Efficient sub 100 nm focusing of hard x rays

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An x-ray beam with energy of 20.5 keV has been efficiently focused down to a spot size as small as 90 nm × 90 nm by a Kirkpatrick–Baez reflecting mirrors device. The first mirror, coated with a graded multilayer, plays both the role of vertical focusing device and monochromator, resulting in a very high flux \(2 \times 10^{13}\) photons/s and medium monochromaticity \(\Delta E/E \sim 10^{-2}\). Evaluation of the error contributions shows that the vertical focus is presently limited by the mirror figure errors, while the horizontal focus is limited by the horizontal extension of the x-ray source. With a gain in excess of a few million, this device opens up new possibilities in trace element nanoanalysis and fast projection microscopy. © 2005 American Institute of Physics. [DOI: 10.1063/1.1928191]

I. INTRODUCTION

Focusing of hard x rays has long been considered unfeasible. This is a consequence of the weak interaction of this type of electromagnetic radiation with matter, resulting in a refractive index \(n\) very close to unity. The difference with respect to unity is usually denoted the refractive index decrement \(d = 1 - n \approx 10^{-6}\) and scales as \(1/E^2\) with \(E\) the x-ray energy. Today, several focusing solutions exist including refractive, diffractive, and reflective optics or combinations of them. More original, but less efficient approaches were also proposed recently.\(^1\)\(^2\)\(^3\) Refractive optics suffer from long focal distances (scaling as \(1/E^2\)). This limitation is overcome by compound refractive lenses\(^4\) that consist in a succession of a large number of individual parabolic shape lenses. Absorption in the lens material puts restrictions on the achievable spot size and gain. The smallest spot sizes for this type of optics are presently achieved with one-dimensional microlenses.\(^4\) Diffractive optics, mainly Fresnel zone plates (FZP), exploit a phase effect that scales as \(1/E\). To obtain submicron spots the lens should be laterally structured on a similar scale. However, to preserve the efficiency at high energies, the thickness of the structures should exceed several microns. Fabrication of diffractive lenses therefore requires sophisticated nanofabrication techniques to achieve structures with high aspect ratios. Submicron focusing with FZP’s is used at several synchrotron facilities using lower energies \((E < 10\text{ keV})\).\(^5\)\(^6\)\(^7\) On compact synchrotron radiation beamlines, the focal length of the lens should be accordingly short resulting in a limited working distance. Both refractive and diffractive optics are strongly energy dispersive, scaling respectively as \(E^2\) and \(E\). As a consequence they are not practical in experiments where the energy needs to be scanned. In general they require a crystal-based monochromator as upstream optical element, which provides monochromaticity of the order of \(10^{-4}\). This represents a loss in intensity of two orders of magnitude compared to the natural line width of an undulator beam \((\Delta E/E \sim 2 \times 10^{-2})\). Reflective optics are achromatic over a large energy range and preserve an efficiency close to unity even at high energies. The main requirement is that the angle of incidence is smaller than the critical angle \(\sqrt{E/\Delta E}\) for total external reflection or that the Bragg diffraction condition is fulfilled for a multilayer coated mirror. The shape of the reflecting surfaces is crucial as it determines the quality of the achieved focus. Because of the efficiency and the absence of chromatic aberrations, reflective optics are classically chosen as focusing optical element for spot sizes down to 10 microns. Several teams work on reducing the spot sizes to a few tenth of a micron.\(^8\)\(^9\)\(^10\)\(^11\) Here we report a focus in which both dimensions are smaller than a tenth of a micron.

Third generation synchrotron sources are bright enough to generate strong beams with a large degree of spatial coherence over the acceptance of microfocusing optical elements. Diffraction limited focusing of the beam is thus possible if the quality of the optical components is sufficient. Only recently focused spots, comparable in size to those achieved with FZP’s at low x-ray energies, have been obtained at higher energies with mirrors in the Kirkpatrick–Baez (KB) architecture as the focusing device. In practice longer beamlines, like the 150 m ID19 beamline at ESRF in Europe or the one kilometer beamline at Spring-8 in Japan have been used above 15 keV. The long propagation distance results in a practical working distance compatible with a sample environment a few tens of millimeters in size.

The KB developments have been started at the ESRF 8 years ago and the technology is now widely in use on 18 beamlines at this facility. We present here the latest developments and results obtained with the KB technology at ESRF along with the future perspectives.

II. DEVICE DESIGN

The Kirkpatrick–Baez device\(^13\) consists of 2 orthogonal reflecting surfaces with elliptical shapes. The adaptive system is shown in Fig. 1. Its architecture is similar to the systems previously described.\(^14\)\(^15\)\(^16\) It is composed of a 170 mm long mirror \(M_v\) focusing at a distance \(f_v = 280\) mm from the center of the mirror in the vertical direction and a 92 mm long mirror \(M_h\) with a \(f_h = 93\) mm focusing distance in the horizontal direction. An x-ray sensitive camera (XCCD) is included in the device to align the system automatically. Four...
actuators (Newfocus picomotors) bend the flat polished mirrors (Wave precision Inc.) into the stigmatic elliptical figures required for imaging the synchrotron source. The transmission of the efforts to the silicon mirrors is achieved by a flexure hinge mechanism.\textsuperscript{17} Four stepping motors (microjacks) position the mirrors with respect to the beam in rotation and translation. The vertical mirror $M_v$ is coated with a multilayer resulting in a high energy bandwidth ($\Delta E/E = 6\%$). The multilayer consists of 30 W/B$_4$C layers with a period of $4.7$ nm at the center. It has a $33\%$ nonlinear gradient in period along the length of the mirror, designed so that the Bragg angle condition is kept despite the variation in the angle of incidence. This design is also necessary to achieve a spot size smaller than the penetration depth of the beam within the $140$ nm thick multilayer. The main benefits of the multilayer design are:

1. The full energy bandwidth of one harmonic of the undulator ($\sim 2\%$) can be used, as opposed to the typical $10^{-4}$ bandwidth of a silicon monochromator, resulting in at least two orders of magnitude increase in photon flux.
2. The larger incidence angle of the multilayer increases the acceptance by a factor of $2.5$ with respect to the single layer platinum coated horizontal mirror. This increase in aperture results in either greater flux when using the full mirror length or a reduction in figure error when only a fraction of the mirror surface is used.

III. EXPERIMENTAL SET UP

The results reported here were obtained on the $150$ m long ID19 beamline using an energy of $20.5$ keV, although the device has also been used at an energy of $15$ keV with similar results. No optical elements apart from slits and a $2$ mm thick polished aluminum filter (transmission $18\%$ at $20.5$ keV) were inserted between the source and the KB device. The gap of the $32$ mm period undulator was selected to adjust its third harmonic to $20.5$ keV, the fundamental at $6.8$ keV being effectively suppressed by the aluminum filter, the beamline windows and the air path. The KB device and compact sample stage are mounted together on a honeycomb table that is both rigid and light. The table is designed to avoid amplification of the floor vibrations that have their highest levels at about $10$ Hz. The angle of incidence at the center of the mirrors is $\theta_v = 7$ mrad and $\theta_h = 2.9$ mrad for $M_v$ and $M_h$, respectively. The FWHM source size is $135(h) \times 25(v)$ microns. The source size values are known to be precise to about $10\%$. The SHADOW (Ref. 18) ray tracing program that considers each pencil beam independently (a significantly incorrect assumption considering the coherence of the beam), predicts a FWHM spot size of $s_{wh} = 62$ nm and $s_{hv} = 40$ nm, respectively, in the horizontal and vertical direction, which is therefore the minimum size that could be obtained in the absence of diffraction. The experiments were performed with the synchrotron ring operating in a low current mode (16 bunch mode). The ring current was approximately $80$ mA, as compared with the $200$ mA maximum current delivered at the ESRF.

IV. EXPERIMENTAL RESULTS

A. Vibration levels

The XCCD camera positioned in the focal plane is used as a beam position monitor (BPM) by integrating the image over each direction and using a Fourier transform centroid algorithm. Its intrinsic precision has been measured by evaluation of the distance between $2$ large fixed apertures, resulting in a standard deviation of $4$ nm rms, but is unknown in the case of a small spot encompassing very few pixels as in this application. Figure 2 shows a time scan of the beam position acquired in the focal plane, with a vibration broadening $\sigma_{v\perp} = 29$ nm and $\sigma_{h\perp} = 27$ nm standard deviation in, respectively, the vertical and horizontal direction. It can be estimated that this is an upper bound for camera noise, vibrations, and short-term drift levels of the overall system. The $2$ ms camera integration time ensures that most mechanical vibrations are taken into account in the statistics. Attaching the KB device and camera to the same highly rigid support thus effectively avoids significant vibrations on the system. The absence of a monochromator, which often induces vibrations and drifts, is an asset for such a system, and is only made possible by the use of the multilayer mirror as an energy-selecting device.
B. Wavefront optimization

A scanning procedure of the entrance slits defining the beam size is used to perform a linear optimization of the figuring of the mirrors with the bending actuators. The contribution of a set of portions of the mirror is evaluated by measuring the position on the camera of the corresponding pencil beam selected by the slit. The influence of each actuator on this set of contributions is described by linear equations. Once the interaction matrix characterizing the system is determined, the value to be applied to each actuator can be derived. The scanning procedure also allows centering the illuminated region on the best portion of the mirror and selecting the width of the illuminated region according to the spot size desired (at the expense of reduced flux). The pencil beam positions $\Delta Z_{BPM}$ and $\Delta Y_{BPM}$, in the vertical ($Z$) and horizontal ($Y$) directions perpendicular to the x-ray direction, characterize which portion of the mirrors contributes to the focus spot broadening. They are proportional to the partial derivative of the wavefront error $\Delta W$ with respect to the spherical wavefront centered on the focal point

$$\Delta Z_{BPM} = \frac{\partial \Delta W(\alpha, \beta)}{\partial \alpha}$$

and

$$\Delta Y_{BPM} = \frac{\partial \Delta W(\alpha, \beta)}{\partial \beta},$$

where $\alpha$ and $\beta$ are the angular coordinates in, respectively, the vertical and horizontal direction.

Although we use two linear scans in both directions, the full wavefront error can be measured by a two-dimensional scan. Figure 3 shows BPM scan results over a large portion of the mirrors, with deviations much larger than the spot size goal of our system, meaning that figure error contributions have to be limited by illuminating only a portion of the mirror surfaces. Once the optimum beam acceptance and centering have been chosen, the physical optimization routine is performed again over this range, and then the slits opened accordingly to illuminate the proper zone of the mirrors. An aperture of $400(v) \times 100(h)$ microns is selected for optimum results.

C. Focus spot size measurement

The classical method to measure submicron spot sizes uses a knife-edge which is translated perpendicular to the beam while the transmission or fluorescence signal is analyzed by a detector. The intensity distribution obtained is then differentiated to get an estimate of the spot profile in one direction. Because of the differentiation, small errors in the knife-edge position or temporal variations of the beam translate into large errors in the estimation of the spot shape. In order to directly measure the beam intensity profile in one direction, we designed a nanowire with an equivalent width much smaller than the expected spot size. A 38 nm thick gold layer was sputtered on a polished vitreous carbon substrate. A 3 micron wide structure with Gaussian shape was obtained by ion milling of the gold layer as shown in Fig. 4, leaving a gold wire, which can be detected by fluorescence. At 9 mrad ($\sim 0.5^\circ$) grazing incidence with respect to the carbon substrate, no reflection occurs and the gold structure equivalent width at normal incidence is 28 nm. Both thickness and width of the wire combine into an equivalent slit with a FWHM of roughly $s_{nw} = 43$ nm, about two times smaller than the expected spot size. The gold nanowire is mounted on a lever based translation mechanism actuated by a stepping motor, with mean increments of 10 nm. Equivalent results were obtained with a piezoelectric actuator. The fluorescence signal is acquired by a silicon diode to $2 \times 10^{11}$ photons/s at a ring cur-
rent of 80 mA. The expected flux with a normal ring current of 200 mA is thus \(5 \times 10^{11}\) photons/s. Figure 6 shows the spot size (FWHM) when the size of the beam incident on the KB device is varied. At low apertures, the diffraction is predominant, whereas at higher apertures the mirror figure errors are predominant. The latter induce basically an increase of the tails of the profiles leaving the FWHM initially unaffected. The gain in intensity obtained by opening the slits above a certain level is therefore misleading and contributes essentially to a background increase.

V. DISCUSSION

Several contributions to the measured finite spot size can be considered. The major ones are: the finite source size \(s_{sh}\) and \(s_{sv}\), diffraction, mirror figure errors, mechanical vibrations \(s_{vv}\) and \(s_{vh}\) and the equivalent width of the nanowire \(s_{nw}\). In the following we evaluate the importance of the different contributions to the focus size.

A. Mirror figure and diffraction contributions

Beam profile prediction from the slope errors of the mirrors is not valid since the degree of coherence of the incident beam is large. Instead a wave-optical approach is used. By integrating Eq. (1), the wavefront error \(D_w\) originating from the mirrors figure errors can be computed. A first approximation of the focal spot distribution is the finite Fourier transform of the phase term \(e^{i\varphi}\), with the phase \(\varphi=2\pi\Delta W/\lambda\) and a mean propagation distance corresponding to the focal length \(f\) at a wavelength \(\lambda\). In the absence of wavefront errors, the diffraction limited sinc \(2\) function FWHM amounts to \(0.88\lambda f_w/w_v\) or 38 nm for an acceptance \(w_v\) of 400 \(\mu\)m in the vertical direction. The equivalent diffraction contribution amounts to \(s_{dh}=50\) nm FWHM for a slit width of 100 \(\mu\)m in the horizontal direction. Figure 7 shows the profiles obtained taking into account the figure errors and diffracting the wavefront. The FWHM values are only slightly broadened due to the wavefront errors, the peak intensities are lowered by about 30% with respect to the diffraction limited case, and this power is distributed in the tails that are enhanced with respect to the ideal case.

B. Error budget evaluation

The evaluation of the errors leads to different conclusions for the vertical and horizontal focusing. The contribution of the mechanical vibrations and the finite width of the gold slit were shown to be two times smaller than the measured spot size. For the horizontal focusing, the main factors limiting the spot size are both the horizontal source size of \(135\pm10\%\) \(\mu\)m and mirror figure limitations, prohibiting the use of a larger numerical aperture. An improvement of the horizontal spot size could thus be obtained, at the expense of flux, by slitting down the source or working with a secondary source. In the vertical direction on the other hand, the source size contribution is negligible and the finite spot size is mainly related to the long focal distance of 280 mm. A reduction of the vertical focal distance and/or an improvement of the mirror quality are required to reduce the vertical spot.

FIG. 5. Beam profiles measured in the focal plane by fluorescence and scanning the gold nanowire. The intensity of the 20.5 keV x-ray beam is \(2\times10^{11}\) photons/s at 80 mA storage ring current. The slit size is 400 \(\mu\)m \(\times\) 100 \(\mu\)m \((\mu\)h) corresponding to a gain of approximately \(4\times10^6\).

FIG. 6. Spot size (FWHM of the beam profile) as a function of the slit width. Squares and triangles correspond to the vertical and horizontal focusing direction, respectively.

FIG. 7. Simulated beam profiles in the focal plane considering diffraction and the wavefront error measured by the XCCD camera.
size further below the 50 nm level. Better mirror figuring can be obtained with a finishing method such as ion beam figuring or elastic emission machining, coupled to an ex situ metrology method.

The efficient focusing of hard x rays to a spot smaller than 90 nm in both directions at high energies (15–20 keV) is a major breakthrough in x-ray optics. It is expected to have numerous applications in the field of projection microscopy and microanalysis of trace elements. Extension of the same approach to higher x-ray energies using longer mirrors is in progress.

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18 http://www.xraylith.wisc.edu/shadow/shadow.html