Exploring Novel Physical Properties by Multi-Dimensional Spectroscopy

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Physical and chemical properties of solids, such as conductivity, magnetism, superconductivity, and chemical reactions, originate from microscopic electronic structure, lattice/molecular vibrations, and molecular movements based on quantum mechanics in materials and their interactions. By revealing the microscopic states and their evolution, we can learn about the origin of physical and chemical properties and hidden functionalities. Also, the microscopic information is helpful for the creation of novel functional properties. To visualize hidden microscopic information, we develop novel spectroscopic techniques using synchrotron radiation, high brilliant electron beams, and other so-called quantum beams. We have started a novel electron spectroscopy technique, Spin-Resolved resonant Electron-Energy-Loss Spectroscopy (SR-rEELS), with bulk-sensitive primary energies of 0.3–1.5 keV. At present, we combine it with a time- and angle-resolved technique, shown in Figure 1, to simultaneously observe both the changing electronic structure and collective excitations and the lattice and magnetic structure relaxation. Based on the obtained information on electronic structures, we aim to develop novel physical properties of new materials.

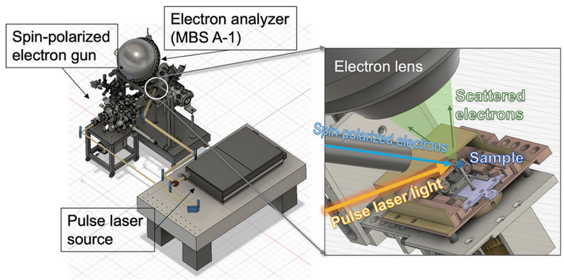

Figure 1. Time-, Spin-, and Angle-Resolved resonant Electron-Energy-Loss Spectroscopy (tSAR-rEELS) apparatus, which our group is now developing. The apparatus comprises a high-brilliant spin-polarized electron gun, a photoelectron spectrometer, and a femto-second pulse laser with an optical parametric amplifier.

Selected Publications

- T. Nakamura, H. Sugihara, Y. Chen, R. Yukawa, Y. Ohtsubo, K. Tanaka, M. Kitamura, H. Kumigashira and S. Kimura, "Two-Dimensional Heavy Fermion in Monoatomic-Layer Kondo Lattice $YbCu_2$," *Nat. Commun.* **14**, 7850 (7 pages) (2023).
- Y. Ohtsubo, T. Nakaya, T. Nakamura, P. Le Fèvre, F. Bertran, F. Iga and S. Kimura, "Breakdown of Bulk-Projected Isotropy in Surface Electronic States of Topological Kondo Insulator SmB_6(001)," *Nat. Commun.* **13**, 5600 (7 pages) (2022).
- S. Kimura, T. Kawabata, H. Matsumoto, Y. Ohta, A. Yoshizumi, Y. Yoshida, T. Yamashita, H. Watanabe, Y. Ohtsubo, N. Yamamoto and X. Jin, "Bulk-Sensitive Spin-Resolved Resonant Electron Energy-Loss Spectroscopy (SR-rEELS): Observation of Element- and Spin-Selective Bulk Plasmons," *Rev. Sci. Instrum.* **92**, 093103 (8 pages) (2021).
- K. Hagiwara, Y. Ohtsubo, M. Matsunami, S. Ideta, K. Tanaka, H. Miyazaki, J. E. Rault, P. Le Fèvre, F. Bertran, A. Taleb-Ibrahimi, R. Yukawa, M. Kobayashi, K. Horiba, H. Kumigashira, K. Sumida, T. Okuda, F. Iga and S. Kimura, "Surface Kondo Effect and Non-Trivial Metallic State of the Kondo Insulator YbB_{12}," *Nat. Commun.* **7**, 12690 (7 pages) (2016).

1. Two-Dimensional Heavy Fermion in Monoatomic-Layer Kondo Lattice $YbCu_2$[1)]

The Kondo effect between localized *f*-electrons and conductive carriers leads to exotic physical phenomena. Among them, heavy-fermion (HF) systems, in which massive effective carriers appear due to the Kondo effect, have fascinated many researchers. Dimensionality is also an important characteristic of the HF system, especially because it is strongly related to quantum criticality. However, the realization of the perfect two-dimensional (2D) HF materials is still a challenging topic. Here, we report the surface electronic structure of the monoatomic-layer Kondo lattice $YbCu_2$ on a Cu(111) surface observed by synchrotron-based angle-resolved photoemission spectroscopy. The 2D conducting band and the Yb 4*f* state are observed very close to the Fermi level. These bands are hybridized at low temperatures, forming the 2D HF state, with an evaluated coherence temperature of about 30 K. The effective mass of the 2D state is enhanced by a factor of 100 by the development of the HF state. Furthermore, clear evidence of the hybridization gap formation in the temperature dependence of the Kondo-resonance peak has been observed below the coherence temperature. Our study provides a new candidate as an ideal 2D HF material for understanding the Kondo effect at low dimensions.

2. Observation of Electronic Structure Modification in the Hidden Order Phase of CeCoSi[2)]

CeCoSi with no local inversion symmetric crystal structure ($P4/nmm$) exhibits a phase transition of unknown origin (Hidden Order: HO) at about 12 K (T_0) above the antiferromagnetic transition temperature (T_N = 9.4 K). The electronic structure change across T_0 was investigated with high-precision optical reflection spectroscopy. The optical spectrum changed from a typical metallic behavior above T_0 to a gap-like structure at around 15 meV below T_0. The gap-like structure was unchanged across T_N except for the narrowing of the Drude component of carriers due to the suppression of magnetic fluctuations. This result suggests a slight change from the typical metallic electronic structure above T_0 to that with an energy gap near the Fermi level in the HO phase. The change in electronic structure in the HO phase was concluded to be due to electron/valence instability.

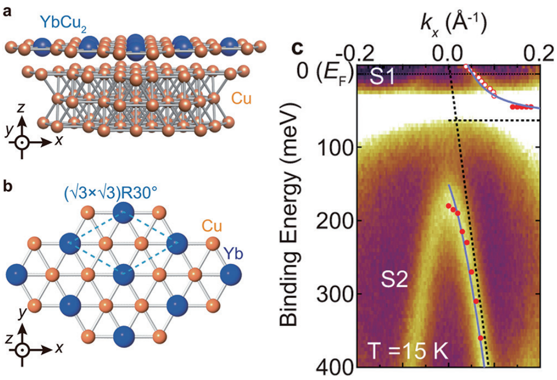

Figure 2. (a) A surface atomic structure of $YbCu_2$/Cu(111). (b) Top view of monoatomic-layer $YbCu_2$. The dashed line indicates the unit cell of $YbCu_2$. (c) ARPES image near the center of the surface Brillouin zone taken with circularly polarized 35-eV photons at 15 K. ARPES intensities are divided by the Fermi–Dirac distribution function convolved with the instrumental resolution. The filled and break lines indicate the simulated *c-f* hybridization band dispersions with the hybridization energy of 120 meV (solid lines) and 0 meV (dashed lines) by the periodic Anderson model. The open and filled circles indicate the peak positions from energy distribution curves (EDCs) and MDCs, respectively.

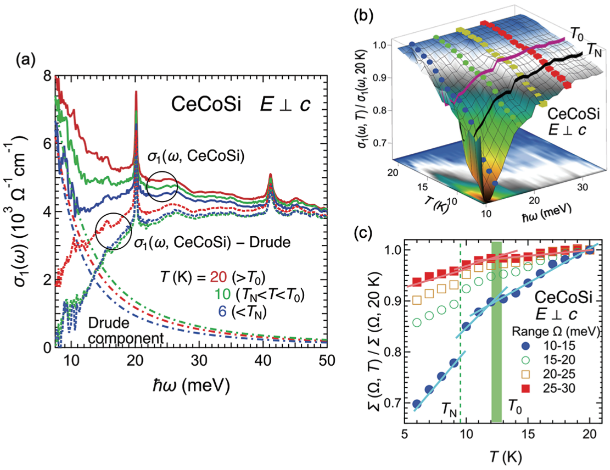

Figure 3. (a) Optical conductivity [$\sigma_1(\omega)$] spectra of CeCoSi at representative temperatures of 20 K (> T_0), 10 K ($T_N < T < T_0$), and 6 K (< T_N) (solid lines). Dot-dashed lines are Drude curves obtained from the values of electrical resistivity and the $\sigma_1(\omega)$ values at $\hbar\omega \sim 5$ meV. The interband components evaluated by subtracting the Drude components from the $\sigma_1(\omega)$ spectra are shown by dashed lines. (b) Temperature-dependent $\sigma_1(\omega)$ spectra normalized by that at 20 K of CeCoSi. The marks are the same as those in (c). (c) Temperature dependence of the spectral integrations of the range Ω of 10–15 meV (solid circle), 15–20 meV (open circle), 20–25 meV (open square), and 20–40 meV (solid square) in (b), which are representative regions of relative $\sigma_1(\omega)$ spectra in (b). The integrated intensity at each Ω is normalized by the value at T = 20 K. Solid lines are guides for the eye. T_0 and T_N are shown by vertical solid and dashed lines, respectively.

References

1) T. Nakamura, H. Sugihara, Y. Chen, R. Yukawa, Y. Ohtsubo, K. Tanaka, M. Kitamura, H. Kumigashira and S. Kimura, *Nat. Commun.* **14**, 7850 (7 pages) (2023).
2) S. Kimura, H. Watanabe, S. Tatsukawa and H. Tanida, *J. Phys. Soc. Jpn.* **92**, 043704 (5 pages) (2023).