# MAGNETISM AND PHOTON POLARIZATION TECHNIQUES

We078

# Morphology and magnetic properties of thin films of Rh on Highly Oriented Pyrolitic Graphite

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The structure and magnetic properties of ultra-thin Rh layers deposited on highly oriented pyrolitic graphite (HOPG) have been investigated by means of core-level photoemission and scanning tunneling microscopy. The Rh growth on HOPG follows the Volmer-Weber mode at 300 K for any coverage, while Rh may form a commensurate p(1x1) ordered structure when deposited at 150 K for a coverage up to one monolayer. For sub-monolayers or monolayer Rh films on HOPG the linear magnetic dichroism in the angle distribution of photoelectrons shows no evidence of in-plane long range magnetic ordering.

Besides, when thick Rh islands were grown we observed the presence of a surface component in the Rh  $3d_{5/2}$  core level photoemission spectra and the appearance of a net magnetic moment in the surface atoms, like in the case of the Rh(100) surface. This suggests that the magnetic properties of Rh atoms are not only related to the reduced dimension of the system, but also to the interaction (orbital hybridization) with the surrounding atoms.

# Precursor non-magnetic states of $PrFe_4P_{12}$ and $CePd_3$ detected by core excitation MCD

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MCD signals can be detected even for states without magnetic order, because the MCD signals can be induced by the external magnetic field. An example on a Kondo-like semiconductor was reported in ref.[1]. Therefore it is interesting to measure MCD signals for magnetic materials at temperatures higher than Tc or for Kondo-like materials.

In general magnetic properties without magnetic order are well visualized when the inverse of the magnetic susceptibility is plotted against temperatures. In this study we have assumed that the magnitude of MCD signal is proportional to atom-selective magnetic susceptibility. It is one of the advantages of core excitation MCD that atom-selective information on magnetic states is obtained for compounds or alloys.

We have measured the MCD signals on  $PrFe_4P_{12}$  and  $CePd_3$  in the rare earth 3d-4f excitation region at several temperatures and then estimated the magnitudes of the obtained MCD signals. The inverse of the magnitude of MCD is plotted against the measured temperature.

Figure 1 shows the results on  $PrFe_4P_{12}$  with arbitrary units of the vertical axis. The curve is completely different from the behavior of the inverse of the susceptibility that is almost constant in this temperature range. This is an example to suggest that the atom-selective precursor state is different from the ordinary averaged magnetic susceptibility.

Figure 2 shows the results on CePd<sub>3</sub>, which is a Kondo-like material with  $T_{K}$  around 150 K. Though the susceptibility of this material is almost constant, the present material shows a remarkable change dependent on the temperatures. This again suggests difference between the atom-selective MCD magnitude and the ordinary averaged magnetic susceptibility.

The above two results clearly indicate that the local atomic state is magnetized by the external field. Of course this magnetization is under both thermal and quantum mechanical fluctuations. These fluctuations contribute the precursor state without magnetic order



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# MAGNETIC CIRCULAR DICHROISM OF 4D-4F RESONANT X-RAY EMISSION FOR GADOLINIUM

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Magnetic circular dichroism (MCD) of x-ray emission spectroscopy (XES) for rare earths in 4d-4f excitation region is very attractive, since it is expected to show a variety of spectra due to the strong Coulomb interaction between 4d hole and 4f electrons. We have measured XES-MCD for a Gadolinium bulk sample at the undulator beamline BL-28A of the Photon Factory, KEK. The sample was cooled to liquid nitrogen temperature during measurements and 1.2 T magnetic fields in the opposite direction were alternately applied. Figure 1 shows total photoelectron yield (TEY) and TEY-MCD spectra, which are very consistent with the XAS result taken by S. Muto et al [1]. For excitation energies shown in the TEY spectra, we have measured the XES and XES-MCD spectra and clear Ramman scatterings appeared at energy losses of -22 and -4 eV. The ground state of Gd<sup>3+</sup> is 4d<sup>10</sup>5p<sup>6</sup>4f<sup>7</sup> (<sup>8</sup>S) configuration and the final states of these Ramman scatterings are 4d<sup>10</sup>5p<sup>5</sup>4f<sup>8</sup> (<sup>8</sup>D) and 4d<sup>10</sup>5p<sup>6</sup>4f<sup>7</sup> (<sup>6</sup>D, <sup>6</sup>P, <sup>6</sup>G) configurations, respectively [2]. The former Ramman scattering extremely enhanced for <sup>8</sup>D resonant excitation (B) and the XES-MCD was much larger than the TPY-MCD. The magnitude of the magnetic moment evaluated with the TPY and XES spectra are 0.2  $\mu_B$  and 0.8  $\mu_B$ , respectively. This indicates the great difference of the magnetic states at the surface and in the bulk.



Figure 1: TEY and XES spectra for Gadolinium bulk sample. Energy loss in the figures of XES is defined as (emission energy – excitation energy).

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# Magnetic Circular Dichroism in the Total Ion Yield of Atomic Iron and Chromium

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We performed an experiment on polarized iron and chromium atoms in the gas phase. For iron we observed a remarkably large polarization dependence of the total ion yield of Fe-vapor far away from any absorption edge. Similar measurements have been performed on chromium below the 2p threshold. They do not show this behaviour. In solid state studies the MCD shows a large effect only at the 2p- and 4f-absorption edges of the 3d-transition metals and rare-earth compounds, respectively. In the non-resonant region several eV away from absorption edges MCD vanishes in absorption measurements of thin iron films i.e. the absorption of left- and right handed radiation differs by less than 0.1% [1]. Additionally our data shows an increase of the MCD-asymmetry with photon energy. This effect can not be understood in an independent particle model: At higher photon energies a stronger contribution from the 3p shell must cause an attenuation of the MCD effect. An explanation for this observation needs more sophisticated theoretical calculations



Figure 1: Results for the MCD asymmetry for radiation with positive (solid circles) and negative (open circles) helicity. The dotted line is a fit to the data points below 220 eV to illustrate the trend to higher values of the asymmetry at higher photon energies.

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# HIGH ENERGY-RESOLUTION MAGNETIC CIRCULAR DICHROISM MEASUREMENT OF FERRITE FAMILY

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Magnetic circular dichroism (MCD) in the soft x-ray absorption spectroscopy has been used as a powerful technique to investigate the electronic state of magnetic materials. The magnetism of ferrite family denoted with  $MFe_2O_4$  (M : a transition metal element), has been studied intensively [1]. In this study, we measured Fe 2p MCD and XAS spectra of  $MFe_2O_4$  to study the 3d electronic states with spin states.

The measurements were performed at the newly constructed soft x-ray beamline, BL23SU, at SPring-8. The light source is an APPLE-2 (Sasaki) type variably-polarizing undulator. The periodic phase shift up to 0.1 Hz of the magnet rows of the undulator provides a switching of the hilisity of circularly polarizing soft x-ray synchrotron radiation. The MCD measurement system, which is synchronized the monochromator control with the phase shift, has shown good signal-and-noise

ratio with the high energy-resolution [2].  $MFe_2O_4$  powder sample was pasted on a sample holder using a carbon tape. During the measurements, a permanent magnet applied 0.4 Tesla magnetic field at room temperature.

The Fe  $L_3$  MCD spectra of MnFe<sub>2</sub>O<sub>4</sub> and NiFe<sub>2</sub>O<sub>4</sub>  $\cong$  are plotted in Fig. 1. The high-energy resolution spectra show the fine structures due to final state multiplets affected by crystal field. The local symmetry of Fe in NiFe<sub>2</sub>O<sub>4</sub> is tetrahedral and octahedral, while that in MnFe<sub>2</sub>O<sub>4</sub> is only octahedral. The origin of the positive peak of NiFe<sub>2</sub>O<sub>4</sub> MCD at 706 eV is attributed to the tetrahedral  $3d^5$  state. The spectra will be analyzed with a multiplet atomic model calculation.

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Figure 1: Fe  $L_3$  MCD spectra of NiFe<sub>2</sub>O<sub>4</sub> and MnFe<sub>2</sub>O<sub>4</sub> at magnetic field 0.4 T and R. T.

# THE XAS AND MCD STUDIES IN CrFe<sub>2</sub>O<sub>4</sub>

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Magnetic Circular Dichroism (MCD) in the core-level absorption spectroscopy (XAS) has become quite a powerful tool for the investigation of the electronic state of ferro- and ferri magnetic materials.  $CrFe_2O_4$  is one of the spinel systems of  $FeCr_2O_4$ -  $Fe_3O_4$ . In  $CrFe_2O_4$ , Cr ions sit octahedral site, while Fe ions sit both tetrahedral and octahedral site.  $CrFe_2O_4$  has been studied extensively about the magnetic and electronic properties.

We have performed XAS and MCD at the soft X-ray beamline BL23SU of SPring-8. XAS

were measured with the total electron yield method. MCD was obtained by switching the right-handed and left-handed circularly polarized X-rays at each photon energy. MCD measurements were carried out at T = 300 K with a magnetic field of H = 0.4 Tesla.

In Fig. 1, Cr 2p XAS and MCD spectra are shown. The overall line shape of the XAS is quite similar between CrFe<sub>2</sub>O<sub>4</sub> and Cr<sub>2</sub>O<sub>3</sub> [2]. The XAS and MCD of CrFe<sub>2</sub>O<sub>4</sub> shows multiplet structures clearly.

The origin of structures seen in the 2p XAS spectra is the combination of the electron-electron interaction in the Cr atom, the crystal field applied to the Cr ion by O<sup>2-</sup> and the hybridization between the Cr<sup>3+</sup> 3*d* orbital and O<sup>2-</sup> 2*p* orbital. Because of the small spin-orbit splitting of the Cr  $2p_{1/2}$  and Cr  $2p_{3/2}$  states the Cr L<sub>2,3</sub>-edges are not separated clearly, so that the spin sum rules could not be applicable. We, however, could estimate the contribution of the orbital moment by means of the orbital sum rules. Our results indicated



Figure. 1: The experimentally observed XAS and MCD for Cr 2p XAS.  $I_+$  (solid line) represents the XAS for magnetization parallel to the photon-spin and  $I_-$  (broken line) represents that for anti-prallel.

that the orbital moment was very small.

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## THE MCD STUDIES IN 2P AND 3D XAS OF LASRCOO3

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Magnetic Circular Dichroism (MCD) in the core-level absorption spectroscopy (XAS) has become quite a powerful tool in the investigation of the electronic state of ferro- and ferri magnetic materials. The family of Perovskite compounds  $La_{1-x}Sr_xCoO_3$  has been extensively studied for their properties related to semiconductor-metal transition as a function of doping concentration. For the compositions of x>0.1,  $La_{1-x}Sr_xCoO_3$  shows itinerant ferromagnetism.[1]

MCD of Co 2*p* in La<sub>1-x</sub>Sr<sub>x</sub>CoO<sub>3</sub> (x = 0.3, 0.4, 0.5) was measured at the soft X-ray beamline BL25SU of SPring-8. XAS was obtained by measuring the total photoelectron of the sample. MCD was evaluated by alternating the magnetic field with respect to the spin of the circularly polarized soft X-ray.

Fig. 1 shows Co 2p MCD spectra of La<sub>1</sub>  $_{x}$ Sr<sub>x</sub>CoO<sub>3</sub> (x = 0.3, 0.4, 0.5). MCD spectra of  $La_{1-x}Sr_{x}CoO_{3}$  were shown multiplet structures and these spectra were quite similar. The origin of structures seen in the 2p XAS spectra is the combination of the electron-electron interaction in the Co atom, the crystal field applied to the Co ion by  $O^{2-}$  and the hybridization between the  $Co^{3+} 3d$ orbital and  $O^{2-}2p$  orbital. We define the integrated intensities of the  $2p \rightarrow 3d$  absorption in the  $L_3$  and  $L_2$  region for the RCP (LCP) beam by  $I_{+}(L_3)$  and  $I_{+}(L_2)$  [I\_(L\_3) and I\_(L\_2)], respectively. In our result,  $R_{MCD} = [I_{(L_3)} - I_{+}(L_3)] / [I_{(L_2)} - I_{+}(L_3)]$  $I_{\perp}(L_2)$ ] became bigger as Sr compositions increased. This fact means that the orbital moment is bigger as a function of Sr concentration.

For the analysis of the 3*d* electronic state, the XAS and MCD spectra will be compared with theoretical calculation of the spectra using the ligand field multiplet model.

# References



Figure 1: Co 2p MCD spectra of La<sub>1-x</sub>Sr<sub>x</sub>CoO<sub>3</sub> (x=0.3, 0.4 and 0.5)at magnetic field 1.4 T and T=50 K.

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# **Re-investigation of electronic structure and ferromagnetism** of non-reconstructed Cr(001) 1x1 surface

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Cr(001) surface is of great interest because of the possible existence of ferromagnetism despite of its antiferromagnetic behaviour in bulk, and intensive studies have been carried out with various electron spectroscopies with and without electron spin analysis, so far [1-3]. The results suggest that the long range ferromagnetic order of Cr(001) surface is strongly influenced by oxygen incorporation, lattice distortion, existence of defects in the surface, etc., the origins of which are not fully understood.

We have measured spin- and angleresolved photoemission spectra of nonreconstructed Cr(001) 1x1 surface and investigated the electronic structure and its dependence upon oxygen and carbon incorporation. The valence band structure obtained by angle-resolved photoemission spectra (Figure 1) shows a good agreement with previous experimental and theoretical investigations including surface electronic states [1, 4]. In spin-resolved photoemission experiments, we have observed that the ferromagnetic order of the Cr(001) surface is strongly influenced not only by a small oxygen concentration but also by carbon. Carbon atoms of a few atomic percent of total volume probed by Auger spectroscopy substantially reduce the ferromagnetic order of Cr(001) surface.



Figure 1. Experimentally obtained valence band structure of Cr(001) along  $\Delta$  and  $\Sigma$  direction in Brillouin zone. Solid curves represent the results of theoretical band calculation.

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# Electronic structure and re-orientation of perpendicular magnetic anisotropy of Co/Au(111) and Co/Pd(111)

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Epitaxially grown magnetic thin films in a monolayer regime have attracted many interests because they embody magnetic properties with low dimensionality such as perpendicular magnetic anisotropy (PMA), enhanced magnetic moments at surfaces and interfaces, etc. [1]. Since the magnetism of thin films depend on details of geometric structures, many studies have been performed using various experimental techniques which are accessible to their structural and magnetic properties. In case of Co thin films, the magnetization direction of Co depends on the film thickness and changes from perpendicular to parallel to the surface at a certain thickness [2]. So far, only a few experimental works were devoted to directly observe the electronic structures of Co films, and the connection between the structural and hence magnetic properties of Co films is not fully understood.

We have measured spin- and angle-resolved photoemission spectra of Co thin films epitaxially grown on Au(111) and Pd(111) substrates and investigated their electronic and magnetic properties. In the Co/Au(111) system [3], we have observed strong mixing between Au 5d and Co 3d states at the interface resulting a considerable spin polarization of Au 5d states and the increase of Co 3d orbital parallel to the surface, which causes the PMA of the Co film. The electronic structure of the Co film in low coverage region is different from that of bulk hcp Co and it becomes closer to those of the bulk as the film thickness increases. The magnetization direction changes from perpendicular to parallel to the surface at about 6 ML. The origin of the reorientation is qualitatively explained by the increasing contribution of Co 3d orbitals perpendicular to the surface due to the relaxation of the atomic distance of Co perpendicular to the surface. In the Co/Pd(111) system [4], the reorientation of the magnetization direction of the Co film occurs at about 4 ML. The origin of the reorientation is qualitatively explained as due to the increasing contribution of the upper  $\Lambda_3$  band with increase of the film thickness as in the case of Co/Au(111) system. The stronger hybridization between Co 3d and Pd 4d states causes lager binding energies of the  $\Lambda_3$  states than in the Co/Au(111).

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# Polarisation Dependence of Soft X-Ray Resonant Magnetic Scattering at the 2p Edge of Fe

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We report on resonant magnetic scattering experiments of circularly, elliptically and linearly polarised soft X-rays in L- and T-MOKE geometry at the Fe-2p absorption edge of an *ex-situ* grown ferromagnetic Fe/C multilayer. We focus on the polarisation dependence of the asymmetry parameters  $A_L$ ,  $A_T$ , which are the normalised differences of the reflected intensities upon reversal of the magnetic field. The asymmetry is investigated in detail as a function of photon energy, angle of incidence, azimuthal angle and magnetic field strength. The measurements were performed with the BESSY polarimeter /1/ exploiting the tunability of the polarisation state of synchrotron radiation from the BESSY-II undulator beamline UE56/2 PGM /2/.

The longitudinal magneto-optical Kerr effect (L-MOKE) is observed predominantly with circularly polarised light and magnetic moments lying in the sample surface parallel / anti-parallel to the light direction. The asymmetry  $A_L$  is found to be directly proportional to the degree of circular polarisation for all investigated energies and angles of incidence. The T-MOKE is observed predominantly with linearly polarised light with polarisation vector in the plane of incidence (p-geometry) and magnetic moment perpendicular to the scattering plane. For the asymmetry we find  $A_T \approx a + bP_L$  where a and b are constants and  $P_L$  is the degree of linear polarisation. This shows that magnetic information can be obtained in T-MOKE even with unpolarised light. In both L- and T-MOKE the asymmetry dependence obeys the theoretical predictions based on the reflection matrix and on the Stokes formalism.

Based on these experimental data set a method was developed for a selfcalibrating determination of both, the polarisation state of light and of the magneto-optical constants of the sample in use.

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# DICHROISM EXPERIMENTS IN THE 3P PHOTOELECTRON SPECTRUM OF FREE LASERPOLARIZED FE ATOMS

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Dichroism experiments are widely used in solid-state physics to investigate magnetic properties. For the most important ferromagnetic element, Fe, no data were available for free atoms yet. We present experimental results on dichroism in the 3p-photoelectron spectra of either oriented or aligned atoms. The ionisation was done by means of synchrotron light from the undulator beam line BW3 at HASYLAB.

The necessary ground state polarisation of the iron atoms was achieved by laser optical pumping around 372nm. This near-UV light was generated by a combination of a narrowband, single-mode cw laser and a subsequent frequency doubling in an external ring cavity [1].



The figure shows the results of the magnetic orientation dichroism. The atomic orientation was generated by optical pumping with either left or right handed circularly polarized laser light.

Further experimental results as well as a detailed comparison with the standard theoretical approach for photoelectron angular distribution from polarised atoms [2] will be presented including the influence of higher order atomic multipole moments.

We welcome and encourage everyone to stop by at our booth to talk about this research field and discuss future experimental or theoretical ideas.

Figure 1: Orientation dichroism of the high spin components <sup>6</sup>F and <sup>6</sup>D of the Fe 3p spectrum.

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### High-resolution angle-resolved photoemission spectroscopy on US

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US has been intensively studied by experiment and theory since it shows the anomalous physical properties such as the para-ferro magnetic phase transition ( $T_C = 180$  K) which is regarded to originate in the U 5*f* electrons. [1] To study the electronic structure near the Fermi level ( $E_F$ ) and temperature dependence of the U 5*f* states, we have performed high-resolution angle-resolved photoemission spectroscopy (ARPES) on US. As a result, we have observed the highly-dispersive bands at the high-binding energy of 4 - 7 eV as well as several bands which approach  $E_F$  around  $\Gamma(X)$  and W point. From the comparison with the band structure calculation [2, 3], the former bands are ascribable to the occupied S 3*p* bands and the latter to the U 6*d* bands. We found that the overall dispersive feature in the experiment is qualitatively well reproduced by the calculation. In the vicinity of  $E_F$ , we found an anomalously sharp non-dispersive peak just below  $E_F$  with a broad shoulder structure at higher-binding-energy side. We assign these peaks as the U 5*f* states from the photon-energy dependent PES and the comparison with the calculation. Upon increasing temperature across  $T_C$ , the midpoint of leading edge of ARPES spectrum suddenly shifts toward  $E_F$ . Our results suggest that the magnetic-phase transition of US is described with the itinerant-ferromagnetism model.

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#### Co L emission spectra of LaCoO<sub>3</sub> at low temperatures

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A nonmagnetic to paramagnetic transition takes place in LaCoO<sub>3</sub> with increasing temperature. The magnetic susceptibility exhibits a broad peak at around 100 K. In the present study, the spinstate transition in LaCoO<sub>3</sub> is studied by the use of x-ray emission spectroscopy. Figure 1 shows the emission spectra of LaCoO<sub>3</sub> taken at the excitation photon energy of 782 eV corresponding to the Co  $L_3$  absorption peak. The emitted photons have been detected in the so-called depolarized configuration [1]. The main peak shifts toward higher energy by 1.5 eV upon heating from 35 to 110 K, whereas x-ray absorption and photoemission spectra do not show distinct change [2, 3]. The observed peak shift results from decreasing population of Co in the low-spin state by heating. According to the selection rule of resonant x-ray emission for the depolarized configuration [1], if the Co<sup>3+</sup> ion is totally symmetric, that is, in the low-spin state with filled  $t_{2g}$  band, the elastic and antibonding inelastic scatterings are forbidden contrary to other high- or intermediate-spin states. Figure 2 shows  $CoL_3$  emission spectra calculated within a full-multiplet  $CoO_6$  cluster model. It is assumed that the high-spin state is thermally excited, and the population is estimated by the magnetic susceptibility data [3]. The calculated spectra also exhibit the energy shift of the main peak similar to the experimental spectra. Even if the intermediate-spin state is thermally excited in place of the high-spin state, it is expected that the calculated spectra show similar peak shift.





Figure 1: Co  $L_3$  x-ray emission spectra of LaCoO<sub>3</sub> taken at different temperatures. The excitation photon energy is 782 eV. The emitted photons are detected in the depolarized configuration.

Figure 2: Co  $L_3$  x-ray emission spectra of LaCoO<sub>3</sub> calculated with the cluster model. The low- to high-spin state transition is assumed.

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## Magnetic Microspectroscopy by a Combination of XMCD and PEEM

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The Study of magnetic nanostructures has been attracting considerable attention as one of the most important fields in nanotechnology, as a result of the rapid advance of information technology. It has become necessary not only to observe magnetic domains and their response to applied magnetic fields but also to evaluate magnetic properties on a microscopic scale.

Element-selective imaging of magnetic domain structures has been enabled by combining X-ray magnetic circular dichroism (XMCD) for core-level absorption with microscopy methods such as photoelectron emission microscope (PEEM) or soft X-ray microscopy. In this presentation, we will demonstrate the powerfulness of XMCD microscopy by utilizing fully the spectroscopic aspect of XMCD. The resolution of spin and orbital magnetic moments based on the sum-rule of XMCD will be proved to be efficient in applying the method to studies of properties such as magnetic anisotropy and oscillatory exchange coupling. Experiments were performed by setting a photoelectron emission microscope at the twin-helical undulator beamline for soft X-ray spectroscopy BL25SU of SPring-8 in Japan[1-3].

Magnetism of fcc-Fe thin films show a wide variety of phenomena. We studied the magnetic coupling across Fe film in epitaxial crossed wedge 0-6 ML Ni/0-14 ML Fe/6 ML Co/Cu(001). PEEM image was taken at 121 energies in the 2p XAS region for both photon spins. By analyzing the spectrum of each pixel by XMCD-sum-rule, Fe spin and orbital magnetic moment distribution was obtained. In the region without Ni overlayer, three thickness ranges were observed with respect to the Fe 3d spin moment: 0-3.5 ML (~2.5  $\mu_B$ , phase I), 3.5-11 (~0.7  $\mu_B$ , phase II), and >11 ML Fe (~2.0  $\mu_B$ , phase III). In phase II, where Fe is expected to be ferromagnetic only at the interface and non-ferromagnetic in the bulk, it was found that the Ni overlayer and the Co substrate couple antiferromagnetically through the Fe layer in the Fe thickness range of ~3.5-7.5 ML. The mechanism for the antiferromagnetic coupling is expected to be the oscillatory exchange coupling through non-ferromagnetic interlayer.

Next, we focused on the spin reorientation transition in Co/Ni bilayer in order to discuss the mechanism in terms of the magnetic anisotropy energy. Here, we have prepared a doublewedged 0-4 ML Co/0-14 ML Ni/ Cu(001) sample. The distribution of the Fe spin moment indicated that a spin-reorientation occurs as functions of both Ni and Co thicknesses. Furthermore, the orbital magnetic moment was shown to be larger in the out-of-plane region. This is consistent with the perpendicular anisotropy of the Ni film alone.

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# O AND N K-EDGE X-RAY MAGNETIC CIRCULAR DICHROISM OF CO AND NO ADSORBED ON MAGNETIC THIN FILMS

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Adsorption of molecules is known to influence strongly the magnetic properties of thin films: suppression and enhancement of magnetization, rotation of magnetic easy axis and so forth. In order to understand the effects of adsorption, it is essentially important to investigate the magnetization not only of magnetic substrates but also of adsorbed atoms and molecules. The x-ray magnetic circular dichroism (XMCD) technique is potentially useful to measure weak signals from adsorbates because of its element-specific character. We have measured O and N *K*-edge XMCD of CO and NO adsorbed on ultrathin Ni and Co films on Cu(001).

All the experiments were carried out at bending-magnet Beamline 11A ( $P_c$ =0.60–0.75 depending on the beam time) in Photon Factory. Evaporated Ni and Co films were grown on clean Cu(001) at room temperature by monitoring the oscillations of reflection high energy electron diffraction. The films were exposed to saturated amounts of CO and NO at 200 K. We have examined the systems of CO/Co(//) [1], CO/Co/Ni( $\perp$ ), CO/6ML-Ni(//) [2], CO/10ML-Ni( $\perp$ ) [2], CO/thick-Ni(//) [2], NO/Co(//) and NO/Co/Ni( $\perp$ ), where // and  $\perp$  denotes the in-plane and perpendicular magnetization, respectively. O and N *K*-edge XMCD spectra were taken by means of the partial electron yield mode and by reversing the magnetization directions.

We have observed systematical features of the O *K*-edge XMCD of CO adsorbed Ni and Co films. There exist two resonances in O *K*-edge absorption spectra of CO, which are ascribed to  $\pi^*$  and  $\sigma^*$ . All the XMCD spectra exhibit meaningful signals at the  $\pi^*$  resonance while do not at the  $\sigma^*$  one. This implies significant hybridization between CO  $2\pi^*$  and metal 3*d* bands. The XMCD signals at the  $\pi^*$  resonance provide negative signs for perpendicularly magnetized films of 10 ML Ni and 3ML-Co/Ni ("negative" means  $\mu^{\uparrow\uparrow}(\pi^*)-\mu^{\uparrow\downarrow}(\pi^*)<0$ ), while the signs are positive for all the in-plane magnetized films. When we apply the *K*-edge sum rule to the observed findings, we can remark that the perpendicular magnetization induces the O orbital moment parallel to the substrate magnetization while the in-plane magnetization leads to the antiparallel O orbital moment. The N and O K-edge XMCD spectra of NO on the Co(//) and Co( $\perp$ ) films show similarities and dissimilarities to the CO case. The XMCD signs were found to be negative for all the spectra, implying that the orbital moments induced at the N and O atoms are always parallel to the Co magnetization, irrespective of the magnetization directions.

Possible qualitative interpretations for the observed *K*-edge XMCD features will be presented within the framework of the local chemical bonding picture.

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## Bandstructure and Correlation Effects in Gd

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Gd is a prototypical Heisenberg ferromagnet with large magnetic moments due to the highly localized 4f subshell. While the 4f-electrons maintain an atomic character in the solid state, the (5d6s)-valence electrons form the metallic bonding and mediate the indirect exchange interaction (RKKY-type) among the 4f moments. Despite the importance of the (5d6s)-valence states, their band structure is yet not well understood.

The width of the (5d6s)-bands derived from photoemission experiments [1] is significantly smaller (by more than a factor 2 for the  $\Delta_2$  states) than that predicted by the stateof-the-art calculations [2]. This discrepancy has been attributed to band narrowing due to strong correlation effects. Notably, such band narrowing in Gd appears to be much larger than in the strongly correlated 3d transition metals [3], in contrast with the comparatively delocalized character of the (5d6s)-states, an unusual fact that has until now no explanation.

We investigated by spin and angle resolved photoemission the Gd band structure in order to address the apparent inconsistency between the theoretical and experimental description. Spin resolved spectra reveal an evolution of the spectral line shape as a function of the photon energy that can not be visualized in the spin-integrated measurements. The weak dispersion and the unusual broadening of the spin-integrated (5d6s)-features is found to be due to superposition of strongly dispersive and non-dispersive components in each of the two spin-resolved photoemission channels. The presence of non-dispersive components is explained by strong momentum broadening [4], due to the extremely short photoelectron lifetime in Gd. Consideration of the photoelectron lifetime convoluted to the LSDA bandstructure [5] results account well for the observed spectral evolution. This study thus resolves the long standing controversy between theory and experiment on the electronic band structure of rare earths and significantly reassess the role of correlation effects in these systems.

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# Dichroism in Angular Resolved Photoemission from Paramagnetic Materials: Pt(111)

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We report on the dichroic photoemission from paramagnetic platinum excited by linearly and circularly polarized light. We measured the difference in the angle-resolved photoemission intensities from the Pt(111) valence band and 4f core level excited by photons of opposite polarization. These differences are usually termed as dichroism in the angular distribution of photoelectrons.

For the case of VUV photoemission, we will show the dependence of the dichroism on details of the electronic band structure. The measurements are compared with relativistic single step photoemission calculations. In particular, the influences of hybridization on the observed circular dichroism will be discussed.

For the case of soft X-ray excitation the influence of the dichroism on other photoemission quantities like the spin-orbit branching ratio will be discussed. We will present first results from a *full relativistic single step core-level* photoemission model. This model will be compared to three step cluster calculations most oftenly used to describe diffraction effects in core level photoemission from solids and surfaces.

One usually distinguishes between LDAD and CDAD depending on the type of polarization of the applied photons (linear or circular). The otical properties of a particular material play an important role for VUV photoemission. It is shown how the metal optics influences all kinds of dichroism in VUV photoemission by comparing the excitation with linearly and circularly polarized photons. We show that it will be generally impossible to measure a pure dichroism of one or the other type.

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# AN EXPERIMENTAL PROOF OF THE BACK-SCATTERING MODEL FOR DICHROIC EFFECTS IN VUV-PHOTOEMISSION

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We investigated the energy dependence of the circular dichroism in the angular distribution of photoelectrons (CDAD) from the shallow Cs-5p core-levels. Cesium was prepared in a hexagonal ordered monolayer on tungsten.

The results are mainly influenced by scattering of the photoelectrons within the adlayer. We are able to describe the observed behavior by means of a known model that treats only the back-scattering of photoelectrons at the potential step between the adlayer and the substrate [1]. It leads to a simple method determining the height of the monolayer above the substrate.

In this approach, the back-scattering model is matched with the measured energy dependence of the CDAD. This procedure yields results already for a single off-normal angle of the photoelectron detection. We determined the distance between cesium and tungsten to be  $2.2\pm0.1$ Å (see Fig.1). It should be mentioned that this corresponds to the effective distance between the center of the adsorbed layer and the potential step between the layer and the substrate. Finally, the fitted result was proven to meet the complete angular dependence of the CDAD at different photon energies.

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Figure 1: Experiment and theory for the energy dependence of the CDAD using different distances between adsorbate and substrate.

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# MAGNETIC COUPLING OF ALKALI AND RARE-GAS FILMS ADSORBED ON A FERROMAGNETIC SURFACE

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We report on the observation of magnetic circular dichroism in the angular distribution (MCDAD) from alkali and rare-gas films adsorbed on a thin remanently magnetised Co layer. The measurements give evidence for an induced magnetic moment of the adsorbate from the ferromagnetic substrate. We observed for Co(0001) - Cs 5p - semi core level an antisysmmetric MCDAD near normal emission (NE) as shown in Fig. 1. The reversed magnetisation leads to a mirror image like distribution. The second system under investigation, physisorbed Co(0001) - Xe 5p, does not give such an obvious asymmetric result, but shows significant differences in the MCDAD signal for both directions of magnetisation. A magnetic coupling between Xe and Co is proved by comparison to spin resolved photoemission data from Ref. [1].

The measurements were obtained using the 6.5 m normal incidence monochromator NIM at Bessy I. The results will be compared to calculations using a spin dependent three step photoemission model [2].



Figure 1: Angular distribution of the near NE-MCD for magnetisation parallel and antiparallel to the projection of the photon propagation on the surface. The photon energy was 22 eV with  $\alpha = 45^{\circ}$ .

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# SPIN ARRANGEMENT OF THE Mn / Fe(001) SYSTEM INVESTIGATED BY THE SPIN POLARIZED PHOTOELECTRON DIFFRACTION

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Recently magnetic properties of ultra thin films on magnetic substrates, particularly the Mn/Fe(001) system have been widely investigated both experimentally and theoretically[1]. In spite of such efforts the magnetic coupling between Mn and Fe interface or between Mn itself in this system, however, is the controversial problem. In order to help understanding of this problem, we applied the spin-polarized photoelectron diffraction method to Mn(1ML)/Fe(001) thin epitaxial film which was grown on the Ag(001) substrate.

All the experiments have been done at the undulator beamline BL-19A of Photon Factory (KEK, Tsukuba). Fe(001) thin film(~12ML) was grown at room temperature(R.T.) on the Ag (001) single crystal substrate which is cleaned by usual procedure. The cleanliness of the Ag(001) substrate and the epitaxial growth of Fe film were checked by the low energy electron diffraction and Auger electron spectroscopy. 1ML or less Mn was deposited on to the Fe(001) thin film and the sample was magnetized along [010] direction. Linearly polarized light of hv = 127 eV was injected to the sample along surface normal direction and the emitted photoelectrons are detected along the [010] direction by the polar angle scan with the electron analyzer equipped with small Mott-type spin detector[2].

Figure shows the spin and angle resolved photoemission spectra (upper part) and the polarization (lower part) at  $\theta = 22.5^{\circ}$ . In this angle, the sign of the polarization is the same between Mn and Fe 3p core levels. Comparing the angular dependence of the polarization of each component with that of calculated for several models  $(p(1\times 1) \text{ ferro}(F), p(1\times 1) \text{ anti}$ ferro (AF),  $c(2\times 2)$ ,  $p(2\times 2)(F)$ , and  $p(2\times 2)(AF)$ ; our experimental see ref.[1]), angular dependence of the polarization is in qualitatively agreement with the calculation for the model having  $c(2\times 2)$  spin-arrangement of Mn atoms.



Figure. Spin polarized spectra (upper part) and the polarization (lower part) at  $\theta = 22.5^{\circ}$ measured at hy = 127 eV.

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# *In situ* photoemission spectroscopy of localized Mn 3d states in (Ga,Mn)As and nanoscale MnAs dots

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We discuss the electronic structure of the Mn 3d states in the ferromagnetic diluted magnetic semiconductor (DMS) (Ga,Mn)As and nanoscale MnAs dots (10nm size) using an *in situ* photoemission system with molecular beam epitaxy (MBE) at BL-1C, Photon Factory, KEK, Japan. Both (Ga,Mn)As and nanoscale MnAs dots have a high potential for new device applications using giant magnetoresistatce [1]. Correlation between the Mn 3d electronic structure and the local environment of the Mn atoms gives us a key to reveal the microscopic mechanism of "carrier-induced ferromagnetism" in (Ga,Mn)As and the size effect in the MnAs dots.

The Mn 3d partial density of states (PDOS) were deduced using the  $3p\rightarrow 3d$  resonant photoemission technique. As shown in Fig. 1, the Mn 3d PDOS is deduced by the subtruction between the 50 eV and 48 eV spectra. The PDOS in *in situ* prepared (Ga,Mn)As is similar to the previous reported spectra [2]. The nanoscale MnAs dots also shows spectra similar to (Ga,Mn)As, which is quite different from metallic MnAs film spectra as shown in right pannel of Fig. 1. The results indicate that the local environment of Mn strongly affects the localized and delocalized character of the Mn 3d states.

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Figure 1. Valence-band spectra of (Ga,Mn)As (left), MnAs dots (middle), and MnAs film (right).

# MAGNETIC CIRCULAR DICHROISM OF CORE ABSORPTION IN Fe-Pt AND Fe-Pd ALLOYS

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A magnetic circular dichroism (MCD) study of Fe 2p core level x-ray absorption (XAS) has been performed for FePt, Fe<sub>3</sub>Pt, FePd and FePd<sub>3</sub> ordered alloys. The FePt ordered alloy has high uniaxial anisotropy related to the tetragonal CuAu I-type structure and then thin films of FePt are attracting much interest as high-density recording media with perpendicular magnetization. FePd also has CuAu I-type structure, but its uniaxial anisotropy constant is smaller than that of FePt [1]. Therefore it is interesting to compare the contributions of orbital magnetic moment ( $m_o$ ) between FePt and FePd because the spin-orbit coupling could be a reason for magnetic anisotropy.

The experiment has been done at the second experimental station of the soft x-ray beamline BL25SU at SPring-8. The light source of BL25SU is the Twin Helical Undulator [2], which provides high brilliant SR with high degree of circular polarization (>99 %). The grazing incidence monochromator employs varied-line-spacing plane gratings and covers an energy range from 220 to 2000 eV [3]. The samples were scraped with a diamond file in the UHV chamber just before the measurements. The total electron yield method was employed for the XAS measurement. The XAS intensities were measured at each photon energy for both magnetization directions (1.4 T) in the Faraday geometry. The difference of XAS provides the MCD.

From the results of experiment, the ratios of A/B, where A(B) represents the integral value of the MCD in the  $2p_{3/2}(2p_{1/2})$  region, have been estimated to be -1.1, -1.1 and -1.2 for Fe metal, Fe<sub>3</sub>Pt and FePt, respectively. In the case of Fe-Pd alloys, the estimated A/B ratios are -1.1 and -1.2 for FePd and FePd<sub>3</sub>. This result means that the contribution of m<sub>o</sub> to the total magnetic moment at the Fe site of FePt is larger than that of FePd and comparable with that of FePd<sub>3</sub> [4,5].

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# DIRECT EVIDENCE FOR AN ITINERANT MAGNETITE ABOVE AND BELOW THE VERWEY TRANSITION TEMPERATURE

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Magnetite (Fe<sub>3</sub>O<sub>4</sub>) is a mix-valence compound that exhibits the so-called Verwey transition at  $T_V \sim 120$  K, in which the electric conductivity abruptly decreases by two orders of magnitude on cooling through  $T_V$ . It has been a general belief that the transition is caused by an ordering of the Fe<sup>2+</sup> and Fe<sup>3+</sup> ions at the octahedral *B* sites of the inverse spinel lattice due to electron and/or electron-phonon correlation effects. [1] Recently, however, new evidence has been reported by diffraction anomalous fine structure experiments [2] that shows no charge ordering of the *B* site ions below  $T_V$ , and that the Fe atoms are electronically equivalent in a time scale lower than 10<sup>-16</sup> sec above  $T_V$ , which indicates the itinerant nature in the conduction of magnetite.



Figure 1: Difference spectra between the majorityspin and minority-spin partial density of states (DOS) taken at hv from 25 to 50eV at 100K, along the surface normal [111] direction.

Here we report a temperature-dependent, spin- and angle-resolved photoemission study of the valence electronic structure near  $E_F$  of a well-characterized, epitaxially grown Fe<sub>3</sub>O<sub>4</sub> film on Pt(111). The majority- and minorityspin partial density of states (DOS) has been obtained at 300 and 100 K, well above and below  $T_V$ . The spectra still show substantial kdependence at 100 K in the 3d region (Figure 1). This provides the most direct experimental evidence that the 3d electrons of the B sites in magnetite are itinerant even at  $T < T_V$ . Magnetite should therefore be considered as an itinerant magnet above and below  $T_V$ . Here we note that being itinerant means that the electrons are not confined to any particular ion in a time scale much faster than the hoping process ( $\sim 10^{-10}$  sec), and that the dispersion observed in photoemission provides the most direct proof as the photoemission process generally takes place within  $10^{-15}$  sec.

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# Transverse Magneto-optical Kerr-effect in the soft X-ray regime at ultrathin metals films and islands on W(110)

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Tunable linearly and circularly soft X-ray radiation opens the possibility for powerful methods in investigating magnetic thin films, islands, and nanoparticles on surfaces, e.g., magnetic dichroism in angle resolved photoemission (MDAD), X-ray magnetic circular dichroism in photoemission (XMCD) and in-situ Magneto-optics at core levels.

Here, we will focus on new results from recent measurements using the transverse Magneto-optical Kerr-effect (T-MOKE) at in-situ prepared iron and cobalt films and islands on W(110). The measurements have been carried out at the U49 undulator beamline at BESSY II using linearly polarized radiation. For recording hysteresis curves we have used an external electromagnet (B<0.5T). Close to the Fe and Co 2p core levels the reflectivity and the Kerr rotation is strongly enhanced by resonant forward scattering, cf. Fig. 1. We could observe huge intensities in the specular reflection although the reflectivity is usually small in the soft X-ray regime. Moreover, we have detected intensity differences up to 50% e.g., at iron films with less than 5ML. The experimental data will be compared to calculations from Oppeneer and coworkers.



Fig. 1: T-MOKE spectra of bcc(110) Fe films taken with p-linearly polarized radiation at the Fe 2p levels for opposite magnetization directions. The solid line (with full circles, lower part) denotes the intensity difference.

When annealing epitaxially grown Fe(110) films on W(110) above 500°C a well oriented Fe island structure can be created. We have analyzed the rotation of the easy magnetization axis of such self-organized islands with respect to flat bcc(110) iron films, where the surface anisotropy determines the remanent magnetization along W[110]. Our experimental data clearly show a remanent magnetization parallel to W[001] depending on the Fe coverage before annealing and on the temperature during annealing. Moreover, we have investigated the magnetic properties of cobalt films on a clean W(110) single crystal and two adsorbate-induced reconstructions which lead to different growth processes of the cobalt overlayer.

# **RESONANT MAGNETIC SCATTERING IN GMR MULTILAYERS**

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The origin of giant magneto-resistance (GMR) in metallic multilayers is frequently related to coupling between adjacent magnetic layers and to spin dependent scattering in the electron transport process.

By simultaneous measurements of sample resistance and of x-ray resonant magnetic scattering (XRMS), we tried to establish a direct correlation between GMR and antiferromagnetic (AF) order.

The sample was a  $(Co_{11}\text{\AA}/Cu_{21}\text{\AA})_{20}$  multilayer sputter-deposited on Si(111). The experiment was performed on beamline 6.3.2 (soft x-ray metrology) of the storage ring ALS (Berkeley).

Field dependent scattered intensity has been measured in a geometrical configuration corresponding to the Bragg peak coming from the AF coupling (order  $\frac{1}{2}$ ), at a photon energy close to Co L<sub>3</sub> absorption edge (hv = 776.5 eV). At the same time, the resistance of the sample was measured in a two wire mode. The applied field was varied over loops of increasing amplitude, always starting from as-prepared samples. This last condition is of particular importance, since we observed strong and irreversible changes upon magnetic cycling the sample.

The comparison between scattered intensity and resistance values as function of **H** clearly shows a direct relation between antiferromagnetic order and magneto resistance.

# Two-band magneto-optical elements for soft x-ray polarization analysis at the Fe, Co 2p and Gd 3d edge

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The magneto-optical effects resonantly enhanced in the vicinity of an absorption edge can be utilized for a quantitative determination of the polarization state of synchrotron radiation (SR) at the 2p edges of the transition metals (TM) [1] and the 3d edges of the rare-earth elements (RE) in the energy range from 700eV to 1500eV.

Gd/Fe and Gd/Co multilayer structures with d-spacing in the order of 1nm were prepared by alternating deposition of Fe (or Co) and Gd using the triode sputtering system at Sincrotrone Trieste. The techniques of magnetic circular dichroism (MCD) and x-ray resonant magnetic scattering (XRMS) have been used to investigate the magneto-optical properties of the Gd/TM multilayers in the regions of the 3d edge of Gd and the 2p edge of TM at room temperature. The MCD effect was measured as a function of the degree of circular polarization in order to calibrate the magneto-optical response from the multilayers with regard to the polarization state of incident light.

A significant dichroic signal from magnetically saturated samples was observed in specular reflection measured at grazing incidence in the geometry of longitudinal magneto-optical Kerr effect (L-MOKE). The MCD signal is derived from the reflectivity curves as an asymmetry ratio  $A = (R^+ - R^-)/(R^+ + R^-)$ , where  $R^+$  and  $R^-$  are the intensities of the reflected light detected by reversing the in-plane magnetization of the sample.

The variation of the degree of circular polarization results in corresponding modulation of both spectral and angular dependent MCD asymmetry ratio [2]. The analysis of experimental data obtained at both Gd 3d and TM 2p edges shows that the degree of circular polarization does not modify the shape of the MCD spectra and the magnitude of the MCD asymmetry signal is proportional to  $P_{circ}$ .

These measurements confirm the possibility to use the multilayert structures Gd/TM after calibration as two-band magneto-optical elements for polarimetry purposes in the energy regions close to the 3d edge of Gd and the 2p edge of TM.

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# **MAGNETISM OF 1D COBALT ATOMIC WIRES**

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We present the first investigation of the magnetic properties of 1D structures in the monatomic limit. Periodic arrays of uniformly-spaced Co monatomic wires have been obtained by controlled step decoration of the vicinal Pt(997) surface [1-3], as shown in Fig. 1. X-ray magnetic dichroism (XMCD) measurements show that the Co wires are superparamagnetic with a blocking temperature of about 15 K. Magnetic ordering in 1D structures is thus demonstrated despite contrasting theoretical predictions [4,5].



Figure 1: STM image of monatomic Co wires on Pt(997). The inset shows the wire periodic structure. Figure 2: XMCD spectra at the  $L_{II}$ ,  $L_{III}$  Co edges for monatomic wires and one monolayer on Pt(997).

The XMCD spectra of monatomic wires and one monolayer Co on Pt(997) reported in

Fig. 2 reveal strong differences in the electronic structure of such systems due to their reduced dimensions. The high values of the orbital to spin magnetic moment ratio and of the magnetic anisotropy energy reduce considerably as the system becomes 2D.

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## **EFFECT OF OXYGEN ON COBALT FILMS ON PALLADIUM (111)**

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Physical properties of Co films on nonmagnetic metallic substrates such as Pd and Pt have been studied very extensively because they show perpendicular magnetic anisotropy [1]. But for practical application situations, gas such as oxygen may adsorbe at the surface, which can affect the electronic structure and magnetic properties. We have studied the changes of electronic and magnetic properties of Co films on Pd(111) single crystal substrate after oxygen exposure and annealing using X-ray photoemission spectroscopy (XPS) and surface magneto-optical Kerr effect (SMOKE), *in situ*. When Co is exposed to  $\sim$  300 L oxygen, Co reacts with oxygen and makes the form of CoO. This is consistent with previous results for bulk Co [2]. CoO is known to be antiferromagnetic, and as expected, the oxidized films show no ferromagnetism. When the oxidized film is annealed at temperatures above 700 K, the oxygen is decomposed from CoO and metallic Co reappears. Hence in annealed film, ferromagnetism reappears and again prefers perpendicular easy axis. The oxygen decomposition occurs more rapidly at higher temperature and thinner films. Alloying near the interface may occur for thicker films. We demonstrate the changes of magnetic properties (Figure 1(a)) and Co 2p XPS spectra (Figure 1(b)).



Figure 1: (a) SMOKE intensities of 5.6 ML Co film (b) XPS spectra of Co 2p and oxygen Auger of 6.1 ML Co film after oxygen exposure and subsequent annealing.

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# Induced magnetic profile from a Fe/V superlattice probed by soft X-ray resonant magnetic scattering

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The element selectivity and bulk sensitivity of X-ray magnetic scattering (XRMS) has been used to investigate the induced magnetism of the 3d electronic states of vanadium across the individual atomic layers in a Fe/V multilayer. Reflectivity measurements were performed using the precision reflectometer at beamline 6.3.2 at the Advanced Light Source (ALS) in Berkeley, USA [1]. This is a bending magnet beamline dedicated to EUV and soft X-ray reflectometry and scattering. The measurements were made as a function of photon energy around the Fe and V  $L_{23}$  thresholds at different scattering angles from the multilayer [2]. The structural parameters were obtained by a refinement to the reflectivity spectra and X-ray diffraction data [3]. This makes it possible to determine the composition profile of the Fe and V sublayers as well as the interfacial surface roughness. The induced magnetic moments of the individual atomic V layers are obtained by scaling the magnetic moments of each monolayer to best fit the total extracted spectral asymmetry ratios obtained from the two directions of the applied magnetic field. The description of the induced magnetic polarization in vanadium is found to heavily rely on the simulated average interface roughness within the multilayer period [4]. The distribution of the Fe 3d magnetic moments is found to be almost uniform across the Fe layers. As expected the V 3d polarization is strongest at the interfaces with Fe and a quantitative assessment is made.

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# EPITAXY OF ULTRATHIN NIO AND COO ULTRATHIN FILMS STUDIED BY PED AND GIXRD

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Ultrathin transition metal oxide films grown on metals can show peculiar electronic, magnetic and structural properties, very different from those of their bulk phases [1]. In fact Madelung potential can be lower at the surface and hybridization of electronic states at the interface can come into play. For these reasons these systems have been an outstanding subject of investigation in the field of highly correlated materials, in the field of catalysis and in microelectronics technology in recent years.

We performed synchrotron radiation based structural studies of NiO/Ag(001) and CoO/Fe(001) systems at ALOISA beamline, at ELETTRA synchrotron radiation source. The substrates were chosen to ensure a good lattice matching to reduce the number of dislocation and defects. The characterization included grazing incidence X-ray diffraction measurements for inplane structure determination, combined with photoelectron diffraction giving the vertical lattice parameter of the films. In the case of the NiO/Ag(001) system specular X-ray reflectivity data allowed an accurate thickness determination and gave information on the roughness of the interface and of the film itself. NiO films in the 5-50 ML thickness range were grown in situ. The films showed a rock-salt structure, in registry with the substrate and no significant strain was found in this thickness range. CoO films were also grown in situ in the 2-10 ML thickness range. They were found to have a rocksalt structure rotated by 45 degrees with respect to the Fe substrate unit cell. In this case in the thinner films a significant strain was found, gradually releasing as the thickness increases.

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# Direct Determination of an Antiferromagnetic Surface Spin Structure by (Soft) X-Ray Magnetic Linear Dichroism

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Magnetic multilayer structures exhibit fascinating scientific effects and have important applications in the high-tech industry. An important class of magnetic multilayers contains antiferromagnetic thin films, which are used to pin the magnetization direction of an adjacent ferromagnetic layer, thereby defining a unique magnetization direction. This pinning effect, which is referred to as 'exchange bias', has been discovered more than 45 years ago and although it is of great technological importance, the origin of the exchange bias effect is despite active research still unknown. One of the obstacles preventing a better understanding of its origin is the lack of sensitivity of conventional techniques to address the surface and interface magnetic properties of thin antiferromagnetic films. On the other hand, it is clear that the surface/interface structure should play the key role in the exchange bias effect. Lacking experimental information most common exchange bias models have assumed a bulk like spin structure at the interface, i.e., the possibility that the magnetic structure of the thin film surface might differ from the known antiferromagnetic bulk structure is generally ignored.

We have studied the antiferromagnetic structure in the surface region of structurally wellcharacterized antiferromagnetic LaFeO<sub>3</sub> films. These some ten nm thin films were grown on SrTiO<sub>3</sub> (110) and SrTiO<sub>3</sub> (100) substrates. Plan-view electron-diffraction and conventional TEM have been used to resolve the crystallography. The magnetic spin structure has been investigated via the x-ray magnetic linear dichroism (XLMD) effect. This gives rise to a polarization dependence of the absorption coefficient on the orientation of the antiferromagnetic axis relative to the electric field vector of linearly polarized x-rays. From the experimentally observed polarization dependence one can directly conclude that the magnetic structure in the surface region of the thin films differs significantly from the magnetic structure of LaFeO<sub>3</sub> bulk. In particular, for LaFeO<sub>3</sub> on SrTiO<sub>3</sub> (110) we find that the antiferromagnetic axis is rotated from the in-plane bulk direction into an orthogonal direction pointing out of the film surface. After correcting the data for the non-linear polarization component and for experimental saturation effects, we can quantitatively derive the direction of the antiferromagnetic axis. For both substrate orientations we find the rotated antiferromagnetic axis to lie in an SrTiO<sub>3</sub> (111) plane.

# FARADAY AND MAGNETIC-KERR ROTATION MEASUREMENTS ON Co FILMS AROUND M, 2 EDGES

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In the extreme ultraviolet (EUV) region, magneto-optical effects have been investigated mainly by MCD measurements with the circularly polarized light. On the other hand, magnetic rotation measurements with the linearly polarized light are also promising if adequate polarizers can be developed. Therefore, we developed multilayer polarizers, so that we have carried out Faraday rotation measurements in the 50-70 eV region [1] and recently we began magnetic Kerr rotation measurements.

In this study, Faraday and Kerr rotation measurements were performed on Co films around  $M_{23}$  edges at UVSOR Facility, at room temperature (RT) using Al/YB<sub>6</sub> multilayer polarizers in a magnetic field of 0.82 T generated by a permanent magnetic circuit. The longitudinal Kerr rotation measurement was performed using the s-polarization configuration. The results of the Kerr rotation measurement for an angle of incidence 65° are plotted by closed circles in Fig. 1. In the figure, the Kerr rotation spectra calculated from (i) the present Faraday rotation spectrum (0.82 T, RT) and (ii) the MCD spectrum (1.2 T, 140 K) obtained by the total photoelectron yield

3.5

method [2], using equations derived by Zak et al. [3], are also shown by dashed and solid lines, respectively. The maximum Kerr rotation angle of the present result is larger than the calculated one from (i). The main reason for the difference may be due to the difference of the magnetization Kerr between the longitudinal (saturated) and Faraday (unsaturated) configurations under applied magnetic field of 0.82 T. It is consistent with the fact that the present result is almost the same as the calculated one from (ii) in which magnetization may be saturated (1.2 T).

#### Longitudinal Kerr Rotation Angle(deg) Co 100nm/Si wafer 3.0 $(\theta = 65^{\circ}, \text{ s-pol.})$ 2.5 Exp. (0.82T,RT) Cal. from Faraday 2.0 rotation (0.82T,RT) Cal. from MCD 1.5 (1.2T,140K) 1.0 0.5 0.0 -0.5 -1.0 54 55 56 58 59 53 57 60 61 62 63 64 65 Photon Energy(eV)

Figure 1: Longitudinal Kerr rotation angle spectra on Co 100nm/Si wafer around Co  $M_{23}$  edges.

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# MEASURING THE ORBITAL MOMENT OF THE ANTIFERROMAGNET COO WITH SPIN-RESOLVED PHOTOEMISSION

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The measurement of the separate spin and orbital contributions to the magnetic moments of transition metal materials is experimentally and theoretically challenging. Besides the sum rules for magnetic soft x-ray absorption dichroism (for ferromagnetic materials) and magnetic x-ray scattering (for magnetically ordered antiferromagnetic systems like NiO [1]), we have explored the feasibility of another spectroscopic technique potentially applicable to antiferromagnets and ferromagnets. Spin-resolved photoemission using circularly polarised soft x-rays can provide quantitative information on the magnetic moments both in ordered and disordered systems.

By measuring the spin-resolved spectra of the valence states, and by integrating over the whole valence states we can determine the total difference between photoelectrons whose spin is parallel or antiparallel to the photon angular momentum. We call  $\Pi$  the ratio between this integrated difference and the integrated valence band photoemission intensity. We take advantage of the sum rule derived by van der Laan and Thole [2], which relates  $\Pi$  to the expectation value of a spin-orbit operator of the system in the initial state.

We have tested the technique for CoO at beam line ID12B of the ESRF. CoO is an antiferromagnetic insulator with an intriguing magnetic structure. The sample was a single crystal kept at T=390K, above  $T_{N\acute{e}el}$ . The ratio of the integrals of the curves measured with parallel and with antiparallel orientations of the photon angular momentum and photoelectron spin converges (going from the Fermi level  $E_F$  towards higher binding energies) to the quantity  $\Pi$ = 0.045±0.005. Applying the sum rule appropriate for the isotropic cases we can thus get a rough estimate of  $\langle L_z \rangle / \langle S_z \rangle \approx 0.84$  at 390K, directly from the measurements.

In order to extract further numbers, as far as local properties are concerned, we have performed model calculations using a CoQ cluster in  $O_h$  symmetry [3], which reproduces the experimental value at T=390K. By combining the calculation results with the known total magnetic moment of  $3.81\mu_B$  (at 0K) we can easily derive  $4_z \ge 1.31h$  and  $4S_z \ge 1.25h$ .

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## Quadrupolar transitions evidenced by resonant Auger spectroscopy

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The hamiltonian describing the interaction between a photon and matter can be written to second order as a sum of electric dipolar and quadrupolar terms [1]. If most of the simple absorption experiments can be interpreted within the electric dipolar approximation, the introduction of quadrupolar transitions is necessary for the interpretation of magnetic effects in absorption measurements. For instance, the magnetism of selected orbitals can be probed by X-ray Magnetic Circular Dichroism; for experiments at the L<sub>2,3</sub> edges of rare earths, quadrupolar transitions from the 2p levels towards the 4f orbitals (which generally carry most of the magnetic moment in rare earths compounds) give a signal of the same order of magnitude as the one due to the dipolar 2p $\rightarrow$ 5d transitions [2]. In Resonant X-ray Magnetic Scattering, the signal due to quadrupolar transitions is predominant at the L<sub>3</sub>-edge of rare earths (2p $\rightarrow$ 4f) [3] or at the K-edge of transition metals (1s $\rightarrow$ 3d) [4]. For a better understanding of the experimental data obtained from these techniques, it is of key importance to be able to bring to the fore the occurrence of quadrupolar transitions in absorption spectra, as well as to quantify their intensities.

From absorption spectra, quadrupolar transitions can only be studied by angular-dependent measurements [5]. Resonant spectroscopies offer a new opportunity to get more insight into excited states of atom by studying lineshapes and intensities of decay processes. We show that resonant Auger spectra carry a clear signature of an additional electron promoted in localized empty states via a quadrupolar transition. In our measurements on  $TiO_2$ , we were able to determine the relative weight of quadrupolar transitions at the Ti K-edge, as well as the symetries of the orbitals reached by the photoexcited electron.

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# MAGNETIC PROPERTIES OF NANOMETRIC MAGNETITE LAYERS : A Resonant Magnetic Scattering Study

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Large magnetoresistance effects in tunnel junctions are expected when using halfmetallic ferromagnets as the electrodes of the junction. Half-metallic ferromagnets are metallic for one spin direction and insulating for the other. This means that the Fermi level is fully polarised. This property is highly dependent on the magnetic ordering in the material.

Fe<sub>3</sub>O<sub>4</sub> is theoretically predicted to be a half-metal ferromagnet, and it is in principle very attractive for applications in magneto-electronics for its high Curie temperature ( $T_C$ = 850 K). However, there are several reports of the anomalous magnetic behavior of Fe<sub>3</sub>O<sub>4</sub> in the thin layer form [1,2]. In this framework, we investigated the local magnetic properties of Fe<sub>3</sub>O<sub>4</sub>(111) layers with thicknesses from 0.5 to 25 nm, by means of resonant scattering of polarized soft x-rays (ALS synchrotron facility, Berkeley). Reflectivity spectra recorded at the Fe 2p resonance are analysed within the framework of ligand field atomic multiplet calculations.

The magnetic properties of Fe<sub>3</sub>O<sub>4</sub> are explained by a collinear antiferromagnetic ordering of spins in the tetrahedral (A) and octahedral (B) sites, which are in the high spin configuration. The Fe<sup>3+</sup> ions with  $5\mu_B$  are located in the A sites as well as on half of the B sites. The remaining half of the B sites is occupied by the Fe<sup>2+</sup> ions with  $4\mu_B$  of magnetic moment. Thus the total moment of Fe<sub>3</sub>O<sub>4</sub> is  $4\mu_B$  per formula unit. The half-metallic character is due to the presence of solely minority spin electrons from the Fe<sup>2+</sup>-B ions above the Fermi level.

In high energy spectroscopy, we can analyse separately the properties of the three different iron sites (Fe<sup>3+</sup>-A, Fe<sup>3+</sup>-B and Fe<sup>2+</sup>-B) present in Fe<sub>3</sub>O<sub>4</sub>, because their contributions are shifted by crystal field effects. For thicknesses larger than ~8 nm, the layers exhibit the magnetic properties of bulk Fe<sub>3</sub>O<sub>4</sub> described before. In contrast, the 0.5 nm-thick layer present specific magnetic properties. The main difference with respect to standard Fe<sub>3</sub>O<sub>4</sub> is the fact that the Fe<sup>2+</sup> ions are in the low spin configuration. The consequence is that the Fe<sup>2+</sup> ions become non-magnetic and that the half metallic character vanishes. This result can be linked to the presence of a magnetically "dead" layer evoked by several authors to explain the anomalous magnetic behavior of Fe<sub>3</sub>O<sub>4</sub> thin films [2]. We also find that the local exchange field between the Fe<sup>3+</sup> at the A and B sites is significantly reduced in comparison to bulk.

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# X-Ray Magnetic Circular Dichroism Study of Size Selected Iron Clusters

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Soft X-ray magnetic circular dichroism (XMCD) is a valuable tool to investigate magnetic properties because of its element specificity and the possibility to determine spin and orbit magnetic moments separately /1/. At third generation synchrotron sources, this technique can even be used to study highly diluted systems such as sub-monolayer coverages of clusters on a substrate. These supported nanoclusters have recently received considerable interest for their potential future application in high-density information storage media or as novel catalysts.

Magnetic properties of size selected transition metal clusters have so far been studied in Stern-Gerlach type experiments on free cluster beams, where magnetic moments have been observed which are considerably larger than the respective bulk values /2/. For potential applications, however, these clusters have to be supported on substrate materials, which may substantially alter their properties.

In recent experiments at BESSY II (Berlin) and ELETTRA (Trieste), we have used XMCD at the Fe  $L_{2,3}$  edges to study the magnetic behaviour of small, deposited Fe<sub>n</sub> clusters



(n=2-9). Small Fe<sub>n</sub> clusters generated by a sputter source were size selected in а magnetic dipole field and deposited under "soft-landing" conditions onto ultrathin Ni films (≈ 20 layers) grown in Cu(100) situ on and remanently magnetised perpendicular to the surface /3/ by using a small coil. Experimental details about the cluster source and the deposition process can be found in two recent papers on deposited Cr<sub>n</sub> clusters /4/.

All X-ray absorption spectra were taken in normal incidence geometry at a temperature of 20 K. The resulting XMCD spectra of the magnetised Fe<sub>n</sub> clusters show large asymmetries at the  $L_{2,3}$  edges. All Fe<sub>n</sub> clusters are ferromagnetically coupled to the Ni/Cu(100) underlayer. As an example, an Fe<sub>8</sub> XMCD spectrum is shown in Fig. 1. Careful analysis of the data allows us to determine the ratio of orbital to spin magnetic moments for the different clusters. These results can be compared to related experiments on ultrathin films and multilayer systems as well as to theoretical predictions, from which an increased orbital magnetic moment is expected in systems with reduced dimensionality.

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# Magnetic Circular Dichroism in the Soft X-Ray Absorption Spectra of Co-Based Heusler Alloys

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In ferromagnetic Heusler alloys, many experimental studies have been carried out in order to discuss the magnetic moments. The Mn-based Heusler alloys  $X_2$ MnZ have the magnetic structure with moments of approximately 4  $\mu_B$  on the Mn atom, which is predominant in the total magnetic moment. On the other hand, Co atoms mainly carry magnetic moments in Co-based Heusler alloys except for Co<sub>2</sub>MnSn. The Co<sub>2</sub>YZ are of particular interest because the local magnetic moment carried by the Co atom is known to have values between 0.3 to 1.0  $\mu_B$ . [1] According to the theoretical descriptions, the orbital angular momentum component on the Co atom is not quenched and contributes to the magnetic moment.[2] In order to investigate the orbital angular momentum on the Co atom, we have carried out measurements of magnetic circular dichroism (MCD) in the soft x-ray absorption (XAS) spectra of the Co 2*p* to 3*d* core excitation for Co<sub>2</sub>TiSn, Co<sub>2</sub>ZrSn, and Co<sub>2</sub>NbSn at BL25SU beamline of SPring-8 in Japan.

The Co 2p to 3d XAS spectra show clear MCD in these three alloys at 50 or 100 K which is much lower than their Curie temperature. We have estimated the absolute values of the spin and orbital angular momentum components in the magnetic moment on the Co atom by using the magneto-optical sum rules with use of the reported total magnetic moments obtained by magnetization measurements in the high magnetic field. The estimated  $\langle L_z \rangle$  is not varied by the substitution of Ti with Zr and Nb. The  $\langle S_z \rangle$  depends also on these Y atoms, that is, on the occupation number of the Co  $e_g$  minority spin bands near the Fermi. These features are consistent with the theoretical descriptions based on the band picture.[2]

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## **Electronic structure of bcc Mn**

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Understanding magnetism has been an important driving force for studying the electronic structure of the magnetic transition metals in the past decades. Using angle-resolved photoemission and inverse photoemission, almost complete band structures have experimentally been obtained for Cr, Fe, Co, and Ni. For Mn metal, on the other hand, no experimental band structure data is available.

We have recently noticed that deposition of, e. g., 4 monolayers (ML) Mn leads on epitaxial Fe(110) films on W(110) to a clear  $p(1\times1)$  LEED pattern [1]. On pure W(110), STM has recently shown epitaxial growth of bcc Mn up to a local thickness of 3 ML [2]. To explore epitaxy further, we have tried to stabilize thick, bulklike films of Mn on W(110). By room temperature deposition and post-annealing we obtain sharp  $p(1\times1)$  LEED patterns for Mn films between 15 and 30 ML, i. e., thick enough to serve as samples for photoemission studies of the Mn bulk electronic structure.

We have studied these films by angle-resolved photoemission. The electron wave vector has been varied parallel and perpendicular to the surface plane. Normal emission spectra have been taken from 6 eV to 300 eV photon energy. Each spectrum is dominated by a broad intensive peak at -2.7 eV and a peak near the Fermi energy. The peak at -2.7 eV does not disperse while the peak at the Fermi energy shows a finite dispersion with angle and photon energy and changes its intensity. Oxygen adsoprtion has been used to assign the peak at -2.7 eV to Mn bulk-derived emission. We interpret our results along similar lines as our previous data taken on fcc Mn grown on Cu<sub>3</sub>Au(100) [3]. In particular, the dispersionless structure at -2.7 eV reflects the strongly correlated electronic structure of bcc Mn.

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# Angle Resolved Photoemission Spectroscopy and Magnetic Circular Dichroism in Fe-Intercalated TiS<sub>2</sub>

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The 3*d* transition metal intercalation compounds  $M_x \text{TiS}_2$  have actively been studied to understand their variety of physical properties. Fe-intercalated TiS<sub>2</sub> have shown various interesting magnetic behaviors and revealed an itinerant electron magnetism associated with the intercalated Fe 3*d* electrons.[1] It has been known for x=1/3 that the Fe  $e_g$  orbitals make covalent bonds with the 3*p* orbitals of the surrounding S atoms and the Fe  $t_{2g}$  orbitals hybridize appreciably with the  $t_{2g}$  orbitals of the nearest neighbor Ti atoms.[2] We have carried out measurements of angle resolved photoemission spectroscopy and magnetic circular dichroism (MCD) in the soft x-ray absorption spectra (XAS) for Fe-intercalated TiS<sub>2</sub>.

The energy dispersion of the photoemission structures of  $Fe_{1/3}TiS_2$  is measured by means of the angle resolved photoemission spectroscopy in the soft x-ray region (~ 460 eV) as well as in the vacuum ultraviolet region. The resonance photoemission technique is employed for the Ti and Fe 2p core excitations. We have found a clear Fermi cut of the spectra and interpreted the origin of the structure lying in the vicinity of the Fermi level. As for the Fe 2p to 3d XAS, clear MCD are seen for x=0.10, 1/4, and 1/3. The Ti 2p to 3d XAS spectra also show MCD, revealing that a magnetic moment is induced on the Ti atom through the hybridization with the Fe t <sub>2g</sub> states.

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# COHERENT RESONANT SOFT X-RAY SCATTERING FROM MAGNETIC DOMAIN PATTERNS

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The spatial arrangement and the dynamics of domain patterns in both antiferromagnets and ferromagnets can be studied by coherent resonant soft x-ray scattering. Interference in coherent scattering gives rise to characteristic speckle patterns, which can be used to (a) track the dynamics of the system by following the intensity fluctuations at a given point in reciprocal space as a function of time and (b) reconstruct the spatial arrangement of the scattering objects, using iterative phase retrieval algorithms. Exploiting XMCD and XMLD, we present experimental results on coherent magnetic scattering from ferromagnetic and antiferromagnetic domain patterns in transmission and reflection geometries. The experiments were performed at the ALS (BL 8) and BESSY II (UE56/1 SGM). The coherence of the spatially filtered x-ray beam is analyzed by diffraction from pinholes and the soft x-ray analog of Young's double slit experiment. First results from domain pattern reconstructions will be presented.



# IN-PLANE MAGNETIC ANISOTROPY IN FE-NI NANOSTRUCTURES

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The correlation between magnetic anisotropies and the microscopic structure of nanostructures and ultrathin films is of growing interest. Compared to bulk materials, these low-dimensionality structures show various effects in magnetism due to reduced symmetry and cluster size effects. Although the reduced symmetry introduced by monoatomic steps on the surface leads to an anisotropy (since the different crystallographic directions are not anymore equivalent), the azimuthal dependence of the in-plane anisotropy has been generally neglected up to now. In this work we focus on the effect of the substrate steps on the growth of ferromagnetic nanostructures and on the related magnetic in-plane anisotropy measured by angle dependent X-ray magnetic circular dichroism (XMCD).

Magnetic and structural proprieties of Fe-Ni nanostructures grown on Cu(111) stepped surfaces are investigated in order to correlate the in-plane step-induced magnetic anisotropy deduced from X-ray induced magnetic circular dichroism (XMCD) to the structure and morphology of the films obtained by scanning tunneling microscopy (STM) and surface extended X-ray absorption spectroscopy (SEXAFS).

We observed a strong in-plane anisotropy in morphology (*cf. figure1*), structure and magnetism (*cf. figure 2*). We demonstrate the importance of the step induced in-plane anisotropy by measuring the orbital magnetic moment dependence as a function of the in-plane azimuth angle. In the submonolayer regime an in-plane magnetic anisotropy is observed related to the step decoration growth mode. In the thickness range 2 - 4 equivalent monolayers, 2D coalescence induces anisotropic in-plane strain and leads to a strong in-plane magnetic anisotropy of the magnetic orbital moment ( $M_L$ ). The microscopic origin of this strong in-plane variation of  $M_L$  has been attributed to magnetocrystalline effects.



## MAGNETIC CIRCULAR DICHROISM IN HEUSLER ALLOYS

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Figure 1: Mn L<sub>23</sub> x-ray emission spectra.

Heusler alloys are of great interest since their discovery in 1903 [1] because they are ternary intermetallic compounds with magnetic properties that can be altered by changing the degree or type of chemical order. Spin-polarized calculations showed that the electronic structure of Heusler alloys has a metallic character for majority spin-electrons [2].

The Mn 2p x-ray photoemission spectra shows the magnetic splitting of Mn  $2p_{3/2}$  core level, that reflects the high 2p-3d exchange interaction [3]. Mn L<sub>3</sub>, L<sub>2</sub> x-ray emission spectra, following the excitation with circularly polarized -x-rays, were carried out on beam-line ID12B, ESRF (Fig.1). The magnetic dichroism was detected in emisson and absorption spectra of NiMnSb and Co<sub>2</sub>MnSb.

It was shown, that the differences at Mn  $L_3$  and Mn  $L_2$  magnetic spectra are linked with non-symmetry character of Mn  $3d_{3/2}$ ,  $3d_{5/2}$  states in valence band. The sharp re-emission peak *B* at Mn  $L_3$  x-ray spectrum corresponds to maximum of empty minority spin DOS Mn

states [4], that is supposed to be a specific feature of Half-metallic ferromagnets [2].

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