# Ultraviolet Synchrotron Orbital Radiation Facility

## VIII-FF Development of the UVSOR Light Source

### VIII-FF-1 UVSOR Upgrade Project

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UVSOR was successfully converted to UVSOR-II, which has eight straight sections and small emittance of 27 nm-rad. The magnetic lattice was modified without changing the circumference of 53.2 m. All the magnets and their beam ducts except for the bending magnets were replaced. An undulator and a super-conducting wiggler were replaced with two new in-vacuum undulators. Some parts of the injector were replaced and upgraded. Some beam-lines were reconstructed. All the reconstruction works were completed within three months, from April to June 2003. In July, UVSOR-II was successfully commissioned. Some preliminary measurements on the beam parameters suggested that the design goal of the emittance, 27 nm-rad, was likely achieved. Vacuum conditioning with beams are in progress. Users experiments will start in September.

### VIII-FF-2 UVSOR Free Electron Laser

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Q-switching operation of the storage ring FEL provides high peak power and is therefore very attractive to application experiments. On the UVSOR, the Q-switching is performed by modulation of RF frequency. In the operation, an excitation of a coherent synchrotron oscillation of electron beam is observed. However, observed damping of the oscillation is faster than one due to synchrotron radiation by factor 100 and, therefore the influence on the FEL lasing is very small. Detailed analysis reveals that the phenomenon is explained with the Robinson damping.

### VIII-FF-3 Ion Trapping at UVSOR

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A vertical tune shift depending on a beam current in multi-bunch condition was observed in the UVSOR electron storage ring. The vertical tune increased as decrease in the beam current, and the slope of the tune shift depended on the condition of the vacuum in the ring. Such change in the vertical tune was explained by change in stability condition of trapped ions on the beam current. The experimental results were consistent with the results from analytic and tracking calculations.

### VIII-FF-4 Improvements of the Vacuum System for the UVSOR II

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In the upgrading to the UVSOR II, all straight chambers and three bend chambers except some elements such as RF cavities and insertion devices should be replaced to new ones. Those were designed and fabricated considering to increase the pumping speed as much as possible in the bend section and the just downstream. The reconstruction work of the ring was started in April 2003. New beam position monitors were additionally equipped for the reinforcement of the focusing magnets in the UVSOR II. After the installation of all chambers, the ring was re-evacuated and baked. Beam injection to the ring was started in the middle of July. The following vacuum conditioning so-called beam scrubbing has been satisfactorily advanced for the user run scheduled to resume in September.

### VIII-FF-5 A New Auger Electron Spectroscopy in Coincidence with Photoelectrons

**ITO, Kenji; SHIGEMASA, Eiji**

Auger photoelectron coincidence spectroscopy, which involves measuring Auger lines in coincidence with the corresponding photoelectron line, reveals a new trend for doubly charged atomic and molecular ions. The experimental resolution in this spectroscopy can be made as good as the instrument resolution permits; the spectral resolution is limited by a lifetime of the inner-shell hole produced by the photoelectron emission. However, the simultaneous measurements of the two electrons can make the situation such that, due to the energy conservation of the whole process, the instrumental resolution instead of the core-hole lifetime governs the spectral resolution. We are preparing for the experimental investigation that will be carried out at a newly constructed undulator beamline BL3U; a high-resolution and high-efficiency analyzer for threshold photoelectrons with a penetration field, and a high-resolution hemi-spherical analyzer or high-resolution time-of-flight spectrometer for Auger electrons. We have already tested the threshold photoelectron analyzer at UVSOR, and found that the energy resolution of a few meV can be attained easily.

# VIII-GG Researches by the USE of UVSOR

## VIII-GG-1 Angle-Resolved Photoion Spectroscopy of $NO_2$ and $SO_2$

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[*Chem. Phys.* **289**, 15–29 (2003)]

Based on recent conceptual and technological improvements for soft x-ray monochromators, a varied-line-spacing plane grating monochromator of the Hetrrick type is installed on the bending-magnet beamline BL4B in the UVSOR facility with a second generation VUV ring of the beam energy of 0.75 GeV. The BL4B has enabled us to realize various spectroscopic investigations under high resolution in the energy range of 90 to 800 eV. High-resolution angle-resolved photoion-yield spectra (ARPIS) of $NO_2$ and $SO_2$ have been measured in the N and O *K*-shell excitation regions. The fragment-ion yield spectra measured at 0° and 90° relative to the electric vector of the light reveal excitation symmetries of complicated electronic states. The spectral features are interpreted in comparison with other transition systems and quantum chemical calculations show strong or weak Rydberg-valence mixing depending on the excitation site in the molecule.

## VIII-GG-2 Atmospheric Lifetime of $SF_5CF_3$

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[*Geophys. Res. Lett.* **29**, 10.1029/2002GL015356, 7-1–4 (2002)]

The vacuum ultraviolet (VUV) absorption spectrum of $SF_5CF_3$ was measured over the range 106–200 nm. At 121.6 nm, $\sigma(\text{base } e) = (7.8 \pm 0.6) \times 10^{-18}$ cm$^2$ molecule$^{-1}$, in which quoted uncertainty includes two standard deviation from the least-square fit in the Beer-Lambert plot and our estimate of potential systematic errors associated with measurements of the reactant concentrations. The VUV spectrum and literature data for electron attachment and ion-molecule reactions were incorporated into a model of the stratosphere, mesosphere, and lower thermosphere. This information provides better constraints on the atmospheric lifetime and hence on the potential of this highly radiatively-active trace gas to influence the climate system. The atmospheric lifetime of $SF_5CF_3$ is dominated by dissociative electron attachment and is estimated to be approximately 950 years. Solar proton events could reduce this to a lower limit of 650 years.

## VIII-GG-3 Pump /Probe Experiments with FEL and SR Pulses at UVSOR

**GEJO, Tatsuo[1]; SHIGEMASA, Eiji; NAKAMURA, Eiken; HOSAKA, Masahito; MOCHIHASHI, Akira; KATOH, Masahiro; YAMAZAKI, Jun-ichiro; HAYASHI, Kenji; TAKASHIMA, Yoshifumi; HAMA, Hiroyuki[2]**
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Storage Ring Free Electron Laser (SRFEL or FEL) has been developed at many synchrotron radiation (SR) facilities all over the world as a powerful light source owing to its high power, high coherence and unique temporal feature. Pump and probe experiments using FEL and SR pulses have been tried to perform for the last decade, since the FEL pulse naturally synchronizes with the SR one. Recently, we have successfully carried out the two-photon double-resonant excitation on Xe atoms, utilizing an SR pulse as a pump and an FEL pulse as a probe light.

In the present work, separate experiments were implemented at two different beamlines of BL3A1 and BL7B at UVSOR. At BL3A1, no monochromator is installed. Therefore, an LiF filter was employed to suppress higher order harmonics of the undulator radiation. The FEL pulses were extracted through the backward mirror of the optical klystron at BL5 and transported to experimental stations through 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. Fine-tuning 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. During the experiment, there were serious background signals due to scattered stray light of SR pulses (typically about $10^5$ counts/sec), which made it difficult to detect the real ion signals. In order to overcome such a difficulty, we temporarily employed the Q-switching technique. With use of this technique, much larger peak power of FEL than that in the normal operation is provided, although the duration of lasing becomes relatively short (~ 0.2 msec). However, if events are selected only during this duration, the improvement of signal to noise ratio (S/N) by a factor of 100 can be achieved.

Figure 1 shows the excitation spectrum near the Xe* $5p^5 5d$ resonance region obtained by setting the FEL wavelength to the maximum of the $5d \rightarrow 4f'$ transition. The background pressure indicated in the figure was kept constant during the measurement. The clear enhancement just below the Xe* $5p^5 5d$ resonance around 117.5 nm is observed in Figure 1, which has not been detected in the previous measurements at a lower pressure. This result strongly suggests that the newly found structure indicated by the arrow is relevant to the formation of Xe clusters, mainly dimers. From the consideration of the excitation energy, it seems to be safe to say that an excited state of $Xe_2$ exists near the $5p \rightarrow 5d$ transition of the Xe atom, which act as intermediate states in the present experiments.

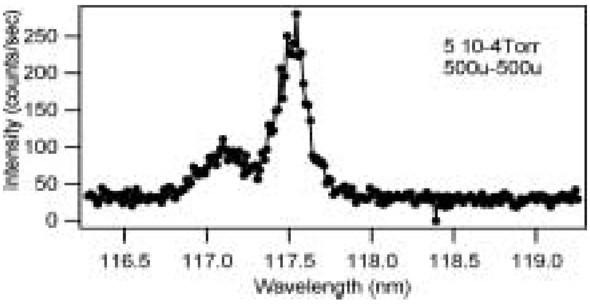


**Figure 1.** Two-photon ionization signal of Xe as a function of the wavelength of SR.

## VIII-GG-4 Photoemission Study of Mixed-Valent Tm-Monochalcogenides: Evidence of Electron-Correlation Effect in Different Tm-Core Levels

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*([1]JAERI.; [2]Univ. Cukurova, Turkey; [3]Himeji Inst. Tech.; [4]Univ. Tokyo.; [5]Tohoku Univ.)*

[*J. Phys. Soc. Jpn.* **72**, 1792–1799 (2003)]

Systematic results of the photoemission spectra for different core levels such as, Tm 4*p*, 5*p* and 3*d* in the mixed valent Tm-monochalcogenides (TmS, TmSe and TmTe) obtained by both experimentally and theoretically are reported. The effects of the electron–electron correlation due to the interaction between core-holes and 4*f*-electrons or the interaction between different configurations are considered to explain the spectral features, depending on the principal (*n*) and orbital quantum numbers of the core levels. Any sharp peak corresponding to a spin–orbital term is not observed especially from the $n = 4$ states. Instead, the multiplet structures are dominantly observed, and the physical identity of the spin–orbital peaks is totally or partially dissolved into the multiplets. Moreover, the electron-correlation effect is found to be dependent on the valence components ($Tm^{2+}$ and $Tm^{3+}$). In the case of the Tm 4*p* core level, in which the principal quantum number is identical with the valence 4*f*, the correlation effect is stronger, and the configuration interaction is therefore considered to explain the spectral features. The photoemission spectra for the shallow core level Tm 5*p* also show the effect of electron correlation, but weaker than that for Tm 4*p*. This has been confirmed bythe resonant photoemission spectroscopy taken at Tm 4*d*–4*f* absorption edges. In addition, the feature of Tm 3*d* photoemission spectra is discussed. All the experimental spectra are compared with the calculated ones. It is therefore understood that the electron correlation effect plays an important role on determining the various features in Tm 4*p*, 5*p* and 3*d* photoemission spectra.

## VIII-GG-5 Infrared Spectroscopy under Extreme Conditions

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[*Physica B* **329-333**, 162–1626 (2003)]

We constructed a magneto-optical microspectroscopy apparatus in the infrared region using a synchrotron radiation, SPring-8. In the apparatus, an infrared microscope with the spatial resolution of 11 mm is combined with low temperatures of 3.5 K and high magnetic fields of 14 T. The purpose is to investigate the electronic structure under extreme conditions of tiny materials such as organic conductors and of small region and the spatial distribution of electronic structures. After the installation of high pressure cells, optical measurements under multiple-extreme conditions is available.

## VIII-GG-6 Collapse of Kondo Lattice in $Ce_{1-x}La_xPd_3$ ($x$ = 0, 0.03)

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The change of the electronic structure as well as the hybridization between the localized 4*f* state and the conduction band (cf hybridization) of $Ce_{1-x}La_xPd_3$ ($x$ = 0, 0.03) due to the La-substitution has been studied by the optical conductivity spectra in the infrared region. The width of the optical transition of Pd $4d \rightarrow$ Ce 4*f* states that was mainly observed in the energy region shrinks by the La-substitution. This means that the cf hybridization is strongly suppressed by the absence of the periodicity of the Ce-ion.

## VIII-GG-7 Optical Reflectivity of the Clathrate Compound $Ba_6Ge_{25}$

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[*Acta Physica Polonica B* **34**, 613–616 (2003)]

We report optical investigations of the electronic properties of the clath-rate compound $Ba_6Ge_{25}$ in which at room temperature Ba atoms "rattle" in Ge-network cavities. When lowering the temperature across $T_S$ ~ 200 K a lock-in of the Ba-atoms to split-site positions in the cages is observed. The low energy Drude type of reflectivity is characterized by a low charge carrier density which smoothly varies with temperature. However, the Drude relaxation time of the charge carriers is found to be almost temperature independent, especially when cooling below $T_S$ where, according to thermopower data, the effective mass is enhanced. This behavior could indicate a formation of polaronic quasi particles below $T_S$ which is also supported by previous measurements of magnetic susceptibility of $Ba_6Ge_{25}$.

## VIII-GG-8 Infrared Magneto-Optical Imaging of κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br

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Since the ground state of κ-(BEDT-TTF)$_2$Cu-[N(CN)$_2$]Br locates in the vicinity of the boundary of the Mott transition, the weak perturbations of the deuteration and the rapid cooling make the phase transition from the superconductor to the antiferro-magnetic insulator. The phase separation is expected to appear on the boundary by the magnetic susceptibility data. Then we observed the spatial distribution of the optical spectrum in the infrared region because the peak at around $h\nu$ = 0.3 eV reflects the physical properties. At the result, the spectral distribution on the sample surface at $T$ = 4 K was observed in spite that no distribution appears at 50 K. The result indicates that the phase separation appears at 4 K.

## VIII-GG-9 Construction of Angle-Resolved Photoemission Apparatus for Solids

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We have constructed a new high-energy-resolution angle-resolved photoemission apparatus shown in Figure 1 for the helical undulator beam line, BL5U. The main purpose is the investigation of the electronic structure near the Fermi level as well as the topological shape of the Fermi surface, so-called "fermiology," of solids, thin films and surfaces. The apparatus consists of a photoelectron analyzer (MBS-Toyama A-1), a main chamber, a sample preparation chamber, a liquid-He flow-type cryostat (JANIS ST-400 UHV) with a manipulator, a He lamp with UV monochromator (GAMMADATA VUV5000 + VUV5040) and several vacuum pumps. Samples are transferred to the prepara-tion chamber from a load-lock chamber, a molecular beam epitaxy system or other chambers that can be replaced by users.

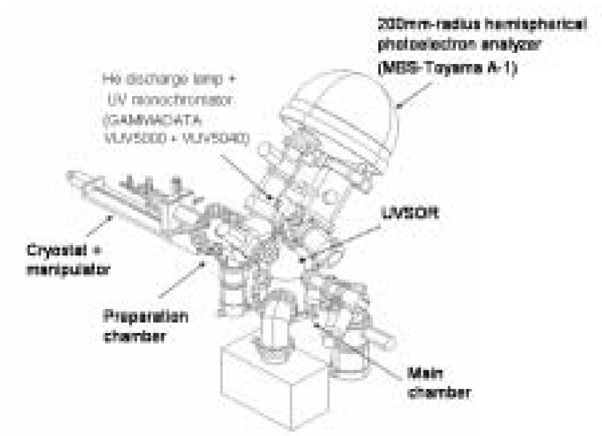


**Figure 1.** High-energy-resolution angle-resolved photo-emission apparatus for BL5U.