Synchrotron Radiation Spectroscopy on Strongly Correlated Electron Systems

UVSOR Facility
Division of Advanced Solid State Physics

KIMURA, Shin-ichi	Associate Professor
ITO, Takahiro	Assistant Professor
TERASHIMA, Kensei	JSPS Post-Doctoral Fellow
IM, Hojun	Post-Doctoral Fellow*
TSUNEKAWA, Masanori	Post-Doctoral Fellow†
MIZUNO, Takafumi	Graduate Student
IIZUKA, Takuya	Graduate Student
MIYAZAKI, Hidetoshi	Graduate Student‡
MATSUI, Takafumi	Graduate Student‡

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 a first-principle band structure calculation. Thanks to the physical properties, the materials are the candidates of the next generation functional materials. We are investigating the mechanism of the physical properties of SCES, especially rare-earth compounds, organic superconductors and transition-metal compounds, by infrared/THz spectroscopy and angle-resolved photoemission spectroscopy using 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. Direct Observation of Dispersive Kondo Resonance Peaks in a Heavy-Fermion System[1)]

In heavy-fermion Ce compounds, it is widely accepted that a localized Ce 4f electron due to the strong correlation forms a sharp Kondo resonance (KR) peak just above the Fermi level through the hybridization with an itinerant conduction electron. As a function of this hybridization, the ground state varies from a magnetic to nonmagnetic heavy-fermion separated by a quantum critical point, revealing two characteristic energy scales: the Kondo temperature (T_K) and coherent temperature (T^*). However, an accurate electronic structure for the Ce 4f state has remained a long debated issue since the discovery of the heavy-fermion system in 1970's. Then we carried out the Ce 4d-4f resonant angle-resolved photoemission spectroscopy to study the electronic structure of strongly correlated Ce 4f electrons in a quasi-two-dimensional nonmagnetic heavy-fermion system $CeCoGe_{1.2}Si_{0.8}$. For the first time, dispersive coherent KR peaks of an f state crossing the Fermi level are directly observed together with the hybridized conduction band. Moreover, the experimental band dispersion is quantitatively in good agreement with a simple hybridization-band picture based on the periodic Anderson model. The obtained physical quantities, *i.e.*, T^*, T_K, and mass enhancement, are comparable to the results of thermodynamic measurements. These results manifest an itinerant nature of Ce 4f electrons in heavy-fermion systems and clarify their microscopic hybridization mechanism.

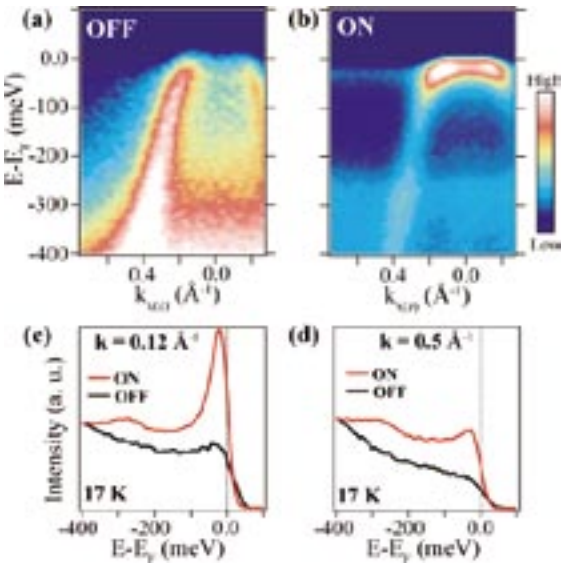

Figure 1. (a), (b) Intensity plots of off- and on-resonant ARPES spectra of $CeCoGe_{1.2}Si_{0.8}$ represent the band dispersion of conduction and f electrons, respectively, along the RT line. (c), (d) EDCs of on- and off-resonant ARPES spectra at the momentum k = 0.12 and 0.5 Å^{-1}, respectively.

2. Excitonic Instability in the Transition from the Black Phase to the Golden Phase of SmS under Pressure Investigated by Infrared Spectroscopy[2,3)]

In the translocation process from the local to itinerant

character of carriers in strongly correlated electron systems, the physical properties drastically change due to the complex relation of the transport to magnetic properties. A strongly correlated insulator, samarium monosulfide (SmS), is a semiconductor with an energy gap size of ~1000 K (~90 meV) and its color is black (namely, the "black phase") at ambient pressure.[2] Above the critical pressure (P_c) of 0.65 GPa, the sample color changes to golden-yellow (the "golden phase") and the electrical resistivity then suddenly drops to one-tenth that in the black phase. To investigate the origin of the pressure-induced phase transition from the black phase to the golden phase, we measured the pressure-dependent optical reflectivity spectra of SmS in the far- and middle-infrared regions. The energy gap becomes narrow with increasing pressure in the black phase. A valence transition from Sm^{2+} in the black phase to mainly Sm^{3+} in the golden phase accompanied by spectral change from insulator to metal were observed at the transition pressure of 0.65 GPa. The black-to-golden phase transition occurs when the energy gap size of black SmS becomes the same as the binding energy of the exciton at the indirect energy gap before the gap closes. This result indicates that the valence transition originates from an excitonic instability.

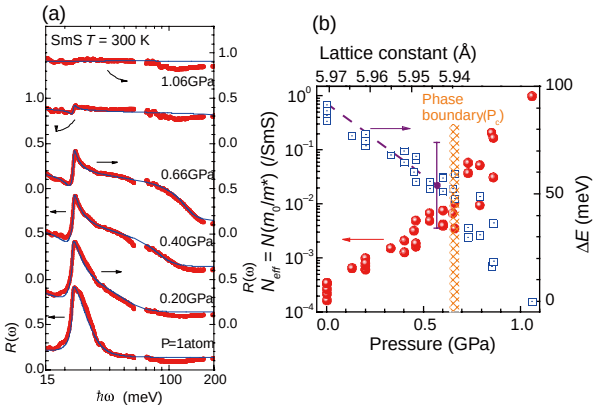

Figure 2. (a) Pressure dependence of the reflectivity spectrum [$R(\omega)$] of black SmS (thick lines) in the energy region of $\hbar\omega$ = 15–200 meV at 300 K. The fitting curve of the combination of Drude and Lorentz functions (thin lines) are also plotted. Successive curves are offset by 0.5 for clarity. (b) Pressure dependences of the effective carrier density (N_{eff}, solid circles) evaluated from the Drude and Lorentz fitting in the left figure and energy gap size (ΔE, open squares) evaluated from N_{eff} and ΔE at 1 atm. The lattice constant that is proportional to the pressure is also denoted in the figure for reference. The energy shift of the exciton peak of the direct transition at the X point at around 0.5 eV (dashed line) normalized at the energy gap size at ambient pressure are plotted as well.

3. Valence Electronic Structure of Cross-Linked C_{60} Polymer: In situ High-Resolution Photoelectron Spectroscopic and Density-Functional Studies[4]

When a C_{60} film is irradiated with a 3 keV electron beam, a cross-linked C_{60} polymer is formed and exhibits metallic electron-transport (I-V) properties in air at room temperature. To elucidate the origin of the metallic I-V characteristics of the cross-linked polymer, we examined the valence photoelectron spectra of the polymer using in situ high-resolution ultraviolet photoelectron spectroscopy (UPS) and found that the spectrum for the cross-linked C_{60} polymer came across the Fermi level (E_F). To understand the UPS results for the C_{60} polymer, we performed first-principles calculations of the band structure for three kinds of optimized three-dimensional unit cells of one-dimensional (1D) cross-linked C_{60} polymers with a cross-linkage consisting of both six- and seven-membered rings and of five- and eight-membered rings (P58). It was found that one quasi-1D P58 cross-linked C_{60} polymer shows semimetallic properties, which provides one possible explanation of both previous (metallic I-V characteristics) and present (valence photoelectron spectra) experimental results.

References

1) H. J. Im, T. Ito, H. -D. Kim, S. Kimura, K. E. Lee, J. B. Hong, Y. S. Kwon, A. Yasui and H. Yamagami, *Phys. Rev. Lett.* **100**, 176402 (2008).
2) S. Kimura, T. Mizuno, K. Matsubayashi, K. Imura, H. S. Suzuki and N. K. Sato, *Physica B* **403**, 805–807 (2008).
3) T. Mizuno, T. Iizuka, S. Kimura, K. Matsubayashi, K. Imura, H. S. Suzuki and N. K. Sato, *J. Phys. Soc. Jpn.* **77**, 113704 (2008).
4) J. Onoe, T. Ito, S. Kimura, K. Ohno, Y. Noguchi and S. Ueda, *Phys. Rev. B* **75**, 233410 (2007).

Awards

KIMURA, Shin-ichi; The Prize for Science and Technology in Research Category, the Commendation for Science and Technology by the Minister of Education, Culture, Sports, Science and Technology.
MIYAZAKI, Hidetoshi; Best Presentation Award, JSR 08.
MIYAZAKI, Hidetoshi; Best Presentation Award, Annual meeting of Tokai Branch of Japan Institute of Metals.

* from Sungkyunkwan University, Korea
† from Nagoya University
‡ carrying out graduate research on Cooperative Education Program of IMS with Nagoya University