A. Beam divergence contribution terms in presence of mirrors

Using a Gaussian approximation, the reflection probability of an X-ray beam incident at a glancing angle θ_i on optically-bent parabolic mirror is derived within the geometrical optics approximation (Susini, 1995). *Optically-bent* mirror means that the source sits on the mirror focal point (first mirror) and that the nearly parallel incident beam is focused at the mirror focal point (second mirror) where the sample is located, as schematically shown in the article's Fig. 1. The profile of real mirrors is, however, always affected by geometrical errors due to gravity, thermal deformation, and/or incorrect bending moments. Referring to the optics setting described in Fig. 1, expressions for the FWHM beam angular divergence $\Delta\tau_p$ and $\Delta\tau_f$, which take explicitly into account the influence of the above mentioned source of errors are derived.

Let us consider the parabola Π_1 in Fig. A with origin in O in the Cartesian reference system OXYZ and be the X-ray source S placed in its focal point. A mirror M_1 of length L_1 with the asymmetric profile Π_1 placed at a distance p_1 from the source ($p_1 = \overline{P_1S}$, $p_1 >> L_1$, $P_1(X_0,Z_0)$ mirror pole) would satisfy the ideal optical conditions for a collimating mirror. Let Π_2 be a second parabola in a new reference system oxyz with origin in the pole of the mirror P_1 defining the shape of mirror M_1 when a bending, symmetric with respect to its pole, is applied. In real cases, and the MS beamline is an example, a symmetric bending Π_2-type often replaces an asymmetric bending Π_1-type to reduce the complexity of the mirror bending mechanisms. Whether the applied bending is symmetric or asymmetric, for manufacturing and metrology purposes it is always more convenient to define the parabolas in the oxyz reference system with origin in the mirror pole and expand the profile in a McLaurin series (Noda *et al*., 1974). In fact, the coefficients of the series can be easily related to the various optical aberration terms (Howells, 1994, p. 381), due to the differences near the pole between Π_1 and Π_2. Similarly, one would define a second pair of parabolas Π_3 and Π_4 to describe geometrical aberrations of the second mirror M_2 of length L_2 and pole P_2 placed at a distance p_2 ($L_2 << p_2$) from the focal point of the Π_3 parabola, where the sample is placed. For most applications in grazing incidence X-ray optics, the main source of error comes from coma and spherical aberrations (Susini, 1995) and a forth-degree polynomial expansion is enough to describe with good approximation hard x-ray mirrors, for which higher order aberrations are negligible. Within this approximation, the ideal optical mirror profile (Π_1 or Π_3) in the mirror coordinate system is:

$$z_{i,0}(x) \approx \alpha_{i,0} x^2 \left(1 + \beta_i x + \gamma_i x^2\right), \tag{A1}$$

where the Maclaurin-expansion coefficients $\alpha_{i,0}$, β_i and γ_i, can be analytically expressed, for any shape and, for a parabolic shape are given by[1]

$$\alpha_{i,0} = \frac{\sin\theta_i}{4p_i}; \qquad \beta_i = -\frac{\cos\theta_i}{2p_i}; \qquad \gamma_i = \frac{5\cos^2\theta_i}{16p_i^2}. \tag{A2}$$

Here: x is the coordinate that locates a generic point along the i[th] mirror (i=1 for M_1 and i=2 for M_2) with respect to the local reference system $\{xyz\}_i$ with origin in the mirror pole $P_i(X_0,Y_0)$; p_1 is the source to first-mirror pole distance, p_2 is the sample to second-mirror pole distance and θ_i is the mirror grazing incidence angle (less than the critical angle of total reflection θ_c) calculated at the M_i pole. The variation of the radius of curvature along the mirror is given by (Susini, 1995):

$$\frac{1}{R_c} = 2\alpha_{i,0}\left[1 + 3\beta_i x + 6\left(\gamma_i - \alpha_i^2\right)x^2\right] \tag{A3}$$

Therefore, the mean optical curvature radius (calculated in P_i) is related to $\alpha_{i,0}$ by the expression $R_o = 1/2\alpha_{i,0}$.

Given a symmetric bending, the mirror profile Π_2 (or Π_4) should, therefore, be as closest as possible to the one of Π_1 (or Π_3). However, at very large radius of curvatures, parasitic effects due to gravity and thermal deformation generally significantly influence the mirror curvature. Therefore, taking into account all the above contributions, the actual mirror figure is given by

$$z_{i,tot}(x) = z_{i,b}(x) + z_{i,g}(x) + z_{i,t}(x) = \alpha_{i,b} x^2 + s_i \alpha_{i,g}\left(\frac{x^2 L_i^2}{2} - x^4\right) + \alpha_{i,t} x^2 + const \tag{A4}$$

with $\alpha_{i,g} = \dfrac{5\rho_i g}{2Y_i T_i^2}$, $\alpha_{i,t} = \dfrac{C_i}{4\pi}\dfrac{a_i}{\kappa_i}\dfrac{P_{i,abs}}{W_i}\dfrac{\theta_i}{h_{i,z}}$

where: $z_{i,b}(x)$, $z_{i,g}(x)$ and $z_{i,t}(x)$ are the mirror profile contributions due to a symmetric bending, gravity sag[2] and thermal deformation, respectively, with the latter approximated to the first-order

[1] The α and β symbols employed here to represent the series expansion coefficients are not at all correlated with those used in Sections 1 and 2. Here, also, we made the choice to maintain Susini's terminology judging that the context where these symbols were used was different enough for not inducing the reader into confusion.

of θ_i (Susini, 1995); the constant term expresses the maximum total deviation (in P_i) due to the sum of all the above-mentioned physical effects; ρ_i is the mirror density, Y_i the Young modulus, T_i the mirror thickness, a_i the thermal expansion coefficient, κ_i the thermal conductivity, W_i the mirror width, C_i the cooling geometry constant, $h_{i,z}$ the vertical half height of the photon beam, P_{abs} the total power absorbed in the mirror and g the gravity acceleration; $\alpha_{i,b}$ can be modified by the user with a suitable bending of the mirror in order to maximize its optical performances and s_i is a sign function (+ for M_1 and – for M_2) which takes into account that M_2 is up side down with respect to M_1.

A1. Beam divergence distribution function width after reflection by the collimating mirror M_1

For mirror M_1, the difference between the $z_{1,tot}(x)$ and $z_{1,0}(x)$ profiles leads to orientation errors of the normal to the mirror surface in any given point x. This slope error distribution along the mirror due to the incorrect profile or, equivalently, to the incorrect orientation of the normal to the mirror surface, is given by:

$$\begin{aligned}\Delta z_1'(x) &= \frac{dz_{1,0}(x)}{dx} - \frac{dz_{1,tot}(x)}{dx} = \frac{d}{dx}\left(z_{1,0}(x) - z_{1,tot}(x)\right) = \\ &= 2(\alpha_{1,0} - \alpha_{1,b} - \alpha_{1,t} - \frac{1}{2}\alpha_{1,g}L_1^2)x + 3\alpha_{1,0}\beta_1 x^2 + 4(\alpha_{1,0}\gamma_1 + \alpha_{1,g})x^3\end{aligned} \quad \text{(A5)}$$

The residual FWHM beam angular divergence $\Delta\tau_p$ after reflection by the first collimating mirror caused by the wrong profile will be, then, given by the maximum angular deviation from the ideal value of the normal to the mirror surface, i.e. by the value of $\Delta z_1'(x = x_{max})$ [3]. However,

---

[2] Starting from a 4th degree polynomial $z_{i,g}(x) = ax^4 + bx^3 + cx^2 + dx + e$ and applying the following boundary conditions:

$$z_{i,g}(x) = z_{i,g}(-x)$$

$$z_{i,g}(0) = z_{i,g,\max} = \frac{5\rho_i g L_i^4}{32 Y_i T_i^2} \text{ (Susini, 1995, Eq.(31))}$$

$$z_{i,g}(L/2) = 0 \text{ and } z_{i,g}'(L/2) = \frac{dz_{i,g}}{dx}\Big|_{L/2} = 0$$

the latter justified by a 4-point bending holding of the mirror, one easily obtains the expression in (A4).

[3] Note that the maximum angular deviation from the ideal value of the normal to the mirror surface directly gives the FWHM beam angular divergence $\Delta\tau_p$ (and analogously $\Delta\tau_f$). The corresponding width $\Delta\tau_p'$ (and analogously $\Delta\tau_f'$) of the beam divergence probability distribution function that appears in equation (3) of the main article is, then, obtained by simply dividing the FWHM by $2\sqrt{\ln 2}$.

depending on the applied bending, the global maximum $x_{\max}$ of the $\Delta z_1^{'}(x)$ function can be reached on the mirror or outside it. When $x_{\max} \notin \left[-\frac{L}{2}, \frac{L}{2}\right]$, the maximum angular deviation is assumed reached at the end of the mirror $\Delta z_1^{'}\left(x = \frac{L}{2}\right)$, e.g. at the point that is farthest away from the mirror pole. Therefore, we write:

$$\Delta\tau_p \cong \begin{cases} \Delta z_1^{'}(x_{max}) = \dfrac{\left\{4\alpha_{1,g}\left(2\alpha_{1,b} + 2\alpha_{1,t} + L_1^2\alpha_{1,g}\right) + \alpha_{1,0}^2\left(3\beta_1^2 - 8\gamma_1\right) + 4\alpha_{1,0}\left[2\left(\alpha_{1,b} + \alpha_{1,t}\right)\gamma_1 + \alpha_{1,g}\left(L_1^2\gamma_1 - 2\right)\right]\right\}^{3/2}}{24\sqrt{3}\left(\alpha_{1,g} + \alpha_{1,0}\gamma_1\right)^2} \\ \Delta z_1^{'}\left(\dfrac{L}{2}\right) = L_1\left(\alpha_{1,0} - \alpha_{1,b} - \alpha_{1,t}\right) + \dfrac{3}{4}L_1^2\alpha_{1,0}\beta_1 + \dfrac{1}{2}L_1^3\alpha_{1,0}\gamma_1 \end{cases} \qquad \text{(A6)}$$

We observe that, when the gravity sag can be neglected, the equivalence $\Delta\tau_p = \Delta z_1^{'}\left(\frac{L_1}{2}\right)$ holds[4]. In this case the first term of $\Delta z_1^{'}\left(\frac{L_1}{2}\right)$ describes differences between the real and ideal radius of curvature, whereas the two additional terms in L_1^2 and L_1^3 describe aperture effects and spherical aberrations (Susini, 1995).

So far, we have been considering the source as a point-like source. However, using purely geometrical considerations, the finite source dimensions *S* can be taken into account and the (A6) generalized as follows (Howells, 1994; Susini, 1995):

$$\Delta\tau_p \rightarrow \Delta\tau_p + \frac{S}{2p_1} + \frac{SL_1}{8p_1^2}. \qquad \text{(A7)}$$

The expression above is directly obtained from Susini's equation (9) (Susini, 1995) simply dividing the spot size by the image distance q (in order to calculate the maximum angular divergence) in the limit of $q \rightarrow \infty$ (for a parabola).

Defining $\alpha_{1,b} = c_1\alpha_{1,0}$, one can evaluate the bending degree c_1 (calculated with respect to the ideal optical curvature) necessary to compensate as much as possible gravity sag, thermal expansion and other aberrations. For negligible gravity and thermal effects, the optimal $\Delta\tau_p$

---

[4] When the third-order term of the mirror profile induced by the thermal expansion cannot be neglected, the (A6) needs to be further generalized. However, since this third-order contribution is proportional to θ_i^3 (Susini, 1995) it can be usually neglected.

value, at the first order in x, would, then, require $c_1 \cong 1$. When gravity and thermal effects are not negligible, for a collimating mirror oriented as in the article's Fig.1, one would instead expect that $c_1<1$ for an optimal bending, and $c_1>1$ for an overbending. For such a mirror, in fact, gravity and thermal effects would already cause a mirror bending (concave mirror surface). The optimal condition $\alpha_{1,b} = \alpha_{1,0}$ would, therefore, be already reached for $c_1<1$ and definitely past for $c_1>1$ corresponding to the symmetric profile above the asymmetric one. The optimum c_1 values for 10, 13 and 25 KeV have been estimated and found equal to 0.8, 0.73 and 0.5, respectively. Note that the minimum IRF FWHM value at different energies is reached at different c_1 values. This is related to the fact that the optical radius of curvature increases as a function of the energy. On the other hand, the gravity radius of curvature is always the same. Thus, starting from the mirror bent by gravity, in order to reproduce the optical radius of curvature, one needs to apply smaller bending moments at higher energies. The theoretical optimum value of $\Delta\tau_p$ for a collimating mirror like the one at the SLS MS beamline is the same for all photon energies and equal to 15 μrad. As we said, this optimum value corresponds to different $c_{1,optimum}$ values since it is a function of the photon energy.

A2. Beam divergence distribution function widths after reflection by the refocusing mirror M_2

An analogous derivation can be made for the second refocusing mirror described in the article's Fig. 1. Since a nearly parallel beam impinges on M_2, the evaluation of the FWHM of the beam divergence $\Delta\tau_f$ after reflection by M_2 only requires the differentiation of (A4) with respect to *x*:

$$z'_{2,tot} = \frac{dz_{2,tot}(x)}{dx} = x\left(2\alpha_{2,b} - \alpha_{2,g}L_2^2 + 2\alpha_{2,t}\right) + 4\alpha_{2,g}x^3 \qquad \text{(A8)}$$

Similarly to what was done for the first mirror, the value of $z'_{2,tot}$ in x(max) would give the maximum variation of the normal to the mirror surface due to its curvature and, therefore, a good approximation of the FWHM beam angular divergence $\Delta\tau_f$ after reflection by the second refocusing mirror in the case of a bent second mirror[3]:

$$\Delta\tau_f \cong \begin{cases} 2z'_{2,tot}(x_{max}) = 2\dfrac{(-2\alpha_{2,b} + L_2^2\alpha_{2,g} - 2\alpha_{2,t})^{3/2}}{2\sqrt{3\alpha_{2,g}}} \\ 2z'_{2,tot}\left(\dfrac{L}{2}\right) = 2L_2(\alpha_{2,b} + \alpha_{2,t}) \end{cases} \tag{A9}$$

where the factor of 2 takes into account that the normal variation has opposite sign in the mirror extremes and this doubles the divergence. Again, the first value applies when the $|x_{max}|$ falls within the interval $\left(0, \dfrac{L_2}{2}\right)$ and the second when it falls outside it.

Defining, as done for M_1, $\alpha_{2,b} = c_2\alpha_{2,0}$, the parameter c_2 gives the bending degree of M_2, whereas the condition $\alpha_{2,b} = c_2\left(\dfrac{L_2^2\alpha_{2,g}}{2} - \alpha_{2,t}\right)$ holds for M_2 in a flat configuration and define the optimal choice of c_2 in order to reduce residual divergences due to gravity sag and thermal deformation in a flat mirror. These effects cannot, in this case, be completely compensated by a parabolic bending and the parameter c_2 can be referred to as the gravity and thermal curvature.

The condition for the minimum value of $\Delta\tau_f$ when M_2 is in a flat configuration is reached when $z'_{2,tot}(x_{max}) = z'_{2,tot}\left(\dfrac{L}{2}\right)$ that is for:

$$\frac{(-2\alpha_{2,b} + L_2^2\alpha_{2,g} - 2\alpha_{2,t})^{3/2}}{2\sqrt{3\alpha_{2,g}}} = L_2(\alpha_{2,b} + \alpha_{2,t}) \tag{A10}$$

If the thermal deformation induced curvature is negligible $(\alpha_{2,t} \approx 0)$, the solution of (A10) gives $\alpha_{2,b} = \dfrac{L_2^2\alpha_{2,g}}{8}$ and the condition $\alpha_{2,b} = c_2\left(\dfrac{L_2^2\alpha_{2,g}}{2} - \alpha_{2,t}\right) = c_2\dfrac{L_2^2\alpha_{2,g}}{2}$ would, then, be satisfied for $c_2 = \dfrac{1}{4}$. The value $\dfrac{1}{4}$ would, then, be the optimal choice of c_2 to reduce residual divergences due to gravity sag in a flat mirror.[5] For bent configurations one should expect $c_2 \cong 1$ in ideal situation and values >1 (<1) indicating overbending (underbending). The two coefficients c_1 and c_2 introduced here are the only free parameters of the model, all the other quantities being experimentally measured.

[5] Flat configurations of the first mirror can be modeled in the same way.

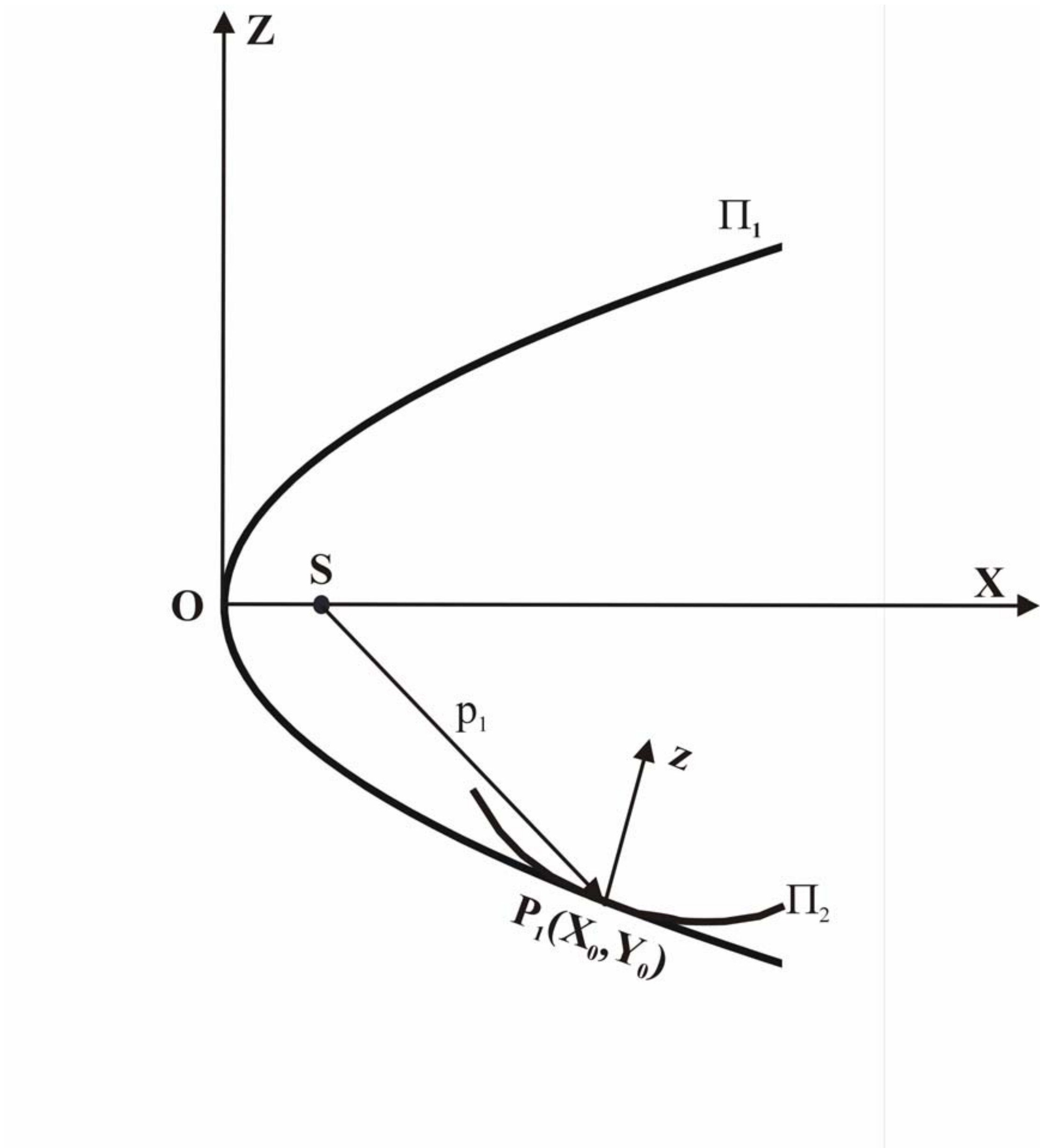

Figure A Symmetric versus asymmetric bending in parabolic mirrors.

References

Howells, M. R., in New Directions in Research in Third-Generation Soft X-Ray Synchrotron Radiation Sources, 359-385; Schlachter A.S. and Wuilleumier F.J. editors, 1994 Kluwer Academic Publishers (Netherland).

Noda, H, Namioka, T. & Seya M. (1974). J. Opt. Soc. Am. **64**, 1031-1036.

Susini, J. (1995). Optic. Eng. **34**, 361-376.

B. Summary of the full pattern FullProf fitting parameters

	$10\,keV$	$13\,keV$	$25\,keV$
U	0.000588 ($focus$ & $flat$)	0.000230 ($focus$ & $flat$)	0.000861 ($focus$ & $flat$)
V	-0.000119 ($focus$ & $flat$)	-0.000035 ($focus$ & $flat$)	-0.000068 ($focus$ & $flat$)
W	0.000167 ($focus$) 0.000010 ($flat$)	0.000067 ($focus$) 0.000004 ($flat$)	0.000018 ($focus$) 0.000003 ($flat$)
X	0.009548 ($focus$) 0.007591 ($flat$)	0.003754 ($focus$) 0.004485 ($flat$)	0.00085 ($focus$) 0.002904 ($flat$)
Y	0.001688 ($focus$) 0.002844 ($flat$)	0.002274 ($focus$) 0.002523 ($flat$)	0.000318 ($focus$) 0.000567 ($flat$)

Table B Full pattern FullProf fitting parameters U, V, W and X, Y for all experimental data sets discussed in the article. The U, W and W parameters were derived according to equation (5) (see Section 7.4, 3rd paragraph) and kept fixed (or only slightly refined) during the FullProf refinements, whereas the X and Y parameters were left free to vary. It should be noted that, according to equation (5), the U and V parameters are the same for the bent-bent to the bent-flat optical configurations.