Ultraviolet Synchrotron Orbital Radiation Facility

VIII-J Development of the UVSOR Light Source

VIII-J-1 UVSOR Upgrade Project

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Towards upgrading UVSOR, we have designed a new magnetic lattice, in which the beam emittance would be reduced and the number of the straight sections available for insertion devices would be doubled. The accelerator components necessary for the upgrade project are under development. A combined function magnet was designed, which is capable of producing both quadrupole field and sextupoles filed. A prototype was constructed and the field measurements are in progress. Some early results have shown that the required field strengths could be achieved. The beam position monitor system was replaced and successfully commissioned. The new system can measure one orbit in a second with resolution of a few microns. It will be a powerful tool in stabilizing the low emittance electron beam. An in-vacuum undulator is under construction. This will be installed in the ring, in spring, 2002, to check the performance and the effects on the electron beam. This type of device is expected to provide SR beams of higher brilliance in higher energy region above 100 eV.

VIII-J-2 Storage Ring Free Electron Laser

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The storage ring free electron laser (FEL) at UVSOR has successfully oscillated in the wide spectral region from 590 nm to 240 nm. In these years, we have made many efforts to realize stable oscillation and high average power. As the result, the average power has exceeded 1 W, which is the world highest record as a storage ring FEL. One of the unique features of the storage ring FEL is the natural and perfect pulse-to-pulse synchronization with the synchrotron radiation pulses, which are produced in the same storage ring. By utilizing this, we have performed a pump-probe experiment, in which SR pulses from an undulator excite Xe atoms and the excited state with 1 nsec lifetime was probed by the FEL pulses.

VIII-J-3 Vacuum System Remodeling for the UVSOR Upgrading

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The beam lifetime of the UVSOR depends strongly upon both bunch size and pressure. Because a high brilliance is obtained by reducing the bunch size in the UVSOR upgrading plan designed at present, the lower operating pressure should be achieved to maintain a long beam lifetime. This project purposes to improve the present vacuum condition and to achieve the required operating pressure in the upgraded ring. We have started observation and inspection of the vacuum situation of the ring. Also the design of new beam chambers for the upgraded ring has been undertaken, where reinforcing the pumping speeds and increasing the beam position monitor are considered.

VIII-K Researches by the USE of UVSOR

VIII-K-1 Photo-Induced Phase Transition of Spin-Crossover Complex Studied by Photoelectron Spectroscopy

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The photo-induced phase transition (PIPT) of an organometal spin-crossover complex $[Fe(2\text{-pic})_3]Cl_2\cdot$EtOH has been studied by photoelectron spectroscopy. The Fe-3d and N-1s spectra showed remarkable changes due to the photo-excitation at low temperatures, indicating that the electron charge is transferred from nitrogen to Fe atoms in the PIPT. The electronic structure of the photo-induced phase is very different from that of the high-temperature phase, which is caused by thermally induced phase transition (TIPT). The valence-band spectra of the photo-induced phase is in good agreement with the cluster calculation involving the E_g-distortion of Fe-octahedrons, indicating that the symmetry lowering in the excited state plays an important role to cause the PIPT. It was also found that the valence-band spectra are enhanced around T_c's, indicating the dynamics of the PIPT competing with the

TIPT.

VIII-K-2 Beam-Line Systems for Pump-Probe Photoelectron Spectroscopy Using SR and Laser

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[*SRI proceedings* (2001)]

Since the first report by Saile, several groups have been developing new spectroscopy based on the combination of synchrotron radiation and laser light. The new spectroscopy are very attractive and interesting, since both synchrotron radiation and laser light are useful light sources with different characteristics: Synchrotron radiation provides high photon energy to investigate core-levels, while laser light is so intense to produce excited valence states with high density. Various combinations of two powerful light sources may open new scientific achievements. Combined systems for photoelectron spectroscopy using synchrotron radiation and laser light have been constructed at BL 5A and BL6A2 in the UVSOR facility, Okazaki. The systems consist of high-performance photoelectron spectrometers and mode-locked lasers. The performance of the systems is reported with a few examples.

VIII-K-3 Experiments with Combined Laser and SR at the UVSOR Facility

KAMADA, Masao

[*LSWAVE proceedings* (2001)]

There are several ways to use SR and Laser. One is to use SR as pump and laser as probe. This is powerful to investigate many interesting phenomena concerning with core-level excitations. Second is to use laser as pump as SR as probe. This is very useful to investigate many phenomena relating with valence excitations. Third one is to use SR and laser simultaneously as 2-photon excitation. This will open the new science relating with core-level excitation too. In recent years, brilliance of SR is increased and also short- wavelength lasers are under progress. So, many persons may expect more combinations in near future such as SR-Pump + SR-Probe, Laser-Pump + Laser-Probe, Laser 2-Photon-Pump, and SR 2-Photon-Pump.

VIII-K-4 Cesiumoxide-GaAs Interface and Layer Thickness in NEA Surface Formation

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[*PES proceedings* (2001)]

Negative electron affinity (NEA) surfaces have found applications as efficient photocathodes and the NEA surface of GaAs(100) and its superlattice is known to be a useful emitter of spin-polarized electrons with a high degree of polarization and efficiency. This can be achieved by the ‘jo-jo’-technique, where Cs deposition and subsequent oxidation are repeated several times. The details of NEA surface formation, however, are not fully understood: previous reports differ considerably both in describing the method of production as well as the underlying chemical and physical mechanism. In this paper we present a systematic study of various sample treatments on GaAs(100) using photoemission spectroscopy. Analyzing the influence of the both the cesiation and oxidation on the bandbending and monitoring the photoemission yield on bulk GaAs(100), we have been able to distinguish three different regimes of activation, depending on the total thickness of the overlayer, the Cs:O ratio and the resulting chemical interaction with the substrate.

VIII-K-5 Surface Photovoltage Effects on *p*-GaAs (100) from Core-Level Photoelectron Spectroscopy Using Synchrotron Radiation and a Laser

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[*Phys. Rev. B* (2001) in press]

The surface photo-voltage (SPV) effect on laser-excited *p*-GaAs (100) has been investigated using core-level photoelectron spectroscopy with synchrotron radiation (SR). The energy shift of the Ga-*3d* photo-electrons due to the SPV effect was remarkably dependent on the sample temperature and the laser photon-flux. The dependence in each case was well interpreted on the basis of a simple SPV formula derived from the band-bending scheme with excess photocarriers. The magnitude of the band bending was about 0.8 eV for clean *p*-GaAs (100) surfaces having no electrodes. Similar core-level shifts were observed in the Ga-3*d* and Cs-4*d* spectra of Cs/GaAs (100), indicating an unpinned behavior of the electronic states of the Cs surface layer. The time response of the SPV effect was also investigated in the nano-second range using a pump-probe method with SR and laser.

VIII-K-6 Performance Tests for the Newly Constructed Varied-Line-Spacing Plane Grating Monochromator at BL-4B

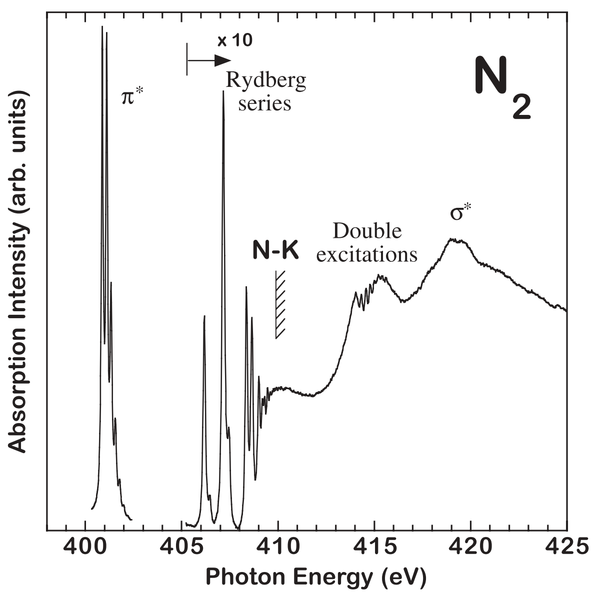
SHIGEMASA, Eiji; NAGASONO, Mitsuru; OJI, Hiroshi; HATSUI, Takaki; GEJO, Tatsuo; KOSUGI, Nobuhiro

The practical construction of the new Varied-line-spacing Plane Grating Monochromator (VLS-PGM) on BL4B at the UVSOR has begun, in order to realize various spectroscopic studies with high resolution in the soft x-ray range (100~1000 eV). The installation has been successfully finished in October 2000. The vacuum condition was ready for obtaining the first SR light before the end of December 2000, and the first performance tests for the monochromator have been carried out.

The absolute photon flux for two gratings available so far (267 and 800 l/mm) has been measured using a Si photodiode supplied by IRD Inc. With the entrance and exit slit openings set at 25 and 10 µm, corresponding to the resolving power of 10000 at 400 eV with the 800 l/mm grating, the photocurrent from the photodiode was measured after the sample position, and converted into the absolute photon flux, taking account of the quantum efficiency of the photodiode. In this case, the resolving power in the regular spectral region for each grating is more than 3000. The throughput photon flux measured ranges from 10^8 to 10^9 photons/sec for the ring current of 100 mA, which is a little smaller than that estimated.

The inner-shell photoabsorption spectra of atoms and molecules were measured, to examine the instrumental resolution. The *K*-shell photoabsorption spectrum of N_2 is presented in Figure 1. The entrance and exit slits were set for achieving the resolving power of 10000. From the comparison with all available spectra of the N 1s → π* resonance of N_2, it seems to be reasonable that the resolving power obtained here is more than 5000. The photoabsorption spectra in the vicinity of the 2p ionization thresholds of S (~170 eV) for H_2S, OCS, SO_2, and CS_2 were also recorded using the 267 l/mm grating. Through the performance tests, it turned out that the resolving power more than 3000 is achieved at the entire photon energy region of interest.

Figure 1. K-shell photoabsorption spectrum of N_2.



VIII-K-7 High-Resolution Symmetry-Resolved K-Shell Photoabsorption Spectra of N_2

SHIGEMASA, Eiji; NAGASONO, Mitsuru; OJI, Hiroshi; HATSUI, Takaki; GEJO, Tatsuo; KOSUGI, Nobuhiro

High-resolution angle-resolved ion-yield spectra of N_2 have been measured using linearly polarized synchrotron radiation. The ion yield spectra (I_0 and I_{90}) measured at 0 and 90 degrees with respect to the electric vector of the light relate to the Σ (ΔΛ = 0) and ∏ (ΔΛ = ±1) symmetry components in the conventional photo-absorption spectrum, respectively. The Σ and ∏ symmetry spectra in the vicinity of the *K*-shell ionization threshold of N_2, except for the π* resonance are shown in Figure 1. Clear decomposition of the conventional photoabsorption spectrum into its symmetry components is observed. Concentrating on the *K*-shell ionization region, structures labeled from A to F, which are considered as being due to the multiple excitations, can be seen in the Σ and ∏ symmetry spectra. A strong and broad enhancement due to the σ* shape resonance, which is a typical single electron transition in the continuum, is found only in the Σ symmetry spectrum. A very weak structure B just above the ionization threshold, and a shoulder structure E of the shape resonance enhancement are observed in the Σ spectrum. In the ∏ symmetry spectrum, a structure A just above the ionization threshold, rather strong features C and D at the double excitation region, and a clear enhancement F just at the shape resonance position are detectable.

In order to elucidate the electronic structures of the spectral features observed in Figure 1, ab initio SCF-CI calculations have been performed, taking account for the transitions from $2\sigma_u$, $3\sigma_g$, and $1\pi_u$ orbitals into $1\pi_g$ (π*) orbital. As a consequence, it is found that the structure labeled F in the ∏ spectrum just at the shape resonance position is attributable to the triple excitations, where a simultaneous excitation of two valence electrons follows the inner-shell excitation.

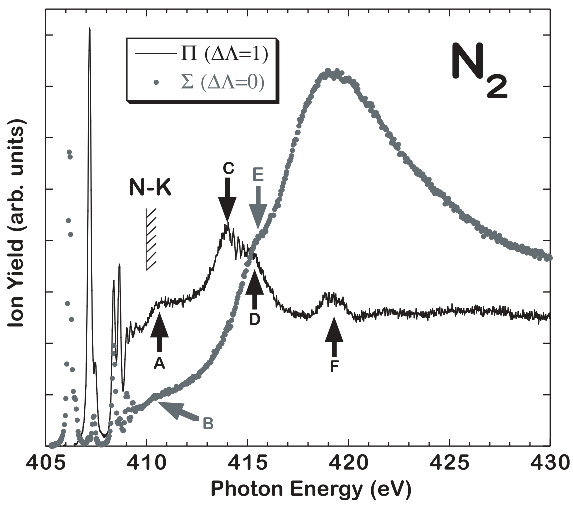


Figure 1. Symmetry-resolved K-shell photoabsorption spectrum of N_2.

VIII-K-8 Dynamical Angular Correlation in Molecular Auger Decay

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([1]LURE)

The first measurements of the angular distribution of Auger electrons from fixed-in-space molecules have been performed in the C *K*-shell ionization region of CO, for both parallel and perpendicular orientation of the molecular axis with respect to the light polarization vector. The ions emitted parallel or perpendicular to the electric vector of the incident radiation determine the possible Σ or Π symmetries in the ionization channels, respectively. The angular distributions obtained for the CO^{2+} $B^1\Sigma$ Auger final state show dramatic spectral variations, which also depend on the initial ionization channels, Σ or Π. The result strongly suggests the breakdown of the two-step model in which the Auger decay is treated independently of the initial photoionization process.

VIII-K-9 Pump /Probe Experiments with FEL and SR Pulses at UVSOR

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Synchrotron radiation free electron lasers (SRFEL or FEL) has been used as a light source because of high power, high coherence and its unique temporal feature. Pump and probe experiments using FEL and synchrotron radiation (SR) pulses have been tried to perform for the last decade. This is due to the fact that the FEL pulse naturally synchronizes with the SR pulse. As the first gas-phase experiment combined FEL with SR, we have carried out the two-photon double-resonant excitation on Xe atoms, utilizing a SR pulse as a pump and an FEL pulse as a probe light.

The experiments were performed on the undulator beamline BL3A1 at UVSOR, where no monochromator is installed. An LiF filter was employed to suppress higher harmonics radiation from the undulator. The estimated photon flux is about 10^{13} photons/sec/0.1% B.W. at I = 100 mA. The FEL pulses were extracted through the backward mirror and transported to an experimental station of the BL3A1 through a series of multi-layer mirrors. The flight path of FEL, which was adjusted to synchronize timing between the FEL and the SR pulses, was about 30 m. A focusing mirror (f = 10 m) was placed in the center of the flight path to keep the beam size of FEL small throughout the transport. About 69% of the extracted power was transferred to the experimental station. The fine adjustment of the delay timing was made by using a movable optical delay system (50 cm) at the experimental station. The FEL and SR pulses introduced coaxially crossed an effusive jet of Xe atoms from a gas nozzle. The singly charged Xe ions produced in the interaction region were detected by means of a conventional channeltron.

The first experiment on the two-photon double-resonant excitation of the Xe* $5p^5nf$ autoionization states using the combination of a mode-locked laser and SR has already been successfully demonstrated by Meyer's group at LURE.[1] In the present work, the combination of FEL and the undulator radiation was chosen in place of the former. The fundamental harmonic of the undulator was adjusted to be 10.4 eV, in order to prepare the Xe* $5p^55d$ intermediate states in a first step. The Xe* $5p^54f$ autoionization resonance can be excited within the wavelength region of FEL in a second step. Because the lifetime of the intermediate states is quite short (600 ps), the synchronization between the SR and laser pulses is essential in this experiment.

The ion yield spectrum for the autoionization Xe* $5p^54f$ resonance obtained as shown in Figure 1. In this measurement the wavelength of FEL was swept by changing the gap of the helical optical klystron. During the measurement a newly developed feedback system was operated to stabilize the lasing. The asymmetric line shape described by the Fano formula has been clearly observed in Figure 1.

We are going to perform the same experiment at the beamline BL7B, where a high-resolution monochromator is installed, to improve the spectral resolution.

Reference
1) M. Gisselbrecht, A. Marquette and M. Meyer, *J. Phys. B: At., Mol. Opt. Phys.* **31** L977 (1998).

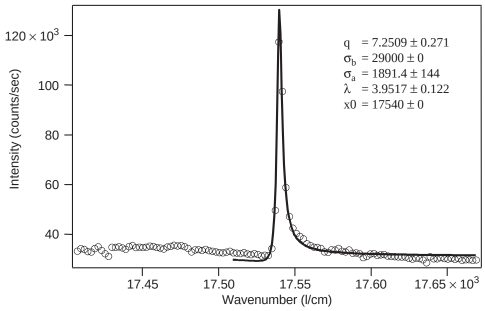

Figure 1. Two-photon ionization signal of Xe as a function of the wavelength of FEL.