Synchrotron Radiation Spectroscopy on Strongly Correlated Electron Systems

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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. Existence of Heavy Fermions in the Antiferromagnetic Phase of $CeIn_3$[1)]

Recently, physics at the quantum critical point (QCP), which is the border between local magnetism and itinerant paramagnetism at zero temperature, has become one of the main topics in the condensed-matter field because new quantum properties such as non-BCS superconductivity appear in the vicinity of the QCP. The ground state of rare-earth intermetallic compounds, namely, heavy-fermion (HF) materials, changes between the local magnetic and itinerant nonmagnetic states through external perturbation by such factors as pressure and magnetic field. The QCP appears owing to the energy balance between the local magnetic state based on the Ruderman–Kittel–Kasuya–Yoshida (RKKY) interaction and the itinerant HF state due to the Kondo effect. In the itinerant HF regime, the conduction band hybridizes with the nearly local 4*f* state, so that a large Fermi surface as well as the hybridization band between them, namely, the *c*–*f* hybridization band, is realized. In the case of a magnetic regime, on the other hand, two theoretical scenarios have been proposed. One is the spin-density wave (SDW) scenario based on spin fluctuation, in which large Fermi surfaces due to *c*–*f* hybridization remain even in magnetically ordered states. The other is the Kondo breakdown (KBD) scenario, in which the *c*–*f* hybridization state disappears in the magnetic state and only small Fermi surfaces due to conduction electrons appear. Many controversies for these scenarios have been performed so far, but the conclusion has not been obtained yet.

Here, we report the pressure-dependent electronic structure as well as the *c*–*f* hybridization state obtained by far-infrared reflectivity [$R(\omega)$] and optical conductivity [$\sigma(\omega)$] measurements of $CeIn_3$ under pressure. $CeIn_3$ has an AFM ground state with a Néel temperature T_N of 10 K. With the

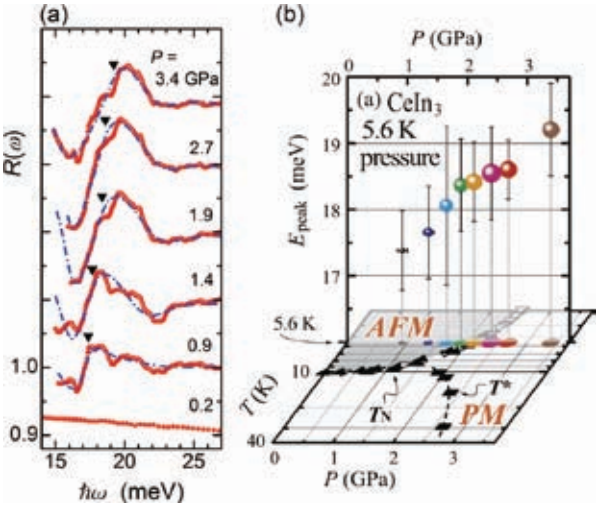

Figure 1. (a) Pressure dependence of the reflectivity [$R(\omega)$] spectrum (solid circles) of $CeIn_3$ and Drude-Lorentz fitting results (dotted-dashed lines) in the photon energy range of 14–27 meV at 5.6 K. The spectra are shifted by 0.1 for clarity. (b) The edges of $R(\omega)$ spectra as functions of pressure. The size of the marks denotes the intensity of the corresponding edge in the $R(\omega)$ spectra. The pressure-dependent Néel temperature (solid and open triangles, T_N) and valence transition temperature (solid squares, T^*) are also plotted at the bottom.

application of pressure, T_N monotonically decreases and disappears at a critical pressure of approximately 2.6 GPa. We observed that the *c–f* hybridization gap appears not only in the HF state but also in the AFM state, and both the energy and intensity of the σ(ω) peak due to the *c–f* hybridization band continuously increase with the application of pressure as shown in Figure 1. Our observations suggest that the electronic structure of $CeIn_3$ in the AFM phase can be explained by the SDW scenario because the *c–f* hybridization state exists even in the AFM phase.

2. Three-Dimensional Electronic Structure and Interband Nesting in the Stoichiometric Superconductor LiFeAs[2)]

Recently discovered iron pnictide superconductors have two-dimensional (2D) Fe–As layers that are similar to the Cu–O planes in high-T_c superconducting cuprates. In high-T_c cuprates, 2D magnetic interaction is important for the origin of the high T_c because of their 2D electronic structure. Such 2D interaction as well as the 2D nesting condition in iron pnictides has long been a focus of discussions. However, the crystal structures of iron pnictides are more three dimensional (3D) than those of cuprates. It is therefore important to clarify the 3D electronic structure of iron pnictides in order to understand the effective interaction of Cooper pair formation.

We reported the electronic structure as well as the orbital characters of a stoichiometric iron-based superconductor LiFeAs (T_c = 18 K) using polarization-dependent 3D angle-resolved photoemission spectroscopy (ARPES) as shown in Figure 2. The obtained band dispersions and orbital characters are qualitatively in good agreement with those derived from local density approximation (LDA) band calculations. Considering a 3D nesting condition, we find that each 2D hole and electron Fermi surface (FS) of d_{xy} orbital character is weakly nested. This weak nesting suggests that (π,π,π) interband scattering is important for the superconducting behavior of LiFeAs.

3. Momentum-Dependent Hybridization Gap and Dispersive In-Gap State of the Kondo Semiconductor SmB_6[3)]

Materials with strong electron correlation have exotic physical properties that cannot be predicted from first-principle band calculations. One example may be seen in a semiconductor with a very small energy gap, which appears in rare-earth compounds such as the Kondo semiconductor or insulator (KI). At high temperatures, KI behaves as a dense Kondo metal, while an energy gap with activation energy of several 10 meV appears at low temperature. The energy gap is believed to originate from hybridization between the nearly localized 4*f* state near the Fermi level (E_F) and the conduction band (*c–f* hybridization).

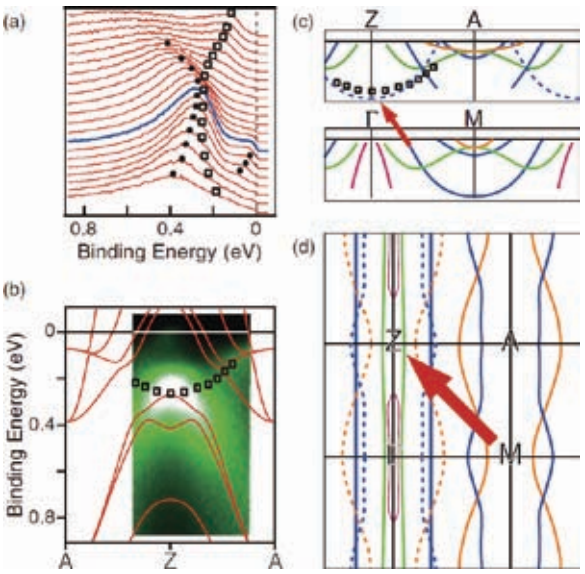

Figure 2. (a)ARPES spectra at the Z point. The solid circles are values that are expected and the open squares are those that are not expected by the band calculation. (b) ARPES image with the band calculation at the Z point. (c) Schematic band dispersions obtained from the experiments. The open squares used in (c) have the same meaning as in (a) and (b). (d) Schematic figure of the 3D FS nesting conditions. The solid and dashed lines indicate the original and nested FSs, respectively. The bold arrow indicates the expected nesting wave vector of (π,π,π).

We reported the temperature dependence of the dispersion curve of the hybridization state using temperature-dependent 3D-ARPES, in order to determine the electronic structure and the reason for the different temperature dependences of the valence transition and magnetic excitation. We found that the hybridization band with a peak at a binding energy of 15 meV near the X point gradually appears on cooling from 150 to 40 K, which has the same temperature dependence as the valence transition. At the Γ point, on the other hand, the peak at E_B ~ 20 meV has the same temperature dependence as the magnetic excitation at Q = (0.5,0.5,0.5), which differs from the 15-meV peak at the X point. This suggests that the magnetic excitation originates from the hybridization band at the Γ point.

References

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