Ultracompact mirror device for forming 20-nm achromatic soft-X-ray focus toward multimodal and multicolor nanoanalyses

X-ray focusing devices play a crucial role in generating X-ray nanoprobes for scanning microscopy. These devices can be characterized by their nanofocusing capabilities, achromaticity, and focusing throughput. Achromatic focusing ensures a constant probe position irrespective of the incident photon energy, whereas high focusing throughput enables efficient conversion of incident X-rays into a nanoprobe with minimal intensity loss. An ideal X-ray focusing device meets all three of these criteria.

In the soft X-ray region, solely total-reflection-based X-ray focusing mirrors offer both achromaticity and high focusing throughput. However, fabricating soft X-ray nanofocusing mirrors with high precision presents significant challenges, as nanoscale smoothness must be achieved on a highly curved (submeter radius) surface. When such surface perfection is attained, the focus size can reach the diffraction limit, which is proportional to the X-ray wavelength. A soft-X-ray focusing mirror has barely achieved a diffraction-limited focus size of 241 nm × 81 nm at photon energy of 0.3 keV [2]. A study found that the size of the achromatic soft X-ray nanoprobe did not decrease with a shorter X-ray wavelength; rather, it expanded [3]. The nanofocusing capability of soft X-ray focusing mirrors can be severely constrained by fabrication process limitations.

To achieve ideal nanoprobes across the entire soft X-ray range, we employed a novel strategy in addition to the straightforward development of new fabrication techniques [4]. With the large grazing angle allowed in the soft X-ray region, millimeter-scale mirrors can moderately accept incident X-rays and achieve focusing throughput that is superior or comparable to that of common soft-X-ray nanofocusing devices (zone plates). These mirrors can be fabricated with high precision, as mirror fabrication and metrology techniques can be optimized for a specific figure frequency range (1 cm^{-1} to 10 μm^{-1}). Additionally, short mirrors allow their focal points to be positioned much closer to the mirror center. Millimeter-scale focal lengths enhance focusing robustness by ensuring that X-rays reach the focal plane before significant spreading occurs due to mirror figure errors. However, fabricating such ultracompact mirrors presents a challenge, as shorter focal lengths proportionally reduce the tangential radii of curvature. A robust focusing strategy, supported by advancements in fabrication technologies, is essential for achieving ideal achromatic soft X-ray nanoprobes.

Figure 1(a) presents a schematic layout of the ultracompact Kirkpatrick–Baez (ucKB) mirror. The KB geometry simplifies a doubly curved focusing mirror into a pair of elliptic-cylindrical surfaces, namely, a vertically focusing mirror (VFM) and a horizontally focusing mirror (HFM). To achieve an extremely short focal length for the component mirrors of the ucKB mirror, the downstream mirror length and focal length were both reduced to 2 mm, an approach that deviates from conventional mirror design principles. The ucKB mirror was specifically designed to focus 1-keV soft X-rays onto a sub-50-nm diffraction-limited spot. Each mirror, designed with a primary grazing angle of 25 mrad, can theoretically achieve a reflectivity of 50.9% at a photon energy of 1 keV.

To efficiently fabricate their highly curved surfaces, angstrom-scale smooth cylindrical surfaces were figure-corrected into the designed elliptic-cylindrical reflective surfaces through Ni deposition. Figure 1(b) shows that the residual figure errors were controlled within 0.5% of

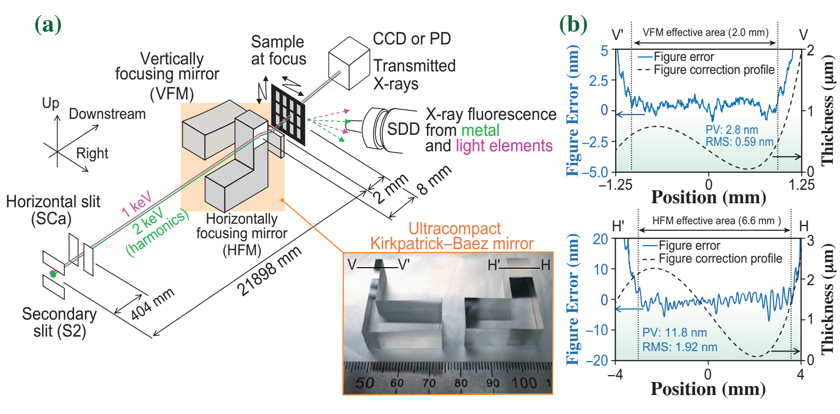

Fig. 1. Experimental configuration using ultracompact Kirkpatrick–Baez (ucKB) mirror. **(a)** Layout of soft-X-ray microscope. Elliptic cylindrical reflective surfaces were fabricated on L-shaped glass cylinder substrates (ruler units: mm). **(b)** Film thickness profile for figure correction of substrates (dashed line) and residual figure errors (solid line). Figure errors within this area are presented in terms of peak-to-valley (PV) and root-mean-square (RMS) values.

the maximum Ni film thickness. Considering the peak-to-valley (PV) residual errors, the VFM is expected to achieve optimal focusing performance for soft X-rays below 2 keV.

The performance of the ucKB mirror was evaluated at SPring-8 **BL25SU** A-branch, which offers photon energy tunability from 0.3 to 2 keV. As shown in Fig. 2(a), the focus spot was evaluated using knife-edge scanning and ptychography. The focus positions remained mostly constant across different photon energies. The difference in focus size between knife-edge scanning and ptychography arises from the bluntness of the knife edge, with the ptychographic measurement providing a more precise focus size. Overall, the ptychographic focus size aligns well with the design specifications. Notably, the focus size decreases with increasing photon energy, reaching a minimum spot size of 20.4 nm × 40.7 nm at 2 keV.

Such achromatic nanoprobes can not only broaden the energy bandwidth but also enable the use of multiple X-ray photon energies for photon-hungry techniques. For instance, the low-energy X-ray fluorescence technique faces challenges due to the inherently low fluorescence yields of light elements, even at micrometer-scale spatial resolution. As shown in Fig. 1(a), with the undulators and monochromator tuned for a 1-keV fundamental and 2-keV second-order harmonics, the ucKB mirror generated a sub-100-nm two-color nanoprobe. This approach allowed simultaneous fluorescence excitation of both light elements and metal elements.

This method was applied to analyze, for instance, hippocampal neurons cultured from rat brains and chemically fixed, as shown in Fig. 2(b). The region where a neuronal structure, known as the spine, seemed to have disappeared was examined using a scanning pitch of 100 nm. Based on the results of soft X-ray fluorescence and transmitted X-ray measurements, distribution maps for the mass of chemical elements and the effective thickness of the samples were produced and applied to concentration analyses, as shown in Fig. 2(c). The analysis revealed that Zn was homogeneously distributed, whereas Cu and Fe exhibited more localized distribution. Such information is not easily obtained through conventional techniques.

The ucKB mirror is an ideal X-ray focusing device in terms of nanofocusing capability, broadband energy, and focusing throughput. These properties make it highly suitable for enhancing multimodal and multicolor techniques in soft X-ray analysis, which can evaluate structural, chemical, elemental, and magnetic characteristics. Additionally, integrating the ucKB mirror with a tabletop X-ray source has the potential to extend the accessibility of scanning X-ray nanoanalyses, which has been limited to synchrotron radiation-based X-rays.

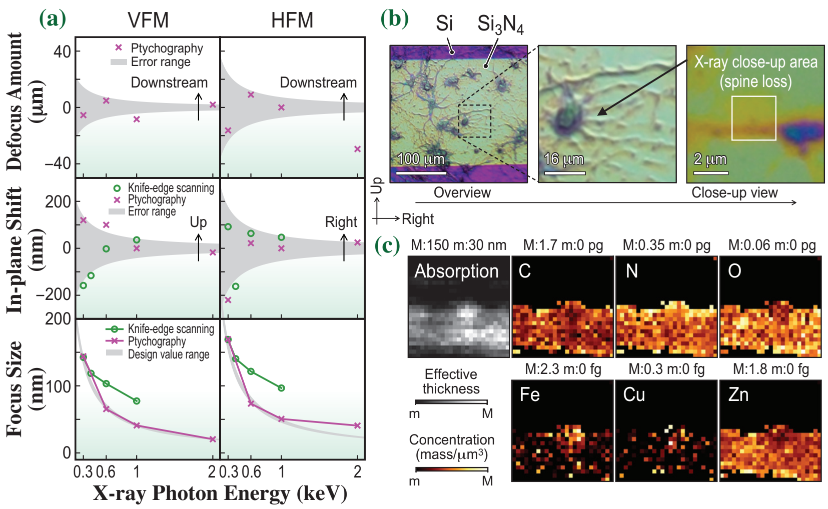

Fig. 2. Achromatic soft-X-ray nanoprobe formed by ucKB mirror. **(a)** Focus position and full width at half maximum (FWHM) focus size *versus* X-ray photon energy. The error ranges for the defocus amount and in-plane shift were calculated based on the ±Rayleigh range and ±FWHM focus size, respectively. **(b)** Visible-light micrographs of primary hippocampal neurons. The close-up area is indicated either by the box or the tip of the arrow. **(c)** X-ray absorption and fluorescence images, with units of nm and mass μm^{-3}, respectively, for primary hippocampal neurons.

Takenori Shimamura[a,*] and Hidekazu Mimura[b]

[a] Institute for Solid State Physics, The University of Tokyo
[b] Research Center for Advanced Science and Technology, The University of Tokyo

*Email: tshimamura@issp.u-tokyo.ac.jp

References

[1] B. Rösner *et al*.: Optica **7** (2020) 1602.
[2] Y. Takeo *et al*.: Appl. Phys. Lett. **117** (2020) 151104.
[3] Y. Takeo *et al*.: Appl. Phys. Lett. **116** (2020) 121102.
[4] *T. Shimamura, Y. Takeo, F. Moriya, T. Kimura, M. Shimura, Y. Senba, H. Kishimoto, H. Ohashi, K. Shimba, Y. Jimbo, H. Mimura: Nat. Commun.* ***15*** *(2024) 665.*