Synchrotron Radiation Spectroscopy on Strongly Correlated Electron Systems

UVSOR Facility Division of Advanced Solid State Physics



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Solids with strong electron–electron interaction, so-called strongly correlated electron systems (SCES), have a various physical properties, such as non-BCS superconducting, colossal magneto-resistance, heavy fermion and so on, which cannot be predicted by first-principle band structure calculation. Due to the physical properties, the materials are the candidates of the next generation functional materials. We investigate the mechanism of the physical properties as well as the electronic structure of SCES, especially rare-earth compounds, organic superconductors and transition-metal compounds, by infrared/THz spectroscopy and angle-resolved photoemission spectroscopy based on synchrotron radiation. Since experimental techniques using synchrotron radiation are evolved rapidly, the development of the synchrotron radiation instruments is also one of our research subjects.

1. SAMRAI: A Variably Polarized Angle-Resolved Photoemission Beamline in the VUV Region at UVSOR-II¹⁾

A novel variably polarized angle-resolved photoemission spectroscopy beamline in the vacuum-ultraviolet (VUV) region has been installed at the UVSOR-II 750 MeV synchrotron light source. The beamline is (shown in Figure 1) equipped with a 3 m-long APPLE-II type undulator with horizontally/ vertically linear and right/left circular polarizations, a 10-m Wadsworth-type monochromator covering a photon energy range of 6-43 eV, and a 200 mm-radius hemispherical photoelectron analyzer with an electron lens of a ±18-degree acceptance angle. Due to the low emittance of the UVSOR-II storage ring, the light source is regarded as an entrance slit and the undulator light is directly led to a grating by two plane mirrors in the monochromator while maintaining a balance between high energy resolution and high photon flux. The energy resolving power $(h\nu/\Delta h\nu)$ and photon flux of the monochromator are typically 1×10^4 and 10^{12} photons/sec, respectively, with a



Figure 1. Photograph of the SAMRAI beamline consisting of the APPLE-II type undulator (U7), the modified Wadsworth type monochromator (M0–S), and the high-resolution photoemission analyzer at the focal point. The monochromator, mainly has five optical components: two plane mirrors (M0 and M1) with water cooling, one set of three spherical gratings (G), an exit slit (S), and one toroidal refocusing mirror (M3). The spherical gratings with a radius of 10 m are located 22 m from the center of the undulator. There is no entrance slit. S is located 6.47 m from G. A second branch for a VUV microscope end station is planned to be constructed after the plane mirror (M2) located between G and S.



Figure 2. Temperature dependence of the normalized magnetization curves (a) and temperature derivative curves of the magnetization as a function of temperature (b) for fabricated La-doped EuO and pristine EuO thin films with a thickness of 100 nm on a SrTiO₃ substrate.

 $100-\mu m$ exit slit. The beamline is used for angle-resolved photoemission spectroscopy with an energy resolution of a few meV covering the UV-to-VUV energy range.

2. La-Doped EuO: A Rare-Earth Ferromagnetic Semiconductor with the Highest Curie Temperature²⁾

We report the fabrication of single-crystalline La-doped EuO thin films with the highest Curie temperature ($T_{\rm C}$) of about 200 K among rare-earth compounds excluding transition metals. Due to a first-principle band calculation and an X-ray diffraction measurement, the observed increase of $T_{\rm C}$ cannot be explained only by the increase of the hybridization intensity due to the lattice shrinking and by the increase of the up-spin electrons in the Eu 5d state by the electron doping. The hybridization between the Eu 4f and donor states due to the La-substitution is a possible origin of the increases of $T_{\rm C}$.

3. Design of Terahertz Pump– Photoemission Probe Spectroscopy Beamline at UVSOR-II³⁾

To elucidate the electronic structure relating to physical properties of solids by using selective excitations of low energy electronic and vibrational structure, a new beamline for novel pump-and-probe spectroscopy experiments combining terahertz coherent synchrotron radiation (THz-CSR) and vacuum-ultraviolet coherent harmonic generation (VUV-CHG) is designed. THz-CSR and VUV-CHG are generated from same electron bunches in the storage ring interacted with an amplitude-modulated pulse laser introduced from the outside of the storage ring. The designed schematic top view of the THz pump–PES probe beamline, BL1, is depicted in Figure 3. The amplitude-modulated Ti:Sa laser pulse (1 kHz, 10 mJ/



Figure 3. Schematic top view of the THz pump – photoemission (PES) probe spectroscopy beamline, BL1, at UVSOR-II. The light sources of PES and THz excitation are the VUV-CHG from "Radiator" and the quasi-monochromatic THz-CSR from the bending-magnet, respectively. For the THz pump – PES probe experiment, the time difference between the THz and VUV lights can be made by the delay line.

pulse) is introduced to the UVSOR-II electron storage ring. Then a periodic energy modulation is created on an electron bunch at one of two undulators, namely "Modulator," located at the straight section. VUV-CHG is emitted from the modulated electron beam at the downstream undulator, namely "Radiator." The modulated electron beam also produces the quasi-monochromatic THz-CSR at the bending magnet located at the downstream of the undulators. Since both THz-CSR and VUV-CHG are monochromatic lights, no monochromator for both lights is needed. However, the spectral feature of the THz-CSR must be confirmed by a Fourier transform interferometer (FTIR). Both VUV-CHG and THz-CSR are directed to the same position on a sample in an ultra-high vacuum chamber with a photoelectron analyzer located at the outside of the radiation shielding wall. The THz pump-PES probe experiment is performed as follows; At first, THz-CSR is irradiated to the sample. Just after that, VUV-CHG is irradiated to the sample and PES measured with the time delay. The time delay between the THz-CSR and VUV-CHG is made by the delay line in the optical pass of VUV-CHG. Since the expected 6-th higher harmonics (~9 eV) is not high photon energy, normal incident optics can be used for the delay line.

References

- S. Kimura, T. Ito, M. Sakai, E. Nakamura, N. Kondo, K. Hayashi, T. Horigome, M. Hosaka, M. Katoh, T. Goto, T. Ejima and K. Soda, *Rev. Sci. Instrum.* 81, 053104 (2010).
- 2) H. Miyazaki, T. Ito, H. J. Im, K. Terashima, S. Yagi, M. Kato, K. Soda and S. Kimura, *Appl. Phys. Lett.* **96**, 232503 (2010).
- 3) S. Kimura, E. Nakamura, M. Hosaka, T. Takahashi and M. Katoh, *AIP Conf. Proc.* **1234**, 63 (2010).

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