

## Probing spin-resolved valence band electronic structures of buried Fe film with hard X-ray photoemission

Photoemission spectroscopy (PES), which is a powerful tool for investigating the electronic structures of solids, has been performed using various excitation light sources in a wide photon energy range from several eV to  $\sim 15$  keV. In particular, hard X-ray PES (HAXPES: photon energy  $> \sim 3$  keV) is recognized as one of the most powerful methods for detecting the bulk and buried interface electronic states of materials owing to the large inelastic mean free path (IMFP) of electrons with a kinetic energy of several keV [1]. The IMFP in HAXPES experiments is several nm, which is larger than that in soft X-ray PES ( $\sim 1$  nm). In contrast, the photoionization cross section for the hard X-ray region is  $\sim 2$ – $4$  orders of magnitude lower than that for the soft X-ray region. Therefore, high-brilliance hard X-rays from undulators at third-generation synchrotron facilities such as SPring-8 and the optimization of the experimental geometry are required to conduct the HAXPES experiments with a total energy resolution better than 0.3 eV [1,2]. To date, X-ray polarization-dependent HAXPES for linear or circular dichroism, angle-resolved HAXPES for band dispersions, and X-ray standing-wave HAXPES for depth profiling have been realized through the development of X-ray optics and electron analyzers as well as synchrotron light sources [1]. Although spin-resolved HAXPES is expected to be suitable for studying the bulk-sensitive spin-dependent electronic structures of magnetic materials, it is still difficult to access the spin resolution in the valence band region by HAXPES owing to two unfavorable aspects: (i) small photoionization cross section in valence electrons and (ii) low efficiency of

a typical spin detector such as a Mott detector [3]. To realize spin-resolved HAXPES in the valence band region, we have developed an ultracompact Mott-type Au spin filter to conduct spin-resolved HAXPES measurements without modifying the HAXPES apparatus [4]. This ultracompact spin filter can be mounted on a sample holder, as shown in Fig. 1(a). The compact sample holder with the spin filter can be easily adapted to the existing HAXPES apparatus without modifying the apparatus. Figure 1(b) shows a schematic illustration of the experimental setup of our proposed spin-resolved HAXPES [4]. The scattering induced by the Au spin filter involves Mott scattering [3], and the Au spin filter partially selects electrons with one spin direction in the geometry shown in Fig. 1(b). To detect electrons with the opposite spin direction, magnetization reversal is necessary [4]. Although magnetization reversal of the sample is essentially required in this method, we can utilize the advantage of two-dimensional (2D) multichannel detection, which enhances the electron detection efficiency, used in a recent high-resolution electron analyzer.

The spin-resolved HAXPES measurements were performed at SPring-8 BL15XU. The sample used for spin-resolved HAXPES was a MgO(2 nm)-capped epitaxial Fe(30 nm) film on a MgO(001) substrate. The photon energy and total energy resolution were set to 5.95 keV and 0.68 eV, respectively. Note that the single-channel figure of merit (FOM) in this spin-resolved HAXPES with the geometry shown in Fig. 1(b) was evaluated to be  $\sim 2 \times 10^{-4}$  when the effective Sherman function ( $S_{\text{eff}}$ ) of  $-0.07$  was

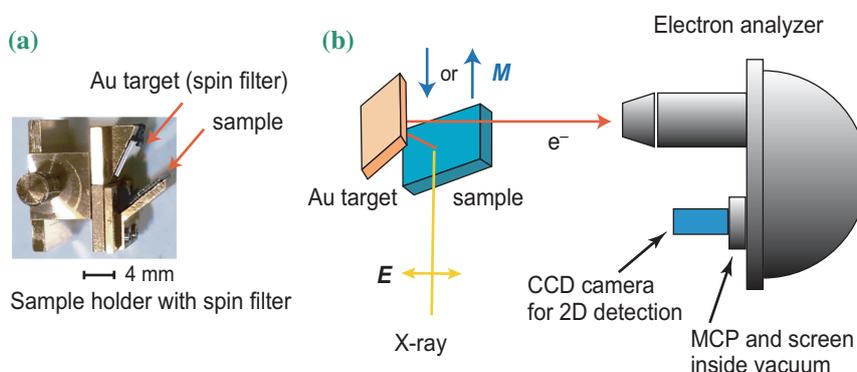


Fig. 1. (a) Photo of the sample holder, which can mount both thin films of the sample and Au target simultaneously. (b) Schematic illustration of the experimental geometry of spin-resolved HAXPES in this work. The photoelectrons emitted from the sample scattered by the Au target are introduced into the electron analyzer. The scattering angle of photoelectrons by the Au target is  $127 \pm 7^\circ$ . The magnetization direction ( $M$ ) is indicated by arrows. The incident angle of X-rays is set to about  $5^\circ$  with respect to the sample surface and  $M$ .

adopted [4]. This value is comparable to that of the typical Mott detector [3]. Since we can utilize the 2D multichannel detector in spin-resolved HAXPES, the effective FOM is enhanced by a factor of  $\sim 4 \times 10^4$ , which is the number of active channels in the 2D detector, in comparison with the case when only one channel of the 2D detector is used. Note that the spin polarization ( $P$ ), majority ( $I_{\text{maj}}$ ), and minority ( $I_{\text{min}}$ ) spin spectra are obtained by using the measured spectra ( $I^+$  and  $I^-$ ) with two magnetization directions shown in Fig. 1(b) as follows:  $P = (I^+ - I^-)/(I^+ + I^-)/S_{\text{eff}}$ ,  $I_{\text{maj}} = (I^+ + I^-)(1+P)/2$ , and  $I_{\text{min}} = (I^+ + I^-)(1-P)/2$ .

Figure 2(a) shows the experimental spin-resolved valence band HAXPES spectra of the buried Fe film. Although the energy resolution in spin-resolved HAXPES (0.68 eV) is lower than that in standard (spin-integrated) HAXPES (<0.3 eV), the spin-resolved HAXPES spectra show a clear difference between the majority and minority spin spectral shapes. The sharp peak and broad structure were found in the majority spin states at the binding energy ( $E_B$ ) of  $\sim 1$  and  $\sim 4$  eV, respectively. In contrast, fine structures were not found in the minority spin states. The background intensity on the high  $E_B$  side differed between the majority and minority spin states owing to the spin-polarized secondary electrons. By comparing the experimental spectra with the spin-resolved partial densities of states (DOSs) obtained by density functional theory (DFT) calculation [5], we see that the sharp peak and broad structure arise from the Fe 3d and 4s majority spin states, respectively. In contrast, the Fe 3d minority spin states are located near the 4s minority spin states, so that the minority spin spectrum shows the broad structure due to the low energy resolution. Note that the Fe 4s states are emphasized in the observed HAXPES spectra owing to the characteristic large photoionization cross-sectional ratio of Fe 4s to 3d in HAXPES [5]. Figure 2(b) shows the spin polarization curve of the buried Fe film. The large spin polarizations at  $E_B$  of  $\sim 0$  and  $\sim 5$  eV are mainly due to the Fe 3d and 4s states, respectively. The spin polarization curve is reproduced by the calculated spin polarization obtained from the spin-resolved cross section weighted DOSs, except for the energy scale of  $E_B$ . Even though the energy resolution is low in the present spin-resolved HAXPES, the spin resolution allows us to detect the deviation between experiments and calculations, which is caused by bandwidth narrowing due to the strong electron correlation effects in Fe [4,5].

Owing to the high-brilliance hard X-rays at SPring-8 and the 2D multichannel detection of the electron analyzer, we have successfully observed the bulk-sensitive valence band spin-resolved HAXPES spectra for the buried Fe film. At the present stage, the energy

resolution is low and the statistical error is large in spin-resolved HAXPES. Further development of spin-resolved HAXPES (effective FOM, energy resolution, and statistical error) is required to study genuine spin-resolved electronic states of advanced magnetic materials such as a half-metal, which shows unique spin-dependent electronic states (e.g., perfect spin polarization at the Fermi level).

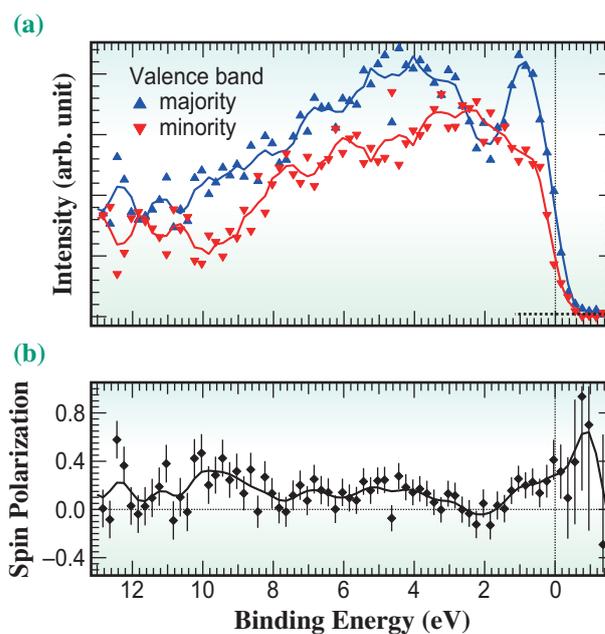


Fig. 2. (a) Spin-resolved valence band HAXPES spectra of the buried Fe film. (b) Spin polarization curve of the buried Fe film. Vertical bars represent statistical errors. The total data acquisition time is  $\sim 13$  hours.

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