BL23SU
JAEA Actinide Science II

1. Abstract

The JAEA Actinide Science II 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 and 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 study of 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. Several important studies have been reported.

The dry oxidation of silicon (Si) surfaces with oxygen gas (O_2) has become an important research topic of interest for producing gate stacks for advanced complementary metal-oxide-semiconductor (CMOS) field-effect transistors. In order to develop next-generation electronic devices, a basic atomic-scale understanding of chemical reactions at the surface and interfaces between oxides and Si substrates has become important. The roles of excess minority carrier recombination and chemisorbed O_2 species at SiO_2/Si interfaces in Si dry oxidation have been studied by comparison of p-Si(001) and n-Si(001) surfaces [1]. This research provides experimental evidence that (1) the excess minority carrier recombination at the SiO_2/p-Si(001) and SiO_2/n-Si(001) interfaces is associated with O_2 dissociative adsorption, (2) X-ray-induced enhancement of SiO_2 growth is not caused by the band flattening due to the surface photovoltaic effect but is ascribed to the electron–hole pair creation due to core-level photoexcitation for the spillover of the bulk Si electronic states to the SiO_2 layer, (3) changes of band bending result from the excess minority carrier recombination at the oxidation-induced vacancy site when turning the X-ray irradiation on and off, and (4) a metastable chemisorbed O_2 species plays a decisive role in combining two kinds of reaction loops of single- and double-step oxidation. On the basis of the experimental results, the unified Si oxidation reaction model mediated by point defect generation is extended from the viewpoints of (a) excess minority carrier recombination at the oxidation-induced vacancy site and (b) trapping-mediated adsorption through the chemisorbed O_2 species at the SiO_2/Si interface [1]. This basic research was described in a press release [2]. In addition to this study, direct observation of chemisorbed O_2 molecules at the SiO_2/Si(001) interface during Si dry oxidation has also been reported to clarify the fundamental aspects of Si oxidation [3].

LaB_6 is well known to have a small work function value compared with other materials. It is widely used as a basic material for electron emission sources employed in many kinds of science and technological apparatus such as those of electron microscopy, electron beam lithography exposure, and electronic accelerator. For the development of more efficient and reliable electron sources, it is important to achieve a smaller work function or to explore novel materials with further smaller work functions. In this research, it was found that a monolayer hexagonal BN (hBN) coating is an attractive method of lowering the work function. Surface analysis by methods such as photoemission electron microscopy (PEEM) and thermionic emission electron microscopy (TEEM) revealed that the hBN-coated region of a LaB_6(100) single crystal has a lower work function than those of the bare (i.e., noncoated) and graphene-coated regions. A larger decrease of work function for the hBN-coated LaB_6(100) than for the graphene-coated LaB_6(100) was qualitatively supported by the density functional theory (DFT) calculations. Adding an oxide layer to the calculations improved the consistency between the calculation and experimental results. The calculations suggest that the presence of an oxide layer on our LaB_6 [4]. This work was also described in a press release [5].

Instrumental developments have also progressed. During the shutdown period after the end of the 2022B user time, the electron energy analyzer was upgraded to realize microscopic photoelectron spectroscopy measurements under near-ambient pressure. Instrument commissioning for utilization studies began after the 2023A user time.

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 station, soft X-ray magnetic circular dichroism (XMCD) station, and scanning transmission X-ray microscopy (STXM) station.

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 4*f* localized ferromagnet $CeRu_2Ge_2$ has been investigated by angle-resolved photoemission spectroscopy (ARPES) experiments [6]. The band structure and Fermi surfaces were observed and were consistent with a calculation based on the density-functional theory for $LaRu_2Ge_2$, thus confirming a localized character of the 4*f* electrons. On the other hand, the Ce 3*d*–4*f* resonant ARPES experiments demonstrated that the 4*f* spectral function is dominated by f^1 components in the vicinity of the Fermi level and thus is different from that expected for a localized electron system. We also revealed that the intensities of the f^1 components have almost no temperature dependence up to at least 100 K, which is two orders of magnitude larger than the Kondo temperature of $CeRu_2Ge_2$; this is in contrast with the results of other ARPES studies for similar heavy fermion compounds using lower photon energies.

In addition, the photoemission studies for the superconductor $La(O,F)BiS_2$ [7], the itinerant ferromagnet $Sr_{1-x}(La,K)_xRuO_3$ [8], a low-dimensional material of the van der Waals itinerant magnet [9], and the Weyl-Kondo semimetal candidate compound $Ce_3Rh_4Sn_{13}$ [10] 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. Recently, materials with low dimensions and/or novel magnetic properties, which are prepared by sophisticated thin-film fabrication techniques, have become good targets to apply XMCD investigations, taking advantage of an element-specific magnetic probe at an atomic level. Unfortunately, the operation of the XMCD station has been suspended owing to a serious malfunction of the insertion device, which was found in February 2022.

In FY2022, papers on magnetism with respect to thin films containing 3*d* and 4*d* transition metals [11,12,13] and a low-dimensional material of the van der Waals itinerant magnet [9] were published. For all the materials, attention was paid to their peculiar functional features. The XMCD experiments on these materials have provided evidential results useful for understanding the electronic states and magnetic properties of these materials.

The initial commissioning of the STXM equipment 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 STXM system will be opened for user experiments from 2023B.

Yoshigoe Akitaka, Tsuda Yasutaka, Takeda Yukiharu, and Fujimori Shin-ichi

Japan Atomic Energy Agency, Materials Sciences Research Center

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