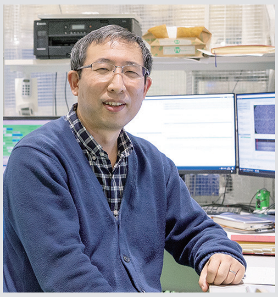
Novel Spin and Chiral Materials Science by Photoelectron Cinemato-Microspectroscopy

UVSOR Synchrotron Facility
Division of Advanced Solid State Physics



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Keywords Photoelectron Spectroscopy, Momentum Microscope, Electronic Spin Structure

Imagine cherry blossom petals fluttering down. Under a uniform gravitational field, the slight asymmetry of the petals creates anisotropy in the airflow, causing the petals to rotate. Capturing the movement of this dynamic 3D-structure as a *cinema* is important for understanding the physics behind this.

When electrons in a material are excited by photons, they are emitted into the vacuum as photoelectrons. Interestingly, the angular distribution of these photoelectrons shows a truly beautiful holographic pattern derived from the motion of valence electrons and the arrangement of atoms in the material. Photoelectron momentum microscope (PMM) is an analyzer that can instantaneously image the behavior of electrons of material and device surfaces. PMM combines imaging-type photoelectron spectroscopy and microscopy techniques to visualize the electronic state (band dispersion, composition, and spin polarization) in reciprocal lattice space of a selected µm-sized area. We have constructed the world's first dual beamline (soft x-ray; SX and vacuum ultraviolet; VUV) PMM system at the IMS UVSOR synchrotron facility!

What we are currently very interested in is chiral phase transition phenomena, for example. The layered material TaS_2 exhibits an attractive behavior in which it undergoes a phase transition to a chiral structure at low temperatures and its electrical conductivity changes by two orders of magnitude. We succeeded in capturing a video of the hysteresis change in the valence band dispersion during this phase transition. The results serve as a benchmark experiment for *in situ* observation of material responses to various external fields.

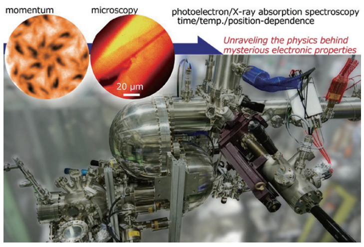

Figure 1. Photoelectron momentum microscope station at UVSOR synchrotron facility. Multimodal measurements hold the key to unlocking the mysteries of the electronic properties of materials.

Selected Publications

- K. Hagiwara, F. Matsui *et al.*, "Development of Dual-Beamline Photoelectron Momentum Microscopy for Valence Orbital Analysis," *J. Synchrotron Radiat.* **31**, 540 (2024).
- F. Matsui *et al.*, "Soft X-Ray Photoelectron Momentum Microscope for Multimodal Valence Band Stereography," *Rev. Sci. Instrum.* **94**, 083701 (2023).
- F. Matsui *et al.*, "Domain-Resolved Photoelectron Microscopy and µm-Scale Momentum-Resolved Photoelectron Spectroscopy of Graphite Armchair Edge Facet," *J. Phys. Soc. Jpn.* **91**, 094703 (2022).
- F. Matsui and S. Suga, "Coupling of k_z-dispersing π Band with Surface Localized States in Graphite," *Phys. Rev. B* **105**, 23526 (2022).
- F. Matsui and H. Matsuda, "Projection-type Electron Spectroscopy Collimator Analyzer for Charged Particles and X-Ray Detections," *Rev. Sci. Instrum.* **92**, 073301 (2021).
- F. Matsui *et al.*, "Photoelectron Momentum Microscope at BL6U of UVSOR-III synchrotron," *Jpn. J. Appl. Phys.* **59**, 067001 (2020).

1. Visualization of Phase Transition and Phase Separation

The most distinctive advantage of PMM[1] is its snapshot-style of measuring photoelectron constant energy intensity distribution pattern in real and reciprocal spaces. Recently, we succeeded in the development of on-the-fly scans for temperature-dependent Fermi surface and valence band measurements during phase transitions, taking advantage of the rapid acquisition (seconds to minutes) of photoelectron patterns.

1T-TaS_2, which exhibits various phase transitions in structural and electronic properties, has fascinated researchers for more than half a century. However, despite many angle-resolved photoelectron spectroscopy (ARPES) studies on 1T-TaS_2, the basic understanding of charge density wave (CDW) formation is far from a common consensus. As for the mechanism of the metal–insulator transition, the Mott insulator model, band insulator model, and one-dimensional metal model have been so far proposed.

In order to solve the mystery of CDW formation in 1T-TaS_2, we conducted the following photoemission measurements.[2] (i) Band-dispersion mapping from selected µm-scale chiral CDW domains, (ii) characterization of 3D CDW nesting vectors by complete Brillouin-zone-dispersion measurements and (iii) on-the-fly observation of valence band and core-level photoelectron spectra during temperature changes.

Figure 2 shows a series of 1T-TaS_2 valence band cross sections scanned in 2D real space in the low-temperature CDW phase. Here, a domain boundary between two different twin domains is recognized. Typical areas of domains are tens to hundreds of µm^2. We exploited the microscopic capabilities of PMM to analyze the electronic structure of selected single domains. By measuring the valence band pattern during temperature change in real time, the hysteresis behavior of the phase transition was visualized in detail.

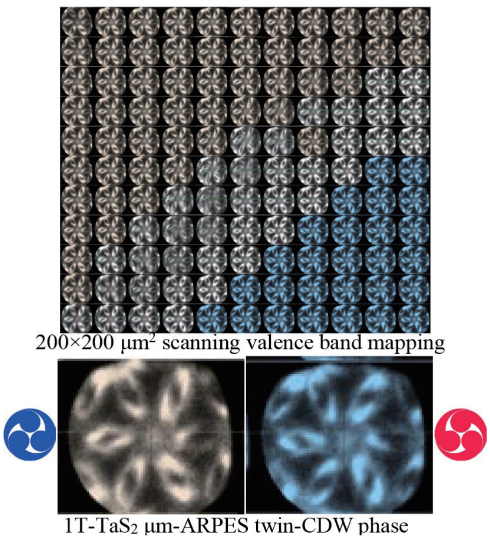

Figure 2. Valence band photoelectron patterns of 1T-TaS_2 surface in the low-temperature CDW phase. The field of view for the ARPES snapshot measurements was a few µm in diameter.

2. Analyzing Polarization of Atomic Orbitals and Electron Spins

The combination of PMM with synchrotron SX and VUV undulators paves the way for comprehensive characterization of atomic orbitals in the Fermi surface and valence band on the µm scale.[3]

Figure 3 shows the 2D momentum (k_x, k_y) distribution of photoelectron intensity of the Au(111) at the Fermi level.[3] Figures 3(a) and (b) show the 2D patterns measured using p-polarized SX light from BL6U, while s-polarized VUV light from BL7U is used in the case of Figures 3(c) and (d). In both conditions, the photoelectron pattern in the entire Brillouin Zone is covered. Shockley surface state centered at the Γ point as small circular contours (Figure 3(a)) is clearly observed with p-polarized excitation with splitting due to spin–orbit coupling (Figure 3(b)), but these features exhibit very weak intensity for normal incidence light (Figure 3(c)). The Shockley surface state mainly comprises 6s and 6p_z orbitals. In the normal-incidence geometry, the transition-matrix element from the initial s and p_z orbitals becomes 0 at the photoemission direction orthogonal to the excited electric vector (Figure 3(d)). The relationship between the orbital angular momentum and the effects of the transition matrix elements can be directly investigated using this normal incidence geometry.

Finally, our main goal is to employ these techniques with spin polarization sensitivity. We have just started obtaining spin-polarized valence band dispersion data for typical materials having spin polarized surface states.

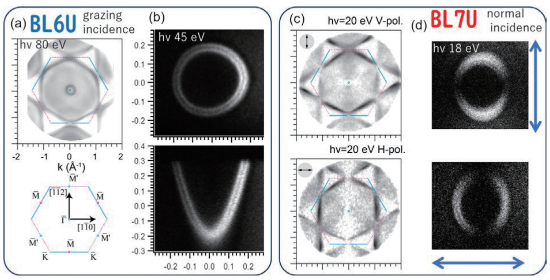

Figure 3. Fermi surface of Au(111) surface measured using grazing incident SX (a,b) and normal incident VUV (c,d) beam. (b,d) The Shockley surface state at the center is zoomed and analyzed. Parabolic band dispersion with splitting due to spin–orbit coupling, *i.e.* Rashba effect, is well resolved.

References

1) F. Matsui, S. Makita, H. Matsuda, T. Yano, E. Nakamura, K. Tanaka, S. Suga and S. Kera, *Jpn. J. Appl. Phys.* **59**, 067001 (2020).
2) F. Matsui, K. Hagiwara, E. Nakamura, T. Yano, H. Matsuda, Y. Okano, S. Kera, E. Hashimoto, S. Koh, K. Ueno, T. Kobayashi, E. Iwamoto, K. Sakamoto, S. Tanaka and S. Suga, *Rev. Sci. Instrum.* **94**, 083701 (2023).
3) K. Hagiwara, E. Nakamura, S. Makita, S. Suga, S. Tanaka, S. Kera and F. Matsui, *J. Synchrotron Radiat.* **31**, 540 (2024).

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