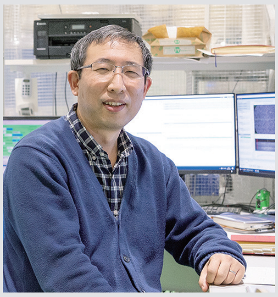
Symmetry Breaking in Crystal Microcosms Reflected in a Photoelectron Kaleidoscope

UVSOR Synchrotron Facility
Division of Advanced Solid State Physics



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The word “*kaleidoscope*” comes from the Greek “*kalos*,” meaning beautiful, and “*eidos*,” meaning shape or form. In a kaleidoscope, light repeatedly reflects off mirrors inside a tube, creating a beautiful pattern. The photoelectron angular distribution we study is similar to this kaleidoscope.

Photoelectron spectroscopy is a technique that uses photon to eject electrons from a solid. By analyzing the ejected electrons’ energy and motion, we can determine the properties of the material. As the photoelectrons travel within a solid crystal, they form a complex angular distribution, much like the patterns in a kaleidoscope. This complex pattern holds astonishing information. The diffraction pattern of core-level photoelectrons reveals the local atomic arrangment of the specific element sites that make up the crystal, and the angular distribution of valence photoelectrons tells us about the fundamental electronic properties of the material.

We have developed a new analytical instrument, the photoelectron momentum microscope (PMM). 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. Specifically, the “broken symmetry” in the kaleidoscopic patterns created by photoelectrons contains crucial information that determines the characteristics of a material.

Our research focuses on a range of intriguing phenomena, including twinning crystal growth, phase transitions, magnetism, and superconductivity. We are particularly fascinated by the interplay between domain boundaries and electronic properties, where the delicate balance between order formation and fluctuations creates a rich and complex environment. We are pioneering a new technique to unravel the mysteries of electronic properties within individual microcrystalline regions (Figure 1).

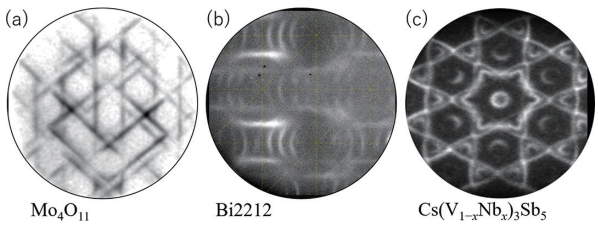

Figure 1. Photoelectron momentum distributions of Fermi surfaces from (a) a quasi-2D crystal, (b) a high-temperature superconducting copper oxide and (c) a Kagome-lattice superconductor.

Selected Publications

- T. Kobayashi, F. Matsui *et al.*, “Temperature-Dependent Electronic Structure of a Quasi-Two-Dimensional Conductor η-Mo_4O_{11},” *Sci. Rep.* **15**, 9034 (2025).
- K. Hagiwara, F. Matsui *et al.*, “Development of Dual-Beamline PMM for Valence Orbital Analysis,” *J. Synchrotron Radiat.* **31**, 540 (2024).
- F. Matsui *et al.*, “Soft X-Ray PMM for Multimodal Valence Band Stereography,” *Rev. Sci. Instrum.* **94**, 083701 (2023).
- T. Kato, F. Matsui *et al.*, “Fermiology and Origin of T_c Enhancement in a Kagome Superconductor $Cs(V_{1-x}Nb_x)_3Sb_5$,” *Phys. Rev. Lett.* **129**, 206402 (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).

1. Photoelectron Feels the Broken Symmetry at the Surface

PMM can also map conduction band dispersion. The conduction band structure is imprinted on the energy-loss background of secondary electrons (SEs) that accompany the primary photoemission process. The interaction of these secondary electrons with the conduction band states reduces their emission intensity into the vacuum, creating a negative contrast pattern of the conduction band.[1,2] This is analogous to the negative photoelectron diffraction patterns seen in the angular distribution of energy-loss electrons accompanying core-level photoemission.[3] Consequently, a standard photoelectron spectroscopy setup, typically used for occupied states, can also visualize the unoccupied conduction band.

Figure 2(a) shows the overall valence band dispersion of graphite, where darker regions indicate a stronger photoelectron signal. We tuned the photon energy to 68 eV to match the bulk L symmetry point. By selectively detecting photoelectrons from a terrace with a single termination type, we confirmed a clear surface symmetry breaking (Figure 2(b)).[4]

Figure 2(c) displays the momentum-resolved kinetic energy distribution of SEs. The unoccupied band dispersion appears as a negative contrast due to the absorption of photoelectrons by the conduction band states. Figure 2(d) presents a series of angular patterns at various kinetic energies. A key finding is that while most patterns exhibit six-fold symmetry, those at kinetic energies around 20 eV show a three-fold symmetry. This suggests that as photoelectrons escape from the solid into the vacuum, they are influenced by the three-fold symmetric structure of the top surface of graphite.

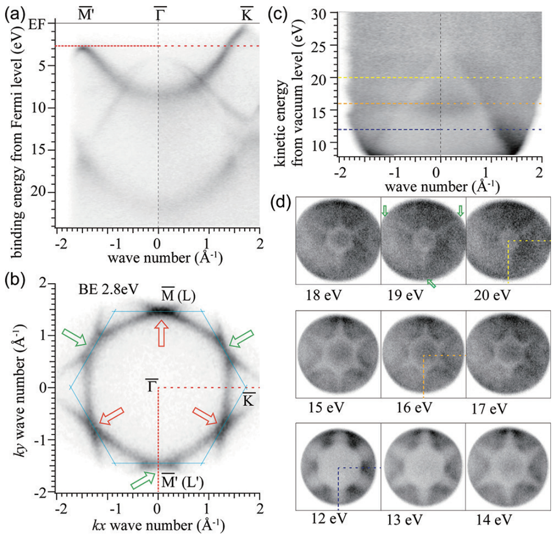

Figure 2. (a) The overall dispersion of the valence band of graphite. Dark areas correspond to larger photoelectron signals. (b) Momentum distribution indicated as a red line in (a). (c) The momentum resolved kinetic energy distribution of SE electrons. The conduction band signature appears as a lighter grey contrast. (d) A series of SE angle patterns at several kinetic energies. Note that at kinetic energies of 19 and 20 eV, the patterns appear as three-fold symmetric.

2. Spins, in Which Direction Are You Oriented?

That's the fundamental question at the heart of spin physics. Spin, a crucial quantum number, enriches the properties of materials, magnetism and superconductivity for instance. To truly understand those phenomena, we need to clarify not only their electronic structures but also the behavior of electron spins.

Spin-polarized photoelectron momentum microscopy (SP-PMM) is a powerful technique for detecting spin polarization in 2D real and reciprocal spaces. Its main advantage is efficiency, providing instant, spin-resolved snapshots of valence band structures. Our SP-PMM at the UVSOR synchrotron facility uses both grazing-incidence soft X-rays and normal-incidence vacuum ultraviolet light. A unique feature of our instrument is a spin rotator placed just before the spin detector, which allows for precise spin manipulation.

The Au(111) surface state is a classic example of spin–orbit coupling. The loss of bulk symmetry at the surface splits the band dispersion into two branches due to the Rashba effect (Figures 3(a), (b)). The in-plane spin components of these split bands are known to point in opposite directions, and observing this state is a common milestone in spin-polarized ARPES.

However, a review of the literature on the Au(111) Rashba state revealed a significant contradiction: The spin orientation of the outer bands is ambiguously assigned as either counterclockwise (ccw) or clockwise (cw). This confusion has been made worse by numerous secondary publications.

To resolve this issue, we performed a case study on the Au(111) surface state using our SP-PMM. By carefully investigating our multichannel detection system, we were able to unambiguously determine the precise spin orientation of the Au(111) surface state (Figure 3(c)).[5]

Our PMM at the UVSOR synchrotron facility uses both grazing-incidence soft X-rays and normal-incidence vacuum ultraviolet (VUV) light with variable polarization. This normal-incidence setup is particularly useful for directly investigating the relationship between orbital angular momentum and transition matrix elements (Figure 3(d)).[6]

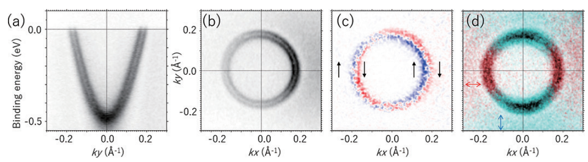

Figure 3. (a) The dispersion of the Au(111) surface state. (b) A cross-section of the surface state at the Fermi level. (c) The determined spin orientation of the surface state. (d) Intensity distributions excited by horizontal (magenta) and vertical (cyan) VUV polarization, respectively.

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