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, 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 are now developing a new electron spectroscopy technique, Spin-Resolved resonant Electron-Energy-Loss Spectroscopy (SR-rEELS), with bulk-sensitive primary energies of 0.3–1.5 keV, as shown in Figure 1, in order to detect spin-selective element-specific bulk plasmons. Based on the obtained information of electronic structures, we aim to develop novel physical properties of new materials.

Selected Publications

1. Optical Study of the Electronic Structure of Locally Noncentrosymmetric CeRh$_2$As$_2$\textsuperscript{1)}

The electronic structures of the heavy-fermion superconductor CeRh$_2$As$_2$ with local inversion symmetry breaking and the reference material LaRh$_2$As$_2$ have been investigated using experimental optical conductivity [$\sigma_1(\omega)$] spectra and first-principles density functional theory calculations. The low-temperature $\sigma_1(\omega)$ spectra of LaRh$_2$As$_2$ revealed a broad peak at $\sim$0.1 eV and a sharp peak at $\sim$0.5 eV after subtracting the Drude contribution of free carriers. The peak features and the background intensity were nicely reproduced in calculated $\sigma_1(\omega)$ spectra from DFT calculations, implying a conventional metallic nature. In CeRh$_2$As$_2$, two mid-IR peaks at about 0.12 and 0.4 eV corresponding to the unoccupied Ce 4$f_{5/2}$ and 4$f_{7/2}$ states, respectively, were strongly developed with decreasing temperature as shown in Figure 2, which suggests the emergence of hybridization states between the conduction and 4$f$ electrons. We compared the temperature dependence of the mid-IR peaks of CeRh$_2$As$_2$ with corresponding data from CeCu$_2$Si$_2$ and CeNi$_2$Ge$_2$ in a ThCr$_2$Si$_2$-type structure to examine the possible impact of local inversion symmetry breaking on electronic structures. We also clarify the local and itinerant character in the electronic structure by investigating the temperature dependence in the $\sigma_1(\omega)$ spectra of various Ce and Yb compounds with a tetragonal ThCr$_2$Si$_2$-type crystal structure.\textsuperscript{2)} The temperature variation in the $\sigma_1(\omega)$ spectrum is still present in the more localized case, even though the Kondo effect is strongly suppressed.

2. Bulk-Sensitive Spin-Resolved Resonant Electron Energy-Loss Spectroscopy (SR-rEELS): Observation of Element- and Spin-Selective Bulk Plasmons\textsuperscript{3)}

We have developed spin-resolved resonant electron energy-loss spectroscopy (SR-rEELS) with the primary energy of 0.3–1.5 keV, which corresponds to the core excitations of 2p–3d absorption of transition metals and 3d–4f absorption of rare-earths, with the energy resolution of about 100 meV using a spin-polarized electron source as a GaAs/GaAsP strained superlattice photocathode. Element- and spin-selective carrier and valence plasmons can be observed using the resonance enhancement of core absorptions and electron spin polarization. The Ni 2p–3d rEELS of nickel monoxide NiO as an example is shown in Figure 3. Furthermore, bulk-sensitive electron energy-loss spectroscopy spectra can be obtained because the primary energy corresponds to the mean free path of 1–10 nm. The methodology is expected to provide us with novel information about elementary excitations by resonant inelastic x-ray scattering and resonant photoelectron spectroscopy.

![Figure 2. Temperature-dependent optical conductivity [$\sigma_1(\omega)$] spectra of CeRh$_2$As$_2$ and LaRh$_2$As$_2$.](image1)

![Figure 3. Resonant EELS spectra of nickel monoxide NiO with the primary energies ($E_i$) of 800–900 eV. The overall spectral intensity is enhanced at $E_i = 850$ and 870 eV near the Ni 2d–3d absorption edge.](image2)

References