High-resolution display-type retarding field analyzer

Photoelectron diffraction (PED) or photoelectron holography (PEH) is a powerful method for studying local atomic arrangements [1]. An advantage of these methods is the utilization of chemical shifts in core-level photoemission spectra. So far, however, such high-energy-resolution measurements have only been performable through time-consuming angular scans of samples using non-display-type analyzers. For more efficient measurements, high-energy-resolution display-type photoelectron analyzers are desired.

Display-type retarding field analyzers (RFAs) are one of the candidates for high-resolution PED or PEH. The simplest RFA optics is composed of three spherical grids, where a retarding voltage (RV) is supplied to the intermediate grid and the two exterior grids are grounded. Because the retarding grid is usually made of a wire mesh, a potential variation occurs across the space between the wires. This generates distortions of electron trajectories near the retarding grid from the normally incident trajectories, which are often called lens effects [2]. These effects usually limit the resolving powers ($E/\Delta E$) of three-grid RFAs to less than 100 [3]. In order to resolve chemical shifts, $E/\Delta E$ as high as or higher than 1000 is required.

In this work, we found a method for obtaining $E/\Delta E$ of 1000 or higher for three-grid spherical RFAs [4]. To find this method, we performed electron trajectory simulations where the trajectories in the vicinity of the retarding grid were calculated in detail. Because an RFA is a high-pass filter, the transmittance function of an RFA has a cutoff at the retarding potential energy. The electrons emitted from a sample with kinetic energy (E_K) values larger than the retarding potential energy can pass through the retarding grid, whereas the electrons with E_K values smaller than the retarding potential energy are repelled by the retarding grid. When using a conventional grid arrangement where the three spherical grids are deployed close together with the same distances, the RFA shows a very gradual transmittance cutoff because of the lens effects, which results in $E/\Delta E$ of about 100. We found that a much steeper cutoff can be obtained by deploying the three grids with very different distances as shown in Fig. 1(a). The lens effects still exist even in this devised arrangement, but they are optimized so as to sharpen the cutoff. The detailed reason for this is given in Ref. 4. $E/\Delta E$ of 3200 was theoretically expected when the mesh number was 250 and the grid radii were 12, 40, and 42 mm. [4].

We constructed an RFA employing the devised grid arrangement at the soft X-ray beamline **BL25SU** [5] as shown in Fig. 1(b). To evaluate $E/\Delta E$ of the RFA, we measured angle-integrated 4*f* core-level photoemission spectra of evaporated gold. A photon energy ($h\nu$) of 700 eV was used. The energy resolution (ΔE) of the beamline monochromator was set at 82 meV. First, we used a conventional RFA, where the grid radii were 32.5, 36.5, and 39.5 mm. The measured spectrum is shown in Fig. 2(a). Although the spin-orbit splitting of $4f_{5/2}$ and $4f_{7/2}$ is barely recognized, the spectral shape is very broad. Then, we used the RFA with the devised grid arrangement in which the grid radii were 12, 40, and 42 mm. The measured spectral widths in Fig. 2(b) are clearly narrower than those in Fig. 2(a), indicating an improved $E/\Delta E$. We compared the Au 4*f* spectrum in Fig. 2(b) with that measured using a concentric hemispherical analyzer with a known energy resolution and estimated $E/\Delta E$ of the RFA to be 1100 [4]. For PEH, photoelectron E_K values of more than 400 eV are required [1]. If the smallest available E_K of 400 eV is used, $E/\Delta E$ of 1100 results in ΔE of 0.36 eV, which allows one to resolve relatively large chemical shifts caused by, for example, different valencies.

To verify the ability to observe PED patterns, we performed PED measurements for single-crystal

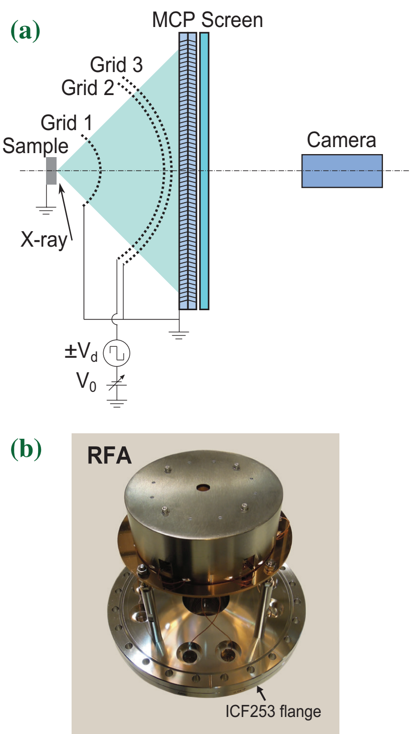

Fig. 1. **(a)** Schematic of the developed 3-grid RFA. **(b)** Photo of the developed 3-grid RFA.

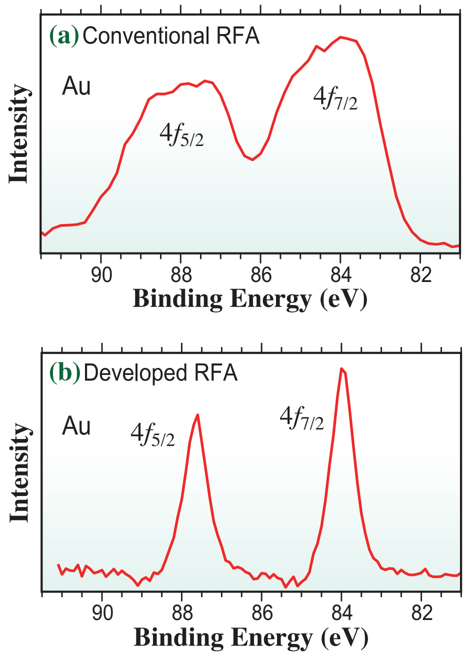

Fig. 2. Comparison of Au 4*f* photoemission spectra measured with **(a)** a conventional three-grid RFA and **(b)** the developed three-grid RFA. *hν* of 700 eV was used.

graphite. *hν* of 900 eV was used, at which C 1*s* photoelectrons had E_K values comparable to those of the Au 4*f* photoelectrons in Fig. 2. Figure 3(a) shows an angle-integrated photoemission spectrum measured in the C 1*s* region. In this figure, the horizontal axis represents the RV. Because $E/\Delta E$ of the RFA was 1100, the ΔE was approximately 0.55 eV in this E_K region. The ΔE of the beamline monochromator was set at 0.18 eV. Thus, the total ΔE was approximately 0.58 eV. Figure 3(b) shows a result of the PED measurements. A C 1*s* PED pattern with sharp Kikuchi lines was clearly observed. Since an RFA is a high-pass filter, to observe a PED pattern in a certain E_K bandwidth, it is necessary to take a difference image between two images corresponding to two different RVs. For the difference image acquisition, as shown in Fig. 1(a), we periodically switched the RV with a rectangular time profile between V_0+V_d and V_0-V_d, where V_0 is the static bias and $\pm V_d$ are voltages superimposed on V_0. The camera observing the screen was synchronized with the periodic RV switching. For the measurement in Fig. 3(b), V_0 was set at the C 1*s* peak and V_d was 0.5 V. The acceptance angle of the RFA was ±49°. The acquisition time of the raw difference image was 80 min. The developed RFA is now open to public use at SPring-8 BL25SU.

Although $E/\Delta E$ of 1100 for our RFA is higher than those of conventional RFAs by one order of magnitude, higher $E/\Delta E$ values are still desired for resolving small chemical shifts of a few hundred meV. Woven wire meshes inherently have inhomogeneities of grid openings, which affect $E/\Delta E$. One possibility is to fabricate a spherical grid by making a large number of radially directed holes through a partial spherical shell like a spherical cap with a sufficient thickness to maintain the spherical shape. We are now developing such a holed grid with a wide acceptance angle.

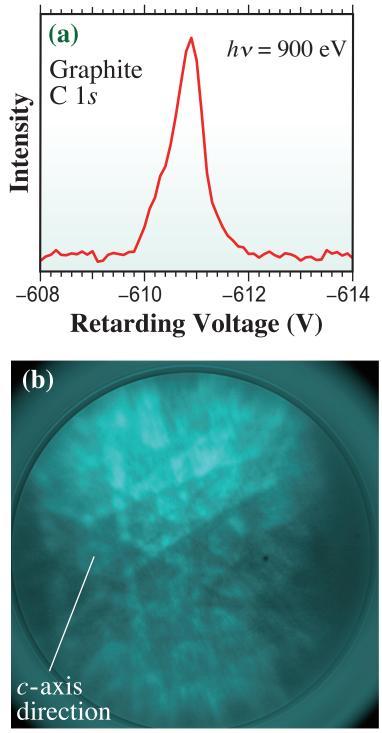

Fig. 3. **(a)** C 1*s* photoemission spectrum measured with the developed RFA for a single crystal of graphite. *hν* of 900 eV was used. The horizontal axis represents the RVs that is, V_0. **(b)** Image obtained by dividing the raw difference image by a background image. The acceptance angle is ±49°. For the difference image measurement, V_0 was set at the C 1*s* peak and V_d was 0.5 V. The *c*-axis direction is indicated in the figure.

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