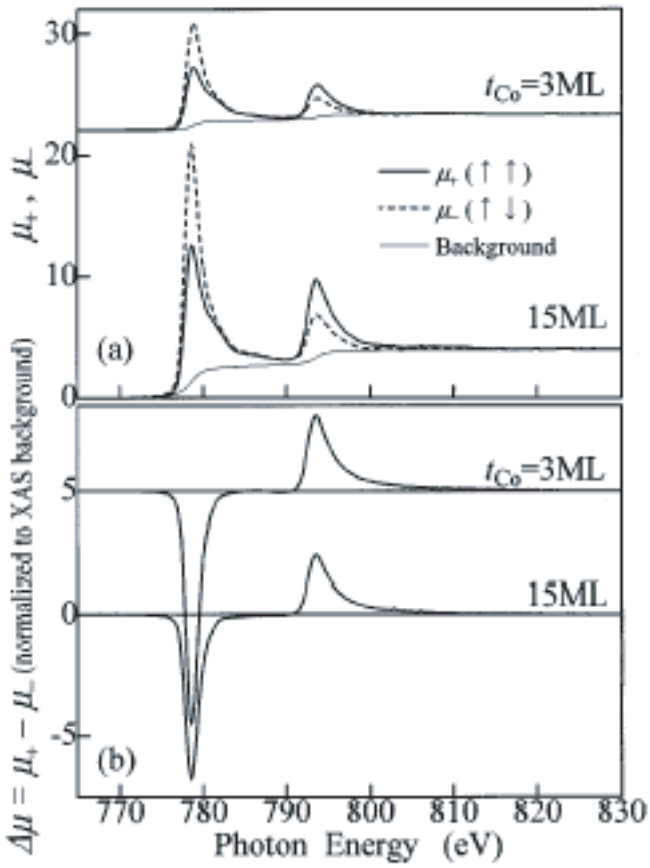
cf. *K* edge

Orbital $$m_l = -\frac{2}{3}\frac{n_h \mu_B}{P_c \cos\theta}\frac{\int_K \Delta\mu \, dE}{\int_K (\mu^+ + \mu^- - 2\mu^{BG}) dE}$$

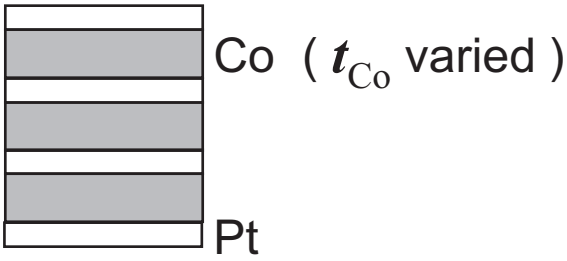
XMCD observation of enhanced orbital moment

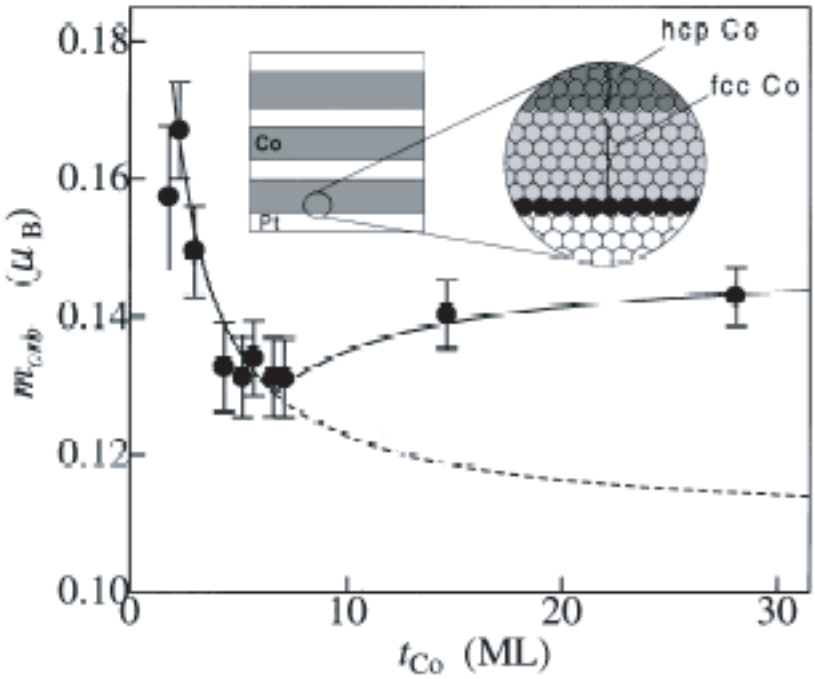
Co/Pt sandwiched films (PMA)

N. Nakajima, T. Koide, T. Shidara, H. Miyauchi, H. Fukutani, A. Fujimori, K. Iio, T. Katayama, M. Nyvlt and Y. Suzuki, *Phys. Rev. Lett.* **81** (1998) 5229.



Normal incidence Co $L_{3,2}$-edge XMCD in H = 2 T





Perpendicular Orbital moments as a function of Co thickness

Orbital moment is enhanced as thickness decreases

Interface anisotropy favors PMA

Effect of adsorption on magnetic anisotropy

Spin reorientation transition (SRT) induced by adsorption

Stabilization of PMA

H/Ni/Cu(001) R. Vollmer, Th. Gutjahr-Loeser, J. Kirschner, S. van Dijken and B. Poelsema, *Phys. Rev.* **B60**, 6277 (1999).

CO/Ni/Cu(001) S. van Dijken, R. Vollmer, B. Poelsema and J. Kirschner, *J. Magn. Magn. Mater.* **210**, 316 (2000).

CO, H, O/Fe/W(110) H. J. Elmer, J. Hauschild and U. Gradmann, *J. Magn. Magn. Mater.* **198-199**, 222 (1999).

O/Fe/Ag(001) J. Chen and J. L. Erskine, *Phys. Rev. Lett.* **68**, 1212 (1992).

CO/Co/Pt(111) O. Robach, C. Quiros, P. Steadman, K. F. Peters, E. Lundgren, J. Alvarez, H. Isern and S. Ferrer, *Phys. Rev.* **B65**, 054423 (2002).

Phenomelogical understanding

Gas adsorption → Decrease in surface anisotropy $|K_{2s}|$, which favors in-plane magnetization

→ Stabilization of PMA

Other examples of SRT induced by adsorption

Structure change of the metal films

H/Fe/Cu(001) R. Vollmer and J. Kirschner, *Phys. Rev.* **B61**, 4146 (2000).

PMA stabilized by H adsorption

SRT within the surface plane

CO/Co/Cu(110)

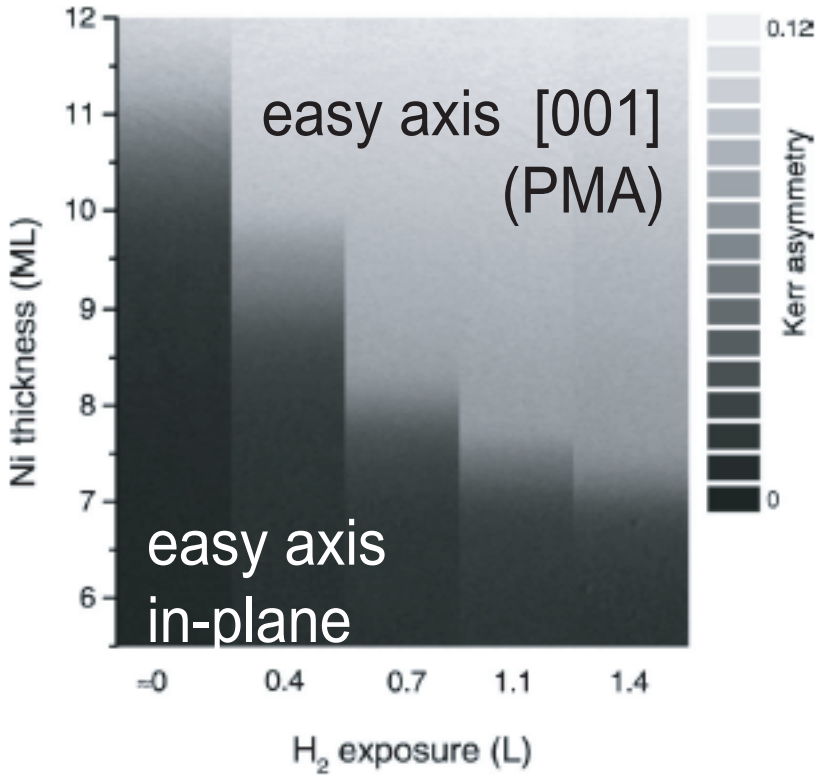
S. Hope, E. Gu, B. Choi and J. A. C. Bland, *Phys. Rev. Lett.* **80**, 1750 (1998).

S. Hope, E. Gu, B. Choi and J. A. C. Bland, *Phys. Rev.* **B57**, 7454 (1998).

Effect of adsorption on magnetic anisotropy

H adsorption on Ni/Cu(001)

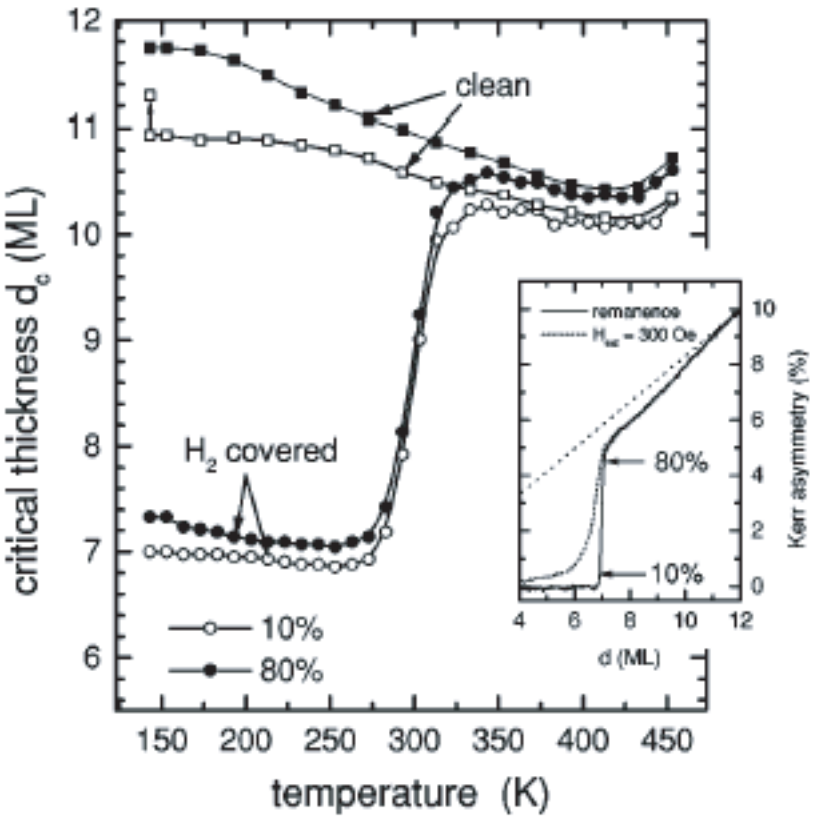
R. Vollmer, Th. Gutjahr-Loeser, J. Kirschner, S. van Dijken, and B. Poelsema, *Phys. Rev.* B60 (1999) 6277.



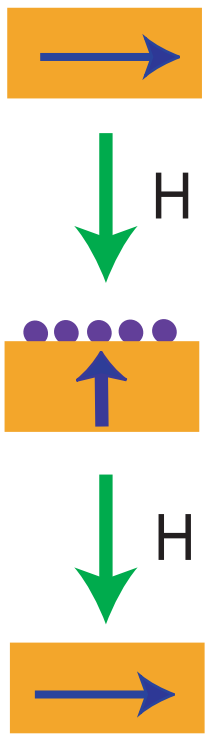
d_c = 10 ML

H ads.

d_c = 7 ML



7-10 ML Ni



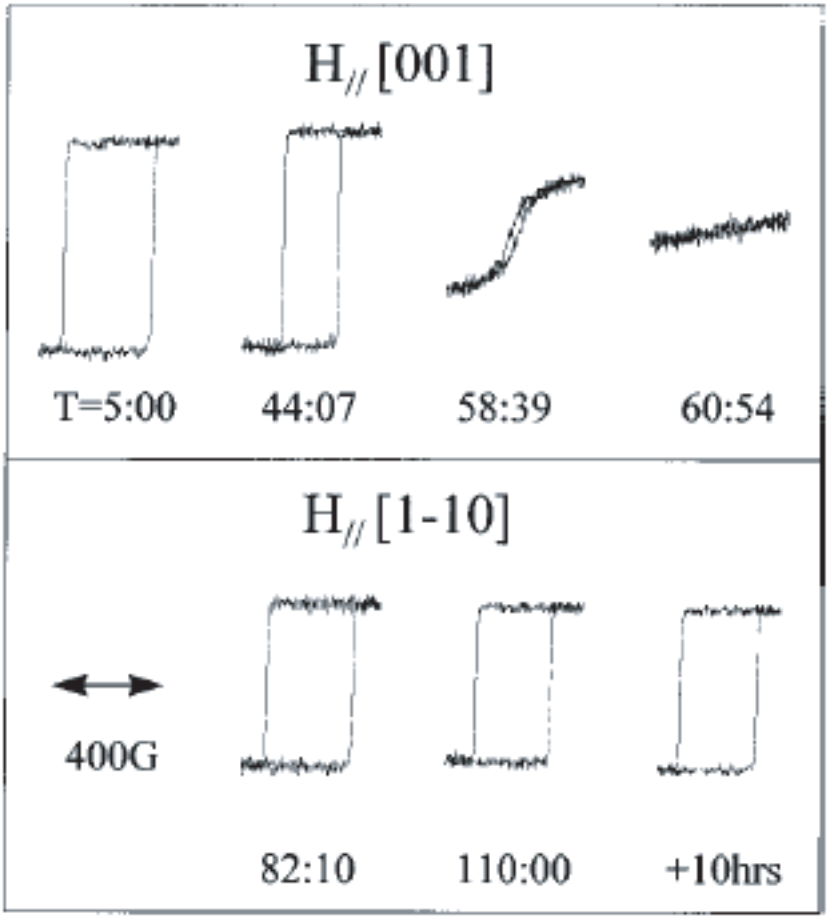
H ads.

H des.

Effect of adsorption on magnetic anisotropy

CO adsorption on Co/Cu(110)

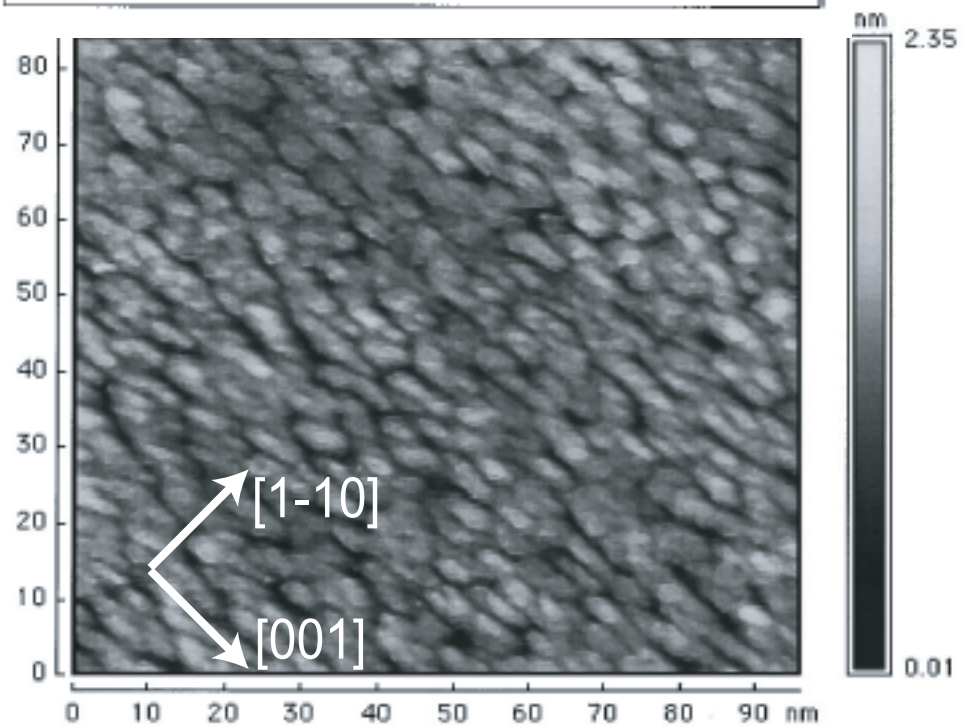
S. Hope, E. Gu, M. Tselepi, M. E. Buckley, and J. A. C. Bland, *Phys. Rev.* B57 (1998) 7454.

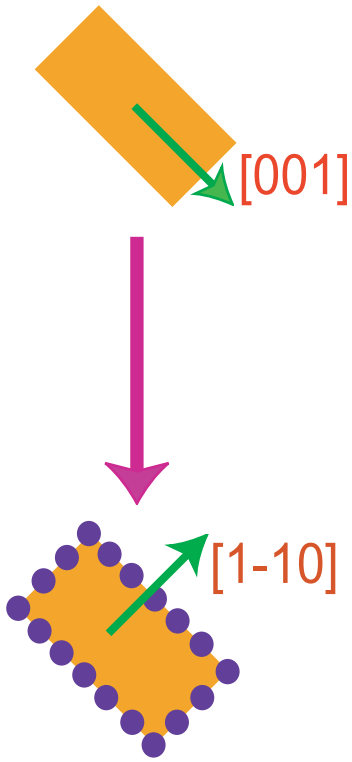


easy axis [001]

time
(CO ads.)

easy axis [1-10]





Scope of today's talk

Origins of the spin reorientation transition (SRT) induced by adsorption

Relation of SRT to the changes of the orbital magnetic moments upon adsorption

1) CO adsorption on Co/Pd(111)

D. Matsumura, T. Yokoyama, K. Amemiya, S. Kitagawa and T. Ohta, *Phys. Rev.* **B66**, in press (Jul. 1, 2002).

Observation of SRT upon CO adsorption

Discussion on the microscopic origin of the SRT

2) NO, H, O and CO adsorption on Co/Pd(111)

Dependence of the adsorbates

Which molecules induce SRT ?

3) CO and H adsorption on Ni/Cu(001)

D. Matsumura, T. Yokoyama, K. Amemiya, S. Kitagawa and T. Ohta, *Phys. Rev.* **B66**, in press (Jul. 1, 2002).

Discussion on the microscopic origin of the SRT

Difference between CO and H

4) Absorbate K-edge XMCD of CO and NO on Co and Ni films

K. Amemiya *et al.*, *Jpn. J. Appl. Phys.* **39**, L63 (2000).

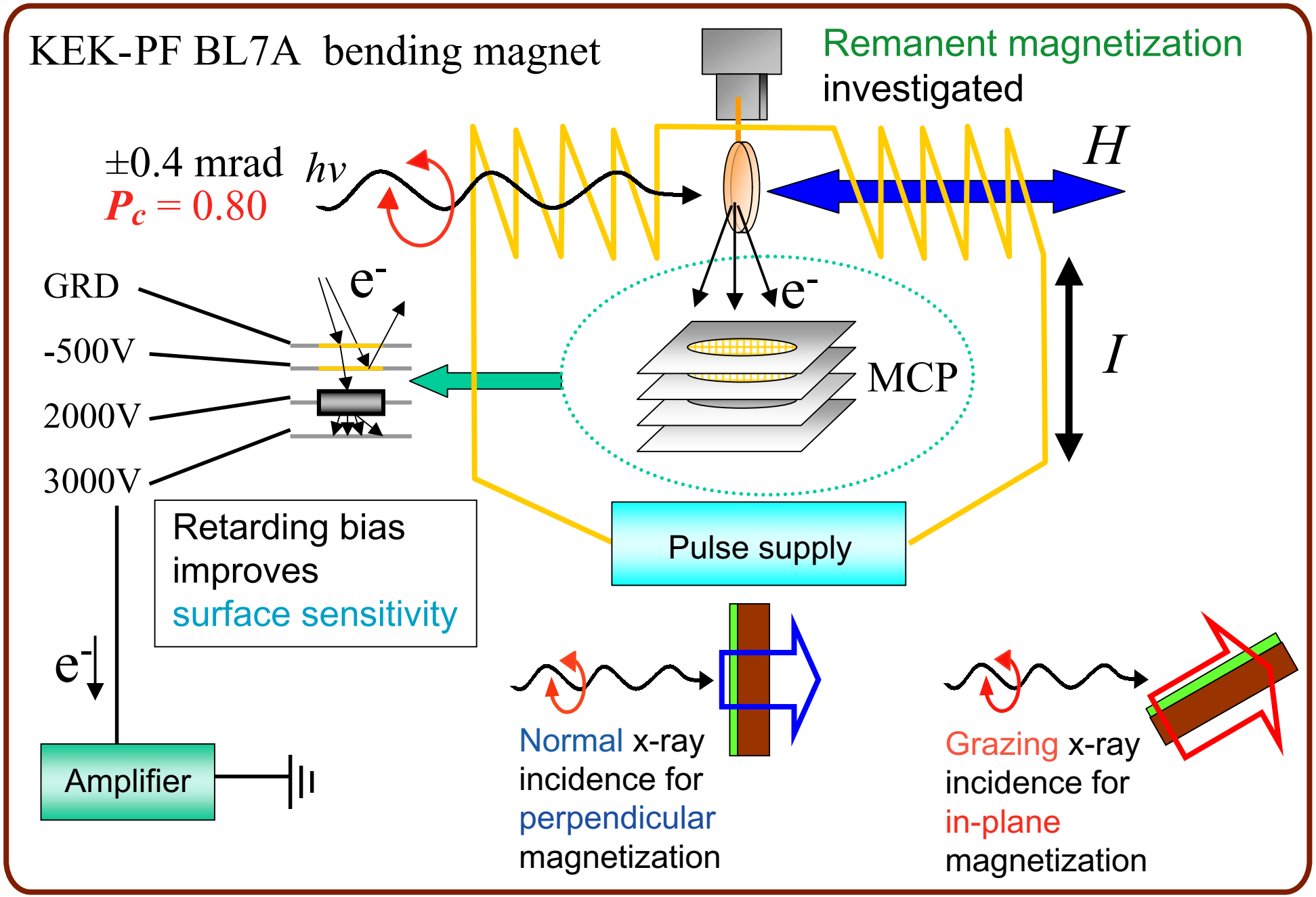
T. Yokoyama *et al.*, *Phys. Rev.* **B62**, (2000) 14191-14196.

T. Yokoyama *et al.*, *J. Electron Spectrosc.* **119**, 207 (2001).

K. Amemiya *et al.*, *Phys. Rev.* **B64**, (2001) 132405.

Y. Yonamoto *et al.*, *J. Phys. Soc. Jpn.* **71**, (2002) 607.

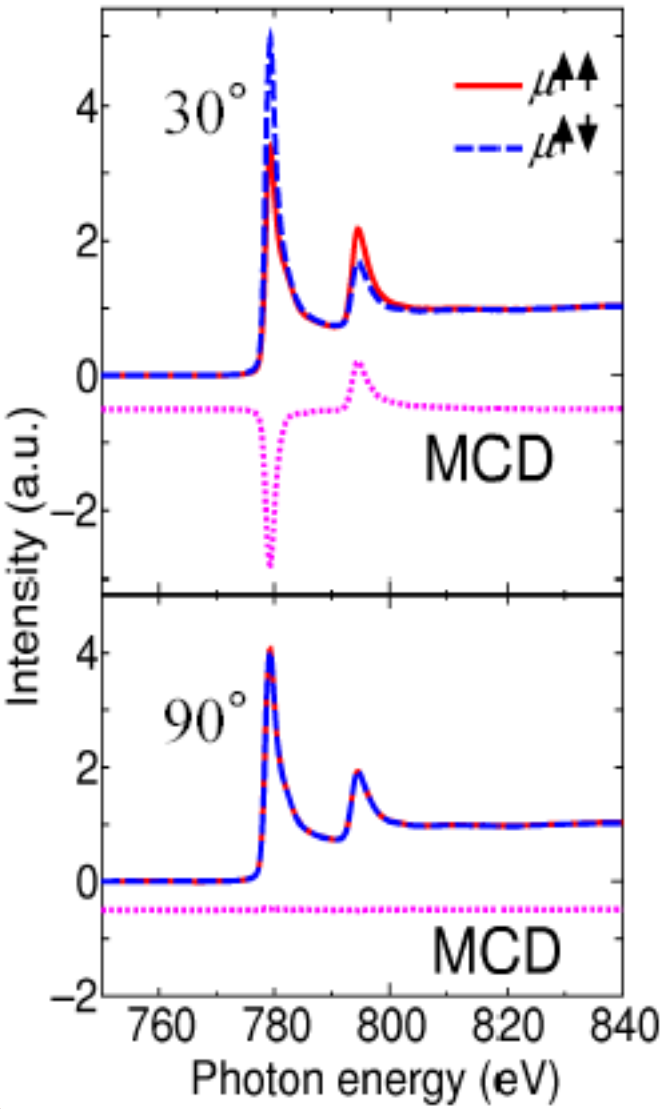
Experimental setup for XMCD measurements

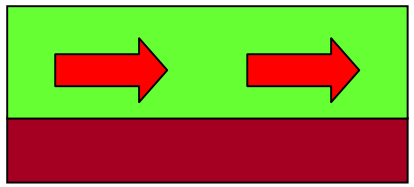


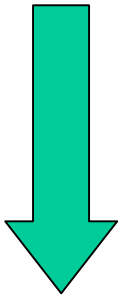
SRT induced by CO adsorption on Co/Pd(111)

Co *L*-edge XMCD of 4.5 ML Co/Pd(111) at 200 K

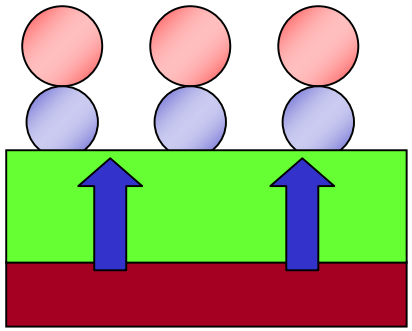
Clean surface



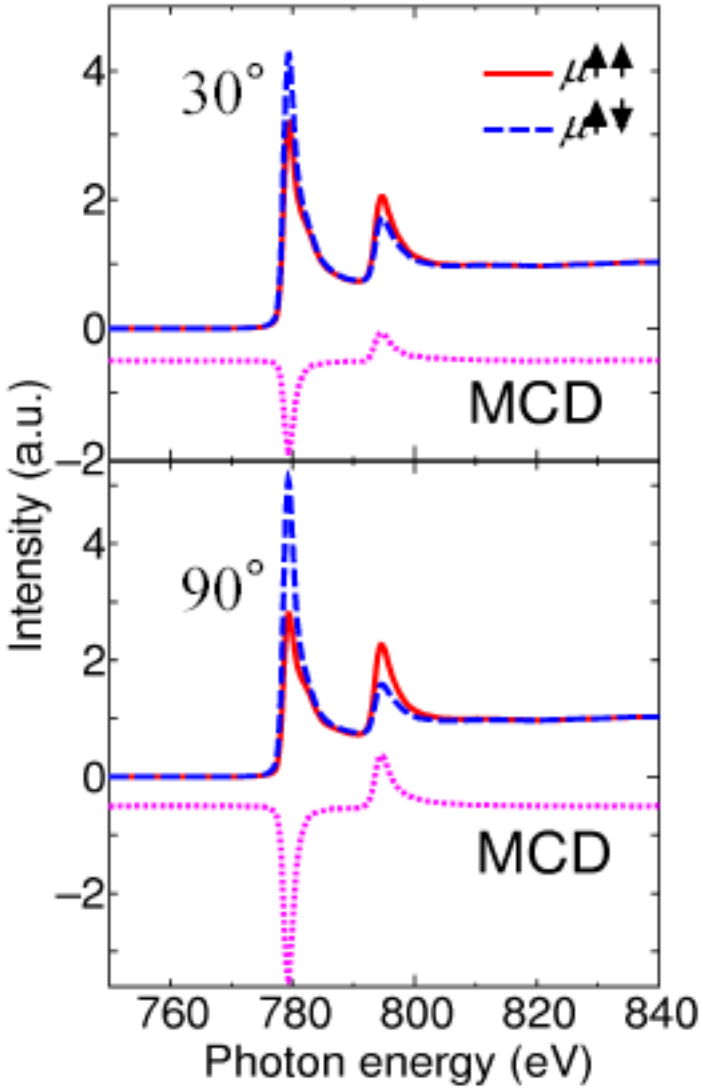




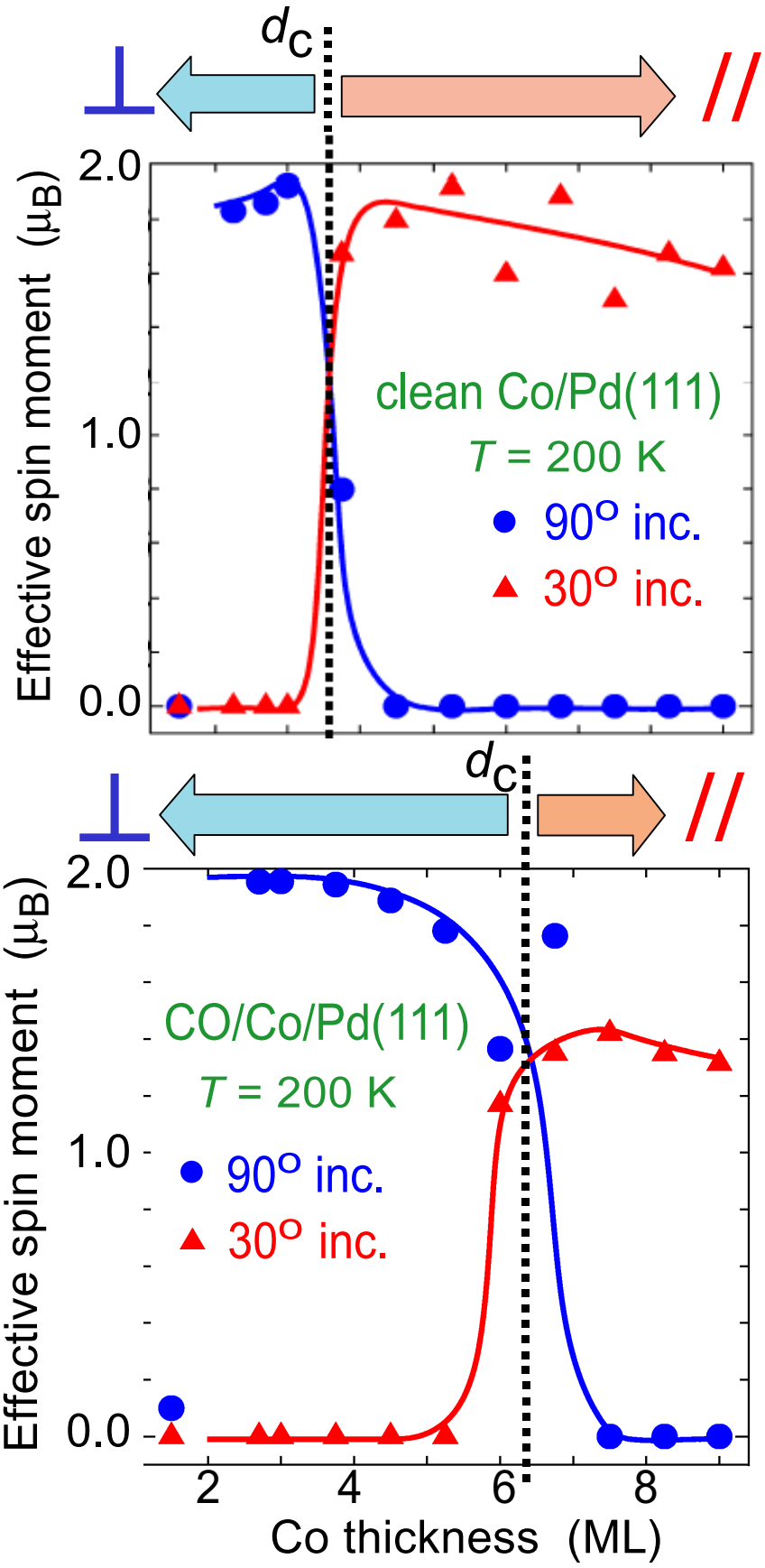
Change of the magnetic easy axis



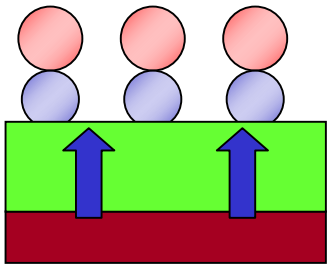
CO adsorbed surface



Co spin in clean and CO-adsorbed Co/Pd(111)

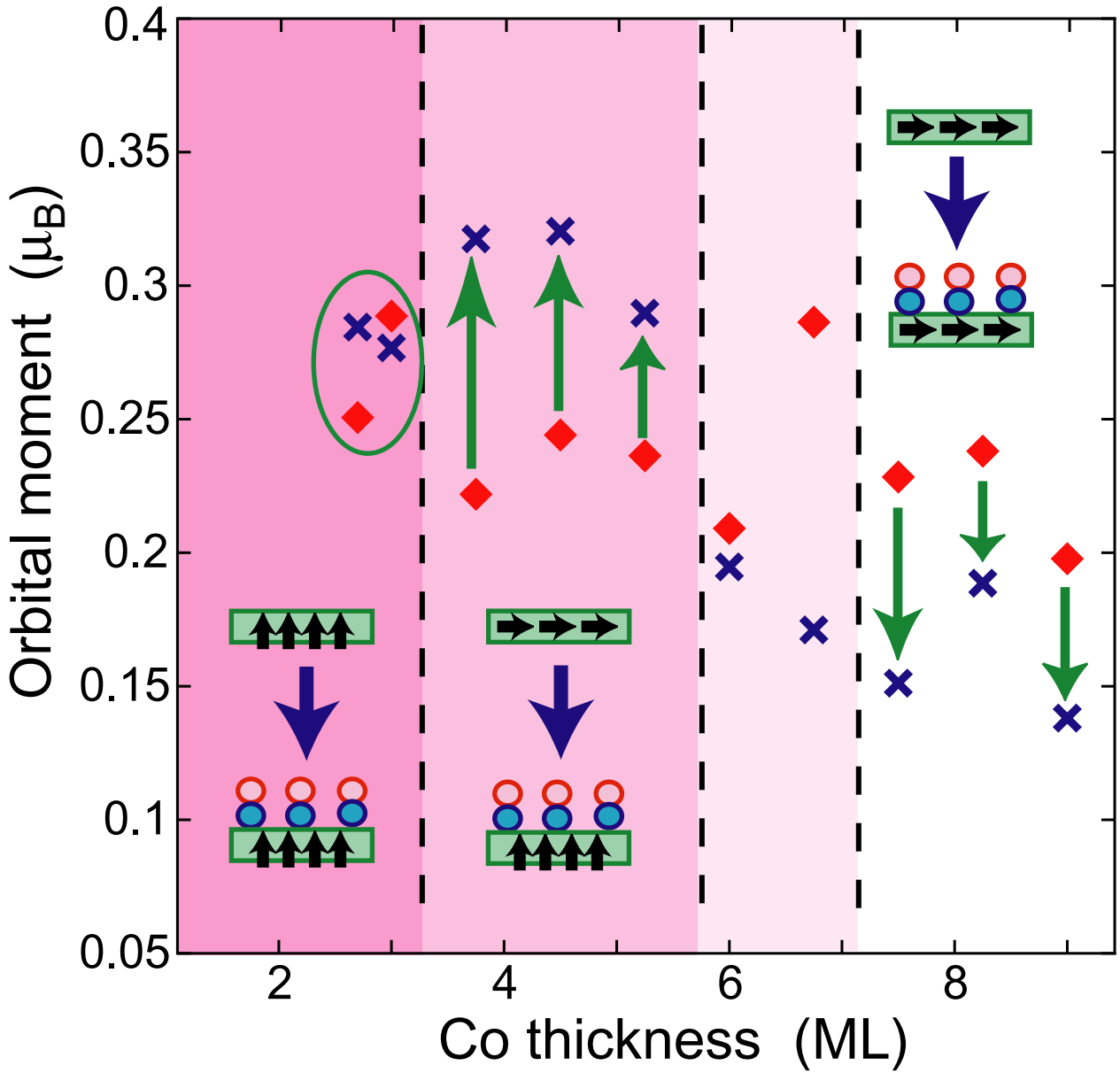


Above the critical thickness d_C, the easy axis is in plane.



CO adsorption extends the PMA stable region.

Co 3d orbital moment in clean and CO-adsorbed Co/Pd(111)



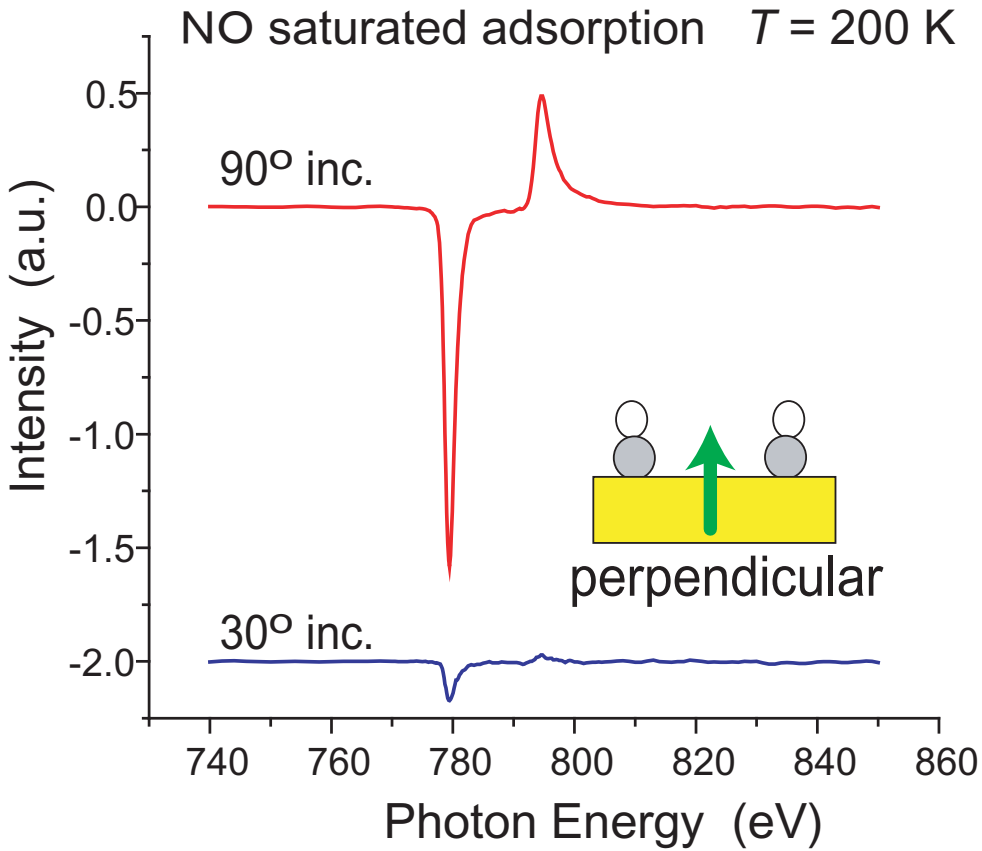
T=200 K

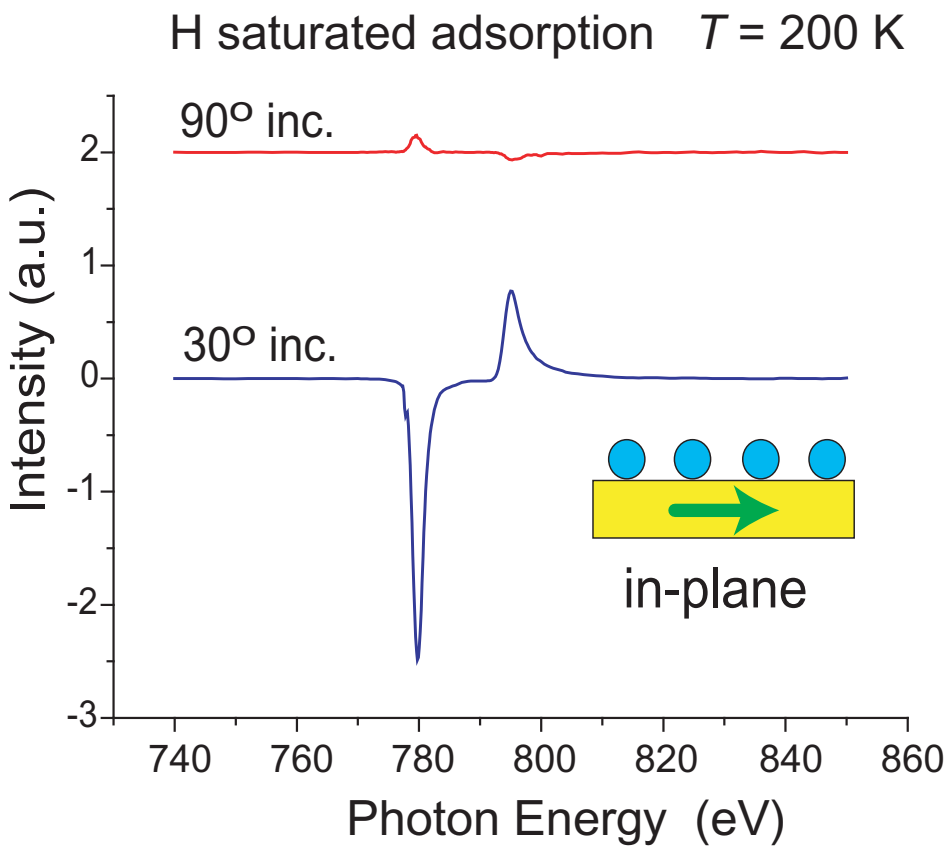
◆ Clean surface

× CO adsorbed

CO adsorption reduces only the in-plane orbital moments.

Co L-edge XMCD of NO- and H-adsorbed 4.5 ML Co/Pd(111)





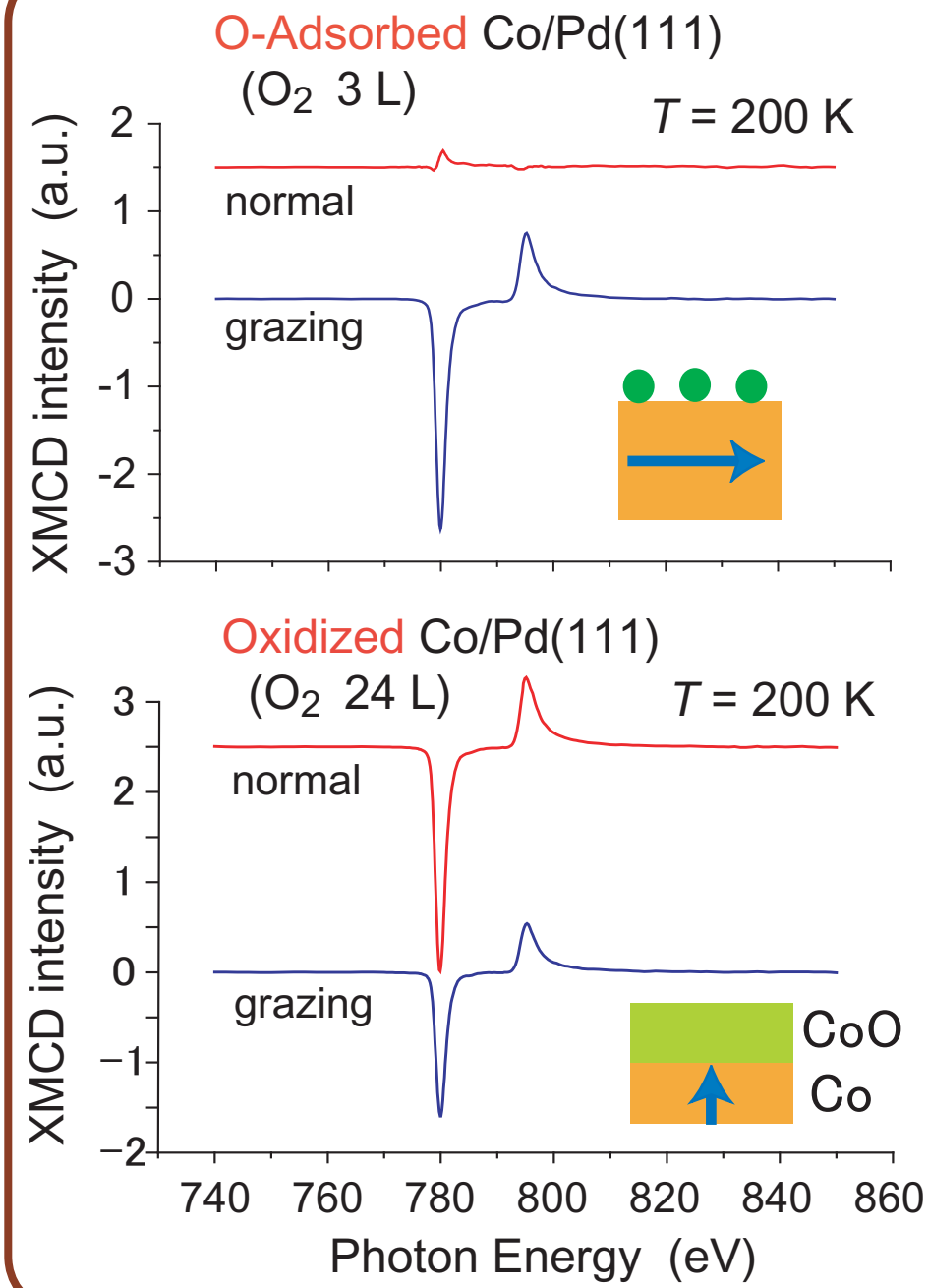
NO induces SRT

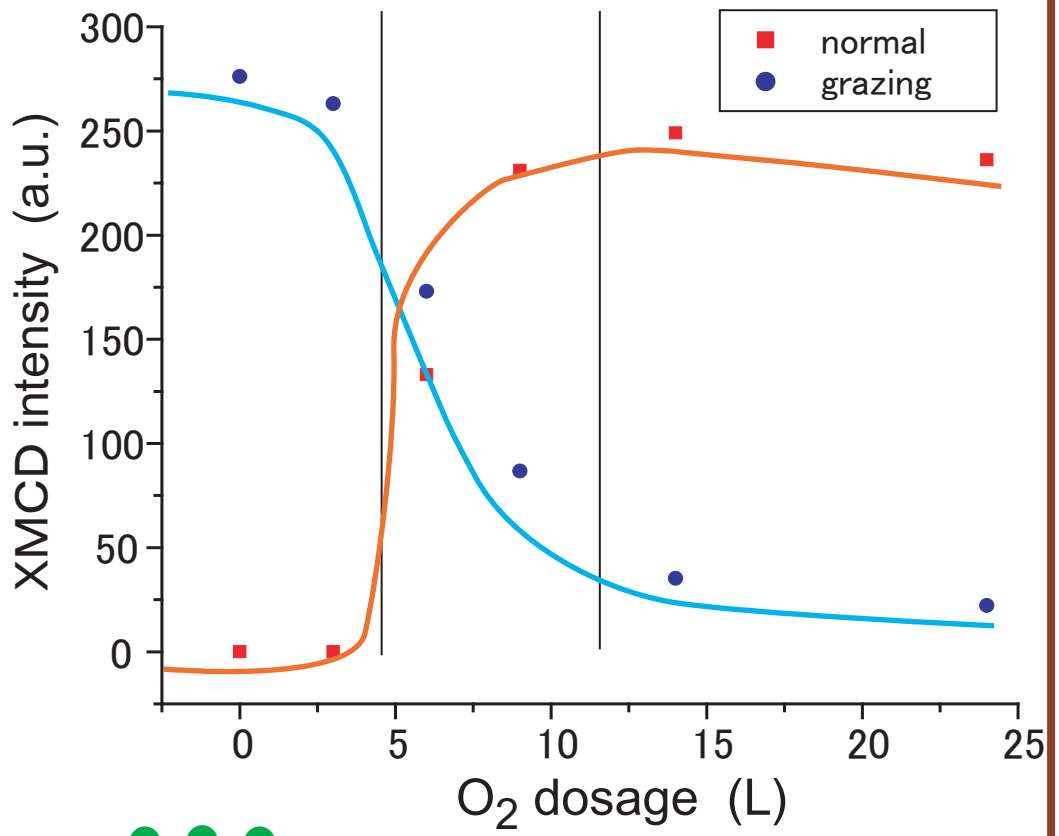
	m_l (μ_B)	m_s (μ_B)	m_l / m_s
Clean	0.25	1.89	0.13
NO ads.	0.16	1.06	0.15

H does not induce SRT

	m_l (μ_B)	m_s (μ_B)	m_l / m_s
Clean	0.30	2.05	0.14
H ads.	0.25	1.78	0.14

Co L-edge XMCD of O-adsorbed Co/Pd(111)

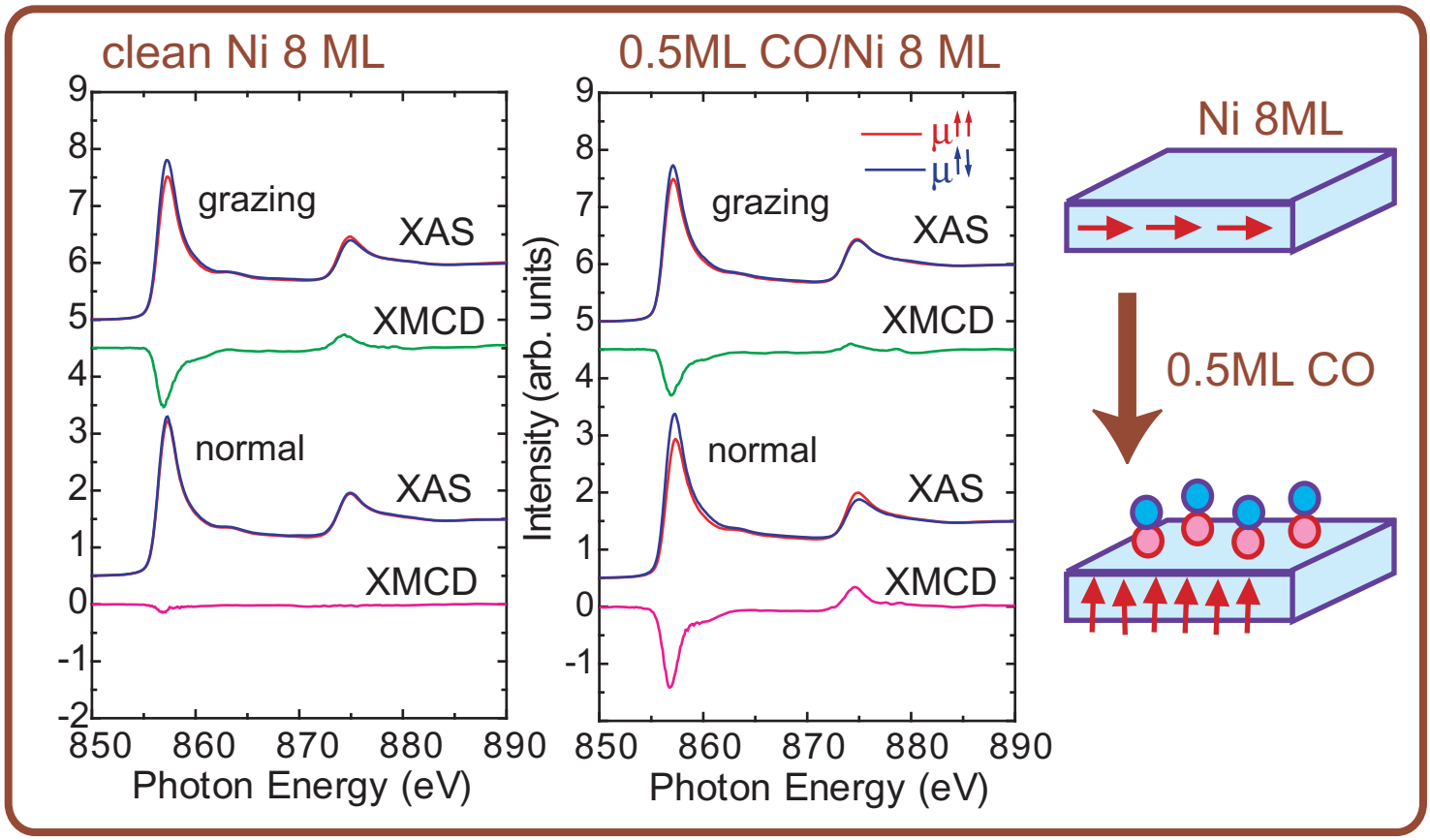


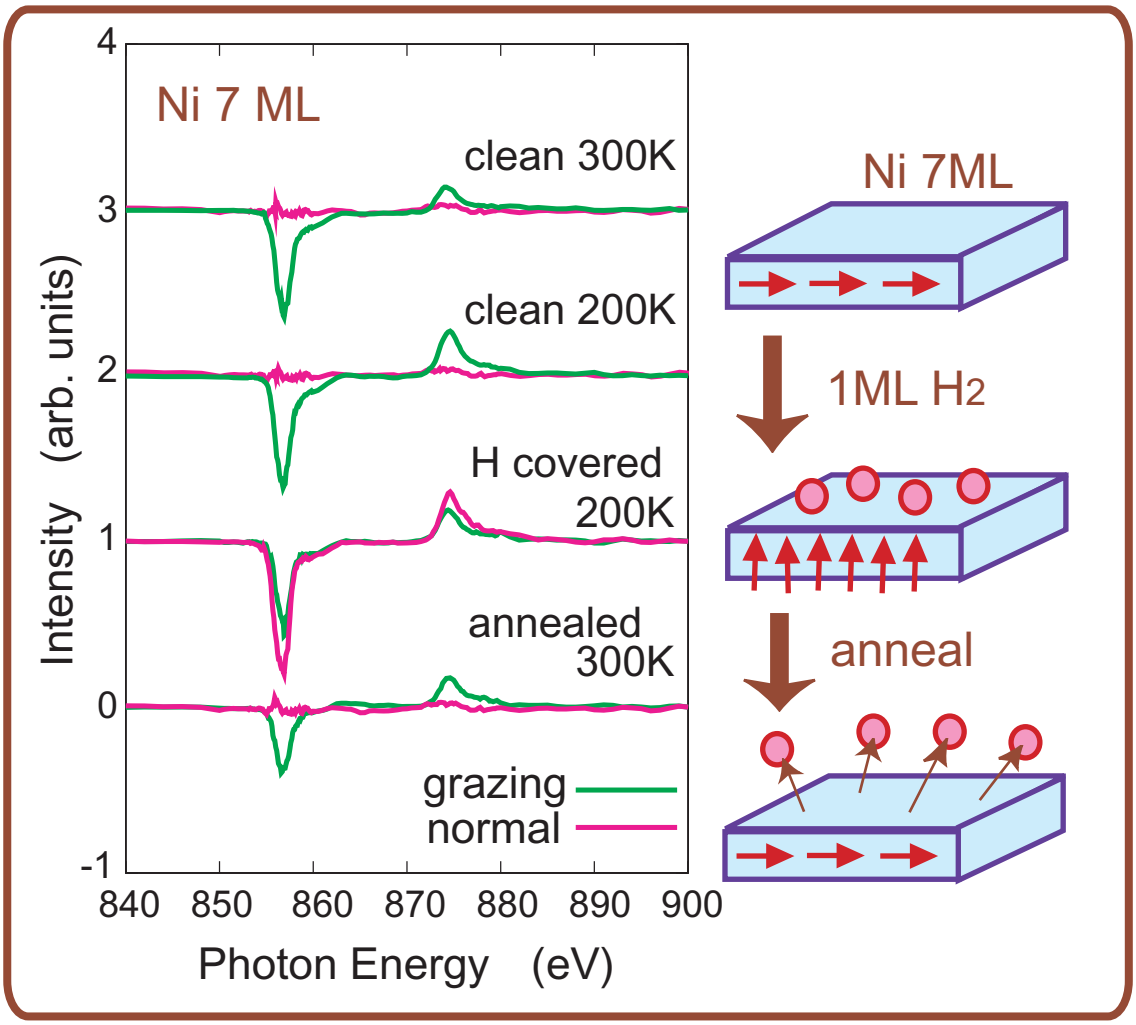


O adsorption does not induce SRT

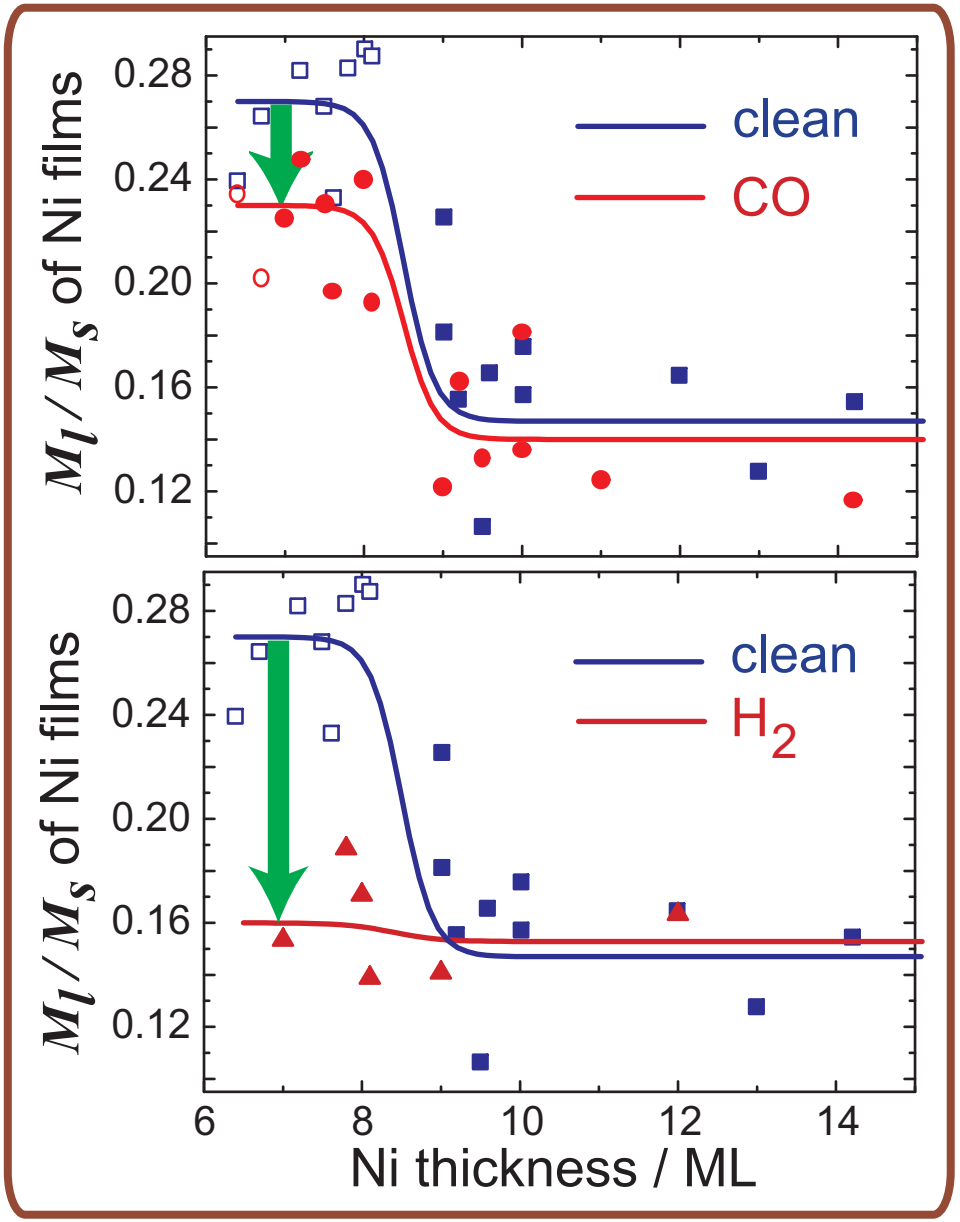
Oxidation leads to SRT due to reduction of Co thickness.

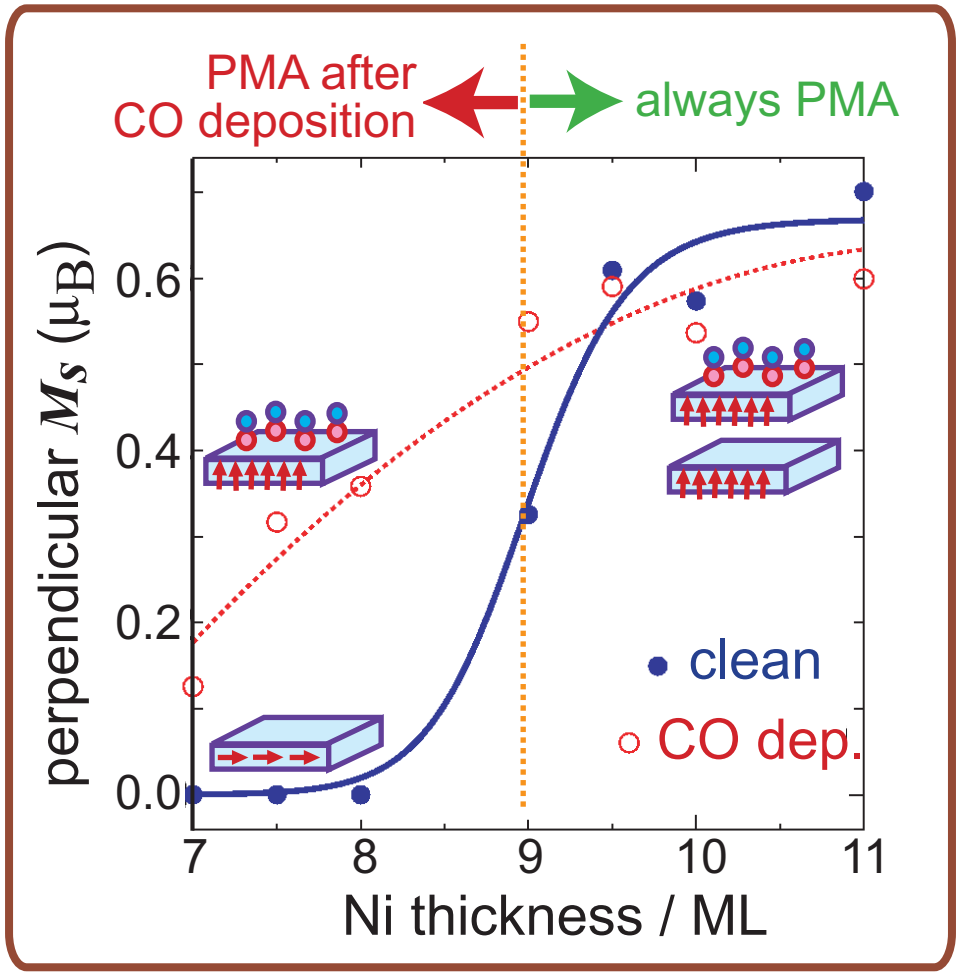
Ni $L_{III,II}$-edge XMCD of CO, H/Ni/Cu(001) films





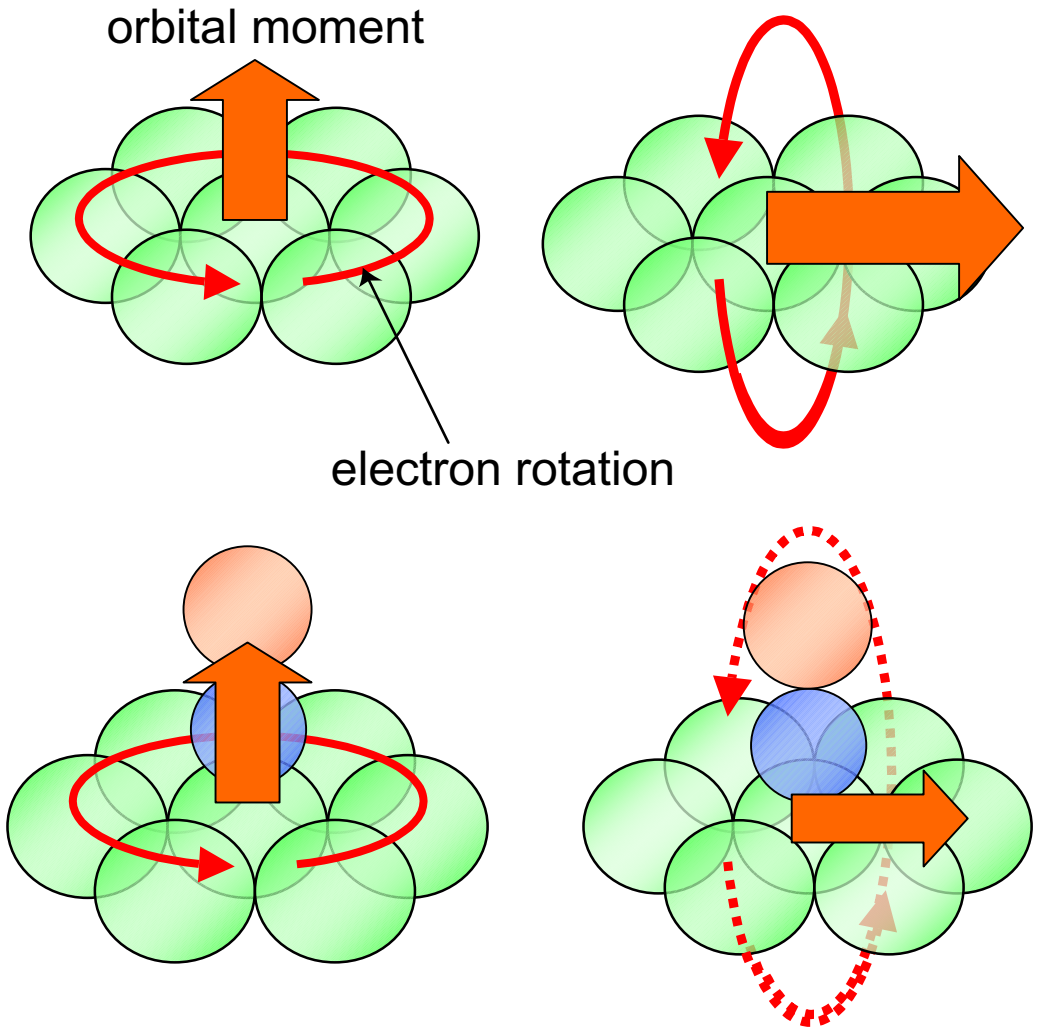
Orbital & spin moments of CO- and H-adsorbed Ni/Cu(001)





H_2 adsorption results in the decrease in both the parallel and perpendicular orbital moments.

Change of orbital moments in Co/Pd(111) upon CO adsorption



Atop site adsorption leads to the reduction of the in-plane orbital moment only.

Spin-orbit interaction

$E_{SO} \propto -\boldsymbol{l} \cdot \boldsymbol{s}$

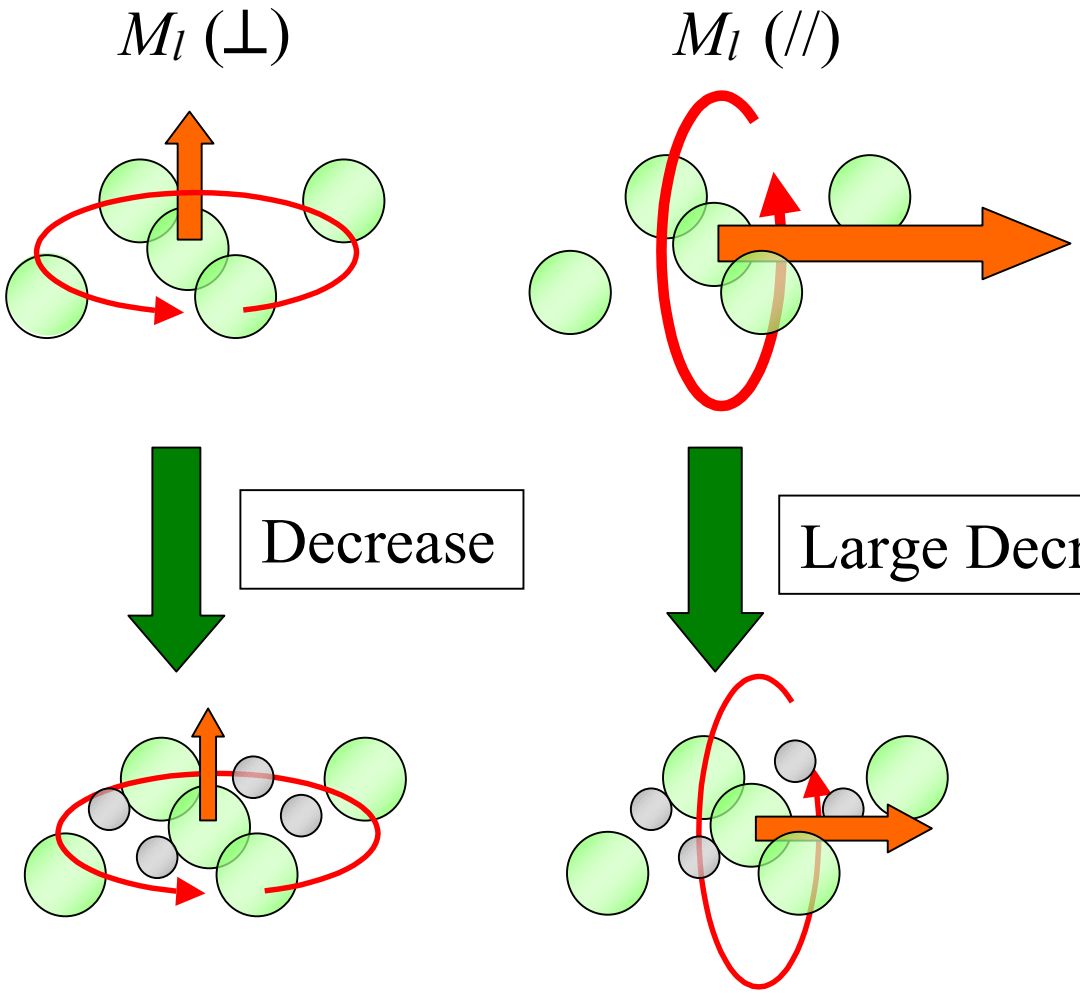
$\boldsymbol{l}$ // $\boldsymbol{s}$ in Fe, Co and Ni

Instabilization of the in-plane magnetization

Stabilization of PMA

NO/Co/Pd(111) and CO/Ni/Cu(001) can be explained similarly.

Change of orbital moments in Ni/Cu(001) upon H adsorption

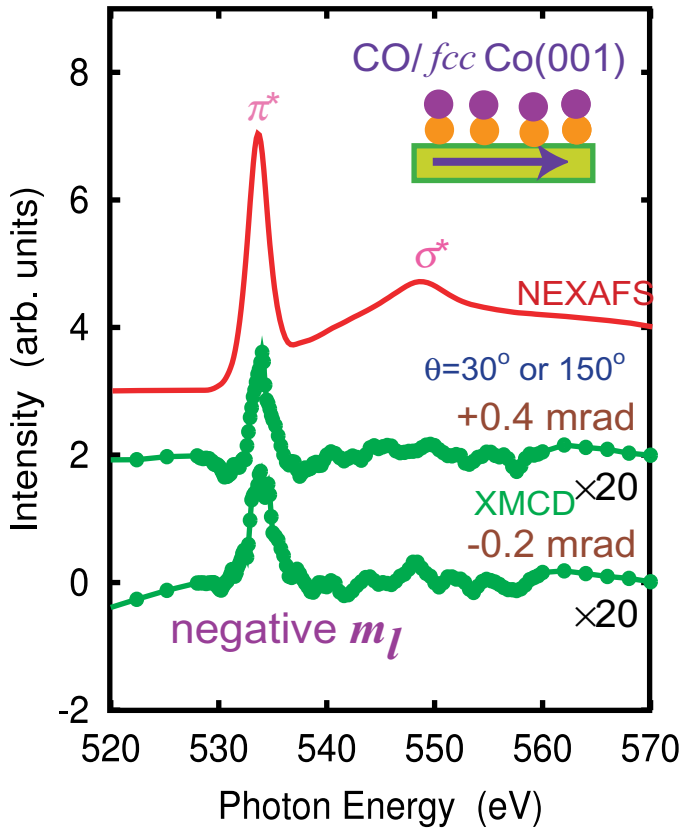


H adsorption reduces both // and ⊥ orbital moments. Since the // moment is larger on the clean film, the effect should be larger.

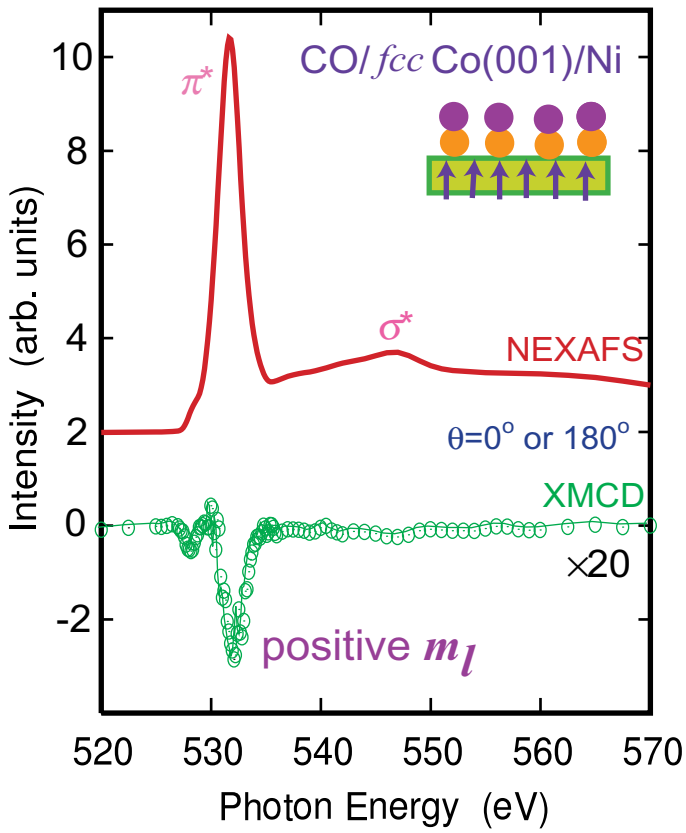
Stabilization of PMA

The reason why SRT does not take place in H/Co/Pd(111) or O/Co/Pd(111) may be smaller coverages: 1 ML H/Ni/Cu(001) vs 1/3 ML O/Co/Pd(111).

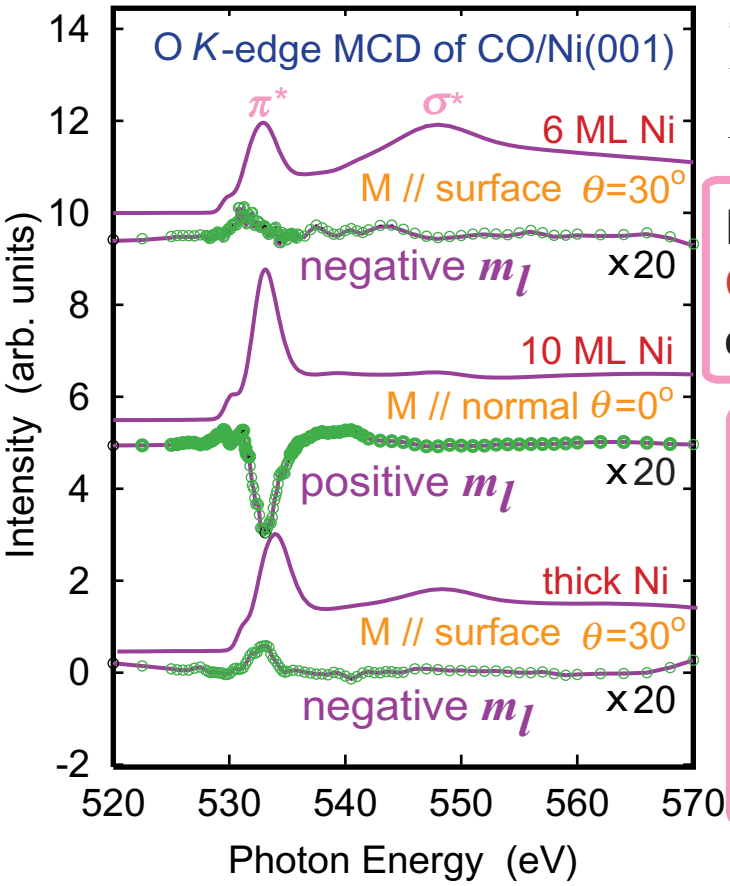
O K-edge XMCD of CO on Co/Cu(001) and Ni/Cu(001)



K. Amemiya, T. Yokoyama, Y. Yonamoto, M. Miyachi, Y. Kitajima and T. Ohta, *Jpn. J. Appl. Phys. Express Letter* **39** (2000) L63.



T. Yokoyama, K. Amemiya, Y. Yonamoto, D. Matsumura and T. Ohta, *J. Electron Spectrosc. Relat. Phenom.* **119** (2001) 207.

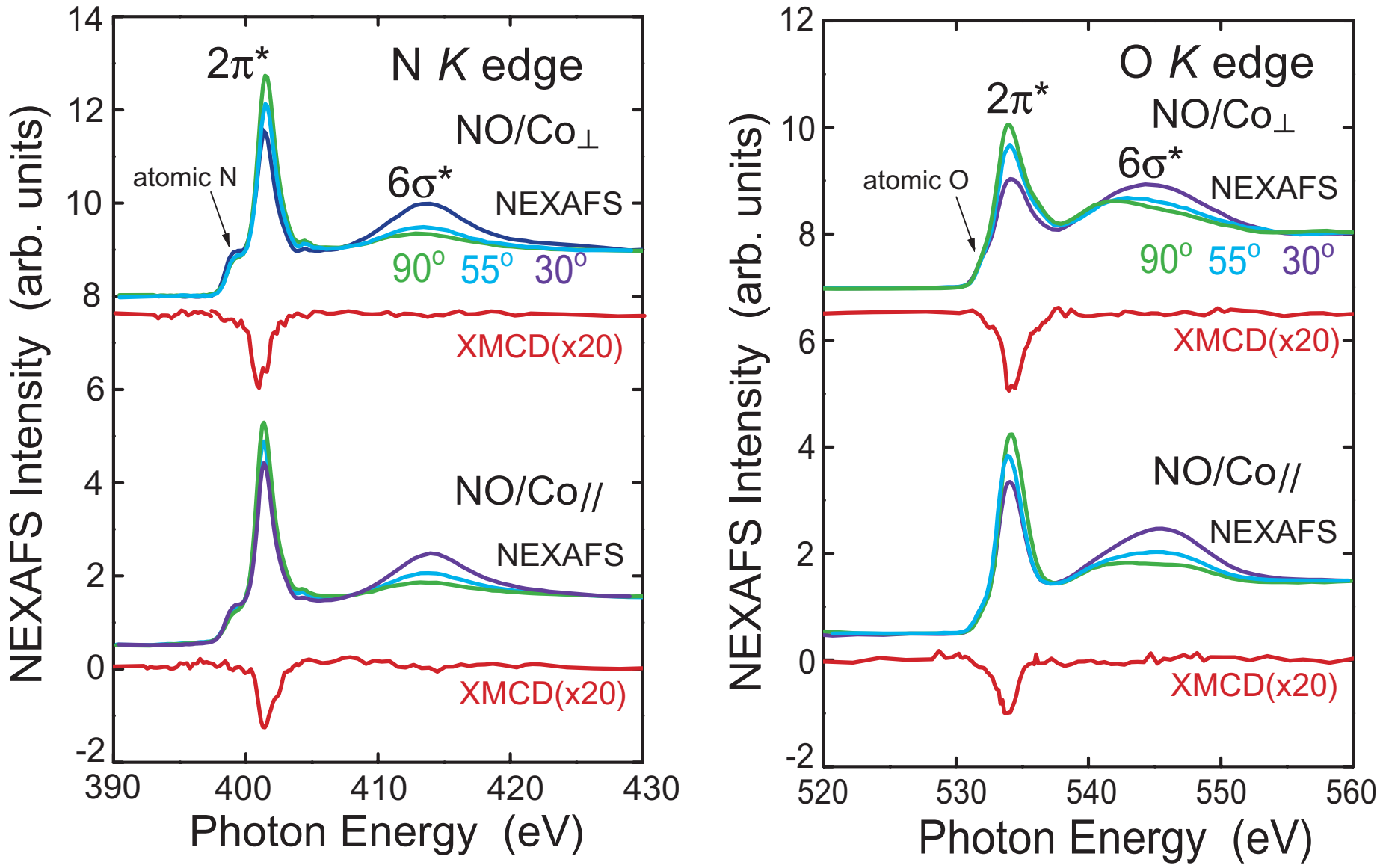


T. Yokoyama, K. Amemiya, M. Miyachi, Y. Yonamoto, D. Matsumura and T. Ohta, *Phys. Rev.* **B62** (2000) 14191.

Induced magnetization on CO 2π* due to the hybridization of metal 3d levels.

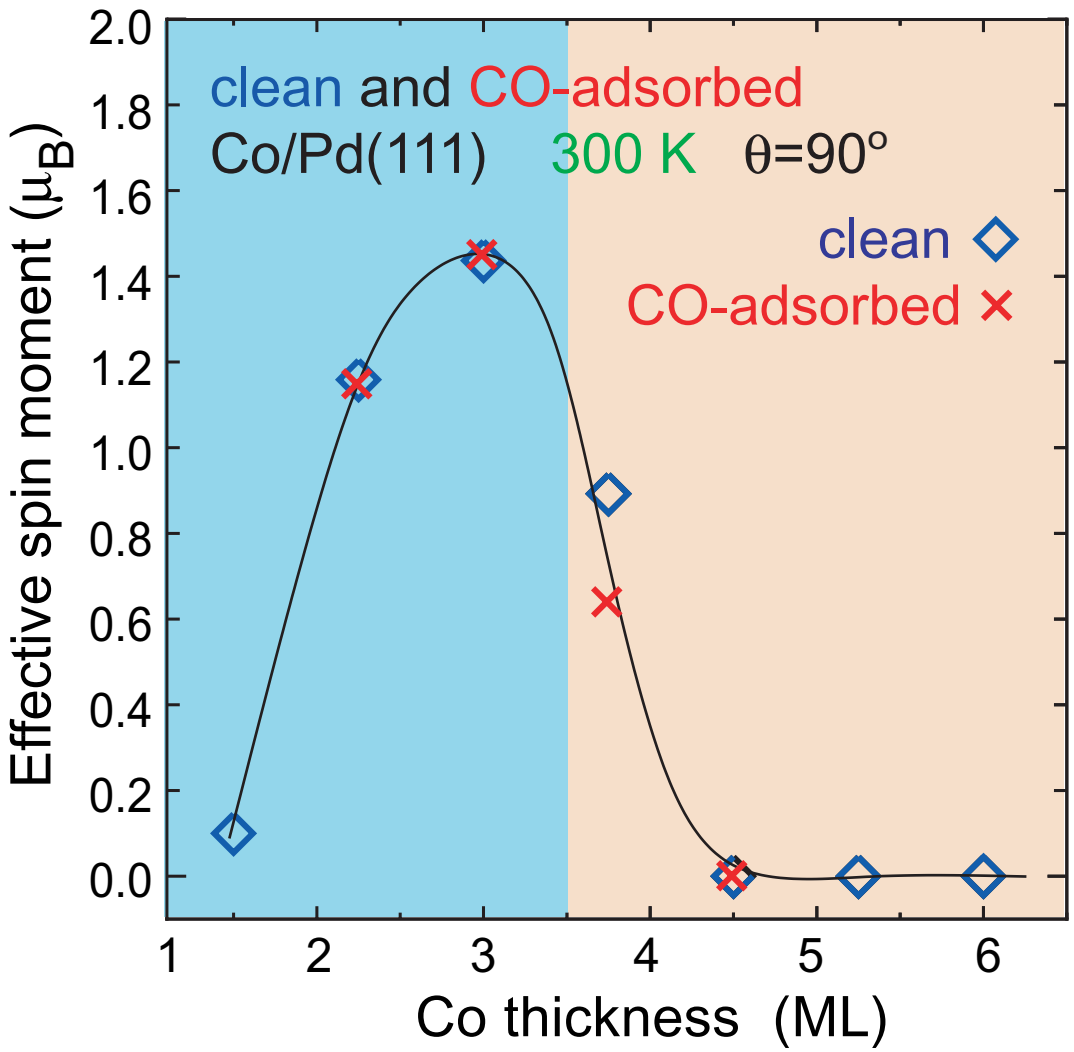
Orbital magnetic moment
in-plane: negative
antiparallel to the substrate magnetization
PMA: positive
parallel to the substrate magnetization

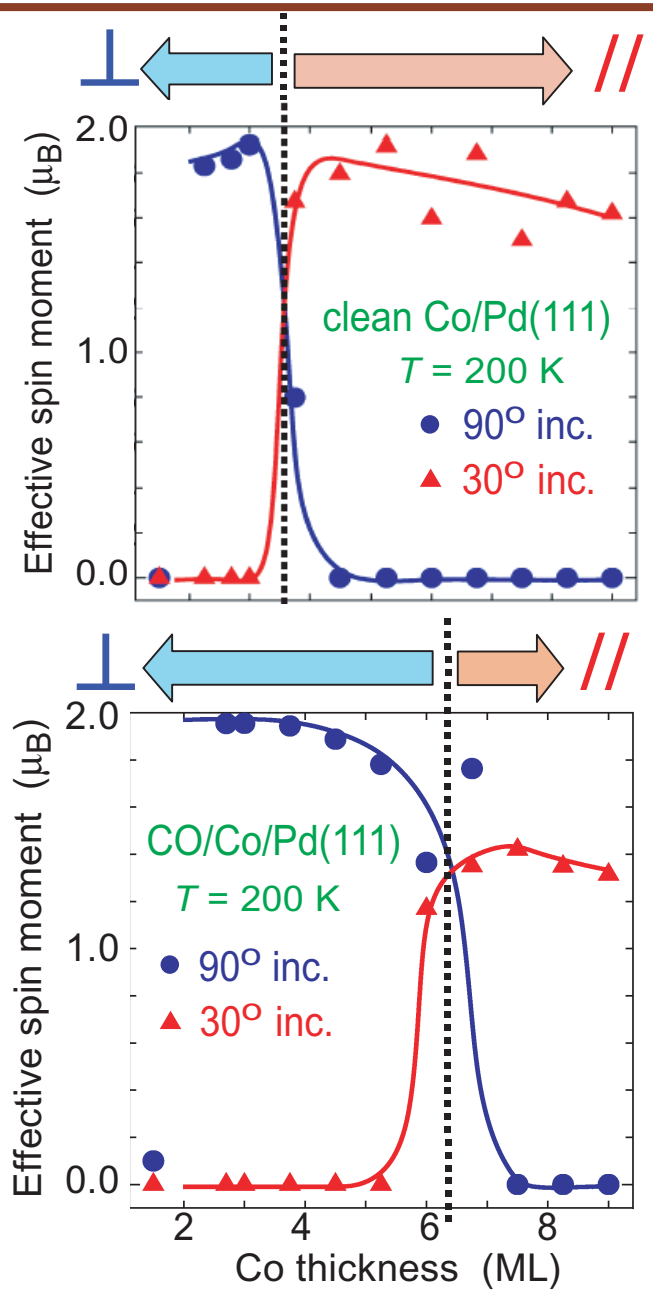
N K- and O K-edge XMCD of NO/Co(001)



negative XMCD at $2\pi^*$ → positive orbital moment

Open question: Why not SRT induced by CO at 300 K ?





No shift in the critical thickness between clean and CO-adsorbed Co/Pd(111) at 300 K, though the saturation coverage of CO at 300 K is greater than the critical CO coverage at 200 K !!

Concluding remarks

1) Experimental findings in NO, H, O and CO adsorption on Co/Pd(111)

Discovered new SRT on Co/Pd(111) induced by CO and NO chemisorption at 200 K

O or H adsorption does not affect d_c of SRT

CO does not affect d_c of SRT at 300 K

2) Experimental findings in CO and H adsorption on Ni/Cu(001)

Confirmed SRT on Ni/Cu(001) induced by CO and H

3) Origin of SRT on Co and Ni films upon CO and NO adsorption

Reduction of the in-plane orbital magnetic moment

Perpendicular orbital moment is not so influenced

Stabilization of PMA through the spin-orbit interaction

4) Origin of SRT on Ni/Cu(001) upon H adsorption

Reduction of both the in-plane and perpendicular orbital magnetic moments

In-plane orbital moment is more influenced

Different adsorption geometry leads to different SRT mechanism

5) Experimental findings of induced orbital magnetic moments on CO and NO on Co and Ni films by means of N and O K-edge XMCD measurements

CO and NO $2\pi^*$ orbitals are magnetically polarized.

PMA: parallel orbital moment to the substrate

In-plane: antiparallel orbital moment