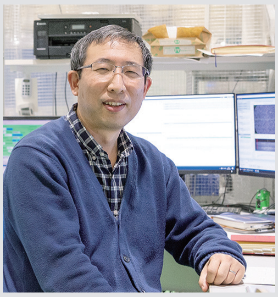
Establishing Advanced Photoemission Methodologies for Novel Spin Materials Science

UVSOR Synchrotron Facility
Division of Advanced Solid State Physics



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Electrons in material are excited by photons and emitted into the vacuum as photoelectrons. Interestingly, the angular distribution of these photoelectrons reveals 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 can 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 and spectroscopy technique for the complete photoelectron analysis. We aim to pioneer cutting-edge spin materials science through comprehensive and detailed characterization of electrons.

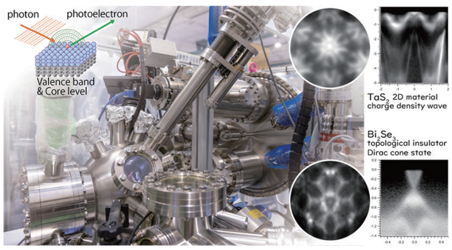

Figure 1. Photoelectron momentum microscope at BL6U of UVSOR synchrotron facility together with valence photoelectron holograms and dispersions of TaS_2 and Bi_2Se_3. Charge density wave phase transition and topological nature can be directly studies in detail.

Selected Publications

- F. Matsui, Y. Okano, H. Matsuda, T. Yano, E. Nakamura, S. Kera and S. Suga, "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).
- F. Matsui, H. Nishikawa, H. Daimon, M. Muntwiler, M. Takizawa, H. Namba, and T. Greber, "The 4π k_z Periodicity in Photoemission from Graphite," *Phys. Rev. B* **97**, 045430 (2018).

1. Single Graphite Step Visualized

Graphite is an incredibly important, versatile mineral, with uses spanning industries. Graphite is an essential component of many batteries, including lithium-ion batteries, and demand is only increasing as new technology is developed. Even though graphite has been thoroughly researched for decades, there is still more to be uncovered. Surprisingly, no photoelectron spectroscopic studies have so far accurately measured the electronic states of the surface and the edge of graphite from a microscopic point of view. It has been "common knowledge" that the electronic structure of graphite is six-fold symmetric, but local observations using microscopy capabilities[1] have revealed the existence of two three-fold symmetric domains that are mirror symmetric by the termination of the alternating stacking structure at the topmost surface (Figure 2).[2] Whereas conventional measurements look at the sum of both, photoelectron momentum microscopy reveals a step-edge structure at the boundary of two terraces of monoatomic layers of graphite with mirror symmetry with respect to each other.

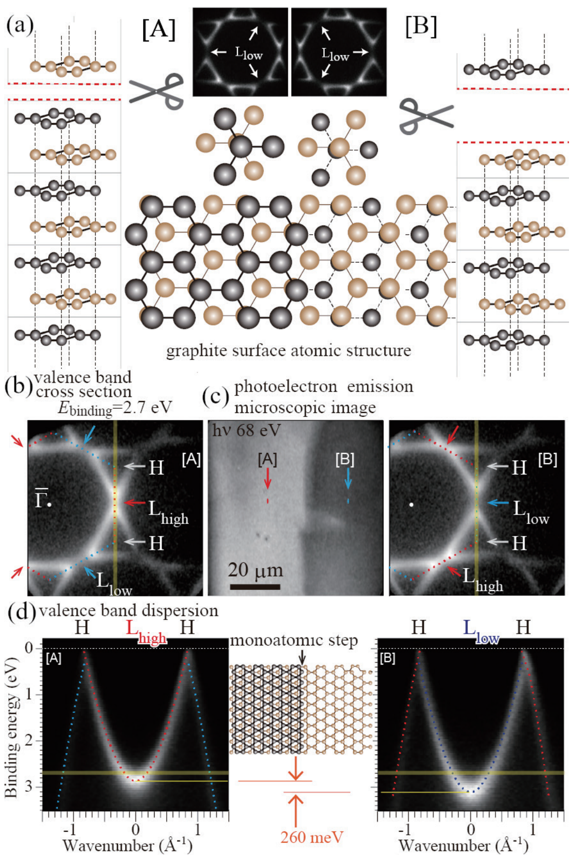

Figure 2. (a) Atomic structure of the cleaved graphite surface. (b) Iso-energy cross section of the graphite surface π band. (c) Graphite single atomic layer step was imaged with different contrast owing to the difference in the π band binding energies in two different terraces as shown in (d).[2]

2. Embed Valence Band in Auger Electrons

Resonant photoelectron spectroscopy highlights certain elemental components of the valence band by adjusting the photon energy to a core-level excitation threshold. However, most of the investigations to date has focused on angle-integrated spectral analysis for elucidating the element-specific density of states. Thus, we explored the condition for the transition of the valence band dispersion information to the Auger electrons by momentum-resolved measurements with a wide-range and high-resolution and realized a new photoelectron spectroscopy with the specificity of elemental and atomic orbitals in band structure analysis.

We performed momentum-resolved resonant photoelectron spectroscopy measurements of graphite crystals using soft X-ray. We identified four different types of resonant pathways at the C K-shell absorption threshold (Figure 3).[3] Fano-resonance-like behavior was confirmed for photoelectron emission from the π band dispersion. The π band dispersion disappeared just below the absorption threshold, and was strongly enhanced at the π* absorption resonance peak photon energy. In addition, two types of resonant Auger electron emission involving the Dirac cone shake-up process were observed. Furthermore, we discovered a peculiar dispersion structure embedded in the normal Auger electron energy region. This phenomenon has also been confirmed with monolayer graphene and adsorbed aromatic molecular species.[4] This resonant valence excitation technique provides a versatile means for characterizing valence band and molecular orbital with element specification.

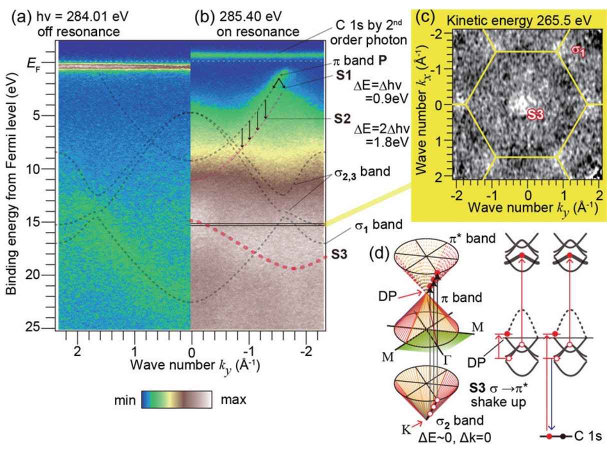

Figure 3. Momentum-resolved valence photoelectron and Auger electron spectra along the direction excited at the photon energy of (a) 284.01 eV and (b) 285.40 eV. Black and red dotted lines indicate the valence band and Auger electron dispersions, respectively. (c) Iso-energy momentum-resolved Auger electron intensity distribution at the kinetic energy of 266.5 eV. (d) Schematic of resonant Auger-electron emission for the pathway S3.[3]

References

1) 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]

2) F. Matsui and S. Suga, *Phys. Rev. B* **105**, 23526 (2022).

3) F. Matsui, S. Makita, H. Matsuda, E. Nakamura, Y. Okano, T. Yano, S. Kera and S. Suga, *J. Phys. Soc. Jpn.* **90**, 124710 (2021).

4) Y. Hasegawa, F. Matsui and S. Kera, *e-J. Surf. Sci. Nanotechnol.* **20**, 174–179 (2022).