BL23SU
JAEA Actinide Science II

1. Abstract

The JAEA actinide science beamline BL23SU is mainly dedicated to actinide material science. The beamline is also utilized for surface chemistry and biophysical spectroscopy. There are three end stations in the beamline: real-time photoelectron spectroscopy station in the experimental hall and actinide science stations in the RI laboratory building.

2. Surface chemistry experimental end-station

The surface chemistry experimental end-station constructed in the experimental hall at BL23SU focuses on research on chemical reactions and functionalities of surfaces/interfaces of solids. Soft X-ray photoelectron spectroscopy (XPS) is mainly employed to conduct *in situ* chemical analyses of surfaces. This station has been used to promote the Advanced Research Infrastructure for Materials and Nanotechnology in Japan (ARIM Japan) of the Ministry of Education, Culture, Sports, Science and Technology (MEXT) of Japan since April 2021.

The instrumental developments have continuously been conducted. During the shutdown period after finishing the 2022B user time, a new electron energy analyzer (Scienta Omicron Hipp-3) was introduced, and the system was upgraded to realize microscopic photoelectron spectroscopy measurements under near-ambient pressure. Although still in the testing phase, we have achieved sufficient energy resolution to observe the spin-orbit splitting in Si 2p photoelectron spectra and have confirmed the capability of performing photoelectron spectroscopy in a nitrogen gas atmosphere at approximately 1 Pa, which is close to the operational limit of the turbomolecular pump. With further advancements in beamline exhaust and gas introduction systems, this system has the potential for photoelectron spectroscopy measurements under gas pressures up to 3000 Pa. Additionally, we have confirmed the feasibility of microscopic spectroscopy measurements using line-space samples with dimensions of several tens of micrometers. The commissioning of the apparatus was progressively advanced through the collection of the aforementioned relevant foundational data. During the 2023A and 2023B user periods, we facilitated user experiments, starting with those enabling chemical state analysis. Here, several important findings obtained using the surface chemistry experimental end-station are presented.

The Rochow–Müller process is widely recognized as the predominant method for the industrial production of organosilicon compounds. To investigate the fundamental mechanisms underlying this process, we explored the interaction between copper oxides, specifically $Cu_2O(111)$ and the bulk Cu_2O precursor "29"-structure on Cu(111), with CH_3Cl under supersonic molecular beam (SSMB) conditions with energies ranging from 0.5 to 1.9 eV. The resultant surface species were characterized using XPS coupled with synchrotron radiation (SR). Our findings reveal the occurrence of two distinct reaction pathways, designated as Reaction I and Reaction II, both involving the dissociative adsorption of CH_3Cl. In Reaction II, Cl emerges as the predominant adsorbed species, surpassing the quantity of carbonaceous species

observed in Reaction I. Within the investigated energy and exposure parameters, Reaction II was identified as the primary pathway for the CH_3Cl interaction on both Cu2O(111) and the "29"-structure on Cu(111) [1]. Additionally, we examined the effects of CH_3Cl bombardment on Cu(111) and Cu(410) using SSMB energies in the range of 0.7–1.9 eV. The results indicated that Cl is the major adsorbed species, significantly exceeding the adsorbed carbonaceous species, and that the threshold kinetic energy for the reaction is influenced by the crystal surface orientation [2].

The precise formation of interfaces between dielectric materials and wide-bandgap semiconductors is critical for advancing electronic devices, particularly power-switching devices. The impacts of nitric oxide interface nitridation on m-face silicon carbide (SiC) metal–oxide–semiconductor (MOS) structures were investigated using electrical measurements and XPS. The results revealed an unexpected increase in Fowler–Nordheim leakage current owing to nitridation, with nitrogen incorporation at the m-face MOS interface being approximately 2.3 times higher than at the Si-face interface. This increased nitrogen content likely reduces the conduction band offset at the SiO_2/SiC interface, leading to elevated gate leakage. These conclusions are further supported by band alignment analysis utilizing SR-XPS.

Gallium nitride (GaN) is a highly promising material for power-switching devices, and GaN MOSFETs are being explored as candidates for such applications. To optimize GaN MOS structures, various oxides have been investigated as potential dielectrics. The formation of a GaO_x interlayer is essential for achieving SiO_2/GaN interfaces with low defect density. However, improper annealing conditions can result in the reduction of the GaO_x layer, leading to the generation of positive fixed charges. It was demonstrated that sputter deposition of SiO_2 effectively minimizes the formation of unstable GaO_x layers, as confirmed by SR-XPS, which showed negligible GaO_x growth. By performing oxygen and forming gas annealing at 600 °C and 400 °C, respectively, we successfully fabricated a MOS device exhibiting minimal capacitance–voltage hysteresis, stable flat-band voltage, and low leakage current.

The surface chemistry experimental end-station, as discussed, has been extensively utilized in the investigation of the physicochemical properties, surface/interface functionalities, and reaction mechanisms of various materials[3-4].

3. Actinide science stations

In the RI laboratory building, there are photoelectron spectroscopy, soft X-ray magnetic circular dichroism (XMCD), and scanning transmission X-ray microscopy (STXM) stations.

At the photoelectron spectroscopy station, photoelectron spectroscopy studies for strongly correlated materials such as actinide and rare-earth compounds have been conducted. As a scientific result, the electronic state of the heavy fermion superconductor UPt_3 has been investigated by angle-resolved photoemission spectroscopy (ARPES) experiments [5]. The band structure revealed by the present ARPES study is consistent with a calculation based on the density-functional theory for $ThPt_3$. This result is reminiscent of Ce-based heavy fermion compounds, whose band structures are often very similar to those of non-4*f* La-based compounds. We also showed that most of

the U 5*f* spectral weight exists not as coherent heavy-fermion bands but as incoherent components that distribute over a wide energy range, spanning from near the Fermi level to approximately 2 eV. This result suggests that the heavy quasiparticle bands are enormously renormalized by the electron correlation effect.

In addition, the photoemission studies for the heavy fermion superconductor $CeIr_3$ [6], ferromagnet semiconductor Mn-doped indium-tin oxide [7], a half-metallic Weyl ferromagnet candidate Co2FeSi [8], and a Dirac semimetal and superconductors a-Sn and beta-Sn [9] were published.

At the XMCD experimental station, we have promoted a wide range of research on strongly correlated electron systems such as topological insulators and functional magnetic materials. Unfortunately, the operation of the XMCD station has been suspended due to a serious malfunction of the insertion device, which was found in February 2022. In FY2023, the XMCD study on the ferromagnet superconductor UCoGe [10] was published.

The commissioning of the STXM installed at the downstream end of BL23SU in the RI laboratory has been completed. We have confirmed that a spatial resolution of about 30 nm with a 25 nm FZP was achieved in 2022A. The first STXM measurement of a radioactive sample was conducted in the 2023B term. Some user experiments have also been conducted since the 2023B term.

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