

Vertical neutron beam focusing with bent mosaic crystals

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Abstract. We report on the performance of bent mosaic crystals when used as a vertical focusing neutron monochromator. High-quality Cu(200) and Ge(335) mosaic crystals with a controlled curvature have been successfully produced at the ILL using plastic deformation at high temperature. As expected from simple geometrical considerations, they exhibit excellent properties for focusing a neutron beam vertically when examined on a high-resolution diffractometer installed on an $m = 1$ thermal neutron guide. Both Cu(200) and Ge(335) curved crystals allow a significant reduction of the focal image size at the sample position compared with a flat crystal with the same defect concentration. As a result, significant gain factors of 6 to 7 in intensity were obtained by replacing a flat crystal of 30 mm with a bent crystal.

1. Introduction

A vertical focusing monochromator provides an efficient mean of producing an intense beam of monochromatic neutrons from a white source [1-2]. To obtain a reasonable diffracted intensity, mosaic crystals are used in preference to perfect crystals, as they can match the incoming beam divergence in the scattering plane [3]. Moreover, the mosaic distribution in the direction perpendicular to the diffracting plane should be as small as possible in order to enable efficient beam compression vertically. Traditionally, a focusing monochromator consists of an assembly of flat rectangular monocrystalline plates mounted onto a mechanical support structure. The orientation of each crystal is adjusted in order to achieve the appropriate curvature, which in the case of symmetric diffraction geometry is related to the lens formula [4]:

$$\frac{1}{L_0} + \frac{1}{L_{MS}} = \frac{2 \sin \theta}{R_v} \quad (1)$$

L_0 is the distance from the source to the monochromator, L_{MS} the distance from the monochromator to the sample position, θ is the Bragg angle and R_v the radius of curvature (figure 1). The curvature is approximated by line segments. The angular step ψ between two segments, i.e. adjacent crystals, is $\psi = h/R_v$, where h is the crystal height. The Bragg angle stays constant over the height of the monochromator. Focusing in the vertical plane leads to a large gain factor in neutron flux, of between 2 and 10, at the sample position without affecting instrument resolution [5-7]. For a fully parallel beam, it produces a focal image of the source with dimensions limited by the height of the plates used to build the monochromator. The choice of the crystal monochromator is therefore of vital importance but not straightforward. A good compromise in practice is to choose the crystal height so that it matches the characteristic height of the sample under study. It will then produce a focus size comparable to the sample dimensions, typically in the range of 20 mm to 60 mm.

Better focusing can be achieved by using curved crystals, as they can perfectly match a given curvature. In the present paper we show that high-quality bent Cu and Ge mosaic crystals can be used as efficient monochromators if properly designed. Based on simple geometrical considerations, the performance of bent crystals in terms of beam compression and intensity gain was compared directly



to that of flat samples with the same characteristics. We also report on our neutron characterisation studies, during which we verified the uniformity of both the curvature and the mosaic distribution. The production of bent Cu and Ge crystals with controlled defect concentration is briefly described.

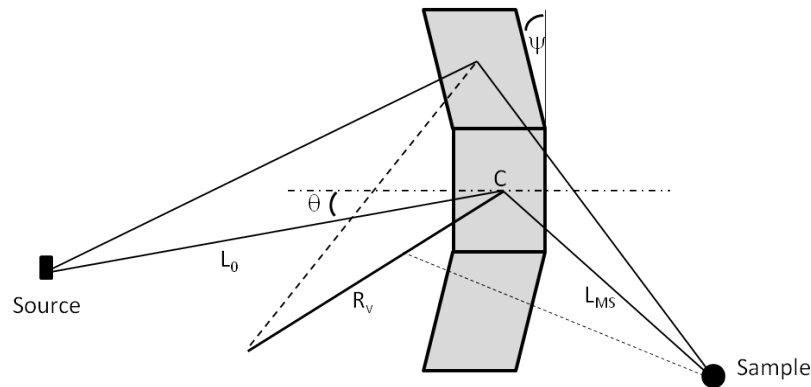


Figure 1. Vertical focusing monochromator composed of flat crystals.

2. Size of the focal image

For a flat crystal, the size of the focal image in the vertical direction is predominantly determined by the dimensions of the plate used to build the monochromator. Additional broadening due to imperfect geometry caused by crystal mosaic spread and incoming beam divergence may also affect focus size. The focus size H_f from a flat crystal of height h_c can be estimated from simple geometrical considerations (figure 2): $H_f = h_c + 2 \gamma L_{MS} \sin \theta$, where 2γ is the total angular deviation of the reflected beam. Assuming Gaussian distributions, we obtain $\gamma = \sqrt{\alpha^2 + \beta^2}$, where α is the beam divergence and β the crystal mosaic along the bending direction. The Full Width Half Maximum of the intensity distribution in the vertical direction is then:

$$H_f(FWHM) = h_c + 2 \sqrt{\alpha^2 + \beta^2} L_{MS} \sin \theta \quad (2)$$

The α value depends on the neutron source (guide, beam tube or virtual point source) and the L_{MS} parameter is defined by the instrument configuration. Since the mosaic β is chosen to be as narrow as possible, it is easy to see that a significant reduction in the focal spot can only be achieved by reducing the value of h_c . However, it may be not practical to use crystals with small dimensions (less than 10 mm) due to the major difficulty involved in orienting the crystallographic planes. Indeed, in a monochromator made of a large number of pieces, the individual crystals need to be accurately aligned with respect to one another to within a fraction of a degree, i.e. 0.05° . For crystals with dimensions of 5 mm, a positioning accuracy better than 5 μm is required. This represents not only a real challenge but is also time-consuming.

In contrast to a flat crystal, a cylindrically bent mosaic crystal matches a given curvature perfectly. The Bragg planes will satisfy the vertical focusing conditions. As a direct consequence, the focus size in the vertical direction H_b is independent of the crystal height and is only affected by imperfect geometry (figure 2). We obtain:

$$H_b(FWHM) = 2 \sqrt{\alpha^2 + \beta^2} L_{MS} \sin \theta \quad (3)$$

The gain factor G in beam compression for a curved crystal compared with a flat crystal can be expressed as:

$$G = \frac{H_f}{H_b} = 1 + \frac{h_c}{2 \sqrt{\alpha^2 + \beta^2} L_{MS} \sin \theta} \quad (4)$$

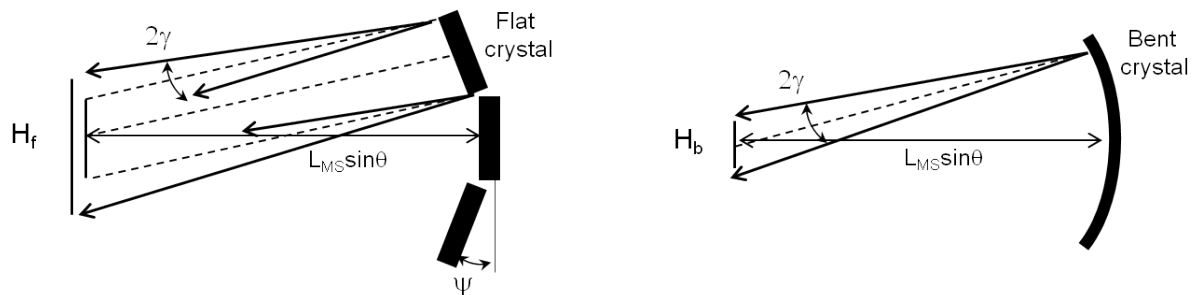


Figure 2. Geometry of focusing in the vertical plane illustrating the broadening of the focal image due to imperfect geometry. γ is the angular deviation from the perfect geometry, taking into account the beam divergence and the crystal mosaic distribution. It should be noted that length scales are not respected (H_b and $H_f \ll L_{MS} \sin \theta$).

As an example, some typical values are reported in table 1. The gain in beam compression has been estimated for a Bragg angle of 30° , a mosaic β of 0.1° and for crystal heights of 10 mm and 30 mm. Calculations were performed for incoming beam divergences of 0.24° and 1° . These values were chosen to be representative of the neutron beams transported by neutron guides at the ILL: $\alpha = 0.24^\circ$ and $\alpha = 1^\circ$ are the divergences for an $m = 2$ thermal guide at a wavelength of $1.2 \text{ \AA}$ and $5 \text{ \AA}$, respectively. The contribution of the crystal height was evaluated for monochromator-sample distances L_{MS} of 2 m and 3 m. The calculations show that in the case of low beam divergences the focal image size can be reduced vertically by a factor of 2 to 4 by using bent crystals. Obviously, the gain factor decreases when the beam divergence or focal length is increased. Nevertheless, the use of bent crystals remains worthwhile, especially when large single pieces are used.

	beam divergence $\alpha = 0.24^\circ$				beam divergence $\alpha = 1^\circ$			
$h_c(\text{mm})$	30	10	30	10	30	10	30	10
$L_{MS}(\text{mm})$	2000	2000	3000	3000	2000	2000	3000	3000
$H_f(\text{mm})$	39,1	19,1	43,6	23,6	65,1	45,1	82,6	62,6
$H_b(\text{mm})$	9,1	9,1	13,6	13,6	35,1	35,1	52,6	52,6
Gain factor G	4,3	2,1	3,2	1,7	1,9	1,3	1,6	1,2

Table 1. Theoretical sizes of the focal image produced by a flat crystal (H_f) and a bent crystal (H_b) as a function of crystal height and focal length. α is the incoming neutron beam divergence. The crystal mosaic β was fixed at 0.1° and the Bragg angle set to 30° . The gain factor G is defined as the ratio H_f/H_b .

3. Experimental approach

3.1 Production of bent Cu(200) and Ge(335) mosaic crystals

Plastic deformation is a well-known technique for producing copper and germanium crystals with controlled mosaic distribution [8]. By applying a suitable pressure perpendicular to the surface of a

high-quality single-crystal plate at high temperature, the mosaic spread can be increased up to the desired value, in the range of 0.1° to 0.5° [8-9]. This technique was developed recently for preparing mosaic crystals with a controlled curvature. Bending was achieved without affecting the quality of the crystals by using smooth plastic deformations applied with a cylindrically shaped anvil with the desired radius of curvature. In addition, through careful positioning in the hot-pressed furnace, it was possible to obtain a mosaic distribution which was reasonably homogeneous throughout the length of the crystal. Starting from high-quality single crystals, several Cu and Ge samples were successfully prepared.

3.2 Neutron experiments

Neutron diffraction properties were studied on the double-crystal test instrument T13A at the ILL. A perfect Ge(hkl) monochromator crystal selected the desired wavelength from an $m = 1$ thermal guide tube with a cross-section of 0.3 (width) $\times$ 0.5 (height) cm^2 . An adjustable vertical slit was positioned in front of the sample allowing the incoming beam height to be varied from 0.1 to 5 cm. The beam width was 25 mm for all experiments. Two detectors were used for this work: a single high-efficiency ^3He detector to record rocking curves and a 64×64 mm^2 two-dimensional area detector (32×32 pixels) to study focusing properties. The 2D detector was mounted on a y translation stage in order to be able to adjust the distance between the sample and the detector.

The effective neutron mosaic spread was determined as the full width at half height of the rocking curves recorded in reflection geometry. The double-crystal configuration was parallel and the arrangement almost dispersion-free (d-spacing monochromator = d-spacing sample). Thus the width of the rocking curve directly represented the mosaic distribution of the sample under study. The mosaic distribution was measured along a direction parallel ($FWHM_H$) and perpendicular ($FWHM_V$) to the bending direction. The curvature of the single-crystal plates was calculated from the peak shift $\Delta\theta$ of the angular positions along the sample. For these measurements the width of the incoming beam was limited to 0.1 cm. To investigate focusing properties, diffraction images were recorded using the 2D detector. The focal point was determined by varying the sample-detector distance L_{MS} and was defined as the smallest image size.

4. Results and discussion

4.1 Crystal quality

The present study focused on two bent Cu(200) and Ge(335) crystals of dimensions $60 \times 20 \times 5$ mm^3 . Single-crystal plates were plastically bent using a cylindrically shaped anvil with a radius of curvature of 1 m. Figure 3 shows the rocking curves from Cu(200) at a neutron wavelength of 1.8 Å provided by a Ge(220) monochromator. The mosaic distribution $FWHM_V$ in the direction perpendicular to the bending is 0.2° . The diffraction pattern indicates a symmetric single peak with a nearly Gaussian shape, demonstrating the homogeneous mosaic structure. Along the bending direction, the global width of the rocking curve is 2.8° . This result is consistent to a peak shift $\Delta\theta = l/Rv$ with $Rv \sim 1\text{m}$, where l is the length of the plate illuminated by the incoming beam ($l = 50$ mm). A narrow beam of 1 mm width was used to determine more accurately the value of the local mosaic spread $FWHM_H$. The rocking curve is wider than the local mosaic spread $FWHM_H$ because the reflection curve is convoluted with the curvature. After deconvolution, provided that distributions are Gaussian, we found that the local mosaic spread $FWHM_H$ is very close to 0.2° . The variation of the peak position as a function of the position y along the length of the crystal indicates a linear shift of $0.06^\circ/\text{cm}$, which is compatible with a uniform curvature. It corresponds to a radius of curvature $Rv = 1.047$ m. Smooth variations in peak intensity are due to inhomogeneities in the mosaic structure.

Similar measurements on a bent Ge(335) crystal at a neutron wavelength $\lambda = 1.1$ Å are shown in figure 3. The mosaic spread $FWHM_V$ is 0.25° . The mechanical properties of germanium make it difficult to introduce a spatially homogeneous microstructure in large single crystals using plastic

deformation. Nevertheless, crystal quality is still acceptable for neutron applications. The peak shift along the bending direction is again linear and varies by 2° over 40 mm, which corresponds to a 1140 mm radius of curvature. The local mosaic spread $FWHM_H$ is 0.2° .

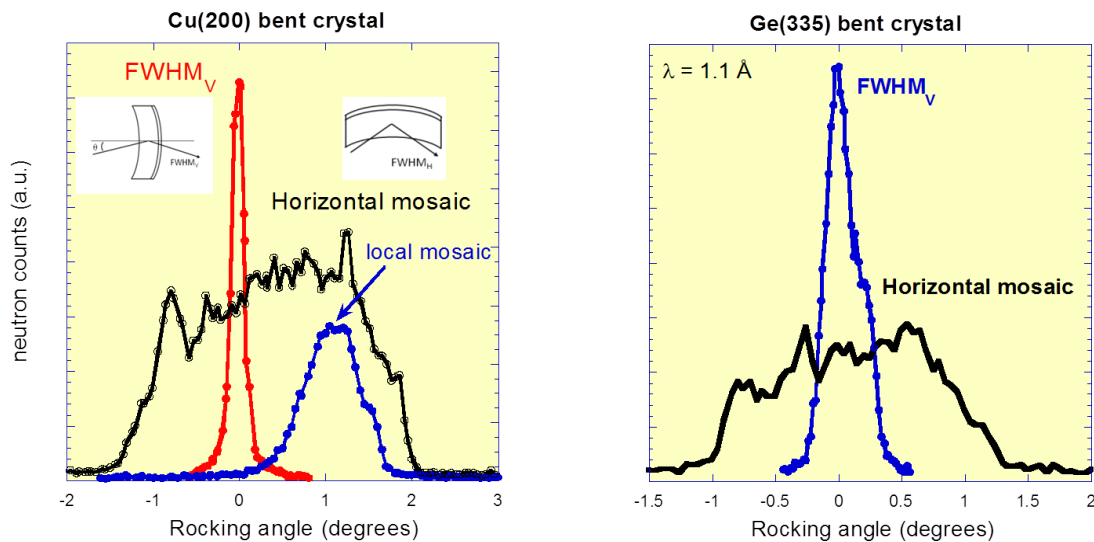


Figure 3. Experimental neutron rocking curves obtained with Cu(200) and Ge(335) mosaic crystals in reflection geometry (for more details, see text).

4.2 Focusing properties of bent crystals

The performance of bent Cu(200) and Ge(335) crystals in terms of peak intensity and beam compression was compared with that of flat crystals with the same mosaic distribution. The vertical size of the incoming beam was 30 mm. Diffraction images from Cu(200) crystals are presented in figure 4. Clearly visible is the focusing effect obtained with the bent crystal when increasing the detector-sample distance L_{MS} from 500 mm (under-focused beam) to 1010 mm (focal point). Beyond the focal point, we observed an increase in the image size resulting in over-focused geometry (figure 4c). The vertical cross-section of the diffraction pattern indicates that the spatial distribution of the intensity is perfectly Gaussian with a Full Width Half Maximum of 5.2 mm (figure 4e). Compared with a flat crystal, the reduction in the focus size is significant. It corresponds to a gain in intensity of a factor 6 resulting in a geometrical beam compression factor of 6, whereas the integrated reflectivity remains constant.

Very similar results were obtained with Ge(335) crystals (see figure 5). The focal point was found to be at a sample-detector distance of 850 mm (figures 5a and 5b). The vertical peak profile is again perfectly Gaussian with a $FWHM$ of 4 mm (figure 5c). The incoming beam size has been reduced by a factor 7 at the focal point, leading to an increase in peak intensity also by a factor 7.

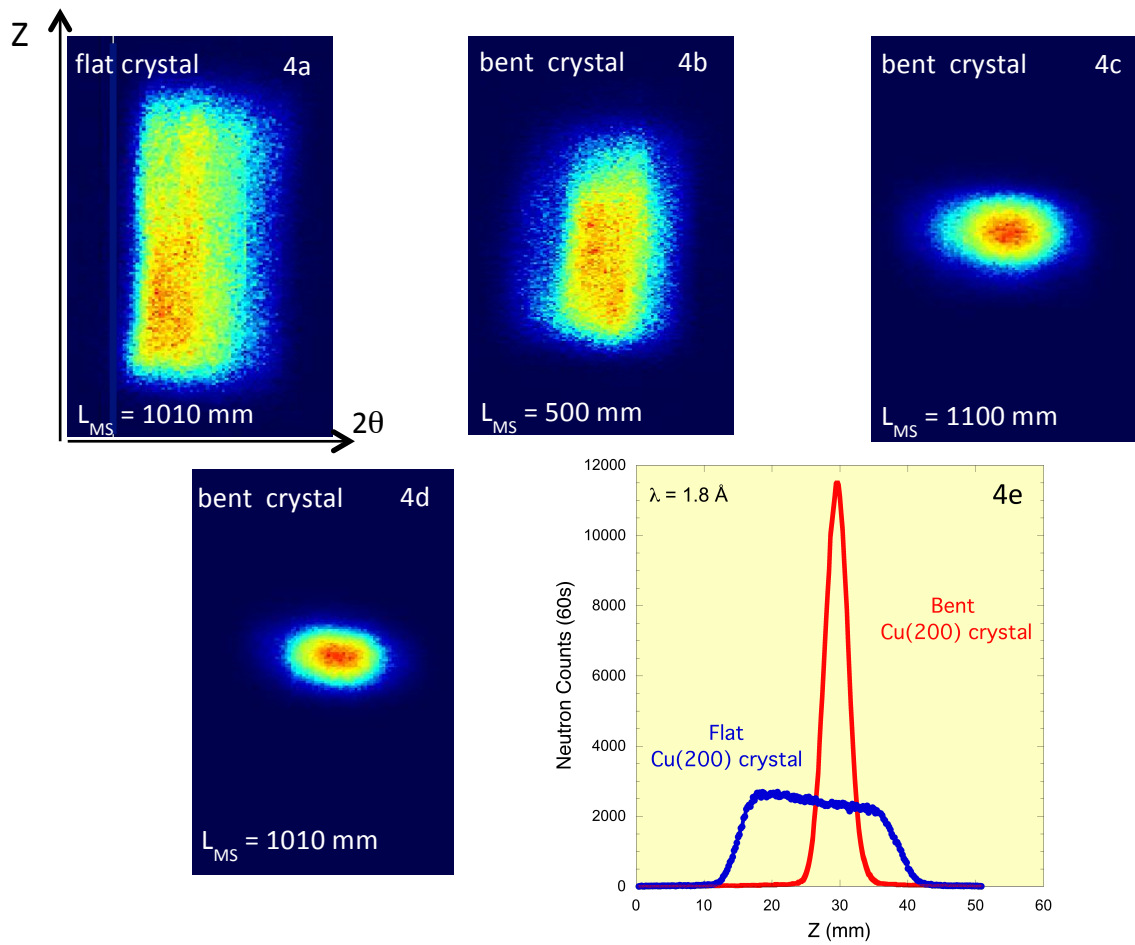


Figure 4. Diffraction images recorded at $\lambda = 1.8$ Å with a flat Cu(200) crystal (Fig. 4a) and with a bent Cu(200) crystal for different detector-sample distances L_{MS} : under-focused (Fig. 4b), over-focused (Fig. 4c) and focused (Fig. 4d) geometries. Spatial distribution of the intensity at the focal point for a flat and a bent Cu(200) crystal (Fig. 4e).

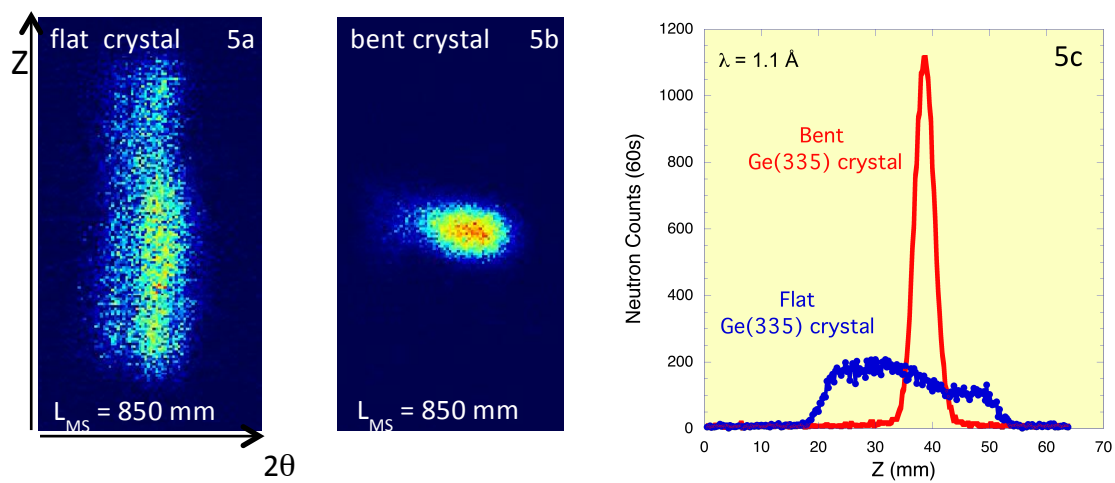


Figure 5. Diffraction images recorded at the focal point ($L_{MS} = 850$ mm) with a flat Ge(335) crystal (Fig. 5a) and with a curved Ge(335) crystal (Fig. 5b) and the corresponding spatial distributions of the intensity along the vertical direction at $\lambda = 1.1$ Å (Fig. 5c).

4.3 Discussion

The main results for focusing properties obtained from bent and flat crystals using neutron diffractometry on T13A are summarised in table 2. Theoretical predictions are also reported for comparison. Equations 1, 2, 3 and 4 were used for the calculations. The incoming beam divergence was set to $\alpha = 0.18^\circ$ at $\lambda = 1.8 \text{ \AA}$ and $\alpha = 0.11^\circ$ at $\lambda = 1.1 \text{ \AA}$ for Cu and Ge samples, respectively. This corresponds to the divergence values provided by an $m = 1$ neutron guide. It must be pointed out that the vertical divergence is unaffected by the reflection from the perfect Ge crystal used as a monochromator on the test instrument. The value of $\beta = FWHM_H$ was determined from the width of the neutron rocking curves, as described in section 3. It was fixed at 0.2° for all samples. Focal lengths were estimated for a 1m radius of curvature when considering a source at infinity, i.e. $L_0 \rightarrow \infty$. As can be seen in table 2, the experimental data are in good agreement with the theoretical predictions. Absolute values of both image size and focal length are very close to the observed ones for all samples. The gain in intensity is proportional to the reduction in beam size.

It must be emphasised that the focus size in the vertical direction produced by the curved Ge(335) and Cu(200) crystals is not affected by the height of the incoming beam. This result is consistent with basic optics, where the crystal acts as a lens. In accordance with the magnification equation, the crystal produces an image of the vertical source of height $h_i = h_s (L_{MS}/L_0)$. For a neutron beam transported by a neutron guide, the source height h_s is not well defined in practice but depends on the incoming beam divergence and crystal properties. In other words, when considering a curved crystal, each point of the crystal only reflects neutrons from the source within a vertical angular distribution γ (see section 2). The effective source height is then $h_s = 2\gamma L_0 \sin\theta$ and finally we obtain equation (3), as presented in section 2, i.e. $h_i = h_s (L_{MS}/L_0) = 2\gamma L_0 \sin\theta$.

	L_{MS} experimental	L_{MS} calculated	Image size ($FWHM$)	Image size (calculated)	G factor (experimental)	G factor (calculated)
Cu(200) flat	∞	∞	34	34.7		
Cu(200) R1m	1010	1003	5	4.7	6.8	7.4
Ge(335) flat	∞	∞	33	34.0		
Ge(335) R1m	850	783	4	4.0	8.3	8.5

Table 2. Experimental and theoretical results for focusing properties of bent Cu(200) and Ge(335) crystals compared with flat crystals with the same characteristics. Both focal length and image size are expressed in mm.

Conclusion

Theoretical predictions based on a simple geometrical approach suggest that a bent mosaic crystal should exhibit better performances in terms of focusing efficiency compared with the flat crystal commonly used to build neutron monochromators. To validate this assumption, Cu(200) and Ge(335) mosaic crystals with a 1 m radius of curvature were successfully produced at the ILL using plastic deformation at high temperature. The uniformity of the mosaic distribution and curvature has been determined by high-resolution neutron diffraction, demonstrating the high structural quality of the samples. The vertical size of the focal spot and the related spatial intensity distribution obtained from bent crystals were studied using a 2D detector. Spot sizes in the range 4-5 mm were then observed for all values of the incoming neutron beam dimension. As a direct consequence, compared with a flat crystal of 30 mm height, a gain factor of 6 and 7 in intensity was obtained with Cu(200) and Ge(335) curved samples, respectively.

Following this promising result, we decided to build a full-scale prototype monochromator for the single crystal diffractometer D10 at the ILL [10]. It is composed of 12 Cu(200) crystals with a 2.5m radius of curvature characterised by a mosaic distribution of 0.2° . It has been designed to replace the existing Cu(200) monochromator with the aim of improving the performance of the instrument at a neutron wavelength of $1.26 \text{ \AA}$.

Looking further ahead, it is important to understand that future neutron scattering instruments will require neutron optical devices that allow experiments to be performed under extreme conditions. Very high pressure, magnetic field or temperature will require the use of small sample volumes in the mm^3 range. As a direct consequence, neutron optics devices such as focusing monochromators will have to be capable of efficiently focusing large neutron beams into very small volumes at the sample position. A monochromator built with bent mosaic crystals will therefore lead to significant improvements in instrument performance in terms of neutron flux and reduction of background.

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