ROBL-CRG	**Experiment title:** **HIGH- PRECISION DETERMINATION OF ATOMIC POSITIONS IN 6*H*- AND 4*H*-SIC CRYSTALS**	**Experiment number**: 20_02_014
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Report:

The measurement of the "quasiforbidden" reflections is a very sensitive method in order to determine very small changes of atomic positions (e.g. relaxations), deformations of the charge density in dependence of the chemical bonding or differences of atomic vibrations . We used this method to determine the atomic positions of 4*H*- and 6*H*-SiC crystals with high precision. This was necessary to examine the polytype-depending displacements of the C- and the Si- atomic positions from the ideal tetrahedron and to compare these results with *ab initio* calculations [1][2]. Such atomic displacements of the carbon atoms ($\varepsilon(j)$) and the silicon atoms ($\delta(j)$) are in order of magnitude of 10^{-4} times the *c*-lattice constant. These small displacements result in a non-vanishing integrated intensity of the "quasiforbidden" reflections. Since these integrated intensities are essentially smaller than the intensities of the occurring "Umweg" reflections, a correction of the influence of the "Umweg" reflections is necessary. To carry on former investigations we made measurements at BM 20 at the ESRF to:

(i) get more precise results by means of refined "Umweg" corrections.

(ii) get unambiguous structure models by measurement of extreme weak reflections (especially asymmetric reflections).

(iii) specify the "absolute" structure by determination of the phase invariant ϕ_{inv} at multiple beam cases, i.e. differentiation of structure models, which only differ from each other in the sign of the atomic displacements.

In order to achieve this goal high primary X-ray intensity ("quasiforbidden" reflections) and a six-circle-diffractometer (rotation about any desired lattice plane normal) was necessary.

Since the evaluation of point (i) and (ii) has not been finished up to now, only point (iii) will be discussed in the following.

Since the relaxed structure is characterized by the atomic displacements ($\delta(j)$, $\varepsilon(j)$) from the ideal tetrahedron structure, both structures $S_+ = (\delta(j), \varepsilon(j))$ and $S_- = (-\delta(j), -\varepsilon(j))$ result in the same integrated

because of $|F_+(\boldsymbol{H})| = |F_-(\boldsymbol{H})|$, where $\boldsymbol{H}$ is the reciprocal lattice vector of the "quasiforbidden" reflection. Therefore the both structures cannot be distinguished by measurement of the integrated intensities only. Because of

$$\phi_+(\boldsymbol{H}) = \phi_-(\boldsymbol{H}) \pm 180°$$

for small displacements δ(j) and ε(j) the "absolute" structure can be determined if the phases of the structure factor of the "quasiforbidden" reflections are known. The measurement of these phases becomes possible by superposition of the "quasiforbidden" reflection $\boldsymbol{H}$ with "Umweg" reflections [3]. When choosing a three beam geometry the two remaining reflections $\boldsymbol{L}$ and $\boldsymbol{H}$ - $\boldsymbol{L}$ have to be strong reflections, because strong reflections are independent of the relaxations. If the phase invariant

$$\phi_{inv} = \phi_{\mathbf{L}} + \phi_{\mathbf{H-L}} - \phi_{\mathbf{H}}$$

is close to 0° or 180°, the wings of the peaks in the azimuth scan (rotation about the lattice plane normal of the weak reflection $\boldsymbol{H}$) of the "Umweg" reflections are strongly asymmetric. The model S_+ shows the inverse asymmetry compared with the model S_- (FIG. 1).

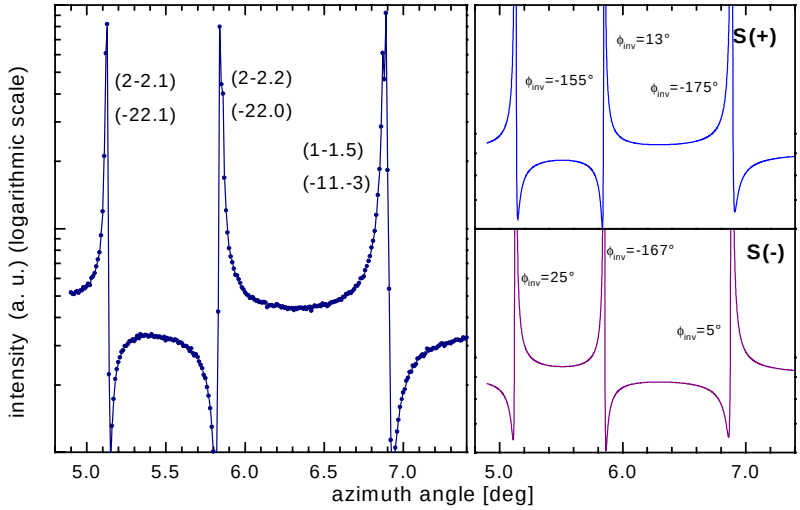

FIG. 1: Left: experimental azimuth scan of the 4*H*-SiC 00.2 reflection. In brackets the Miller indices of the strong reflections $\boldsymbol{L}$ and $\boldsymbol{H}$ - $\boldsymbol{L}$. Top right: simulation of the 4*H*-SiC 00.2 reflection according to Ref. [4] model S_+ with the phase invariant. Bottom right: simulation of the 4*H*-SiC 00.2 reflection according to Ref. [4] model S_- with the phase invariant.

In order to distinguish the model S_+ from S_-, reflections with small Miller indices are particularly useful, since the influence of temperature vibrations can be neglected for silicon carbide in this case.

Comparing the measured data with the simulation of the "Umweg" peaks according to Ref. [4] we found a good agreement with the model S_+ in the case of 4*H*-SiC (FIG. 1) as well as for 6*H*-SiC (without figure). These results also match with the *ab initio* calculations in Ref. [2].

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[2] Käckell P.: *Siliziumkarbid – Strukturelle und elektronische Eigenschaften verschiedener Polytypen*, Dissertation, Friedrich-Schiller-Universität Jena, 1996.

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[4] Shen Q: *A New Approach to Multibeam X-ray Diffraction Using Perturbation Theory of Scattering*, Acta Cryst. (1986), **A 42**, 525-533.