Ultraviolet Synchrotron Orbital Radiation Facility

VIII-N Development of the UVSOR Light Source

VIII-N-1 Influence of Electron Beam Properties on Spontaneous Radiation from an Optical Klystron

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[Nucl. Instrum. Methods Phys. Res., Sect. A 429, 191 (1999)]

Spectrum of the radiation from an optical klystron is

very sensitive to the beam energy spread which is an important factor for the storage ring free electron laser. In order to make use of the spectrum for a measurement of the relative energy spread, we have investigated influences of the transverse beam property on the spectrum using a helical optical klystron on the UVSOR storage ring. From analysis of the experimental results, it has been found that the most important effect comes from inhomogeneous magnetic fields and a horizontal beam size.

VIII-O Researches by the USE of UVSOR

VIII-O-1 Nano-Second Desorption of Alkali Fluorides Excited by Synchrotron Radiation Pulses

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Desorption of the excited-state alkali atoms has been investigated on alkali fluoride crystals in the photon energy region of 20-75 eV, which includes the excitation energy for Li 1s-core excitons, Na 2p-core excitons, and valence excitations. The quantum yield of the excited-state alkali desorption was found to be comparable between valence and core-level excitations. Time response of desorption from LiF and NaF was also observed by using synchrotron radiation pulses. It was found that the time response consists of a nano-second component and slower one, indicating that the fast desorption is due to the lattice instability induced by electronic transition and the slow one is due to the thermal instability of surface defects.

VIII-O-2 Photo-Induced Change in Semiconductor-Vacuum Interface of p-GaAs(100) Studied by Photoelectron Spectroscopy

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The photo-induced change in the semiconductorvacuum interface on GaAs (100) and Cs/GaAs(100) has been investigated by using a photoelectron spectroscopic technique with synchrotron radiation and a mode-locked Nd:YAG laser. Both Ga-3d and As-3d photoelectron peaks showed transient energy shifts under the laser irradiation without any spectral change. The amounts of the energy shifts were strongly dependent on the sample temperature and the laser photon flux. It was found that the dependence fits well to the theoretical curve derived from the surface photo-voltage effect based on the band bending scheme in the surface layer of the semiconductor.

VIII-O-3 Electronic Structures of Organic Salts DMTSA-BF₄ Using Photoelectron Spectromicroscopy

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The electronic structures of 1:1 charge-transfer organic salts DMTSA-BF₄, where DMTSA is 2,3dimethyltetraseleno-anthracene, have attracted the interest of many researchers because of the high electrical conductivity and metallic physical property.¹) The photoemission study for DMTSA-BF₄ has not been performed so far since DMTSA-BF₄ is not large enough to carry out the ordinary photoemission experiments. We succeeded to measure the photoemission spectra for DMTSA-BF₄ using the photoelectron spectromicroscopy.

Figure 1 shows the photon energy dependence of the photoemission spectra for DMTSA-BF₄. From the photon energy dependence of the photoionization crosssection, the atomic orbital characters of the observed spectral features are determined. The Se 4p states are located at ~1 eV, ~3 eV, and ~6 eV. The C 2p and F 2p states are located at ~8 eV. In our photoemission measurements, the clear Fermi edge was not observed. This suggests the importance of the electron correlation effect and/or one-dimensional nature for DMTSA-BF₄. The photoemission spectra for DMTSA were also measured in order to compare the electronic structures for DMTSA-BF₄. The Se 3d core level photoemission spectra did not show the chemical shift but the photoemission spectrum for DMTSA-BF₄ showed the tail at the higher binding energy side as compared with that for DMTSA. The difference of the electronic structures between DMTSA and DMTSA-BF₄ is discussed, based on the valence band and Se 3d core level photoemission results.

Reference

1) J. Dong, K. Yakushi, K. Takimiya and T. Otsubo, *J. Phys. Soc. Jpn.* **67**, 971 (1998).



Figure 1. The photon energy dependence of the photoemission spectra for the organic salts DMTSA-BF₄.

VIII-O-4 Photoemission Study of Si(111) Clean Surfaces at High Temperature Using Laser Annealing

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The electronic structures of Si (111) clean surfaces at high temperature have been studied using the photoemission spectroscopy. In the photoemission measurements, it is not easy to observe the electronic structures at high temperature because of the electric field used to anneal the sample to high temperature. By combining the laser annealing and the photoelectron spectromicroscopy, we demonstrate that the photoemission spectra at high temperature are easily obtained.

Figure 1 shows the temperature dependence of the photoemission spectra for Si (111) clean surfaces from room temperature to ~1200 °C. At room temperature, three surface states were observed at ~0.2, ~0.8 and \sim 1.8 eV. With increasing the annealing temperature, the surface state at ~0.8 eV disappears. The temperature dependence of the spectral features obtained in our measurements is essentially identical to that previously obtained by Yokotsuka et al.¹⁾ In addition, we found that the shift of the photoemission spectra was observed at ~0.2 eV higher binding energy side under 0.04~0.89 W laser irradiation. The shift at ~0.2 eV was also observed at the Si 2p core level photoemission spectra. With increasing the annealing temperature, the shift disappears at the laser power of 2.00 W. The observed shift indicates that the surface photovoltage $effect^{2}$ is caused by the photocarrier, which is induced by the laser irradiation. The disappearance of the surface photovoltage effect may be caused by the metallic nature of the high temperature semiconductor.

References

- 1) T. Yokotsuka, S. Kono, S. Suzuki and T. Sagawa, *Solid State Commun.* **39**, 1001 (1991).
- 2) J. E. Demuth, W. J. Thompson, N. J. DiNardo and R. Imbihl, *Phys. Rev. Lett.* **56**, 1408 (1986).



Figure 1. The temperature dependence for the Si (111) clean surface under laser irradiation.

VIII-O-5 Behavior of the "6eV Satellite" in Ni Thin Film Observed by Valence Band Photoemission

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[J. Electron Spectrosc. Relat. Phenom. 101-103, 257 (1999)]

It is now well established that the electronic structures of thin films are interestingly different from those of bulk. In case of 3d-transition materials, this difference is directly related to the changing of *d*-band width, exchange integral and the magnetic moment with changing of the film thickness.¹⁾ In such situation, any correlation-induced peak in the photoemission, such as Ni-6eV satellite also shows some interesting and varied feature depending on the film thickness. Another important aspect, that has to be considered, is the hybridization effect between the film and substrate. Here, we report the Ni valence band photoemission result for different thickness films on ferromagnetic Co(001) in order to study the 6 eV satellite feature and the resonant effect on magnetic dichroism signal for satellite.

In Figure 1, we show a series of Ni-valence band photoemission spectra taken at hv = 67.2 eV. The peak around the Fermi level is the main peak assigned by $3d^9$ state. Another peak stayed around 6 eV is the satellite assigned as $3d^8$ state. At hv = 67.2 eV, the satellite peak shows resonance enhancement.²⁾ The satellite peak positions for all films are indicated by dashed line. It is clear that satellite peak position is shifted to the higher binding energy for thicker films. The origins of this variation are the thickness dependent *d*-band width and the corresponding *d*-*d* correlation effect. For thicker films, the large width of *d*-band pushes the satellite peak higher binding energy position. In Figure 2, we show the resonance effect on magnetic linear dichroism (MLD) for satellite peak where the sample is 8ML Ni grown on 10ML Co. After comparing with off-resonant result in Figure 2 (b), we found that the in-plane MLD signal for satellite shows resonant behavior.

Reference

W. Clemens *et al.*, *Solid State Commun.* **81**, 739 (1992).
C. Guillot *et al.*, *Phys. Rev. Lett.* **39**, 1632 (1977).



Figure 1. Ni-valence band photoemission spectra for Ni films of different thickness (1-21 Å). The peak around 6.5 eV is the satellite peak, which shows binding energy shift for thicker films as indicated by dashed line.



Figure 2. MLD for 8ML Ni on 10 ML ferromagnetic Co. In (a), MLD at on-resonance shows opposite trend for the main peak and the satellite peak. In (b), MLD at off-resonance where satellite shows weak MLD.

VIII-O-6 Magnetic Stability of Co-Film Grown on Oxygen-Rich Cu(001) Surface

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From the points of view of both technological application and fundamental research, it is a common intention to grow metal film on very clean surface (surface without any contamination). Using of clean substrate ensures the perfect crystalline growth, defect-free interface, and in the case of magnetic film, the magnetic stability. Oxygen is considered to be the most defined contamination from which we want to get ride of for getting good epitaxy. But oxygen can be also used as a surfactant¹) (which promotes the layer-by-layer or well-shaped island growth). In the present study, we report the magnetic behavior of Co films grown on oxygen-rich Cu(001) substrate. We also checked the electronic structures, reconstruction phases, surfactant effect *etc*.

Figure 1 shows the magnetic dichroism (a technique to study the magnetism) results for two systems, (a) 5ML Co on clean Cu(001) and (b) 5ML Co on oxygenrich Cu(001) surface. The oxygen-rich Cu, oxidized by 1200L oxygen, shows ($\sqrt{2}\times2\sqrt{2}$)R45°-O reconstructed phase in LEED pattern. The 5ML Co film was then deposited on that oxidized Cu and was characterized by XPS and LEED. It was found that oxygen was segregated to Co surface and promotes to grow the c(2×2) reconstructed phase. The new state of oxygen on Co is the chemisorbed state. The interesting observation is that this reconstructed phase of Co also shows almost similar magnetic behavior to Co on clean Cu. The order of dichroism signal is nearly same for both cases [as seen in Figure 1(a) and Figure 1(b)]. Therefore, it can be said that the large amount of oxygen on the Cu before Co deposition does not cause any negative effect on the magnetic (ferromagnetic phase) stability of Co film.

Reference

1) C. Tolkes et al., Phys. Rev. Lett. 80, 2877 (1998).



Figure 1. The MUDAD (magnetic dichroism in angular distribution by unpolarized light) for 5ML Co on fresh Cu [in (a)] and on oxidized Cu [in (b)]. The Co 2p peaks were measured for the two-magnetization directions. The almost same order of dichroism signal confirms the magnetic stability of Co films on oxygen-rich Cu.

VIII-O-7 Thickness Dependent Oxidization of Co Films and Observation of Different CoO Phases

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The bulk CoO is a well-known Mott insulator and shows strongly correlated nature. It is known that CoO can be also obtained by oxidization of bulk Co or Cofilm. In case of bulk Co, the entire oxidization process is confined to only hcp structure, the only available phase at room temperature. By using artificial Co thin film, we can vary the oxidization process as growth behavior of Co on other metal, *e.g.* Cu(001), differs depending on the thickness. For example, Co films of less than 2ML on Cu show different sizes of island, whereas more than 2ML Co films grow in layer-by-layer mode and show fct (face center tetragonal) structure. We have studied the core level photoemission by growing CoO for different thickness of Co films.

Figure 1 (a), shows the Co2*p* photoemission spectra for clean and oxidized Co films, where the thickness of the Co is 1.3, 2.3, 3.6, and 5ML, respectively. The films were oxidized by almost same amount of exposure. In (a), spectra with symbols are for oxidized Co and solid lines are for clean ones. Peak at binding energy 778.0 is from elemental Co. Peak at 780.2 eV (dashed line) is assigned to be d^8L (L stands for ligand hole state) and the higher binding energy satellite (dotted-dashed line) is assigned as the final state with a mixture $d^9\underline{L}^2$ and d^7 states on the basis of charge transfer (CT) theory¹) for CoO. The results show that the thinner films are more reactive and show an early formation of CoO. In Figure 1 (b), the difference between the saturated CoO phase for 1ML and 10ML Co films is shown. It is noticed that the features for thinner film seem to be broader. Here we predict that the difference spectrum shown in this figure probably represents the 2D CoO phase, which is different from the bulk CoO phase.

Reference

1) K. Okada et al., J. Phys. Soc. Jpn. 61, 449 (1992)



Figure 1. Co2*p* XPS for different Co films. In (a), the solid spectra for elemental Co and spectra with symbol for oxidized Co films. In (b), the spectra of saturated CoO phase for 1ML and 5 ML Co films and their "difference spectrum."

VIII-O-8 Satellite Structure Observed in 2p XPS of Co Thin Film

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Satellite structures in the photoemission spectra for 3d-transition metals have been frequently reported for more than twenty years.¹⁾ The well-known example of the satellite structure is the 6eV-satellite in Ni valence band and core level photoemission.²⁾ It is now established that the *d*-*d* interaction is the origin of the formation of satellite. This interaction might take a strong value at the surfaces and interfaces where the reduction of dimension exists along the surface normal. Therefore, we may expect the variation of satellite intensity if the thickness of the thin film system is considered to be a variable. At the same time, the hybridization between the film and the substrate might play an important role for observing the satellite.

Figure 1 shows the $\text{Co}2p_{3/2}$ XPS spectra for different thickness of Co films on the clean Cu(001) substrate. As shown in the figure, we observe a regular change in the four spectra for the both main peak (upper inset) and the satellite region (lower inset). In the case of 1 ML film, the film structure is considered not to be homogeneous, but island type. This situation may make different charge distribution (less *d*-hole) in the valence band at the Co-site. As a result, the chemical environment of the

atom from where the core electron is outgoing is changed. Therefore, the charge transfer process with the Cu and the Coulomb interaction may differ form these in thicker film. The situation causes the binding energy shift for the main peak. In case of satellite region (in lower inset), the intensity difference is also apparent. For 1 ML film, the states around 779.5-783.0eV binding energy is more intense than those in other films. This is shown clearly in the difference spectra between 1 ML and 10 ML films. The reasons for intense satellite in 1 ML film are the stronger d-d correlation and less hybridization with the substrate states.

References

S. Raaen, Solid State Commun. 60, 991 (1986).
S. Hüfner et al., Phys. Lett. A 51, 299 (1975).



Figure 1. $Co2p_{3/2}$ photoemission spectra for 1, 3, 5 and 10 ML Co films on Cu(001). **Upper inset.** The binding energy difference in the main peak. **Lower inset.** The intensity difference around the satellite region. This difference is clear in the "difference spectrum" of 1 ML and 10 ML (shown underneath of four spectra).

VIII-O-9 Design Study for a Varied-Line-Spacing Plane Grating Monochromator

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Recent conceptual and technological improvements to monochromators have enabled us to realize various studies on vibrational spectroscopy in the soft x-ray range (100~1000 eV), which contains the K-shell thresholds of chemically important elements like C, N, and O, even without the use of undulator radiation. At the UVSOR, there is only one monochromator for high

resolution spectroscopy in the photon energy region of interest (250~550 eV). A maximum resolving power of 4000 at 400 eV is achievable with this monochromator, but due to a little complicated scanning mechanism, it has difficulty maintaining such high resolution in the entire photon energy range. Design study for a new monochromator has recently been started in order to improve the situation. A Varied-line-spacing Plane Grating Monochromator (VPGM) has been chosen for this work. Thanks to the availability for high quality gratings and simple scanning mechanism, VPGM seems to be one of the most trustworthy monochromators to realizing high resolution in the soft x-ray range. Figure 1 shows a layout of the optical elements of the present VPGM. The resolution of this monochromator was studied by ray-tracing simulation as well as analytical estimation. This study shows that a resolving power of more than 5000 is achievable over the energy range from 250 to 550 eV with one single grating having the groove density of 1000 l/mm.



Figure 1. Schematic layout of the designed monochromator.

VIII-O-10 Photodissociation of Ozone in the K-Edge Region

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[J. Phys. Chem. A 103, 4598 (1999)]

Ozone is one of the most important molecules in chemistry since ozone in the stratosphere absorbs UV light emitted from the sun and prohibits humanity from the exposure by the UV light. We have investigated the photochemistry and photodissociation dynamics of Ozone in K-edge region with angle-resolved time-offlight apparatus for the first time. The β values and kinetic energy distributions were calculated from simulation of the TOF spectra obtained. The β value of O^+ at an excitation energy of 529.0 eV is -0.7, which is consistent our previous assignment that the 529 eV band comes from the $\pi^*(2b_1) \leftarrow 1s(2a_1)$ transition. In contrast to this, the β value at an excitation energy of 535.7 eV is -0.3. This implies that the transition moment does not lie perpendicular to the molecular plane and that this band should be mixed by more than two transitions. They are consistent with the previous assignments¹⁾ that the 529 eV band is the $\pi^* \leftarrow O_T(1s)$ resonance excitation and that the 536 eV band is mainly

consisted of two transition, the $\pi^*(2b_1) \leftarrow O_C(1s)$ and the $\sigma^*(7a_1) \leftarrow O_T(1s)$.

Reference

1) T. Gejo, K. Okada and T. Ibuki, *Chem. Phys. Lett.* **277**, 497 (1997).

VIII-O-11 Infrared Magnetic Circular Dichroism of Strongly Correlated 4f Electron Systems with Synchrotron Radiation

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[Jpn. J. Appl. Phys., Part 1 38, 392 (1999).]

Magnetic circular dichroism (MCD) experiment in the infrared region using synchrotron radiation has been done for the first time. Since the off-axis component of the synchrotron radiation is circular polarization in itself, a MCD spectrum in wide energy range can be measured easily by using the infrared synchrotron radiation. The experimental method is suitable for the investigation of strongly correlated electron systems because the electronic structure, which is made by magnetic many-body interactions, is located in the wide energy region near the Fermi level.

VIII-O-12 Optical Conductivity of the Kondo Insulator YbB₁₂: Gap Formation and Low-Energy Excitations

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[*Phys. Rev. B* **58**, R7496 (1998)] [*Jpn. J. Appl. Phys.* Series 11, 85 (1999)] [*Physica B* **259-261**, 317 (1999)]

Optical reflectivity experiments have been conducted on single crystals of the kondo insulator YbB₁₂ in order to obtain its optical conductivity, $\sigma(\omega)$. Upon cooling below 70 K, a strong suppression of $\sigma(\omega)$ is seen in the far-infrared region, indicating the opening of than energy gap of ~25 meV. This gap development is coincident with a rapid decrease in the magnetic susceptibility, which shows that the gap opening has significant influence on magnetic properties.