

Focusing and reflectivity properties of a parallel double bent crystal (+n,-m) setting

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Abstract. After preliminary results obtained and published recently [1], in our contribution focusing and reflectivity properties of the dispersive double bent-crystal arrangement are presented in much more detail. It has been found that two different bent perfect crystals in (+n,-m) setting can be good candidates for high efficiency neutron microfocusing as well as high-resolution monochromatisation. Due to the (+n,-m) setting of two different bent perfect crystals, a high resolution is expected in both $\Delta(2\theta)$ (2θ is the scattering angle) as well as $\Delta\lambda/\lambda$ (λ is the neutron wavelength). Experimental tests were carried out with the setting employing the bent Si(111) slab and Si(220)-sandwich, which contained either one, or two or four 1.3 mm thin simply stacked slabs. Thanks to a high reflection probability of both bent elements and an easy manipulation with the curvature of the Si(220)-sandwich, an excellently focused intensive monochromatic beam of the width from one to several millimetres was obtained. The properties of the double bent-crystal setting were studied in Řež at the neutron optics diffractometer for the neutron wavelength of 0.162 nm and for various thicknesses and curvatures of the Si(220)-sandwich. It has been also found that besides an excellent focusing and reflectivity properties of the dispersive double bent-crystal setting the obtained monochromatic neutron current is sufficiently high for standard high-resolution diffraction experiments even at the medium power research reactor.

1. Introduction

Nondispersive as well as dispersive double-crystal monochromators are quite common at synchrotron sources. However, in the case of neutrons, at present one can find only several scattering instruments in the world which use nondispersive double-crystal setting for neutron monochromatisation (e.g. HB-1A Triple-Axis Spectrometer at High Flux Isotope Reactor in ORNL, Double Crystal Diffractometer at NIST for USANS, 4F1 and 4F2 spectrometers in LLB Saclay). Such monochromator with two parallel crystals produces monochromatic beam running parallel to incident white beam. For the change of the neutron wavelength such monochromator does not need a movable (rotating) table and the position of the sample is fixed. The problem occurs when using mosaic crystals. Due to the double reflection process monochromatic beam current is much smaller than in the case of one crystal monochromator. For one mosaic monochromator the neutron current is proportional to its peak reflectivity r ($r < 1$). Then for double crystal monochromator it is proportional roughly to r^2 and the situation becomes dramatically inconvenient when $r \approx 0.5$ or less



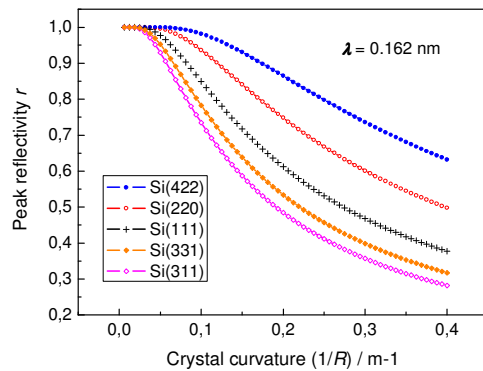


Figure 1. Peak reflectivity vs crystal curvature related to different Si reflections as calculated on the basis of eq. (1) for symmetric reflection geometry ($\psi=0^\circ$).

crystals can be close to 1 and there is a small acceptable loss of neutrons due to the double reflection process. The general formula for the reflection probability (often called as peak reflectivity) of neutron passing through a homogeneously deformed perfect crystal has been derived in the form as [2,3]

$$r(R_M) = \{1 - \exp[-Q_{hkl}(\partial\Delta\theta/\partial s_0)^{-1}]\}, \quad (1)$$

where $(\partial\Delta\theta/\partial s_0)$ is the rate of change of the Bragg angle on the flight path Δs_0 in the crystal in the incident beam direction and Q_{hkl} is the kinematical reflectivity of the crystal unit volume. For cylindrically curved monochromator it has been derived in the general form [3,4] when including also asymmetric diffraction geometry given by the angle ψ of the asymmetry cut of the crystal

$$\partial\Delta\theta/\partial s_0 = (1/R_M)(\cos \psi / \cos \theta_M)[1 - (1 + \sigma) \sin(\theta_M + \psi) \sin(\theta_M - \psi)], \quad (2)$$

where R_M is the bending radius, θ_M the Bragg angle and σ the Poisson constant. An example of several calculated peak reflectivities for Si crystal which perfectly fit with experiment [2,3] is shown in figure 1. Furthermore, curved crystal instruments are known by the fact that they do not use Soller collimators and the resolution can be controlled through the bending of the crystals. Possible employment of the focusing does not necessarily imply focusing in real space, but it can also cover focusing in scattering which is very often used in powder diffractometry, namely, for strain/stress measurements in polycrystalline materials [5-9]. Double-crystal arrangements with the bent perfect crystals provide variety of performances which could be exploited in neutron diffractometry. With two bent crystals in successive reflection, the double crystal rocking curve is generally much broader in comparison with the flat perfect crystals. However, when the phase space windows of both bent crystals match, the double crystal rocking curve is narrow because it is related only to the convolution of the reflectivity curves and thus the intensity of the doubly reflected beam is high. This matching of the phase space windows can be even achieved with the bent crystals having different d -spacings by a proper adjustment of individual curvatures of the crystals [7,10]. In ref. 7 the authors point out the possibility of a rather sharp focusing (up to 1 mm spot) by means of successive reflections on three Si(111) crystals (mosaic+bent+bent) in the $(+n, -n, -n)$ setting, however, with a rather low luminosity. Another proposal of using the antiparallel $(+n, +n)$ setting of two bent Cu(200) crystals (copper plates of small mosaic spread of about $3'$) achieving the spot width of 2.5 mm for residual strain/stress instrument was reported in ref. 11. Very promising results of an extensive investigation of $(n, -m)$ combination of a vertically focusing mosaic Ge(331) crystal in combination with a horizontally bent perfect Si(111) crystal were published in papers [12,13]. Here, it was clearly demonstrated both real

space focusing and focusing in scattering on a standard polycrystalline γ -Fe sample (pin of the diameter of 2 mm) at $\lambda=0.183$ nm. Furthermore, it was found an excellent property that for an optimum curvature of the bent Si(111), *FWHMs* of the γ -Fe diffraction profiles in the 2θ -angular scale for both parallel and antiparallel settings were practically the same. This fact can have enormous impact in powder diffractometry and namely for residual strain/stress measurements when permitting to measure two strain components simultaneously. Of course, that in double-crystal combination of mosaic and bent perfect crystal due to a large mosaicity of the mosaic crystal, the matching of the phase space elements is not as good as it can be in the case of two bent crystals. In this paper we deal with the results of the investigation of the properties of the (+n,-m) setting of the bent Si(111)+Si(220)-sandwich for neutron monochromatisation and its possibility to be employed in neutron diffractometry. Some results of the preliminary investigations with two thin (2x1.3 mm thickness) bent Si(220) slabs were published in ref. 1. On the basis of the obtained promising results we continued in extensive investigations of this setting by using a sandwich of four, two and one slab of Si(220), symmetric or asymmetric diffraction geometry and a large range of curvatures. In our case the (+n,-m) setting can be considered as slightly dispersive one.

2. Experimental arrangement

For this experiment we used neutron optics diffractometer installed at the 10 MW research reactor LWR-15. The diffractometer operates at the fixed neutron wave length of $\lambda=0.162$ nm which is provided by a cylindrically bent perfect Si(111) crystal of the dimensions of 200x40x4 mm³ (length x width x thickness). Schematic sketch of the diffractometer performance is drawn in figure 2. Depending on the experimental requirements, the diffractometer can operate in two or three axis

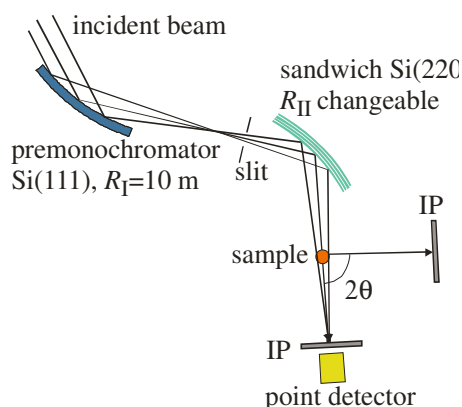


Figure 2. Sketch of the diffraction performance as used for the experimental investigations.

mode. First it should be pointed out that the radius of the curvature of the first Si(111) crystal of the DC setting was fixed ($R_I \approx 10$ m) which was not changed during the experiment. The second bent perfect Si(220) monochromator was in fact sandwich of four slabs, two slabs or one slab each of the dimension of 200x40x1.3 mm³. By using a sandwich instead of a single piece of the same thickness a large range of curvatures without a danger of breaking could be used. Even in the case of diffraction studies with one Si(220) slab, a sandwich of four slabs was used when three additional slabs in the bending device had a different orientation and did not participate in the diffraction process. For neutron detection either a point detector or neutron Imaging Plate (IP) were used. The sample could be put on the third axis of the instrument which is at the distance of 50 cm from the Si(220) crystal.

The distance between the Si(111) and Si(220) crystals was

fixed and equal to 180 cm. No Soller collimator was installed before and after Si(111) crystal. The focal length of the bent Si(111) crystal was about 130 cm and the convergent rays of the extracted monochromatic beam crossed about 50 cm in front of the Si(220) sandwich. The beam divergence of the incident beam on the Si(111) crystal was 40' and had no significant impact on the double crystal performance.

3. Experimental results

3.1. Symmetric diffraction geometry of the Si(220) sandwich

First of all the double crystal rocking curves of the bent Si(220) with respect to the bent Si(111) were investigated. Figure 3 shows a few chosen rocking curves related to different sandwiches, two widths of the incident beam from the bent Si(111) crystal (two widths of the slits) but all for the same radius

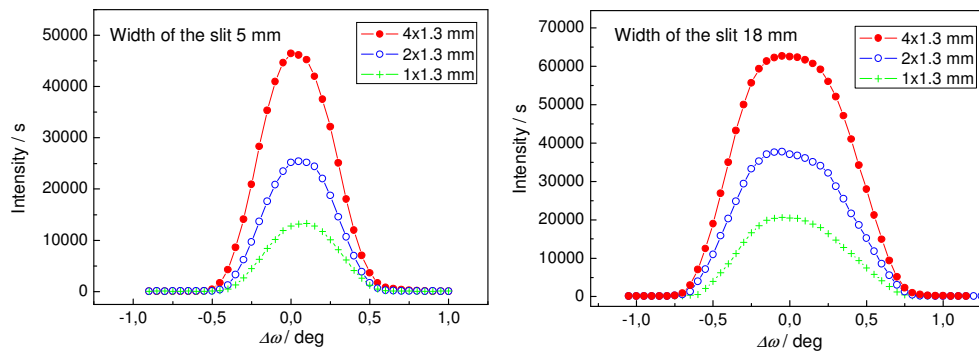


Figure 3. Double-crystal rocking curves obtained by rocking the Si(220) sandwich with respect to the Si(111) premonochromator for the curvature of the sandwich of 0.28 m^{-1} ($R=3.6 \text{ m}$).

curvature of the bent Si(220) sandwich. It can be seen from figure 3 that the *FWHMs* of the rocking curves related to the 18 mm slit are much larger than that related to the 5 mm slit. However, peak intensities differ substantially in favour of using the 18 mm slit. Furthermore, it can be seen from the rocking curves that the double crystal setting provides very good intensities of monochromatic neutrons (see peak intensities of the rocking curves). As to the reflectivity of the double crystal setting, it can be seen from figure 1 that the peak reflectivity of the bent Si(111) is close to 1 and that of the sandwich is estimated to be about 0.6. It means that the peak reflectivity of the system is still high ($r_{111} \times r_{220} \approx 0.6$). For the estimation of the monochromatic neutron current and focusing, the beam profile

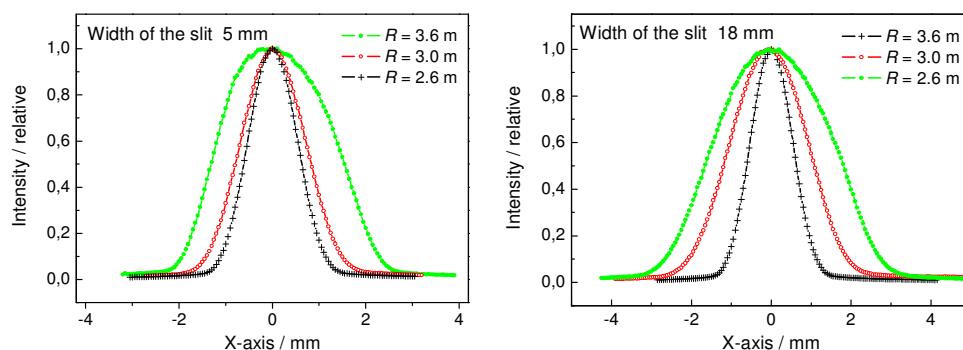


Figure 4 Beam profiles measured by means of IP situated at the distance of 70 cm from the Si(220) crystal for three radii of curvature and for two widths of the incident premonochromatized beam.

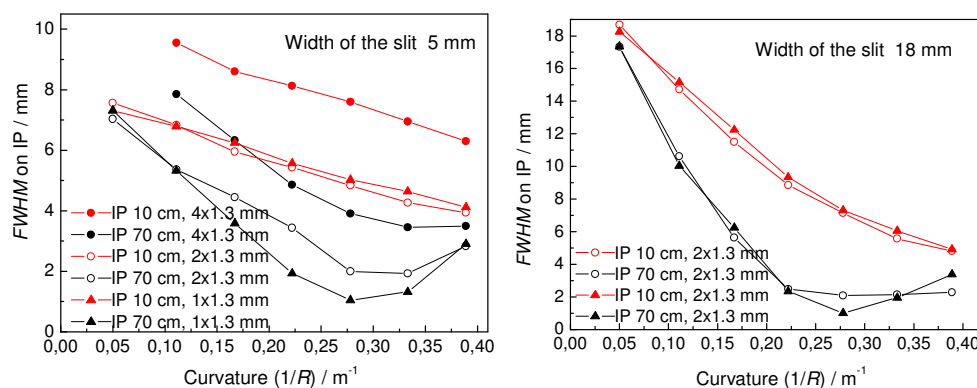


Figure 5. Width of the beam diffracted by the bent sandwich as a function of curvature for two different slits limiting the incident beam on the Si(220) sandwich.

properties were investigated at some distance from the Si(220) sandwich. For the focusing and beam profile studies an Imaging Plate situated either just behind the crystal (distance about 10 cm) or in front of the point detector at the distance of 70 cm. The images of the beam were taken for different radii of curvature of the Si(220) sandwich and again for two widths of the beam impinging on the Si(220) sandwich (5 mm or 18 mm). Figure 4 displays several beam profiles as taken by IP. Thanks to the homogeneous deformation of the crystals, each profile is smooth and has a nearly Gaussian form. Then figure 5 shows the dependence of *FWHM* of the beam profile on the curvature of Si(220) sandwich. It can be seen from figure 5 that a strong focusing effect in the range of curvatures of $0.25 \text{ m}^{-1} - 0.38 \text{ m}^{-1}$ was achieved. The minimum *FWHM* of the diffracted beam of 1 mm and the best focusing effect were found in both cases of the slits with one Si(220) slab in the sandwich for the curvature of 0.28 m^{-1} ($R=3.6 \text{ m}$).

Then, the neutron current related to the focused beam obtained by the (+n,-m) double-crystal performance was compared with the neutron current incident on the bent Si(220) crystal for $R=3.6 \text{ m}$. For this purpose a 2 mm slit was installed in front of the Si(220) crystal (see figure 2) and the passing beam was imaged by the Imaging Plate at the distance of 10 cm. The peak intensity of the beam profile was not dependent on the width of the slit. Vertically integrated signal registered by the IP was then compared with the vertically integrated signal registered by the IP situated at the distance of 70 cm behind the Si(220) crystal, just before the point detector (see figure 6). It can be seen from figure 6 that thanks to the strong focusing effect the peak intensities of the obtained profiles are comparable.

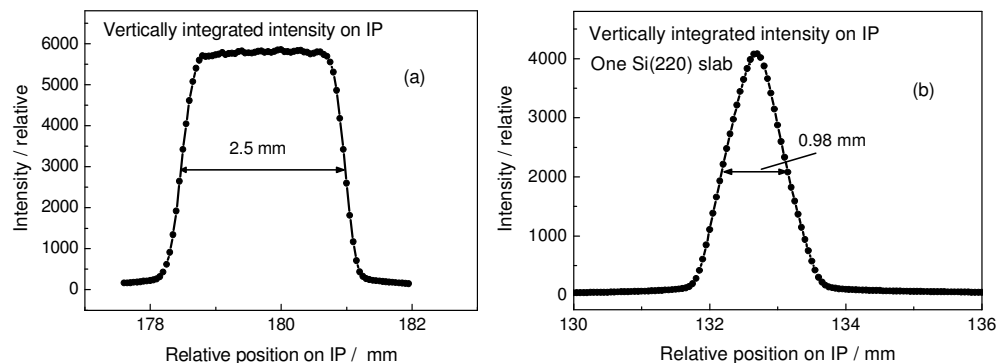


Figure 6. Relative comparison of neutron intensities: (a) - intensity passing through a 2 mm slit measured by IP before the Si(220) crystal, (b) - intensity passing through a 5 mm slit measured by IP at 70 cm behind the Si(220) crystal.

3.2. Asymmetric diffraction geometry of the Si(220) sandwich with three operating slabs

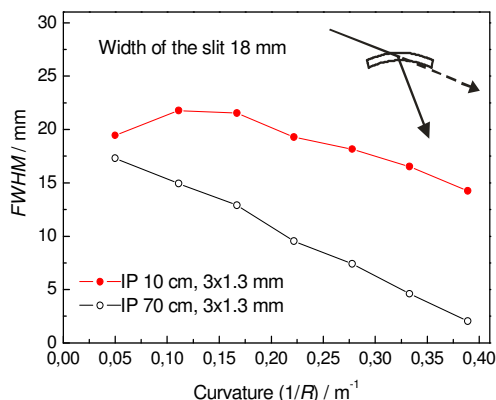


Figure 7. *FWHM* of the beam diffracted by the Si(220) sandwich in the asymmetric diffraction geometry vs its curvature.

The additional slabs in the bending device which were cut with the longest edge parallel to the [001] scattering vector provided us the possibility to study also properties of the Si(220) sandwich in the asymmetric transmission geometry of (see insert in figure 7). Figure 7 shows the dependences of *FWHM* of the beam diffracted by the bent Si(220) sandwich (in this case with three operating slabs) in the asymmetric diffraction geometry as a function of curvature for the slit of 18 mm and two different distances from the sandwich. It can be seen from figure 7 that in this case the minimum *FWHM* (at the distance of 70 cm) achieved in the range of used curvatures was 2 mm. For a larger curvature, *FWHM* can be expected even smaller.

3.3 Luminosity of the diffractometer

Of course, the luminosity of the diffraction performance which is determined by the efficiency of the double diffraction process on the bent Si(111)+Si(220) setting also plays an important role. In this way the dependence of the peak intensities of the double-crystal rocking curves of the bent Si(220) slab/slabs on the curvature with respect to the constantly bent Si(111) slab can appear very helpful. The following figure 8 shows an example of the DC rocking curves for one curvature of the sandwich and the dependence of the peak intensities of the DC rocking curves of the individual sandwiches on the curvature of the slabs for symmetric Si(220) reflection as well as of asymmetric transmission geometry. It can be seen from figure 8 that the sandwich with 3 slabs set in asymmetric transmission geometry provides slightly better reflectivity and *FWHM* parameters that the that the sandwich with 4 slabs set in symmetric reflection geometry.

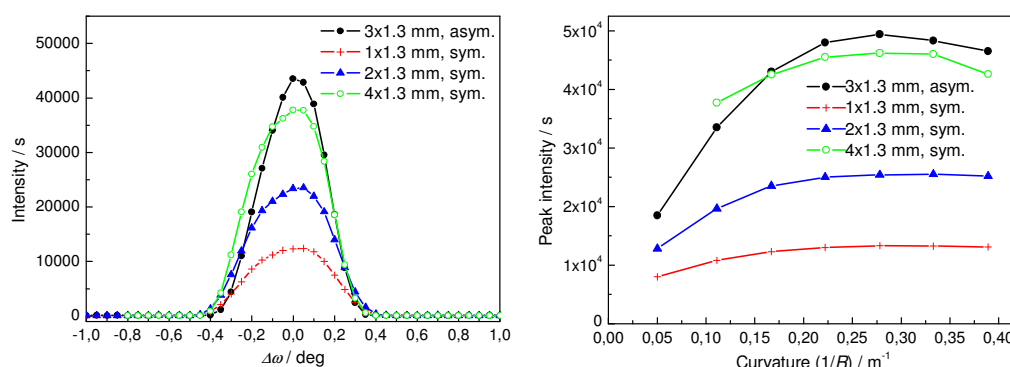


Figure 8. Examples of the rocking curves of the double crystal Si(111)+Si(220) setting for the radius of curvature $R=6$ m ($1/R = 0.167$ m⁻¹) – (a) and the dependence of the peak intensities of the rocking curves on the curvature of the sandwich – (b).

3.4 Diffraction by a polycrystalline sample

Finally, in the next step, diffraction profiles on a well annealed standard α -Fe solid polycrystalline pin of 2 mm diameter were taken for different curvatures of the sandwich with one Si(220) crystal and 5 mm slit for the incident beam. The sample was installed at the third axis of the instrument at the distance of 50 cm from the Si(220) crystal and the Imaging Plate was at 45 cm from the sample (see Fig. 2). Scattering angles on the sample for α -Fe(211) and α -Fe(220) reflections were $2\theta_s=88^\circ$ and 107° , respectively. Figure 9 shows two of several results taken for different Si(220) crystal curvatures

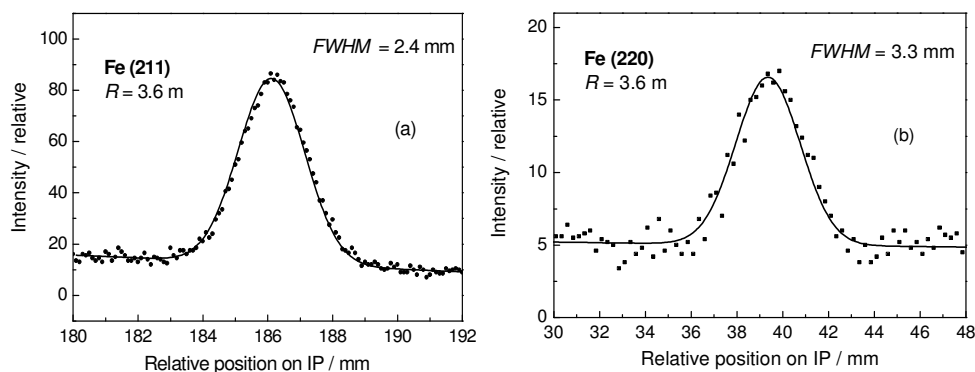


Figure 9. Diffraction profiles related to α -Fe(211) - (a) and α -Fe(220) - (b) reflections obtained by diffraction on a standard polycrystalline pin of the diameter of $\phi=2$ mm and imaged by IP.

which provided minimum *FWHMs*. Inspection of figure 9 reveals that the diffracted beams from the α -Fe pin have very low divergence $\Delta(2\theta_s)$ of about 1×10^{-3} rad for 211 reflection and 3×10^{-3} rad for 220 reflection, respectively and document a high resolution property of the double-crystal performance.

4. Conclusion

It can be said from the obtained experimental results shown in figures 2-6 that contrary to a common view, the presented double-crystal monochromator with crystals in the dispersive setting can provide a highly focused neutron beam of a sufficient current at the sample position that can be used in powder diffractometry; of course, at a reasonably high flux neutron source. One should keep in mind that one works with open beams without any Soller collimators and the (+n,-m) setting behaves as a self collimation assembly producing a highly monochromatized beam. Due to the double diffraction on crystal planes of different lattice spacing, a high resolution is thus expected in both $\Delta(2\theta)$ (2θ is the scattering angle) as well as $\Delta\lambda/\lambda$. In the present case, it is necessary to point out that the instrument could not be fully optimized because of using a fixed curvature of the Si(111) crystal. Furthermore, due to the given experimental conditions, the distance between the bent crystals was also fixed and rather large (1.7 m). When all these parameters in connection with a possibility of an employment of vertical focusing are optimized, we believe that the luminosity of the presented experimental setup could be by one order of magnitude higher. Even at our medium power neutron source when using a high resolution position sensitive detector instead of IP (having rather low efficiency), the time expected for the collection of the data related to individual diffraction profile is expected to be less than 20 minutes. For the design and full optimization of such DDC setting, Monte Carlo simulations are desirable [14]. For this purpose one can use a code which has been already published and which permits an evaluation of the resolution function and intensity properties of neutron diffractometer taking into account any spatial configuration of the experimental setup and the monochromator curvature as well as including also a double-crystal monochromator.

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