Exploring Novel Physical Properties by Multi-Dimensional Spectroscopy

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Awards
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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.

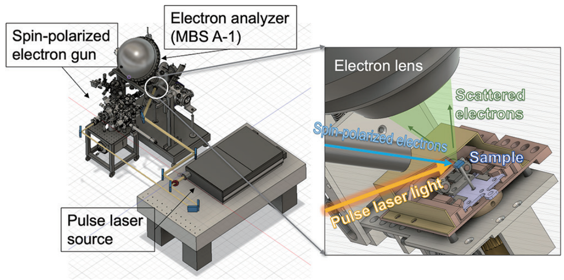

Figure 1. Time-, Spin-, and Angle-Resolved resonant Electron-Energy-Loss Spectroscopy (tSAR-rEELS) apparatus, which our group is now developing. The apparatus comprises a high-brilliant spin-polarized electron gun, a photoelectron spectrometer, and a femtosecond pulse laser with an optical parametric 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 Fèrve, 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. Current- and Photo-Induced Phase Transition on Black Samarium Monosulfide[1,2]

A strongly correlated insulator, samarium mono-sulfide (SmS), is well-known to present a pressure-induced insulator-to-metal transition (IMT) with the color change from black to golden-yellow. Recently, it has also shown current-induced IMT (CIMT) with negative resistance. To clarify the origin of the CIMT of SmS and also the relation to the pressure-induced IMT, the electronic structure change has been investigated by optical reflectivity and angle-integrated photoelectron spectra by applying an electric current. At lower temperatures than about 100 K, where the nonlinear *V–I* curve has been observed, the carrier density rapidly increases, accompanied by decreasing relaxation time of carriers with increasing current. Then, the direct gap size increases, and the mean valence changes from Sm^{2+}-dominant SmS to the mixed-valent one with increasing current. These results suggest that the CIMT originates from increasing the Sm 4*f*–5*d* hybridization intensity induced by the applied current.

One scenario for the pressure-induced IMT of SmS is exciton condensations with decreasing energy gap by pressure. To investigate the role of the excitons, optical reflectivity, Sm 3*d* x-ray absorption spectroscopy (XAS), and x-ray diffraction (XRD) with the creation of excitons by photoexcitation (PE) are reported. In the pump–probe reflectivity measurement, following a huge reflectivity change of about 22%, three different relaxation times with a vibration component were observed. The fast component with the relaxation time (τ) of less than 1 ps is due to the excitation and relaxation of electrons into the conduction band, and the slowest one with $\tau >$ several 100 ps originates from the appearance of the photo-induced (PI) state. The components with $\tau \sim 10$ ps and vibration originate from the appearance of the PI state and the interference between the reflection lights at the sample surface and the boundary between the black-insulating and PI states, suggesting that the electronic structure of the PI phase is different from that of the black insulating state. XAS spectra indicate that the Sm mean valence is shifted from the Sm^{2+} dominant to the intermediate between Sm^{2+} and Sm^{3+} by PE, but did not change to that of the golden metallic phase across the IMT, consistent with the reflectivity data. The XRD result after PE shows that the PI state has much less lattice contraction than the golden metallic phase. These results suggest that the IMT cannot be achieved solely by creating excitons after PE but requires other effects, such as a lattice contraction. The photo-induced phase transition and the golden metallic phase are schematically explained in Figure 2.

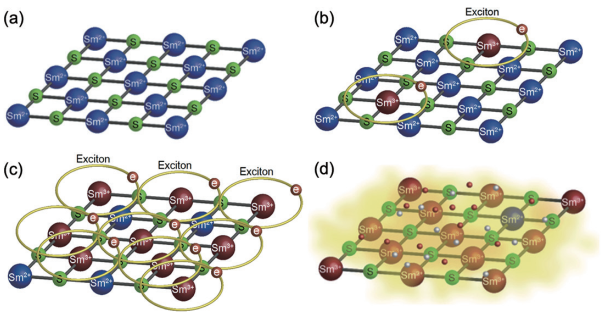

Figure 2. Schematic figure of the photo-induced phase transition along the excitonic instability picture. (a) The black insulating phase. (b) After the photoexcitation (PE) by several pulses irradiated to the sample, 4*f* electrons are excited and become excitons. The created excitons are isolated and localized at the original sites. The electronic structure and optical constants are slightly changed, but the lattice constant is identical before the PE. (c) After many laser pulses PE with a pile-up effect, many excitons are created but still isolated. The valence transition becomes visible by XAS because of many Sm^{3+} states. (d) The golden metallic phase, where excitons are condensed, and the state becomes metallic.

2. Light-Field-Driven Non-Ohmic Current Generation by an Intense THz Pulse in a Weyl Semimetal[3]

In recent years, coherent electrons driven by light fields have attracted significant interest in exploring novel material phases and functionalities. However, observing coherent light-field-driven electron dynamics in solids is challenging because the electrons are scattered within several tens of femtoseconds in ordinary materials, and the coherence between light and electrons is disturbed. This study presents the light-field-driven dynamics by applying a THz pulse (~1 ps) to the Weyl semimetal $Co_3Sn_2S_2$, which has a relatively long coherent time to pico-seconds. As the electric-field intensity of the irradiating THz pulse was increased, the reflected/emitted THz wave changed from being similar to the incident THz wave to an asymmetric electric field. This asymmetric electric field emission suggests the generation of non-Ohmic direct current *via* coherent acceleration, and the fact that its intensity dependence is proportional to the square of the electric field suggests electronic excitation by the Landau–Zener transition, a characteristic of the light-field picture.

References

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