Resonant Photoemission: Selective Access to Surface Atomic and Molecular Orbitals

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The electronic properties and chemical reactivities of materials are closely related to the behavior of valence electrons near the Fermi level. Momentum-resolved valence-band photoelectron spectroscopy is a powerful technique to characterize such electrons. Furthermore, by tuning the excitation energy to the core-level absorption edge, valence electron can be resonantly selected and analyzed. In order to establish the reliability of this method, comprehensive measurement and understanding of the photoelectron emission process are important.[1)] BL6U at UVSOR is a beamline dedicated to valence band dispersion mapping with a practical photon energy range of 40 to 600 eV. Wide-acceptance-angle acquisition system enables measurement of full set of valence band dispersion data over several Brillouin zones.[2)]

1. Graphite Valence Band Dispersion in Resonance Auger Electron Spectra

Figure 1 shows a comparison of the angular distribution of graphite valence band between below and above the C 1s absorption threshold. The intensity of the π band with parabolic dispersion is enhance at the π* resonance due to "participant Auger electron process." Until now, there was a common understanding that the angular distribution of Auger electron intensity is due to diffraction, not band dispersion. Here, we found that the resonant Auger electrons *do* carry band dispersion information. The red dashed curves show that momentum is conserved by the electrons involved in the "spectator Auger electron process."

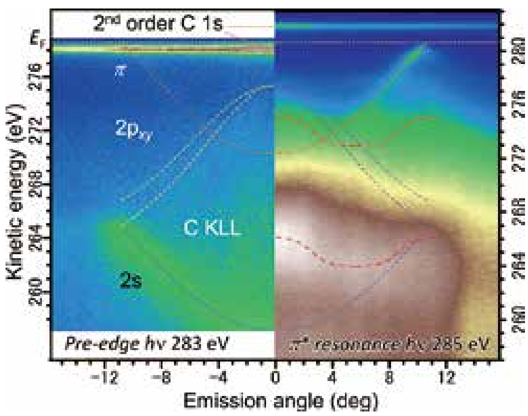

Figure 1. Angle-resolved photoelectron spectra below (left) and above (right) the C K edge threshold. Red dashed lines in the right panel are the newly found participant Auger electron dispersion features.

These experiments can only be performed on soft x-ray beamlines with low carbon contamination. In cooperation with the UVSOR technical team, we removed carbon from the beamline optics by O_2 photochemical etching and successfully experimented with this resonant photoelectron spectroscopy. The spectator Auger process at the C absorption edge was actively explored in the past to try to break specific chemical bonds by selecting antibonding orbitals as the transition destination of 1s electrons. The present success of resonance Auger electron experiments at the C K absorption edge is the basis for expanding the range of application to molecular adsorption systems.[3,4)]

2. Momentum Microscope

Conventionally, azimuthal and polar scans of sample orientation were required for the angle-resolved photoelectron spectroscopy and diffraction measurements. Aforementioned wide-acceptance-angle acquisition system combines an optimized-structure mesh for gathering photoelectrons emitted into wide solid angle and a mechanical lens deflector for two-dimensional data acquisition. Alternatively, display-type analyzers enable the direct observation of wide-solid-angle photoelectron intensity distribution from a selected point without changing the angles of incident light or the sample orientation. By combining a photoelectron emission microscope column and two hemispherical deflection analyzers, *i.e. momentum microscope*, iso-energy photoelectron intensity k_x–k_y distribution can be obtained with high-momentum, energy, and spatial resolutions. A project is underway to install a state-of-the-art momentum microscope in UVSOR and a comprehensive photoemission experiment station.

References

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3) D. Usachov, F. Matsui *et al.*, *2D Materials* **6**, 045046 (2019).
4) D. Usachov, B. Senkovskiy and F. Matsui, *UVSOR activity reports* (2018).