Pendellösung suppression in the diffraction pattern of a set of thin perfect crystals in a Bonse-Hart camera

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Abstract

In a Bonse-Hart camera, a set of two or more grooved (channel cut) perfect crystals are used to provide a very high angular resolution experiment. The resulting multi-reflection profiles can be calculated in a first approximation by the dynamical theory of diffraction, where usually only coherent scattering is considered. In addition, there is experimental evidence of incoherent compton scattering (ICS) and thermal diffuse scattering (TDS) contributions. Such contributions are especially important when performing small angle scattering experiments where intensities in an interval of ten orders of magnitude are often recorded. We propose and analyze here the use of thin (a few micrometers) crystals for a Bonse-Hart camera in order to decrease the TDS and ICS components, thus to increase the performances of the device. However, using thin crystals causes the occurrence of interference fringes (Pendellösung) which degrades the instrument resolution. We study in this paper the possible elimination of Pendellösung fringes by angularly offsetting one crystal with respect to the others. Optimizing the offset value, the Pendellösung oscillations of the crystals interfere destructively, then significantly reducing their contribution to the total resolution function.

1. Introduction

The combination of several perfect single crystals is often used in x-ray optics to achieve an optimum matching of source parameters and experimental requirements. Examples are given for instance by Hart, Rodrigues and Siddons [1] and by

Matsushita and Hashizume [2]. A well-known application of such crystal systems is the so-called Bonse-Hart camera that is used for very high resolution x-ray small-angle scattering [3]. Here grooved crystals provide x-ray beams with a very sharp decrease of the angular intensity distribution that is produced by multiple reflections inside the channel-cut crystals. The first channel-cut conditions the beam emerging from the source whereas a second, identical crystal serves as an analyzer of the beam scattered by the sample positioned between monochromator and analyzer.

It has been shown experimentally that the slope of the instrument resolution function given by the composition of the angular x-ray response functions does not simply follow the curve calculated from the dynamical theory but is affected by incoherent (Compton) scattering (ICS) and thermal diffuse scattering (TDS) (see e.g. Ref. [4]). These scattering processes strongly limit the performance of the Bonse-Hart device by the fact that the slope of the convoluted reflection curves becomes less steep at a certain distance from the main Bragg peak. The quantitative behavior is presently unclear but from a qualitative point of view it is a well-known fact that the intensity of both ICS and TDS are proportional to the volumes in real and in reciprocal space occupied by the photons and that therefore the instrument function could depend on the crystal thickness.

Until now and to our knowledge mostly thick crystals have been used in Bragg (reflection) geometry. For the sake of clearness we call a crystal "thick" when its thickness is much larger than the so-called "extinction length" that is the penetration depth of x-rays into crystals needed to fully reflect the beam. This thickness is typically a few micrometers whereas the absorption length is much larger, in particular for higher energy x-rays [5]. By decreasing the crystal thickness it could be possible to decrease ICS and TDS and thus to increase the performance of the Bonse-Hart device.

Thin crystals present a drawback related to the occurrence of Pendellösung fringes which produce a fine structure on the tails of the Bragg peak. In order to fully benefit from the TDS and ICS decrease the Pendellösung fringes must be eliminated or at least reduced. This can be achieved by creating a destructive interference effect between the crystal reflectivity curves in the Bonse-Hart camera. The aim of the present theoretical study is to assess the feasibility of this approach prior to performing the corresponding experiments. Experimental investigations with thin crystals have shown that their x-ray reflection properties match perfectly well with the theoretical predictions (see e.g. Ref. [6]).

2. Theoretical background

The reflectivity or response function of a single perfect parallel-sided crystal of thickness t_o in Bragg (or reflection) geometry can be calculated from the dynamical theory of diffraction as [7]:

$$R(\Delta) = \frac{1}{b}\frac{I_H}{I_o} = \frac{1}{b}\left|\frac{x_1 x_2 (c_1 - c_2)}{c_2 x_2 - c_1 x_1}\right|^2 \qquad [1]$$

where I_H is the intensity of the external diffracted wave, I_o is the intensity of the incident wave, $c_1=exp(-i\varphi_1 t_o)$, $c_2=exp(-i\varphi_2 t_o)$, $\varphi_1=2\pi k_o \delta'_o/\gamma_o$, $\varphi_2=2\pi k_o \delta''_o/\gamma_o$; γ_o is the direction cosine of the incident wave and the other quantities are defined as:

$$\begin{pmatrix} \delta'_o \\ \delta''_o \end{pmatrix} = \frac{1}{2}\left(\psi_o - z \pm \sqrt{qP^2 + z^2}\right) \qquad [2]$$

$$\begin{pmatrix} x_1 \\ x_2 \end{pmatrix} = \frac{-z \pm \sqrt{qP^2 + z^2}}{P\psi_{\bar{H}}} \qquad [3]$$

$$z = \frac{1-b}{2}\Psi_o - \frac{b}{2}\alpha \qquad [4]$$

$q=b\Psi_H\Psi_{\bar{H}}$, $\alpha = 2\,\Delta\, sin\theta_B$, $\Delta = \theta - \theta_B$, θ being the incidence angle, θ_B the kinematical Bragg angle and P the polarization factor. Ψ_H is the Fourier component of the electrical susceptibility Ψ_0, b is the asymmetry factor = sin(θ–α)/sin(θ+α), $k_0=1/\lambda_0$ and λ_0 is the wavelength. Eq (1) can be written [8] as a function of an oscillating term $sin^2(av)$, where $a=\pi k_0 t_0/\gamma_0$ and $v = Re[(qP^2+z^2)]$. This term is important for thin crystals and causes the reflectivity curve to oscillate. This phenomenon is known as "Pendellösung" and the interference pattern is constituted by these Pendellösung fringes. The diffracted power R(Δ) of a parallel and monochromatic incident beam, given by Eq. 1, is plotted as a function of the incident angle in fig.1. When such a parallel and monochromatic beam is reflected successively by two (different or identical) crystals, the resulting reflectivity curve R(Δ) is the product of the reflectivity curves of both crystals $R_1(\Delta)R_2(\Delta)$, providing both crystals are perfectly aligned. If one crystal is misaligned by an amount δ, the resulting curve is $R(\Delta)=R_1(\Delta)R_2(\Delta-\delta)$. Therefore, the effect of a double reflection is a reduction of the peak reflectivity. A significant reduction of its width is however produced only if $\delta\neq0$. The multiple reflection case can be calculated by applying iteratively the product of individual diffraction curves.

In order to determine experimentally the response function of a crystal or a set of crystals one measures the reflected intensity against the incident angle assuming a perfectly collimated monochromatic incident beam. However, a beam with these characteristics can never be obtained in a real experiment, one has to use another crystal or set of crystals to perform the analysis. A possible experimental setup for this purpose is a double crystal dispersion-free arrangement where the first crystal is kept fixed and the second crystal is rotated while the intensity reflected by the second crystal is recorded. If the Bragg angles of both crystals are the same this configuration is called parallel or non-dispersive, as shown in fig. 2. In this special case the angular width of the total diffraction pattern is independent of both the wavelength and the angular spread of the incident beam [9]. Therefore by rotating the second crystal, it is possible to record the resulting response function of the intrinsic diffraction curves of the first and second element(s), respectively. The measured diffraction pattern P(β) is commonly called a "rocking curve". This principle is used in the Bonse-Hart camera.

The rocking curve of two crystals with the same lattice spacing in non-dispersive configuration is calculated by:

[5]

$$P(\beta) = \frac{\int_{-\pi}^{\pi} R(\phi)\, R(\phi - \beta)\, d\phi}{\int_{-\pi}^{\pi} R(\phi)\, d\phi} \equiv R \otimes R$$

The function in the numerator is the cross correlation integral [10], so P(β) is the normalized cross correlation function of the diffraction pattern. Assuming that the reflectivity curves of the two crystals are identical, both having the typical "Darwin" shapes, the operation indicated by Eq.5 yields a rocking curve that is 1.32 times the full width at half maximum (fwhm) of a single Darwin curve. If both curves have the same Gaussian profile, the overall rocking curve would be 2 times the fwhm of a single Gaussian curve.

If instead of having two single crystals we have two channel-cut crystals with multiple reflections (m and n reflections, respectively) then Eq.5 can be written in a more general way as:

[6]

$$P(\beta) = \frac{\int_{-\pi}^{\pi} R^{m}(\phi)\, R^{n}(\phi - \beta)\, d\phi}{\int_{-\pi}^{\pi} R^{m}(\phi)\, d\phi} = R^{m} \otimes R^{n}$$

If the crystals are mounted in a dispersive way the total diffraction pattern is described by a more complicated expression [7]. In such cases and for any other combination between crystals and other optical elements, the ray tracing method [11] appears to be the most suitable tool in order to compute theoretical rocking curves and instrument efficiencies because it takes into account not only the crystal-specific angle-wavelength coupling, but also the dimensions and depth of the source and the effect of the slits and of crystal curvature in the case of focusing.

3. Pendellösung suppression

ICS and TDS superimpose on the wings of the reflection curve, or on a modulation of the wings like the Pendellösung fringes in thin crystals. This can be detrimental for small angle scattering experiments, and apparently Hastings and Siddons first mentioned the possibility of using thin crystals for Bonse-Hart optics [12]. In this paper we propose to use the effect of destructive interference of the Pendellösung fringes produced by slightly misaligned thin crystals for the Bonse-Hart technique in order to produce a very sharp decrease of the rocking curve tails. We investigated this effect for a set of crystals in both the Laue and the Bragg configuration. A combination of two crystals reduces the intensity of the wings if the second crystal is displaced by an angular amount equal to the semiperiod of about $\delta/2$ of the Pendellösung in order to place the two Pendellösung fringe patterns in anti-phase. This produces a destructive interference effect that tends to smear out the oscillatory behavior. The smoothing of the Pendellösung fringe pattern is not perfect because the semiperiod is a function of the rocking angle, thus no complete cancellation is possible through the whole angular range. An additional reflection can, however, further reduce the residual oscillatory signal.

For estimating quantitatively the Pendellösung contribution in a diffraction pattern one can define the following figure of merit:

$$FM(\delta) = \int_{-\pi}^{\pi} [\ln(R_{thin}(\Delta)) - \ln(R_{thick}(\Delta))]^2 d\Delta \tag{7}$$

The definition of Eq.7 can be intuitively interpreted as the total area subtended between the thin crystal diffraction curve $R_{thin}(\Delta)$ and the corresponding thick crystal curve $R_{thick}(\Delta)$. Squaring is needed to avoid cancellations between positive and negative parts. The logarithmic expression ensures that a higher weight is assigned to lateral oscillations than to the central peak. For a double crystal reflection, when the second crystal is misaligned with respect to the first by a variable quantity $\delta/2$, it is possible to calculate the variation of the FM with δ as:

$$FM(\delta) = \int_{-\pi}^{\pi} \left[\ln\left(R_{thin}(\Delta)\, R_{thin}\left(\Delta - \frac{\delta}{2} \right) \right) - \ln\left(R_{thick}(\Delta) - R_{thick}\left(\Delta - \frac{\delta}{2} \right) \right) \right]^2 d\Delta \qquad [8]$$

In order to determine exactly the angular shift which maximizes the Pendellösung suppression one must take the minima of the function FM(δ), as shown in fig.3 for three different crystal thicknesses. The effect of misaligning crystals with the optimum δ value is shown in fig.4 for different multiple reflections. Appreciable reduction of the Pendellösung is found and compared with the results of a perfectly aligned configuration. Quantitative results are given in Table I. The reduction of Pendellösung also leads to a significant suppression of the third and higher harmonics, as shown in Table II.

The same effect of reduction of Pendellösung fringes can be observed for a set of two or three crystals in the Laue (transmission) case. Fig.5 shows the diffracted power functions of a system of one, two and three identical, 50 μm thick Si(111) crystals, with an appropriate misalignment between them. The effect of oscillation reduction is less important in the Laue case, because the width ratio between the main peak and the side peaks is much bigger in the Bragg case than in the Laue one. As a consequence, the reduction is more effective the bigger the width ratio is. Considering different Laue crystals, the Pendellösung reduction can vary very much between different cases of thickness and polarization.

4. Conclusions

Thin crystals are proposed to be used in order to reduce TDS and ICS in Bonse-Hart-type small angle scattering experiments. In this work we have evaluated the reduction of the Pendellösung fringes in the response function of a set of perfect thin crystals.

From our theoretical study we conclude that in the case of a Bragg multiple reflection monochromator it is possible to find an optimum value of the misalignment of one of the crystals in order to reduce the intensity of the wings given by the Pendellösung fringes. This reduction is, however, always accompanied by some loss in the integrated intensity. The calculations presented here show how to obtain the optimum misalignment for different cases. The quantity FM gives an objective measure of the Pendellösung effect in the response function of the considered Bonse-Hart optics. Future experimental investigations must be performed in order to show whether the sacrifice of intensity is worth the gain in angular resolution.

5. Acknowledgments

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6. References

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thicknesses = 5, 10 and 20 μm (respectively)	FWHM [μrad]	Integral ($.10^{-6}$)	Peak value
$R(\Delta)$	40.0,39.4,37.9	44.3,42.6,42.4	0.93,0.94,0.94
$[R(\Delta)]^2$	37.0,34.9,34.3	31.1,30.2,29.6	0.86,0.89,0.89
$R(\Delta)R(\Delta-\delta/2)$	32.5,34.3,33.7	27.5,28.9,29.2	0.86,0.88,0.88
$[R(\Delta)R(\Delta-\delta/2)]^3$	26.4,27.7,27,7	15.3,17.3,17.8	0.63,0.69,0.69
$[R(\Delta)]^2R(\Delta-\delta/2)x[R(\Delta)]^2R(\Delta-\delta/2)$	34.3,36.7,36.06	23.3,25.8,25.4	0.67,0.70,0.70

Table I. Values of width, integrated intensity and peak for different combinations of Si 111 crystals and three different thicknesses. Energy is 8 keV. The δ value is the optimum one for each case.

thicknesses = 5, 10 and 20 μm (respectively)	FWHM [μrad]	Integral ($.10^{-6}$)	Peak value
$R(\theta)$	5.41,3.61,2.91	2.03,3.02,3.40	0.34,0.75,0.97
$[R(\theta)]^2$	4.01,3.01,2.71	0.48,1.63,2.47	0.12,0.56,0.94
$R(\theta)R(\theta-\delta/2)$	10.4,0.90,1.10	0.043,0.27,1.19	0.007,0.13,0.83

Table II, Values of the third harmonic contribution, and harmonic suppression with the misalignment used in the previous table. Energy is 24 keV and crystal is Si 333.

Figure Captions:

FIGURE 1. Reflectivity curves of a thick (left) and a 10 μm thin (right) silicon (111) oriented crystal for σ–polarization and 8 keV photon beam energy. The solid lines represent a single reflection, the dotted lines a triple reflection and the dashed lines the rocking curves of two sets of triple crystal reflections

FIGURE 2. Different multi-crystal configurations. Configuration A is a double non-dispersive Bragg reflection, yielding a response function $R_t(\Delta)=R(\Delta)R(\Delta-\partial/2)$. The configuration B includes a third reflection in a monolithic channel cut crystal, giving a response function of $R_t(\Delta)=[R(\Delta)]^3$. Configuration C shows a system consisting of two identical sets of crystals, where the second one is rotated with respect to the first one. The resulting response function is proportional to the convolution between the response functions of the individual sets of crystals (Eq. 5)

FIGURE 3. Figure of merit, FM(δ), for a silicon (111) crystal at 8 keV for three different crystal thicknesses: 20 μm (solid lines), 10 μm (dotted line) and 5 μm (dashed line). Minima are found at δ values of -15, -7,5 and -3.8 μrad respectively.

FIGURE 4. Response function for double Bragg reflections in different cases. The continuous lines from the top to the bottom refer to a single reflection R(Δ), a double reflection with the second crystal misaligned of the optimum quantity δ/2, i.e. $R(\Delta)R(\Delta-\delta/2)$, and a sextuple reflection $[R(\Delta)R(\Delta-\delta/2)]^3$, respectively. The dotted line is the double reflection $[R(\Delta)]^2$, and the dashed line is the convolution of two identical sets of three reflections: $[R(\Delta)]^2R(\Delta-\delta/2)$. Figures 4a, and 4b refer to crystal thicknesses of 5, and 20 μm respectively.

FIGURE 5. Response function of a Si111 crystal, 50 μm thick, in Laue geometry (curve at the top). The middle curve shows the effect of a set of two crystals with δ=6 μrad and the one at the bottom shows the effect of three consecutive transmission crystals with $\delta_1 = 6$ μrad and $\delta_2 = 3$ μrad

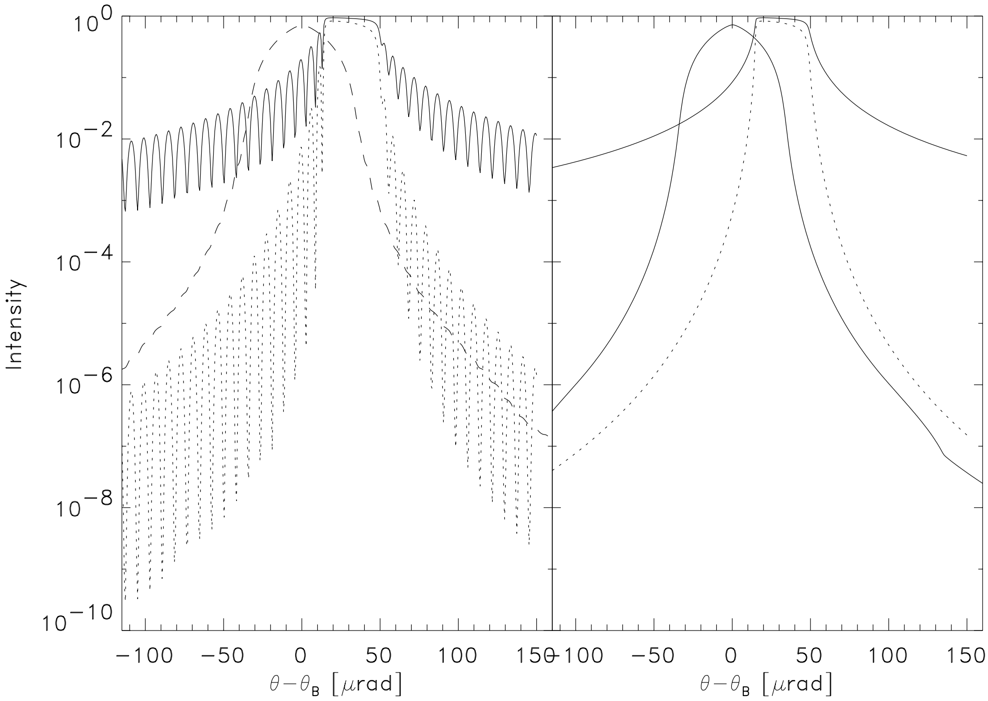

Fig1-ThD64 M. Sanchez del Rio

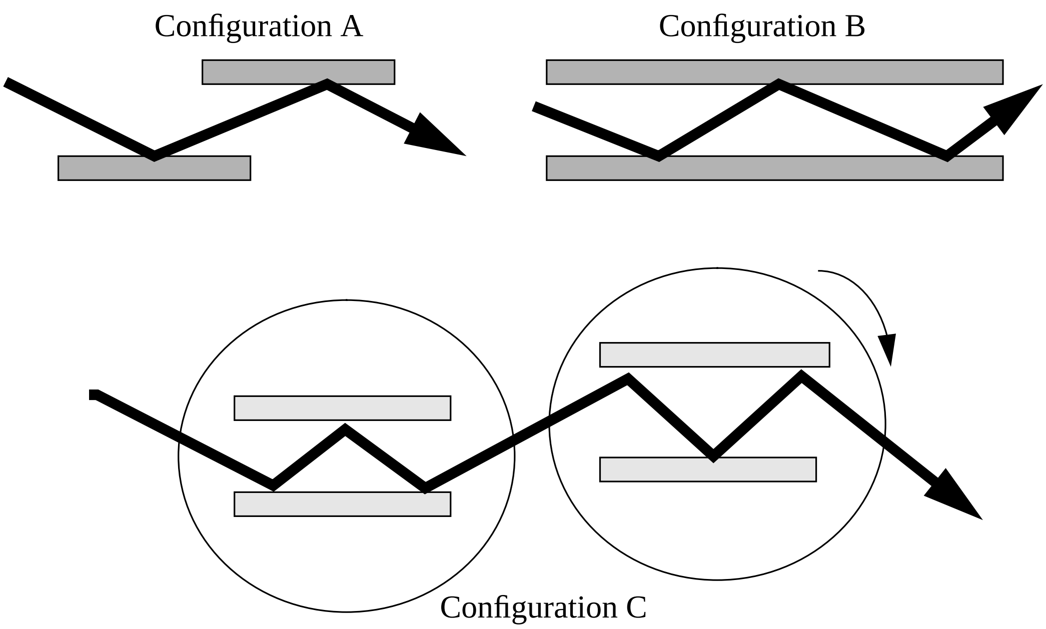

Fig2-ThD64 M. Sanchez del Rio

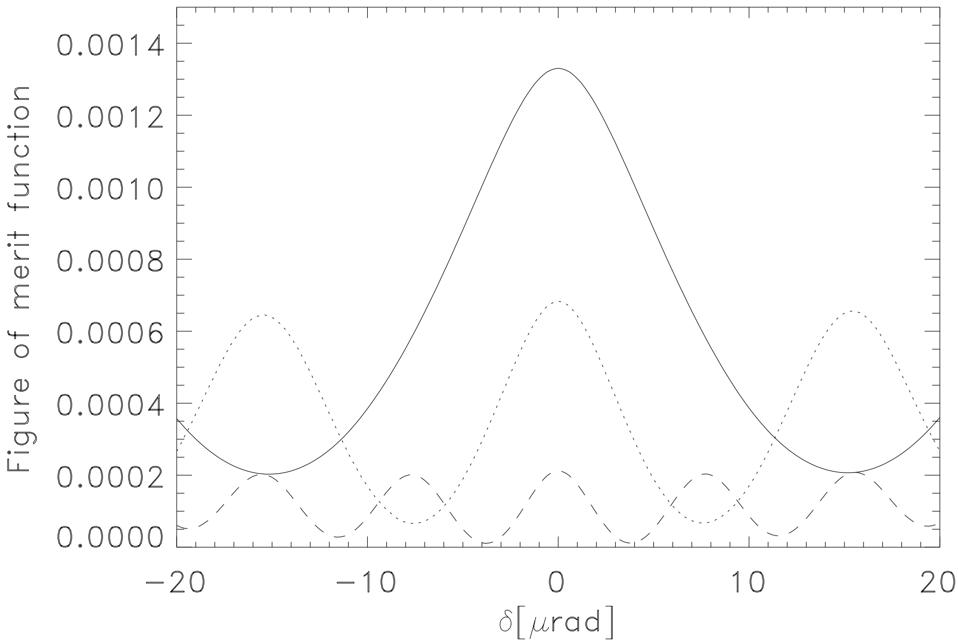

Fig3-ThD64 M. Sanchez del Rio

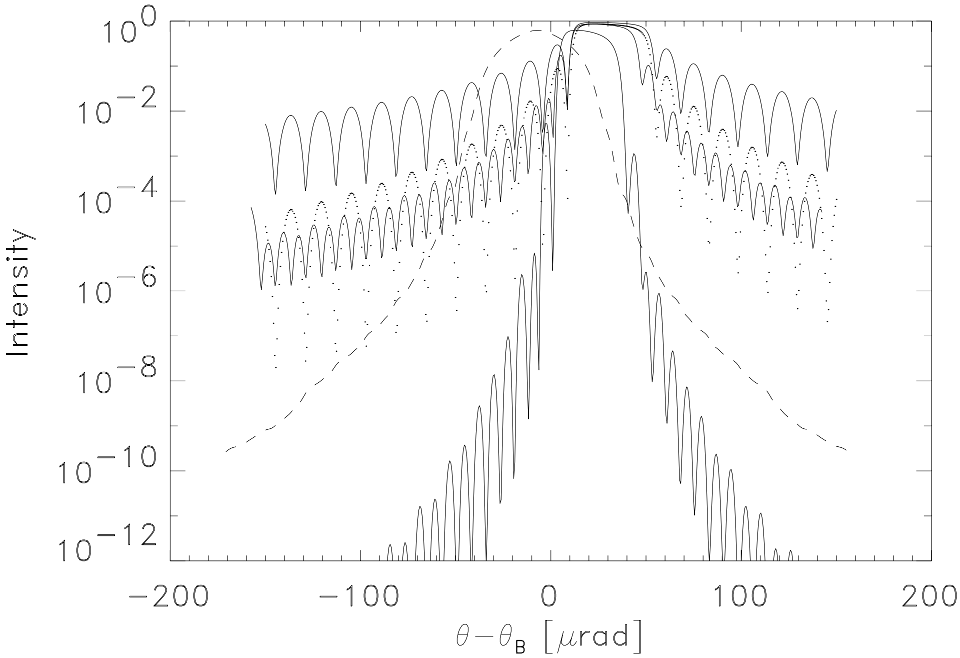

Fig4a-ThD64 M. Sanchez del Rio

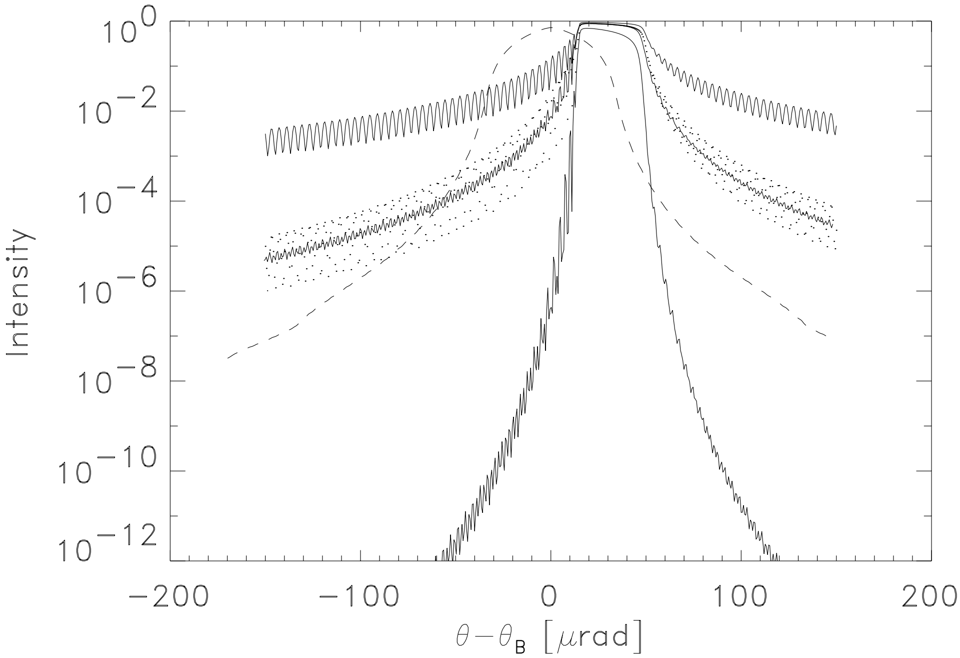

Fig4b-ThD64 M. Sanchez del Rio

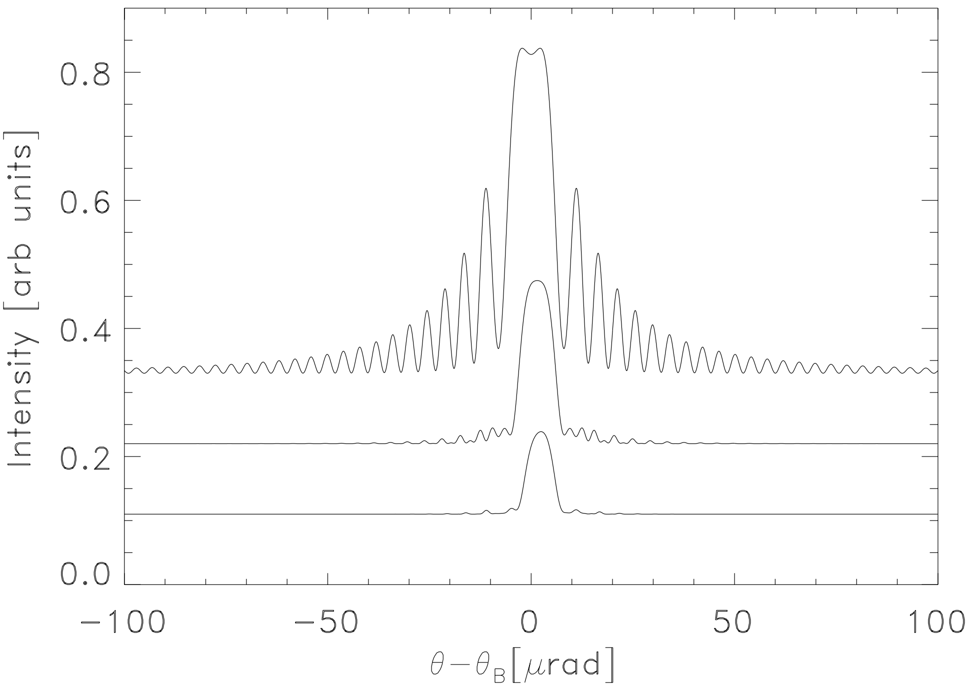

Fig5-ThD64 M. Sanchez del Rio