ICANS-XIII
13th Meeting of the International Collaboration on
Advanced Neutron Sources
October 11-14, 1995
Paul Scherrer Institut, 5232 Villigen PSI, Switzerland

NEUTRON COMPOSITE GERMANIUM MONOCHROMATORS FOR DIFFRACTION INSTRUMENTS AT SINQ

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ABSTRACT

Composite germanium monochromators are in the beginning of their application in neutron diffraction. In order to optimize their properties for high resolution neutron powder diffraction we significantly improved the alignment procedure of the individual wafers used for building the monochromator plates, as we added the tin necessary for soldering with a sputtering technique instead of tin foil spacers. Sputtering could also be replaced by evaporation giving similar results. A layout of the new vertical focusing monochromator is shown.

1. Introduction

Germanium is a widely used material for neutron monochromators, as it has both high scattering length density and moderate absorption when used in the thermal range. A particular advantage is the absence of second order contamination for certain reflections. However, it is also known that the introduction of a defect structure appropriate for an efficient neutron monochromator by plastic deformation has turned out in the past to be a severe problem, especially as an anisotropic mosaic is necessary in order to use this crystal for a focusing monochromator. This problem can be avoided by stacked composite wafers monochromators. Recent work at Brookhaven National Laboratory has demonstrated that a largely improved quality can be obtained when the crystals are assembled from plastically deformed thin wafers of 0.3 mm thickness [1]. First such ideas have been presented in [2]. Based on this achievement several groups are actively engaged in the production of Germanium monochromators along the procedure traced by BNL. We intend to use this monochromators at the new spallation source SINQ presently under completion at PSI Villigen.

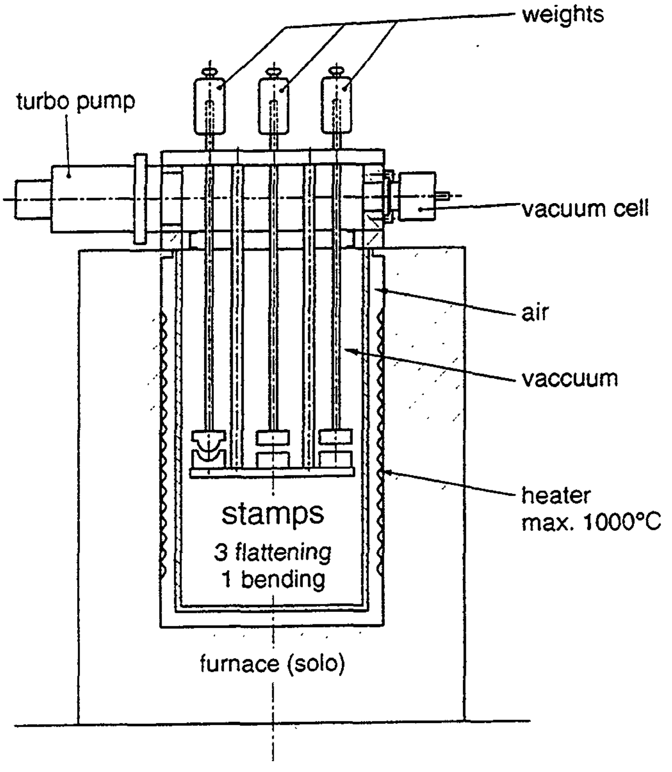
Keywords: Diffraction, Monochromators, Germanium, Composite, Instrumentation
PACS: 61.12.Dw 62.20.Hg

As *hkk* ($k \geq 0$) reflections will be used on the new high-resolution powder diffractometer by simple turning the monochromator around the ω axis, we need a high precision re-assembling of the wafers after the bending and flattening processes necessary in order to introduce the anisotropic mosaic. Such a monochromator was in operation at the reactor SAPHIR [3], however, at this time there was a one block germanium monochromator with anisotropic mosaic available, where re-alignment is not necessary.

2. Materials, Methods and Production

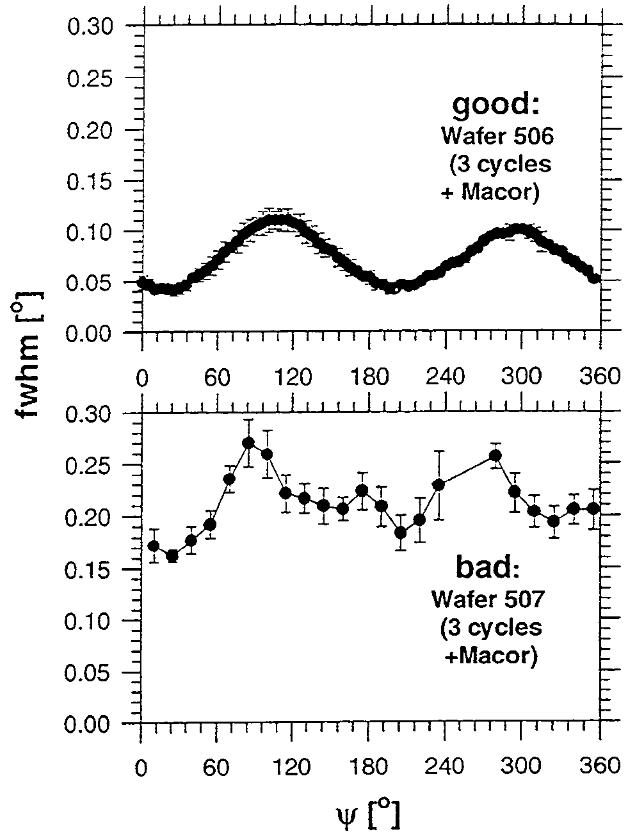
Germanium wafers have been purchased from Eagle-Picher Industries with a thickness of 0.4 mm and a diameter of 76 mm. Before cutting the wafers from the single crystal block, two reference planes with different lengths perpendicular e.g. to *[0,1,-1]* and *[2,-5,-5]* for the Ge$_{511}$ primary orientation have been cutted off from the entire boule in order to allow re-alignment and identification in a unique way. The surface was only etched, but not polished. Each wafer was marked with a diamond and cleaned with alcohol.

Figure 1:
Schematic drawing of the insert to the top-loading vacuum furnace used at PSI



In the first bending and flattening process, we followed the method introduced by BNL [1]. We used stamps made of steel (Steel 304). The bending radius of the stamp and the corresponding matrix was 44.2 mm and 47.6 mm, respectively. The difference compensates for the 6 wafers and the 7 grafoils processed between. All stamps were protected with tantalum in order to avoid reactions between steel and germanium. A pressure of 3 N/cm^2 was loaded manually at 870 °C for 20 minutes on all stamps. We had simultaneously four stamps in the furnace: One for bending 6 wafers separated by 0.2 mm thick grafoils, and three stamps loaded with two wafers each separated by a grafoil. Ge$_{511}$ wafers were bent twice (once to one side), Ge$_{311}$ had to go another cycle. A Schematic cut through the furnace is shown in Fig. 1. This allows to proceed parallel with 12 wafers in one load, which is essential, as we can only operate one cycle per day due to the long cooling time (~20 hours) of our vacuum furnace.

Figure. 2:
Halfwidths of rocking curves (fwhm) of two Germanium 311 wafers (0.4 mm thick) at different ψ-orientations, showing an accepted (top) and a refused (bottom) wafer. The bottom wafer has to be re-flattened.



Flattening has proved to be the most difficult and essential step in the whole bending procedure. Using standard steel stamps protected by grafoil or tantalum has not given acceptable results for final use with neutrons. This step was only used during the bending process described above in order to re-flatten the wafers approximately. Unevenness could be observed by extended mosaics in neutron test measurements as shown in Figure 2 (bottom). We therefore have chosen a 8 mm thick ceramic (MICAR™), which was flattened to reach a parallelity of 20 µm. Such plates should be regularly tested for flatness and eventually re-machined. The germanium was in direct contact with the ceramic up to 870 °C. The flattening step with the ceramic has been done twice, allowing an acceptance of the wafers close to 100%. About 5% got lost due to breakage during all the processes involved. A majority of the wafers was tested with rock scans at different ψ angles using neutrons (Saclay and T13C at ILL).

3. Sputtering Step

In order to reach a uniform coating of the flattened wafers, magnetron sputtering technique (Leybold) was chosen using a in-house built tin target. The goal of this step was to fix a uniform layer of tin directly to the wafers, allowing a precise alignment of the individual wafers against each other, as the package is not disturbed by the soft tin foils. Additionally the amount of tin can be reduced. We have chosen a thickness of 2.6 µm on both sides instead the 25 µm thick tin foils chosen by others [1]. There is still additional tin pressed out of the composite stack during the soldering process. The thickness of the tin layer was measured on a reference glass substrate. The coated wafers were afterwards aligned in a stamp made of graphite and pressed with their long reference cut of 25 mm length to the graphite block machined to 0.02 mm precision, using a excentric mounted bolt. Afterwards, the whole package was heated in our vacuum furnace (10^{-5} bar), loaded with 6 N/cm^2 at 350°C, stabilized at 450°C for approximately 10 minutes and afterwards cooled within 3 hours with the weight on the composite wafer package down to room temperature. First test cuts with a diamond saw show, that we can reach a stable monochromator package this way, which can be machined with standard tools.

Our monochromators will be cutted into pieces of 55 mm width and 25 mm height for the high resolution powder diffractometer HRPT and 55 mm width and 12.5 mm height for the four circle diffractometer SC3. A focusing monochromator which allows individual alignment of each piece is presently under construction. Focusing will be done by one motor only, using a method described elsewhere [4].

4. Results

The typical rocking curves at the extremes and the anistropic mosaic distribution as a function ψ of a single wafer of 0.4mm thickness are shown in Figs. 3 and 4, respectively. A first assembly with tin sputtered wafers has been tested with neutrons. Results are shown in Figs. 5 and 6, showing the almost sinusoidal mosaic spread. In addition, we also could measure a Ge_{004} -reflection in an off-centric beam geometry and therefore reached the goal of changing the wavelength by turning around the ω-axis only. The measured reflectivity of 35% and 30% at the minimal and maximal mosaic of 3.9' and 7.6', respectively, close to the calculated ones of 49% and 34% for λ=1.3 Å, respectively [4,5]. The incoming flux was $3.44 \cdot 10^3$ n/cm^2/s. The permanent quality control is essential, as shown in Figure 2 by two examples.

Figure 3:
Neutron rocking curves of a single wafer (Ge $_{511}$ orientation) at ψ=0° and ψ=90°, using a perfect Ge_{331} monochromator with λ=1.49 Å at Saclay.

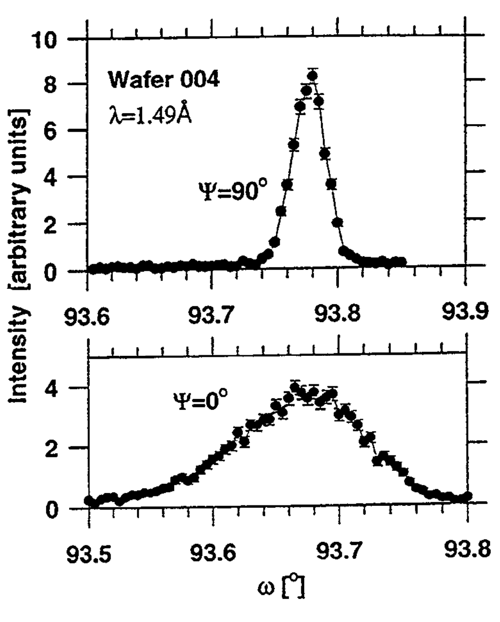


Figure 4:
Halfwidths of rocking curves (fwhm) of a single wafer (0.4 mm thick) at different ψ-orientations.

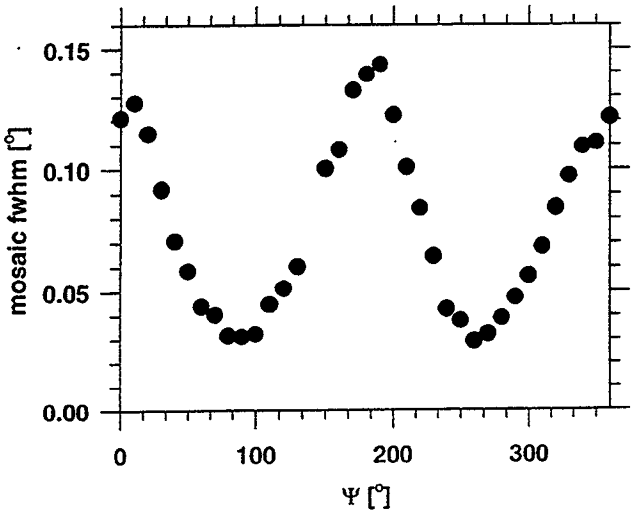


Figure 5:
Neutron rocking curves of a composite Germanium monochromator of 24 wafers (9.6 mm thick, Ge $_{511}$ orientation) at ψ=30° and ψ=120°, using a perfect Ge$_{331}$ monochromator with λ=1.3 Å at T13C/ILL.

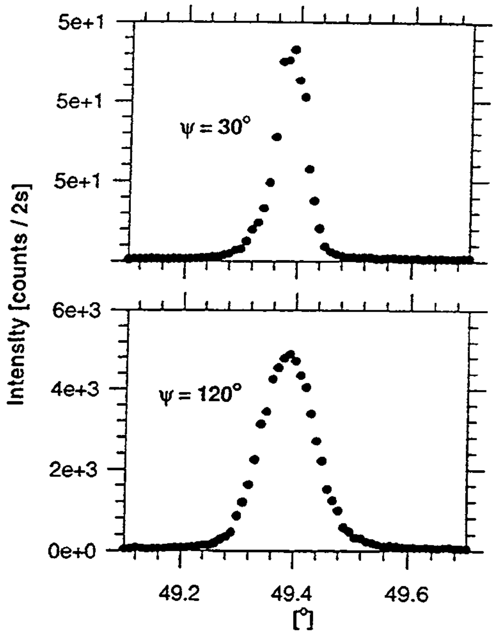
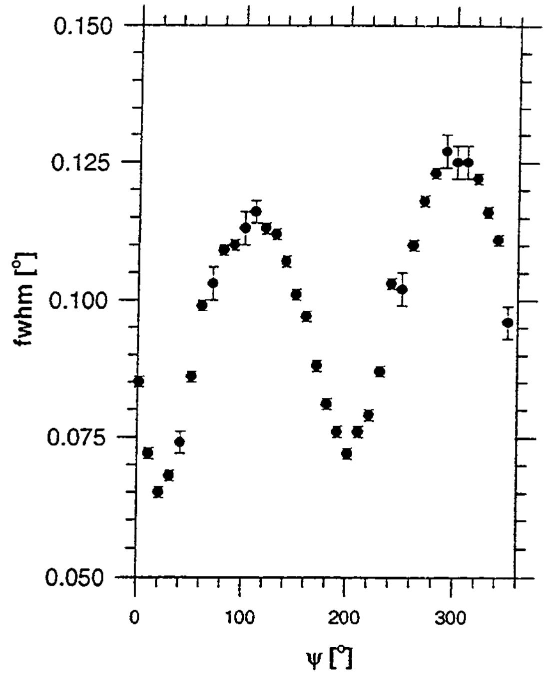


Figure 6:
Halfwidths of rocking curves (fwhm) of a composite Germanium monochromator of 24 wafers (9.6 mm thick) at different ψ-orientations.



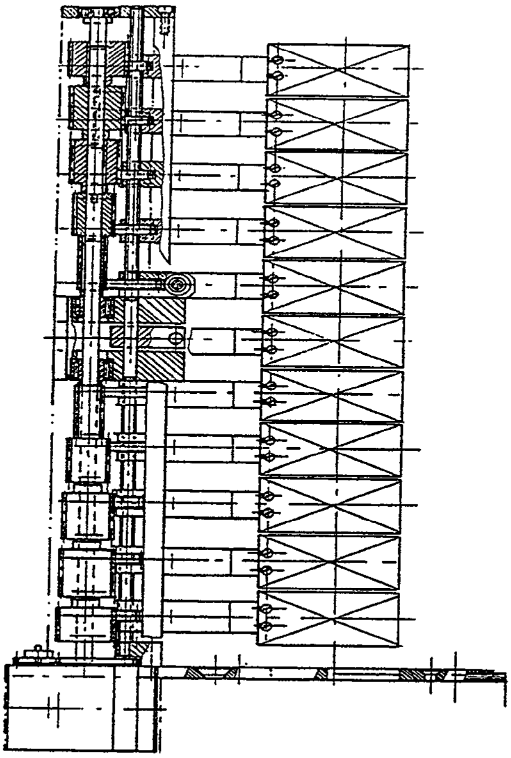
5. Mechanical Layout

The new high resolution powder diffractometer HRPT and single crystal diffractometer SC3 at SINQ will be equipped with a vertical focusing composite germanium monochromator of 280 mm height (Figure 7). Together with a lift system available, this allows a very flexible setting of the wavelengths without changing the instrument position. However, if additional wavelengths are asked for, the instrument may be manually moved from the 120° to the 90° take-off position. The single crystal diffractometer will mostly use the 311 reflection. The lift system allows the use of a high intensity/low resolution C002-monochromator for setting up the orientation matrix or for special applications with a high wavelength of λ= 2.34Å.

6. Summary

The method introduced by BNL [1] yields good Germanium composite wafer monochromators. Improvements by sputtering the tin to the wafers instead of using tin foils increases the accuracy of re-assembling the 'original' crystal. No additional mosaic increase due to misalignment is observed. A small decrease may be due to the better flatness of the composite monochromator compared to the single wafer, which is relatively thin and may be bent marginally due to the mounting pressure from the sample holder necessary for the neutron tests. We expect comparable results if using tin evaporation instead of sputtering. The sputtering technique was used by us due to the availability of such a machine at PSI. A 3 µm thick tin layer is recommended instead of 2.6 μm in order to increase the amount of the tin, as the wafers are soldered on a large numbers of spots but not on the full surface as shown by the cuts with a diamond saw. Additionally, the primary reference planes should be elongated from 25 mm to 30-35 mm in order to lower errors in the alignment further. For the monochromator of the high resolution powder diffractometer, we will add one or two additional cycles to the present two in order increase the mosaic shown in Figs. 5 and 6 to approximately 12' or 16' respectively. Monochromators built in such a way will be especially interesting for instruments using a high take-off angle, as neighboring reflections are accessible by a limited angular motion in ω, therefore still covering the whole beam. Further improvement of the technique may be reached by using an improved bending technique as described by [8].

Figure 7:
Mechanical Layout of the new vertical focusing HRPT monochromator.



Acknowledgment

We would like to thank Dr. T. Vogt from Brookhaven National Laboratory for the generous support with technical information. We are indebted to Dr. A. Magerl from ILL and Dr. W. Paulus from LLB for beam time on their neutron test spectrometers, essential for the permanent quality control of our production. We thank Drs. P. Böni and W. Bührer for stimulating discussions, and last but not least the technique department from PSI for support in their workshops and using their vacuum furnace and J. Allibon at ILL for all the help with the 'specialties' of T13C.

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