

Synchrotron Radiation Spectroscopy on Strongly Correlated Electron Systems

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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 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 Momentum-Dependent Exchange Interaction in a Heisenberg Ferromagnet¹⁾

Local magnetism is generally believed to originate from the direct interaction among isolated electron spins described by the Heisenberg model, in which the electronic states with local magnetic moments are perfectly localized. In rare-earth compounds, instead of direct exchange interaction, superexchange interaction mediated by ligand valence electrons and indirect exchange interaction by conduction electrons among rare-earth ions are dominant because the 4*f* state in rare-earth ions is spatially localized. In the case of the superexchange and indirect exchange interactions, hybridization between the 4*f* electrons with local magnetic moments and the valence/conduction electrons is important. Then we measured the temperature-dependent three-dimensional angle-resolved photoemission spectra of EuO(100) thin film, a typical Heisenberg ferromagnetic semiconductor, to investigate the essential origin of the ferromagnetic transition. We observed sizable

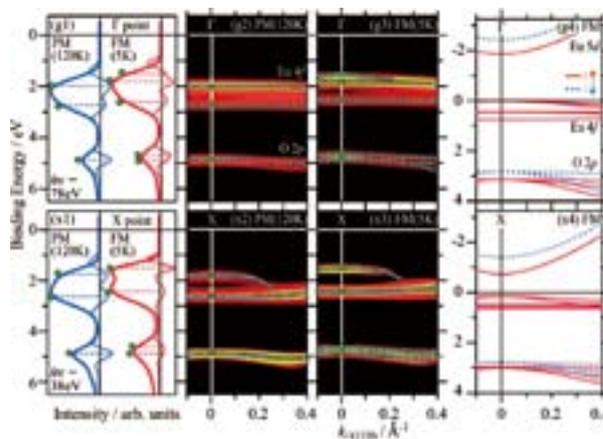


Figure 1. Energy distribution curves (EDCs, thick lines) and their second-derivative EDCs (thin lines) of EuO(100) thin film in the paramagnetic phase (PM) at 120 K and in the ferromagnetic phase (FM) at 5 K at the Γ (g1) and X (x1) points. The dashed lines indicate the peaks in the second-derivative EDCs that correspond to the peaks and shoulders in the EDCs. The 3D ARPES images were derived from the second-derivative EDCs in the paramagnetic phase at 120 K and in the ferromagnetic phase at 5 K near the Γ [(g2), (g3)] and X [(x2), (x3)] points. The open circles in (g2), (g3), (x2), and (x3) indicate the peaks and shoulders in the 3D ARPES images. The band structures calculated by the LSDA+*U* (*U* = 7 eV) method are shown near the Γ (g4) and X (x4) points. The solid and dashed lines in (g4) and (x4) denote the majority (\uparrow) and minority (\downarrow) spin states, respectively.

energy dispersion and large binding-energy shift of the Eu 4*f* state below the Curie temperature only near the Γ and X points, despite the expected Heisenberg-type local magnetism. The band dispersion and temperature dependence of the Eu 4*f* state indicates that the indirect exchange and superexchange interactions have strong momentum dependence. The observed temperature-dependent energy shift of the 4*f* state is the essential origin of the magnetism of EuO.

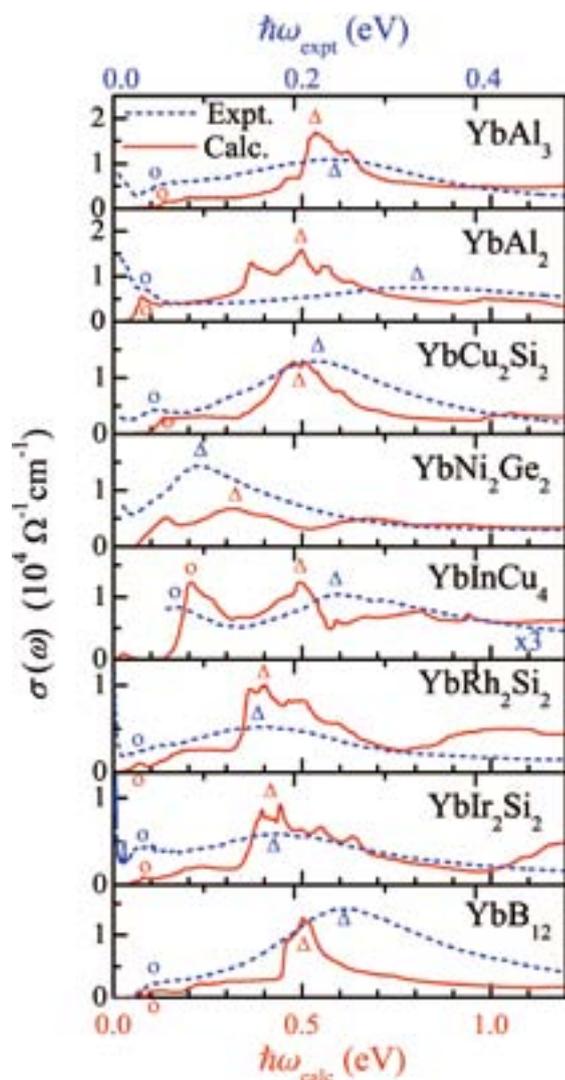


Figure 2. Calculated optical conductivity [$\sigma(\omega)$] spectra compared with experimental spectra of YbAl_3 , YbAl_2 , YbCu_2Si_2 , YbNi_2Ge_2 , YbInCu_4 , YbRh_2Si_2 , YbIr_2Si_2 , and YbB_{12} . The $\sigma(\omega)$ spectra of YbCu_2Si_2 , YbNi_2Ge_2 , YbRh_2Si_2 , and YbIr_2Si_2 were calculated along the a -axis, in the same way as the experimental spectra. The horizontal scales of the experimental and calculated spectra are located at the top and bottom, respectively. Note that the horizontal scale of the calculated $\sigma(\omega)$ spectra is reduced by a factor of 0.42 ($= 0.5/1.2$) compared with that of the experimental spectra. The vertical scales of the calculated and experimental $\sigma(\omega)$ spectra are identical except in the case of YbInCu_4 , in which the experimental spectrum is three times as large as the calculated spectrum. The mid-IR peaks and far-IR shoulders of the experimental and calculated spectra are marked by open triangles and open circles, respectively.

(YbAl_3 , YbAl_2 , YbCu_2Si_2 , YbNi_2Ge_2 , YbInCu_4 , YbRh_2Si_2 , YbIr_2Si_2 , and YbB_{12}) can be explained by first-principle band structure calculation with the spin-orbit interaction from the comparison with the results with the experimentally obtained $\sigma(\omega)$ spectra. On CeX_3 , the mid-IR peak shapes are not identical to one another: CePd_3 , CeSn_3 , and CeIn_3 have a triple-peak structure, a double-peak structure, and a broad single-peak structure, respectively. These peaks can be theoretically explained by the optical transition from the occupied state to the spin-orbit splitted Ce $4f$ state. On the other hand, in Yb compounds, the spectral feature of a peak in the middle-infrared (IR) region (mid-IR peak) and a shoulder structure in the far-infrared region (far-IR shoulder) in the experimental $\sigma(\omega)$ spectra can be described by the band calculation with a common renormalization factor of $z = 0.42$. These results suggested that the infrared spectra of Ce and Yb compounds originate from the own band structure but that the Yb $4f$ state shifts to the Fermi level with strong electron correlation.

2. Origin of Infrared Peaks in the Optical Conductivity of Cerium and Ytterbium Compounds^{2,3)}

We have demonstrated that the middle-infrared (mid-IR) peaks commonly appearing in the optical conductivity [$\sigma(\omega)$] spectra of CeX_3 ($X = \text{Pd}, \text{Sn}, \text{In}$) and ytterbium compounds

Awards

KIMURA, Shin-ichi; Morita Memorial Award, The Alumni Association of Physics Department “Senshu-kai,” Tohoku University.
MIYAZAKI, Hidetoshi; Best Presentation Award, PF Workshop on Recent Research Activities and Future Prospect of Angle-Resolved Photoemission Spectroscopy.

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