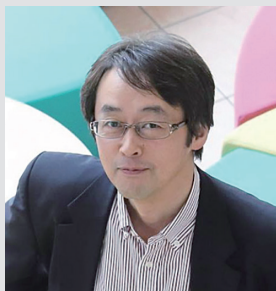


Exploring Novel Physical Properties by Multi-Dimensional Spectroscopy

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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.

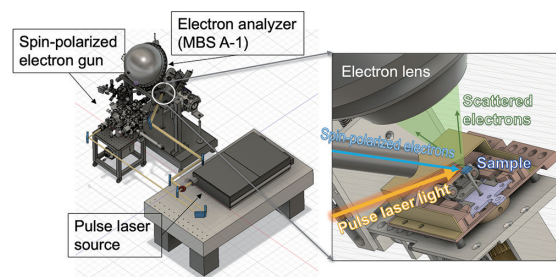


Figure 1. Time-, Spin-, and Angle-Resolved resonant Electron-Energy-Loss Spectroscopy (tSAR-rEELS) apparatus, which our group is now developing. The apparatus comprises a high-brilliant spin-polarized electron gun, a photoelectron spectrometer, and a femto-second pulse laser with an optical parametric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

Selected Publications

- 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 $\text{SmB}_6(001)$,” *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).
- S. Kimura and H. Okamura, “Infrared and Terahertz Spectroscopy of Strongly Correlated Electron Systems under Extreme Conditions,” *J. Phys. Soc. Jpn.* **82**, 021004 (28 pages) (2013). [review]

1. Breakdown of Bulk-Projected Isotropy in Surface Electronic States of Topological Kondo Insulator $\text{SbB}_6(001)$ ¹⁾

The topology and spin-orbital polarization of two-dimensional (2D) surface electronic states have been extensively studied in this decade. One major interest in them is their close relationship with the parities of the bulk (3D) electronic states. In this context, the surface is often regarded as a simple truncation of the bulk crystal. Here we show a breakdown of the bulk-related in-plane rotation symmetry in the topological surface states (TSSs) of the Kondo insulator SbB_6 .²⁾ Angle-resolved photoelectron spectroscopy (ARPES) performed on the vicinal $\text{SbB}_6(001)-p(2\times 2)$ surface showed that TSSs are anisotropic and that the Fermi contour lacks the fourfold rotation symmetry maintained in the bulk. This result emphasizes the important role of the surface atomic structure even in TSSs. Moreover, it suggests that the engineering of surface atomic structure could provide a new pathway to tailor various properties among TSSs, such as anisotropic surface conductivity, nesting of surface Fermi contours, or the number and position of van Hove singularities in 2D reciprocal space.

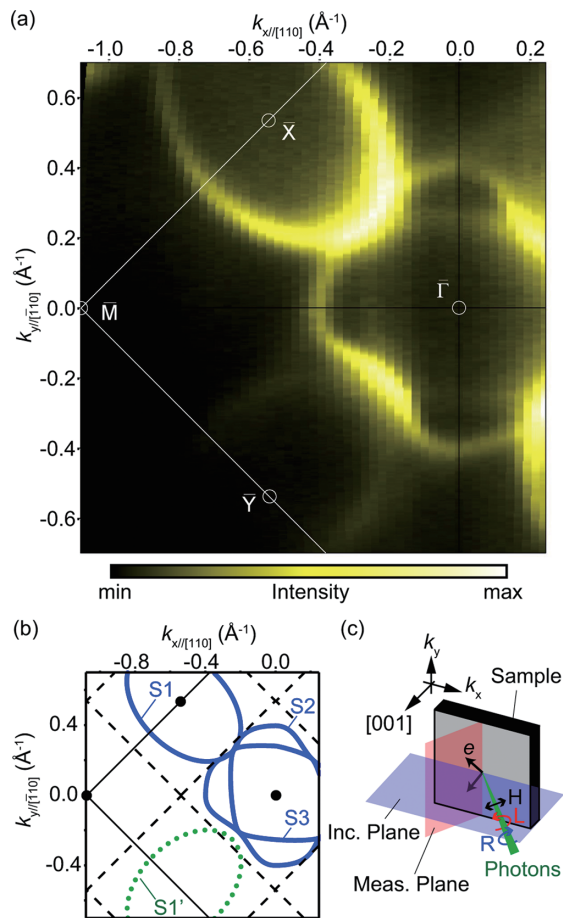


Figure 2. (a) ARPES Fermi contours (FCs) taken at the photon-incident plane of $(\bar{1}10)$ at 13 K. (b) Schematic drawing of the observed FCs together with the border of surface Brillouin zones; solid lines for bulk-truncated (1×1) and dashed for (2×2) . (c) Experimental geometry and definition of the in-plane wavevectors k_x and k_y . k_x and k_y are always in the photon-incident and photoelectron detection planes, respectively.

2. Fluctuating Spin-Orbital Texture of Rashba-Split Surface States in Real and Reciprocal Space³⁾

Spin-orbit interaction in low-dimensional systems, namely, Rashba systems and the edge states of topological materials, has been extensively studied in this decade as a promising source to realize various fascinating spintronic phenomena, such as the source of the spin current and spin-mediated energy conversion. Here, we show the odd fluctuation in the spin-orbital texture in a surface Rashba system on $\text{Bi}/\text{InAs}(110)-(2\times 1)$ by spin- and angle-resolved photoelectron spectroscopy and a numerical simulation based on a density-functional theory (DFT) calculation. The surface state shows a paired parabolic dispersion with the spin degeneracy lifted by the Rashba effect. Although its spin polarization should be fixed in a particular direction based on the Rashba model, the observed spin polarization varies greatly. It even reverses its sign depending on the wave number. DFT calculations also reveal that the spin directions of two inequivalent Bi chains on the surface change from nearly parallel (canted parallel) to antiparallel in real space in the corresponding wave vector region. These results indicate an oversimplification of the nature of spin in Rashba and Dirac systems and provide more freedom than expected for spin manipulation of photoelectrons.

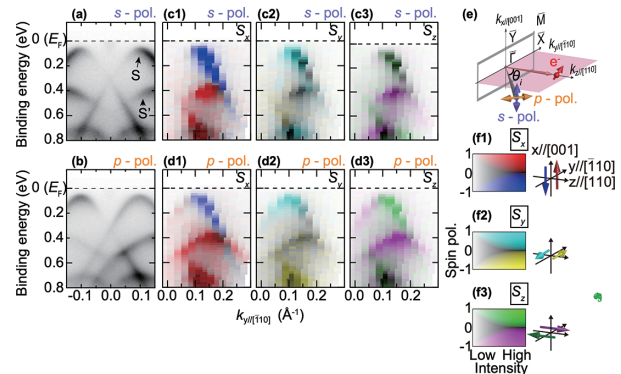


Figure 3. Spin-polarized surface band structures measured by spin- and angle-resolved photoelectron spectroscopy (SARPES) (a) and (b) ARPES and (c) and (d) SARPES 2D plots measured with s-polarized (top) and p-polarized (bottom) photons along $\bar{\Gamma}-\bar{X}$ (parallel to the Bi chains). (a) and (b) Spin-integrated ARPES intensity maps. SARPES maps polarized to (c1) and (d1) S_x , (c2) and (d2) S_y , and (c3) and (d3) S_z . The spin orientations are defined in (f). (e) Experimental geometry of the SARPES measurements and definitions of the coordinates. The (2×1) surface Brillouin zone and the common plane of the photon incidence and photoelectron detection are superposed simultaneously. (f) Definitions of the spin directions of the photoelectrons.

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- 1) Y. Ohtsubo, T. Nakaya, T. Nakamura, P. Le Fèvre, F. Bertran, F. Iga and S. Kimura, *Nat. Commun.* **13**, 5600 (7 pages) (2022).
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