Soft x-ray photoemission and magnetic circular dichroism of correlated systems and nano-materials

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1. Introduction

Use of soft x-rays from third generation synchrotron radiation facilities has open up new opportunities in the high-energy spectroscopic studies of strongly correlated systems. A breakthrough was made in 2000 at a soft x-ray undulator beamline BL25SU of Spring-8 by high-resolution, bulk-sensitive photoemission (PES) experiments on Ce compounds [1]. At that beamline, an endstation for magnetic circular dichroism (MCD) in soft x-ray absorption spectroscopy was also built, and has enabled us to perform high-resolution MCD measurements. Although PES and MCD are standard techniques nowadays, their usefulness in the electronic structure studies of advanced materials is remarkable and their scientific impact is still enormous. In addition to BL25SU, two soft x-ray beamlines are now in operation in Spring-8. In this report, I briefly describe the present status of soft x-ray PES and MCD experiments on strongly correlated system and nano-materials in which I am now involved in Spring-8 and Photon Factory and discuss future prospects.

2. Soft x-ray beamline BL23SU of Spring-8

A JAERI beamline BL23SU consists of a 10 m varied-line-spacing plane-grating monochromator (VLS-PGM) and four endstations, out of which PES and MCD endstations are located in a “hot sample area” where unsealed U samples can be measured [2]. An APPLE-2 helical undulator provides modulated circularly-polarized light. The photon flux after the post-focusing mirror is typically 2-3 $\times$ 10^{11} photons/sec/100mA/0.02%bw in the 500-1000 eV range. The total energy resolution including the monochromator and the spectrometer (Gammadata-Scienta 2002) is ~85 meV at hν ~ 850 eV as measured from the Au Fermi edge. This performance is comparable to that of BL25SU.

3. Photoemission

There are several advantages in using soft x-rays for PES. (i) Photoelectron escape depth becomes longer and hence the spectra become more bulk-sensitive [1]. (ii) Uncertainty in the momentum perpendicular to the surface, i.e., Δk_z, is small, making the ARPES band mapping of three-dimensional materials more reliable. (iii) The photo-ionization cross-sections of transition-metal d and rare-earth/actinide f orbitals become large compared to the other orbitals. In particular, resonant photoemission effect is enhanced for transition-metal 3d and rare-earth 4f orbitals compared to shallower core levels. (iv) The background becomes weaker, making intrinsic spectral features more clearly observed.

For actinides, unfortunately, 5f-electron emission is not enhanced at the 4d → 5f absorption region [3]. Nevertheless, the prominent 5f cross-sections in the soft x-ray region enables us to study the 5f-related electronic in U compounds. For example, U 5f-derived bands and Fermi surface were clearly observed in ARPES measurements at BL23SU of $UFeGa_5$, which is a paramagnetic metal with a layered structure, and agreed well with LDA band-structure calculation [4]. On the other hand, the antiferromagnetic $UPtGa_5$ exhibited a narrow 5f band in the vicinity of the Fermi level, unlike the LDA calculation. Comparison of those data with photoemission spectra taken in the ultra-violet region (hν ~ 20 eV) gives further insight into hybridization between the 5f versus non-f atomic orbitals.

In order to extend the capability of soft x-ray ARPES to three-dimensional materials, a major obstacle has been the difficulty in obtaining atomically flat surfaces by cleaving single crystals. Recently, the pulsed laser deposition (PLD) technique has become a powerful tool to synthesize single-crystal thin films of various transition-metal oxides and their nano-structures. In Photon Factory, a combined PES-PLD instrument was constructed [5] and has been devoted to study three-dimensional perovskite-type transition-metal oxides. At BL23SU, too, we have set up a facility by which measurements on in-situ prepared oxide thin films can be made. Such a facility will become a useful too to perform ARPES measurements of oxides using soft x-rays.

4. Magnetic circular dichroism

MCD is known as an "element-specific" probe of magnetism and therefore is suited to study complex materials. The MCD equipment at BL23SU has the capability of carrying out experiment at high magnetic fields up to ~10 T and at low temperatures down to ~10 K [6]. Our MCD measurements of the ferromagnetic superconductor UGe_2 [7] at the U 4d, 2p and Ge 2p edges revealed that not only the U 5f but also Ge 4s electrons are spin-polarized whereas the U 6d electrons are not [8], in contrast the magnetic Compton scattering result.

If we combine the high magnetic field and the low temperature, one can probe minority paramagnetic impurities in a ferromagnetic material. Such measurements would give unique information about the crucial problem of intrinsic and extrinsic defects in diluted magnetic semiconductors (DMS's). Indeed, the field- and temperature dependent MCD measurements of prototypical DMS, $Ga_{1-x}Mn_xAs$, revealed three Mn signals of different magnetic nature: ferromagnetic, paramagnetic and field-induced ferromagnetic components [9]. In cases where the ferromagnetic moment is only a small fraction of the paramagnetic moment, identification of the ferromagnetic state can be made by high-temperature, low-field MCD measurements, as demonstrated for $Zn_{1-x}Co_xO$ [10].

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