Synchrotron Radiation Spectroscopy on Strongly Correlated Electron Systems

UVSOR Facility
Division of Advanced Solid State Physics

KIMURA, Shin-ichi
Associate Professor (–June, 2013)*

MATSUNAMI, Masaharu
Assistant Professor

HAJIRI, Tetsuya	Graduate Student†	KATO, Masaki	Graduate Student†
SHIMURA, Yusuke	Graduate Student†	KANEKO, Masaki	Graduate Student†
HIRATE, Satoshi	Graduate Student†	MOMIYAMA, Hakuya	Graduate Student†

Solids with strong electron–electron interaction, namely strongly correlated electron systems (SCES), have 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. Strongly Hybridized Electronic Structure of $YbAl_2$: An Angle-Resolved Photoemission Study[1)]

Heavy-fermion or valence fluctuation systems are characterized by the strong correlations of *f* electrons, resulting in an enhanced effective mass of quasiparticles due to the band renormalization effect. Consequently, the band structure and Fermi surface (FS) topology are modified from those predicted in band structure calculations based on local density approximation (LDA). To estimate the band renormalization effect, a direct comparison of the experimental band structure as well as the FS with the calculations is necessary. Angle-resolved photoemission spectroscopy (ARPES), which can probe both band structure and FS, is the most suitable tool for this purpose. Here we reported the electronic structure of a prototypical valence fluctuation system, $YbAl_2$, which is known as one of the prototypical valence fluctuation systems, using ARPES. In a recent hard X-ray photoemission spectroscopy study, the mean valence of Yb ions was estimated as +2.2 below 300 K. Correspondingly, an extremely high Kondo temperature (T_K) exceeding 2000 K has been suggested by the magnetic susceptibility or the inelastic neutron scattering. The mass enhancement factor is particularly small among heavy-fermion systems known so far, implying the small renormalization effect. Thus, $YbAl_2$ can be a suitable system to investigate the applicable limit of LDA calculation in relation with the fluctuating valence for the heavy-fermion systems. The observed band dispersions shown in Figure 1 are well described in terms of band structure calculations based on LDA calculation. Strong hybridization between the conduction and 4*f*

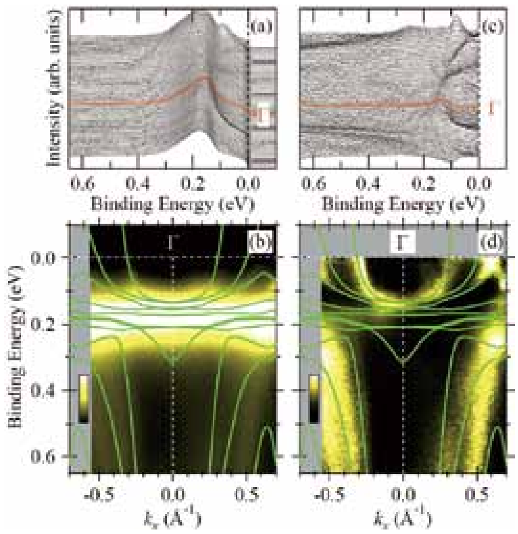

Figure 1. ARPES spectra (a) and intensity plot (b) near E_F along the Γ-K direction. (c), (d) The same data as in (a) and (b), normalized to the angleintegrated spectrum. The band dispersions obtained by the LDA calculation are overlapped in (b) and (d).

bands is identified on the basis of the periodic Anderson model. The evaluated small mass enhancement factor and the high Kondo temperature qualitatively agree with those obtained from thermodynamic measurements. Such findings suggest that the strong hybridization suppresses band renormalization and is responsible for the valence fluctuations in $YbAl_2$.

2. Optical Study of Archetypical Valence-Fluctuating Eu-Systems[2)]

Intermetallic compounds based on rare earth elements with an unstable valence, especially Ce, Eu and Yb, present many unusual properties and are therefore the subject of intense research since many years. Historically, the understanding of these compounds has been dominated by a dichotomy between the Kondo lattice/intermediate valence (KL/IV) scenario for Ce-/Yb-based systems and the valence fluctuating (VF) scenario for Eu-based systems. For Ce- or Yb-based KL/IV systems the nature of the electronic states and the low-energy excitations have been intensively investigated and are relatively well understood (with the exception of the immediate vicinity of the quantum critical point) while for Eu-based VF systems this knowledge is very limited. Then, we have investigated the optical conductivity of the prominent valence fluctuating compounds $EuIr_2Si_2$ and $EuNi_2P_2$ in the infrared energy range to get new insights into the electronic properties of valence fluctuating systems. For both compounds we observe upon cooling the formation of a renormalized Drude response, a partial suppression of the optical conductivity below 100 meV and the appearance of a mid-infrared peak at 0.15 eV for $EuIr_2Si_2$ and at 0.13 eV for $EuNi_2P_2$. Most remarkably, our results show a strong similarity with the optical spectra reported for many Ce- or Yb-based heavy fermion metals and intermediate valence systems, although the phase diagrams and the temperature dependence of the valence differ strongly between Eu- and Ce-/Yb-systems. This suggests that the hybridization between 4*f*- and conduction electrons, which is responsible for the properties of Ce- and Yb-systems, plays an important role in valence fluctuating Eu-systems.

3. Design of a Spin-, Symmetry-, and Momentum-Resolved Electronic Structure Analysis Instruments at UVSOR-III

UVSOR Facility equips two public undulator-beamlines for angle-resolved photoemission spectroscopy, one is BL5U in the photon energy $h\nu$ region of 20–200 eV and the other BL7U of $h\nu$ = 6–40eV. Since the monochromator of the former beamline is an old-style spherical grating type SGM-TRAIN constructed in 1990s and the throughput intensity and energy resolution are poor, the beamline was planned to be replaced to state-of-the-art monochromator and end station. Then we designed a new spin and angle-resolved photoemission spectroscopy instrument with variable photon energy and polarization. We employed a Monk-Gillieson-type variable-line-spacing plane-grating monochromator covering the photon energy of 20–200 eV. The end station shown in Figure 3 equips a VLEED spin detector. The beamline is constructed in FY2013-FY2014 and will be opened to users from FY2015.

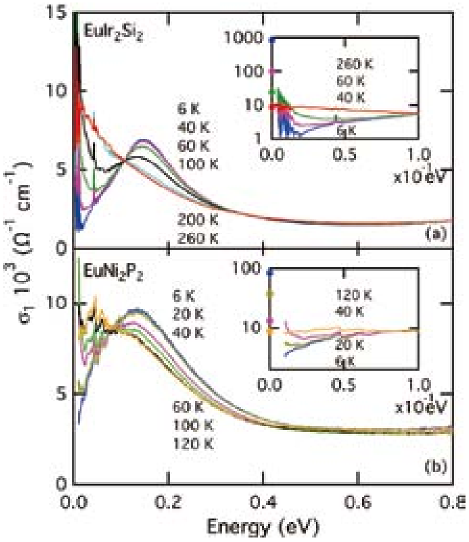

Figure 2. The real part of the optical conductivity spectra of $EuIr_2Si_2$ and $EuNi_2P_2$ at various temperatures. Insets: The low-energy optical conductivity spectra (solid lines) and the corresponding values for the dc conductivity, $\sigma_{dc} = \rho^{-1}$, (symbols) at 6, 40, 60 and 260 K for $EuIr_2Si_2$ and at 6, 20, 40 and 120 K for $EuNi_2P_2$.

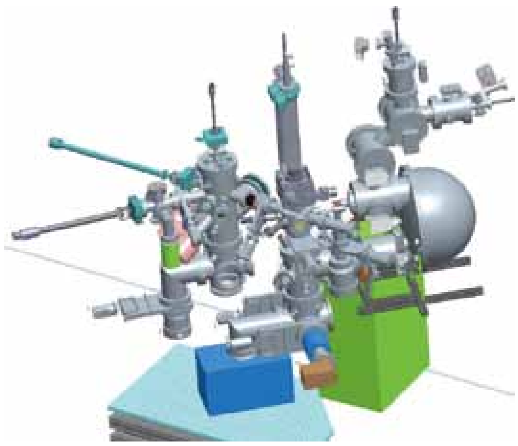

Figure 3. Schematic figure of the spin-, symmetry-, and momentum-resolved photoemission end station of BL5U, UVSOR-III.

References

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* Present Position; Professor, Graduate School of Frontier Bioscience, Osaka University

† carrying out graduate research on Cooperative Education Program of IMS with Nagoya University