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, biophysical 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 the chemical reactions and functionalities of surfaces/interfaces of solids. Soft X-ray photoelectron spectroscopy (XPS) is mainly employed to conduct 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.

Graphene is an attractive two-dimensional material for the application of next-generation electronic devices. Real-time XPS using intense synchrotron radiation and precise spectral analysis were performed to quantitatively evaluate potassium (K) concentration depending on irradiation time [1]. This work was described in a press release [2].

A basic understanding of the oxidation processes of Si(111) substrate with thin Hf films is important to improve Si electronic devices. XPS and a supersonic oxygen molecular beam (SOMB) were employed to study the initial stages of oxidation. The oxidation starts at the outermost Hf layers, producing HfO_2. Hf silicates were generated in the vicinity of the HfO_2/Si interface at 2.2 eV SOMB. The Si substrate was oxidized to produce SiO_2. Si atoms were emitted from the SiO_2/Si interface region underneath the HfO_2 overlayers and allowed to pass through the HfO_2 overlayers to react with the impinging O_2 [3].

TiO_2 is an important material as a functional catalyst. The oxidation of oxygen vacancies at the surface of anatase TiO_2(001) was studied using XPS and SOMB. The oxygen vacancies at the top surface and subsurface can be eliminated by SOMB. Oxygen vacancies are present on the surface when it is untreated before introducing into a vacuum chamber. These stable vacancies can also be effectively eliminated by SOMB [4].

The Cu surface oxidation was reinvestigated by XPS and SOMB. A carefully prepared surface using 0.5 eV SOMB was used. A new structural model was proposed and the previously proposed model was confirmed. From the detailed analysis of O 1s spectra, it was found that the nanopyramidal model is preferable for the $(\sqrt{3} \times \sqrt{3})R30°$ Cu_2O(111) [5]. XPS was also applied to study Pd nanoparticles (NPs), which are important hydrogen-storage materials. The hydrogen absorption and diffusion mechanisms were studied depending on the structure and size of Pd NPs [6].

SiC and GaN are attractive materials for future power electronic devices. The realization of high-performance metal-insulator-semiconductor (MIS) transistors requires a good interface between

the insulator and substrate materials.

Si oxide films on SiC substrates and their interfaces with substrates were studied by XPS. It was found that the electrical properties can be improved by the nitridation of Si oxide films with NO [7,8]. It was also found that a combination of NO annealing and post-nitridation annealing in a CO_2 atmosphere is effective [9].

To improve the performance of SiO_2/GaN metal-oxide-semiconductor (MOS) devices using N-polar GaN $(000\bar{1})$ substrates, the process conditions are clarified by electrical measurements and XPS, in comparison with those on Ga-polar GaN(0001). Although the SiO_2/GaN $(000\bar{1})$ structure was more thermally unstable than the GaN(0001) substrate, excellent electrical properties were obtained by optimizing the conditions for post-deposition annealing. However, the electronic properties for SiO_2/GaN$(000\bar{1})$ were worse than those for SiO_2/GaN(0001), leading to a high gate leakage current. The results implied that caution is needed when using N-polar GaN$(000\bar{1})$ substrates for MOS device fabrication [10].

The oxidation at GaN surfaces depending on GaN crystal planes was studied by real-time XPS. It was found that H_2O vapor has the highest reactivity, suggesting that an oxidant gas other than H_2O and O_2 should be used to avoid unintentional oxidation during $Al_xGa_{1-x}N$ atomic layer deposition [11].

As described above, the surface chemistry experimental end station was widely used for studies on physicochemical properties, functionalities of surface/interface of materials, and mechanisms of surface reactions.

3. Actinide science stations

In the RI laboratory building, there are photoelectron spectroscopy stations and soft X-ray magnetic circular dichroism (XMCD) stations. In addition to them, the scanning transmission X-ray microscopy (STXM) station has been installed and commissioned.

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 structure of $ThPd_2Al_3$, which is isostructural to the heavy fermion superconductor UPd_2Al_3, was investigated by photoelectron spectroscopy [12]. The band structure and Fermi surfaces of $ThPd_2Al_3$ were obtained by angle-resolved photoelectron spectroscopy (ARPES), and the results were well explained by the band-structure calculation based on the local density approximation (Fig. 1). The comparison between the ARPES spectra and the band-structure calculation suggests that the Fermi surface of $ThPd_2Al_3$ mainly consists of the Al 3*p* and Th 6*d* states with a minor contribution from the Pd 4*d* states. The comparison of the band structures between $ThPd_2Al_3$ and UPd_2Al_3 shows that the U 5*f* states form Fermi surfaces in UPd_2Al_3 through hybridization with the Al 3*p* state in the Al layer, suggesting that the Fermi surface of UPd_2Al_3 has a strong three-dimensional nature. The ARPES experiments for the mixed valence Eu compound $EuNi_2P_2$ [13] were also conducted.

At the XMCD station, we have promoted a wide range of research on strongly correlated electron systems such as the 4*f* and 5*f* compounds, as well as topological insulators and functional magnetic materials [14–22]. Recently, low-dimensional and novel materials with magnetic properties, which are prepared by sophisticated thin

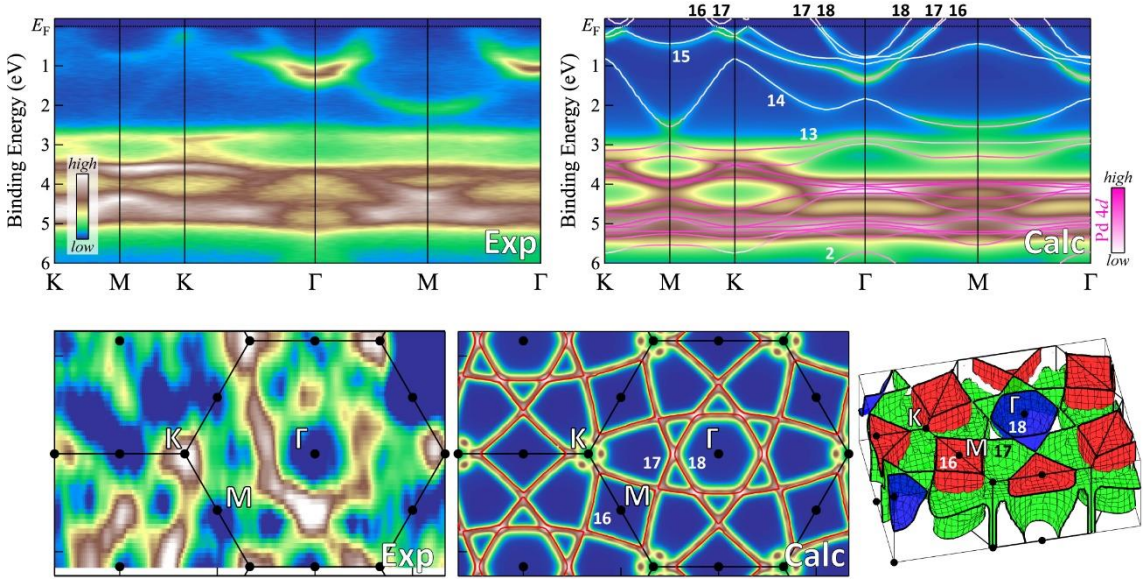

Fig. 1. Band structure and Fermi surface of $ThPd_2Al_3$ obtained by ARPES.

film fabrication techniques, have become good targets to apply XMCD investigations as an element-specific magnetic probe at the atomic level. One of the reasons for the high demands is that XMCD exhibits a powerful characteristic of individual estimation of the spin and orbital magnetic moments at a target element, which is useful for studying the origin of magnetic anisotropy in thin films and investigating whether the target element is truly magnetic or not. Among nine papers published in FY2021, six papers were on magnetism with respect to thin film samples, such as topological insulators [14], transition metal and rare-earth oxides [15,17,19], Heusler compounds [16] and low-dimensional materials [20]. On the other hand, there were interesting spectroscopic findings about magnetic extended X-ray absorption fine structures at Mn $L_{2,3}$ absorption edges, which can provide us with a deep insight into the spin pair distribution function and magnetic structure [16]. However, in the midst of a successful and stable operation of the XMCD station, a serious breakdown of one of the two insertion devices was found in February 2022. Unfortunately, the XMCD experiments cannot be performed for the time being until repairs are completed.

The commissioning of the STXM station installed at the downstream end of BL23SU in the RI laboratory has been progressing smoothly, and we have confirmed that a spatial resolution of about 60 nm has been achieved with 50 nm FZP. The STXM system is now under preparation for future user experiments.

Yoshigoe Akitaka, Takeda Yukiharu, and Fujimori Shin-ichi

Japan Atomic Energy Agency, Materials Sciences Research Center

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