Ultrahigh resolution soft X-ray emission study at BL07LSU in SPring-8

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Soft X-ray emission (SXE) spectroscopy has attracted much attention because of its high potential for probing element-specific electronic states of a wide range of materials such as wide gap insulators, liquids, electrolytes, corrosions and so on. However, the energy resolution of a conventional SXE spectrometer around several hundreds of meV for *K*-edge of light elements and *L*-edge of 3*d* transition metals is not enough to discuss multiplets, charge transfer properties or low energy excitations such as phonons, magnons, orbitons and/or distinguish a particular chemical state that is involved in a specific function of wet materials. In recent years, there have been a lot of efforts to improve the energy resolution of the SXE spectrometer[1-5]. In this paper, we report an ultrahigh resolution SXE spectrometer that features $\Delta E < 100$ meV and/or $E/\Delta E \sim 10000$ in the range of 350 eV to 750 eV. The spectrometer was installed at BL07LSU in SPring-8 where SXE spectra with precise polarization correlation can be obtained using more than 99 % linear polarization in both vertical and horizontal directions of the incident photon.

To achieve the ultrahigh resolution SXE, we have realized an extremely small spot size for the incident beam by applying post focusing mirror optics in a Kirkpatrick-Baez (K-B) configuration with which an aberration free rectangular spot can be obtained at the focal position. To obtain a vertical spot size of less than 1 μm on the sample and to maximize the gain of the photon flux by fully opening the exit slit (> 100 μm), the demagnification factor was set to 150, which corresponds to the exit arm length of around 150 mm for an entrance arm length of 23 m. Thus the vertical refocusing mirror was installed in the measurement chamber. Figure 1 shows the obtained spot image measured using two dimensional scanning of a 1 μm pinhole with an AXUV 100 silicon photodiode supplied by IRD Inc. By virtue of the vertical spot size around 1 μm we can remove the entrance slit of the spectrometer to gain the detection efficiency without losing the energy resolution. We have designed the optical layout applying short entrance and long exit arms for the grazing incidence flat field type optical mount, which achieves high detection efficiency and high energy resolution. In order to eliminate both defocus and small coma aberration that will lose the energy resolution around $E/\Delta E \sim 10000$, we have applied a coma-free operation mode proposed by Strocov *et al.*[6] by adding a horizontal driving axis to adjust the entrance arm length. The resolving power curves simulated by ray tracing adapting the simplified coma-free operation are shown in Fig. 2. The results show $E/\Delta E \geq 10000$ in the range of 350 eV to 550 eV and $E/\Delta E \geq 8000$ in the range of 550 eV to 750 eV.

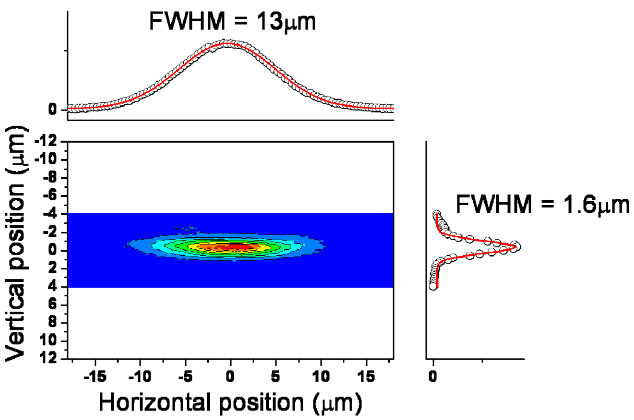

Figure 1 Measured focal spot of the incident beam at the sample position obtained for an exit slit width of 40 μm.

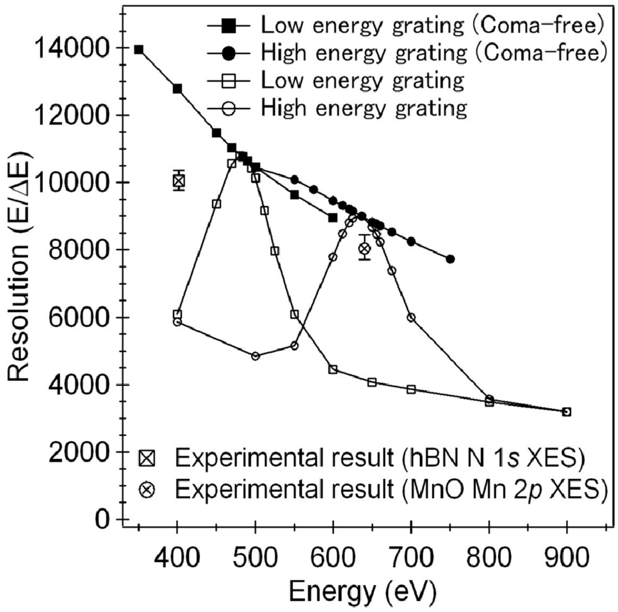

Figure 2 Energy resolution of the spectrometer estimated by ray tracing with lines between open symbols that correspond to the ray tracing without coma-free mode.

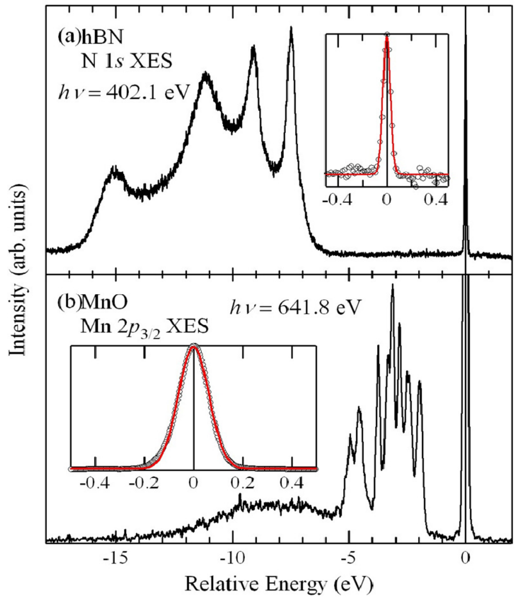

Figure 3 Performance of the SXE spectrometer for the (a)N 1*s* edge of hBN and (b) Mn 2*p* edge of MnO.

Figure 3 demonstrates SXE spectra at the (a) N 1*s* edge of hBN and (b) Mn 2*p* edge of MnO. The total energy resolution estimated from the elastic line ΔE_{total} is 70.0 meV (N 1s edge: 402.1 eV) and 147 meV (Mn 2p edge: 641.8 eV). The effective energy resolution of the SXE spectrometer ΔE_{spec} is obtained by the following deconvolution,

$$\frac{\Delta E_{spec}}{E} = \sqrt{\left(\frac{\Delta E_{total}}{E}\right)^2 - \left(\frac{\Delta E_{exc}}{E}\right)^2} \qquad (7)$$

which results in ΔE_{spec} = 40.0 meV ($E/\Delta E_{spec}$ = 10050) for the N 1*s* edge and ΔE_{spec} = 79.8 meV ($E/\Delta E_{spec}$ = 8046) for the Mn 2*p* edge. The experimental results plotted in Fig. 2 exhibit approximately 90 % and 80 % of the simulated resolving power for the Mn 2*p* and N 1*s* edges, respectively. SXE spectra at Ti 2*p* (~450 eV), O 1*s* (~520 eV), F 1*s* (~670 eV) edges were also obtained with more than 80 % of the simulated resolving power.

We also have measured SXE of liquid H_2O/D_2O water using flow-through liquid cell and successfully obtained their O 1s SXE spectra with the world's highest resolution at present. The $1b_1$ splitting of the water spectra is much more enhanced than previous results[7,8] along with an additional shoulder at higher energy side of the $1b_1$ peaks.

References

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