Supplementary information

1. Heat Dissipation Design for the Harmonic Rejection Mirror

The mirror substrate is a 150 mm long, 35 mm wide and 30 mm high silicon bar, designed to horizontally reflect the incident beam. For the heat dissipation, simulations of temperature distribution and substrate deformation using finite element analysis show that water cooling from the bottom of the substrate can maintain a smooth temperature variation and acceptable height gradient along the substrate length, shown in Figure 1, even with a large slit opening and thus a higher heat load.

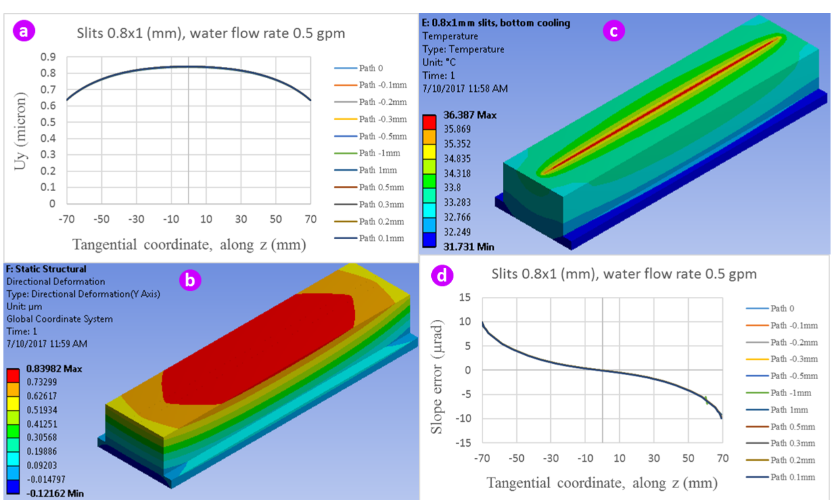

Fig. 1. Temperature distribution and height deformation with cooling from bottom. (slits opening: 0.7 mm x 1.0 mm, 6 mrad grazing incidence, incident power: 48.7 W) c) temperature distribution. upper left and lower right: height deformation. lower left: corresponding slope error along the substrate length.

Based on the simulation results, a water-cooled copper heat sink was built. The heat sink is assembled in three pieces, as shown in Figure 2. The mirror substrate is mounted on the top surface of the top block which has grooves of 5 mm wide and 4.2 mm deep on the other side. The top surface is 8 mm away from the grooves. The middle and bottom pieces have two through holes each such that when the three blocks are brazed together, the grooves constitute a water channel for the cooling water to flow inside the heat sink, with connections on the bottom. Since the copper heat sink also works as one of the vacuum chamber's walls, no in-vacuum water line fittings are necessary. The cooling water was circulated by a chiller (Thermo Scientific SC100), with the temperature set to 20 $^\circ$ C. During the experiment, a vacuum better than $6{\times}10^{-5}$ Torr was achieved.

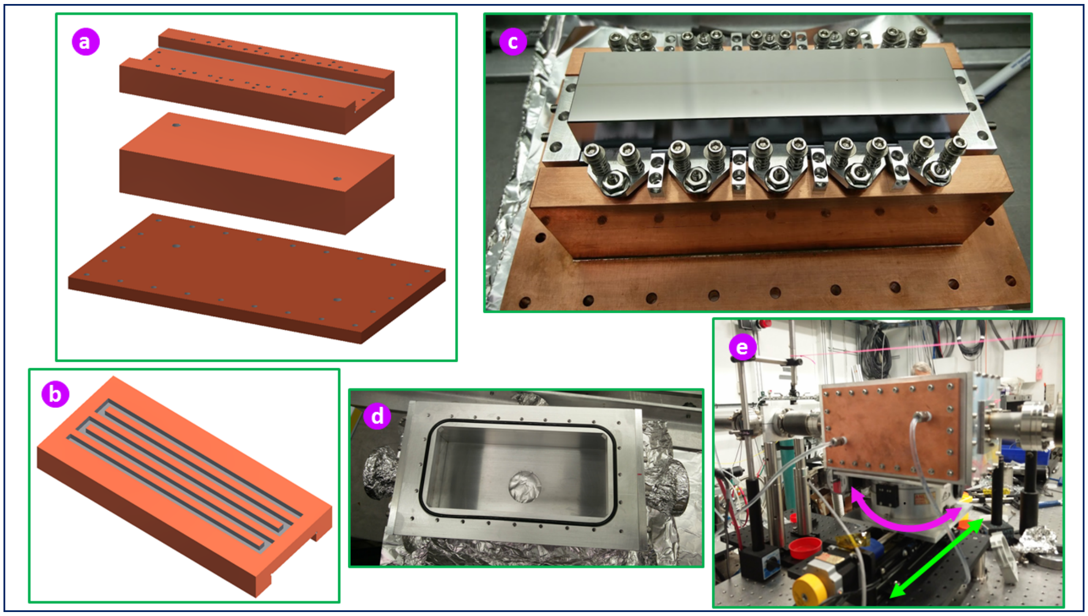

Fig. 2. Harmonic rejection mirror mount. (**a**) the heat sink is made of three copper blocks brazed together. (**b**) grooves on the top block produce a water channel below the substrate for bottom-cooling. (**c**) mirror mounted on the heat sink. A 0.1 mm indium foil is used to improve thermal conductivity. (**d**) vacuum chamber for mirror. The copper heat sink assembly is installed to the chamber and also works as one of the chamber's walls. (**e**) Mirror chamber installed at the beamline, mounted on a rotation stage and a translation stage. Two Swagelok connections are installed to circulate cooling water.

2. CRLs Ray-tracing simulations and mounting

McXtrace, a Monte-Carlo ray-tracing x-ray beamline simulation software, was utilized to determine the optimum number of lenses to focus the pink beam (Bergbäck Knudsen *et al.*, 2013). Figure 3 shows the simulations for different numbers of 200 μm radius of curvature Be lenses at 8 keV. Adding more lenses does not significantly reduce the vertical spot size, at the expense of additional absorption. Using seven lenses is sufficient to reach a vertical size of 14 μm FWHM, compatible with our MHz pump-probe design. This number was thus chosen to determine our x-ray optical setup.

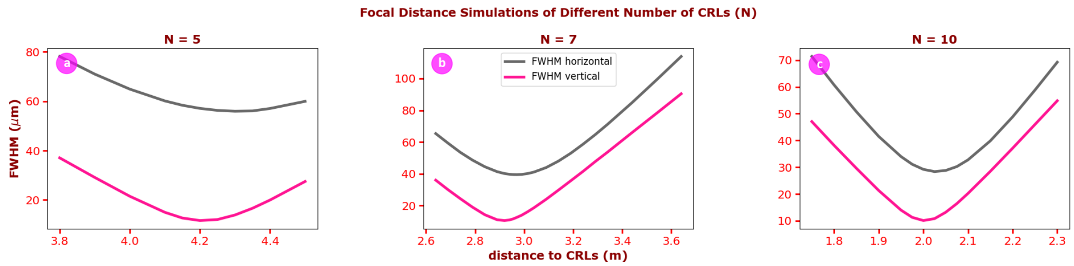

Fig. 3. Focusing simulations for different numbers of CRLs at 8 keV.

In addition to the CRLs, two Cu-Al-Ni alloy pinholes of 0.8 mm and 1.0 mm apertures respectively are installed at both end of the lens stack to limit the incoming beam size and to reduce scattered radiation outside of the paraboloids. Between the CRLs and Cu-Al-Ni pinholes, copper spacers are sandwiched to improve heat dissipation. The stack is presented in Fig. 4, inside the copper adapter that was designed to mount our 2D Be lenses inside the existing water-cooled mount made for 1D Be lenses. The lens stack is installed in a vacuum chamber with XYZ translation and pitch and yaw position controls. The CRLs were cooled in tandem with the harmonic rejection mirror, and a pressure of mid 10^{-5} Torr was maintained during the experiment.

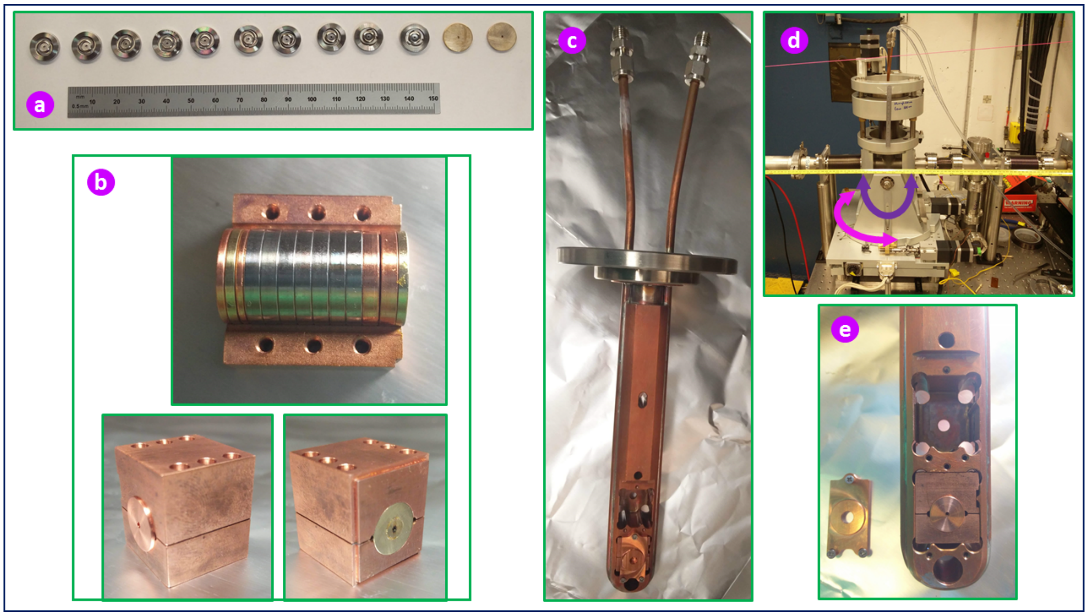

Fig. 4. (**a**) Beryllium lenses and two pinholes from RXOPTICS. (**b**) Stack mounted in adapter cube. (**c**) Water-cooled lens holder made by JJ X-Ray, with room for two lens stacks and through beam. (**d**) The motorized lens stage from JJ X-Ray installed at the beamline in 7ID-B, providing alignment adjustments. (**e**) Tight fit for the adapter maintains good thermal contact.

References

Bergbäck Knudsen, E., Prodi, A., Baltser, J., Thomsen, M., Kjær Willendrup, P., Sanchez del Rio, M., Ferrero, C., Farhi, E., Haldrup, K., Vickery, A., Feidenhans'l, R., Mortensen, K., Meedom Nielsen, M., Friis Poulsen, H., Schmidt, S. & Lefmann, K. (2013). *Journal of Applied Crystallography*, **46**(3), 679–696.