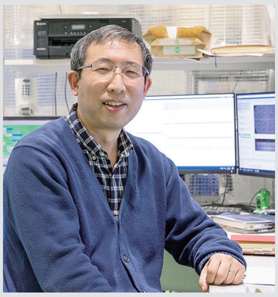
Novel Spin and Chiral Materials Science by Advanced Photoemission Methodologies

UVSOR Synchrotron Facility
Division of Advanced Solid State Physics



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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. Analyzing "art" based on physical laws lead to discoveries that connect the world of atoms with practical technology and applications, and this is what makes us so excited.

We have constructed an advanced photoelectron momentum microscope (PMM) experimental station at the UVSOR Synchrotron Facility of IMS. The PMM is a novel concept analyzer for imaging photoelectron holograms and Fermi surface patterns from the selected µm-sized area. The combination of domain-resolved photoelectron microscopy and µm-scale momentum-resolved photoelectron spectroscopy techniques is essential for the investigation of fragile radiation sensitive materials and complicated phase-separated systems.

Electron spins, which we pay particular attention to, are the source of various physical properties and functions such as magnetism, superconductivity, and topology. We are developing a unique 3D spin vector imaging system and element-selective resonant photoelectron diffraction/spectroscopy technique for the complete photoelectron analysis. We aim to pioneer cutting-edge spin materials science through comprehensive and detailed characterization of electrons.

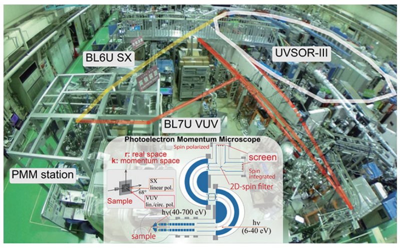

Figure 1. Photoelectron momentum microscope station at UVSOR synchrotron facility. Soft X-rays from BL6U for k_z dispersion and core-level excitations and vacuum ultraviolet light from BL7U at normal incidence for atomic orbital analysis make this station unique.

Selected Publications

- 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, S. Makita, H. Matsuda, E. Nakamura, Y. Okano, T. Yano, S. Kera and S. Suga, "Valence Band Dispersion Embedded in Resonant Auger Electrons," *J. Phys. Soc. Jpn.* **90**, 124710 (2021).
- 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, S. Makita, H. Matsuda, T. Yano, E. Nakamura, K. Tanaka, S. Suga and S. Kera, "Photoelectron Momentum Microscope at BL6U of UVSOR-III synchrotron," *Jpn. J. Appl. Phys.* **59**, 067001 (2020).

1. Chiral Charge Density Wave Revisited

1T-TaS_2 has fascinated researchers for half a century as a system that undergoes a phase transition with three structural changes from high to low temperatures. In the lowest temperature phase, a chiral charge density wave (CDW) structure (√13×√13)-R±13.90° (Figure 2(a) and (b)) is observed by electron diffraction and scanning probe microscopy. Regarding the electronic structure, the first paper on two-dimensional ARPES, which observed the average of both chiral structures,[1] had a strong impact, and it greatly influenced subsequent papers. Only recently has the nesting vector, which is the basis of charge density waves, begun to be accurately discussed in measurements using chiral single domains. In a single layer, unpaired electron spins are isolated in a 13-Ta-atom units, commonly known as the Star of David (six pointed star). Isolated spins are stabilized by interlayer interactions. Recently, STM revealed coexistence of Mott-insulator and band-insulator domains on the cleaved surface,[2] which has attracted attention as a great opportunity to elucidate the CDW mechanism (Figure 2(c) and (d)). Here, spin-resolved micro-ARPES using PMM will be a decisive tool to clariify the physics behind this complex CDW phenomena.

Figures 2(e) and 2(f) shows the constant energy countour and band dispersion of 1T-TaS_2, respectively, measured by UVSOR-PMM[3] at 30 K.[4] The ellipsoidal electron pockets around the M points are modified and exhibit the so-called "windmill" rotational symmetry modulation around the Γ point due to the CDW formation. The microscopic field-of-view enabled selective observation of one of the two types of twinned CDW domains. Although we have expected the formation of a sharp gap on the band dispersion due to CDW, noticeable intensity remained even around the region of $E_{binding}$ = 1 eV and $|k|$ = 1 Å^{-1} (Figure 2(f)). This result may be due to the observed mixture of multiple electronic states in the Mott insulator and band insulator at different surface terminations, which was pointed out in STM.[2] Further detailed PMM works will be described elsewhere.

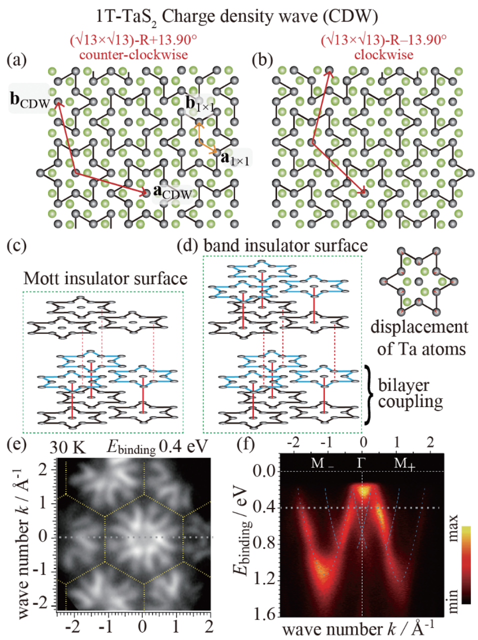

Figure 2. (a) and (b) Lateral atomic structure models of 1T-TaS_2 in the CDW phase. (c) and (d) Structure models of different surface terminations. (e) Constant energy contour at the binding energy of 0.4 eV. (f) Valence band dispersion along the MΓM axis.

2. Original Analyzer for 3D-Atomic Structure Imaging and 3D-Spin Vector Analysis

Photoelectron holography is an element specific 3D atomic imaging technique. Local atomic arrangements of dopant atoms can be characterized. Compositional crossover of multiple-site Ag doping in Bi_2Se_3 from substitution to intercalation was revealed (Figure 3).[5]

We are aiming at highly efficient and comprehensive measurement of atomic structure and spin distribution. Omnidirectional photoelectron acceptance lens (OPAL)[6] together with Projection-type electron spectroscopy collimator analyzer (PESCATORA)[7] enables photoelectron holography measurement of the full hemisphere. Moreover, we invented Right angle deflection imaging analyzer (RADIAN)[8] for spin vector analysis with k/r-space resolution. We are expanding the MM system based on our original inventions.

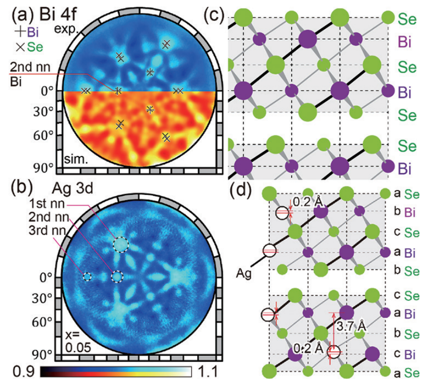

Figure 3. (a) Bi4f and (b) Ag3d photoelectron holograms of Ag-doped Bi_2Se_3. Atomic structure models of (c) pristine and (d) Ag-doped Bi_2Se_3 deduced by holography analysis.

References

1) Clere *et al. Phys. B* **351**, 245 (2004).
2) J. Lee *et al.*, *Phys. Rev. Lett.* **126**, 196405 (2021).
3) F. Matsui, Y. Okano, H. Matsuda, T. Yano, E. Nakamura, S. Kera and S. Suga, *J. Phys. Soc. Jpn.* **91**, 094703 (2022). [Editor's pick]
4) F. Matsui *et al.*, *Rev. Sci. Instrum.* **94**, 083701 (2023). [Editor's pick]
5) F. Matsui *et al.*, *J. Electron Spectrosc. Relat. Phenom.* **264**, 147295 (2023).
6) H. Matsuda and F. Matsui, *Jpn. J. Appl. Phys.* **59**, 046503 (2020).
7) F. Matsui and H. Matsuda, *Rev. Sci. Instrum.* **92**, 073301 (2021).
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