LATTICE-PARAMETER DETERMINATION OF QUARTZ BY MEANS OF THE Ω-SCAN METHOD

H. Berger, H. Bradaczek, G. Hildebrandt
EFG International, Research Center, Berlin, Germany

Abstract - **The lattice parameters of quartz depend on the crystal quality and influence the accuracy of the X-ray orientation determination of resonators. Therefore, they should be known better than 20 ppm. Using the Ω-Scan Method, the lattice parameters can be determined together with the orientation, measuring at least two additional reflections. Preliminary measurements on SC-cut quartz blanks were performed with standard deviations of the lattice parameters of ≤6 ppm in a measuring time of about 3 min. The evaluation of the peak position has to consider its systematic error. The total error is estimated to be <10 ppm. The measuring procedure can also be applied to IT-cut, FC-cut and AT-cut quartz blanks and, moreover, to any single crystal.**

Keywords - **Lattice-parameter determination, quartz, X-ray Ω-Scan Method, correction of systematic errors**

I. INTRODUCTION

The lattice parameters of quartz depend on the impurity content and are influenced by the growth conditions. The problem has been discussed extensively by Brice [1]. Values recommended by this author for various quartz qualities are shown in Table I. Still higher differences were found by Kusters and Adams [2] comparing a number of charges.

TABLE I
LATTICE PARAMETERS OF QUARTZ [1]
25°C; uncertainties in the last digit

Grade of quartz	a(Å)	c/a
premium natural	4.9129 ± 2	1.10010 ± 6
premium synthetic	4.9134 ± 2	1.10012 ± 4
medium synthetic	4.9136 ± 3	1.10008 ± 6
low synthetic	4.9138 ± 5	1.10004 ±10

The lattice parameters describe the crystal quality and are fundamental material quantities for measurements and calculations in solid state physics and of technological properties. However, their exact knowledge is also the precondition for the accurate evaluation of X-ray diffraction measurements like the orientation determination. The angular coordinate Θ of a SC-cut quartz blank is usually determined by the X-ray measurement of the angles of two lattice-planes to the surface. If the lattice parameters of the corresponding quartz charge are unknown and, e.g., those for synthetic premium instead of low quality quartz are used (cf. Table I), errors of 4×10^{-4} Å in a and 8×10^{-4} Å in c result. Then the angular coordinate Θ will also have a systematic error in the order of 10 arcsec. Therefore, for the accurate orientation determination of resonators and the following cutting-angle sorting, the lattice parameters should be known. For highest accuracy of the orientation coordinates, the uncertainty in the lattice parameters should to be smaller than 1×10^{-4} Å or 20 ppm.

In principle, the lattice-plane distances in single crystals of sufficient quality can be determined with a precision up to 0.5 ppm [3], in special cases up to 0.1 ppm [4]. The errors of the lattice parameters of non-cubic crystals calculated from two or more measured lattice-plane distances are usually larger. So the lattice parameters a and c of quartz were determined with total errors of 0.4 ppm and 4 ppm, respectively [5].

Such measurements need special precision diffractometers and the crystal sample has to be adjusted exactly. However, the lattice-parameter determination of quartz should be performed as an automatic process combined with the succeeding orientation determination. Furthermore, the total errors of the lattice parameters need not to be extremely small. It will be shown in the following that these demands can be fulfilled using the Ω-Scan Method as applied in Automatic and Semi-Automatic Cutting-Angle Sorters (Machines produced by EFG International Berlin).

II. MEASURING PRINCIPLE

In the X-ray Ω-Scan Method the quartz blank rotates around an axis perpendicular to its surface (Ω circle). During one rotation, an X-ray beam is successively twice reflected by each of certain lattice planes. From the angular distances of the reflections at two lattice planes the orientation coordinates can be precisely calculated [6-8].

If the orientation of the sample is known, then, in principle, also the lattice parameters can be calculated alternatively from the measured values of the Ω-Scan. If the orientation as well as the lattice parameters are unknown, all these parameters can be determined simultaneously, registering and evaluating at least one additional reflection. However, in order to obtain a sufficient sensitivity for the lattice parameters as well as for the orientation, the reflections selected for the orientation determination should be combined at least with two further reflections. These should have high diffraction angles and opposite sensitivity for both lattice parameters (i.e., the ratios of the indices sums $h+k$ to the indices l in both hkl triples should be opposite.) The orientation coordinates, the incidence angle of the incident X-ray beam, and above all the lattice

parameters a and c then can be calculated by a least-squares procedure.

The measurement of at least two additional reflections at high diffraction angles demands to equip the usual Ω-Scan measuring apparatus with a second X-ray detector. A view of an arrangement with two detectors for the combined orientation and lattice-parameter measurement is shown in Fig. 1.

III. Measurements of SC-cut Quartz Blanks

Preliminary combined measurements of orientation and lattice parameters of SC-cut quartz blanks have been performed using CuKα radiation and a Cutting-Angle Sorting Machine equipped with an additional detector according to Fig. 1. For the orientation determination the reflections 2 1 1 and 2 1 3 were taken as usually [7]. Additionally, the reflections 3 3 1 (diffraction angle 72.05°) and 3–1 6 (78.53°) were measured (Fig. 2). In the Ω-Scan diagram, the peaks for $K\alpha_1$ and $K\alpha_2$ radiation are partially resolved because of the high diffraction angles.

The peak positions can be determined with a standard deviation in the order of 0.03° per 1 rotation of the turntable. This corresponds to errors in a and c, respectively, of 1×10^{-4} Å (20 ppm) and 3×10^{-4} Å (60 ppm). Using, e.g., 100 rotations of the turntable (measuring time 3 min), the errors decrease by a factor of 10, fulfilling the precision demands as quoted above.

The lattice parameters measured at different points of an SC-cut blank are given in Table II. The values are varying in the order of 1×10^{-4} Å or less.

A special problem are the systematic errors and their reliable correction. The problem has been extensively discussed earlier with regard to the orientation determination by means of the Ω-Scan Method [9, 10]. Corresponding estimations of the influence on the lattice parameters show that the refraction error is smaller than 1×10^{-5} Å and, therefore, can be neglected. Temperature deviations of 1°C would lead to errors in a and c, respectively, of 7×10^{-5} Å and 4×10^{-5} Å. As the temperature can be kept constant to better than 1°C, the concerned uncertainties can also be neglected. The main systematic error is due to the uncertainties in the determination of the peak positions. These uncertainties are the differences between the values calculated from the measured reflection curves and their unknown real values, which would deliver the real orientation coordinates and lattice parameters. The error depends on the applied evaluation procedure of the reflection curves. As estimated by simulation calculations, the errors in the lattice parameters a and c may reach some digits in 10^{-4} Å. Therefore,

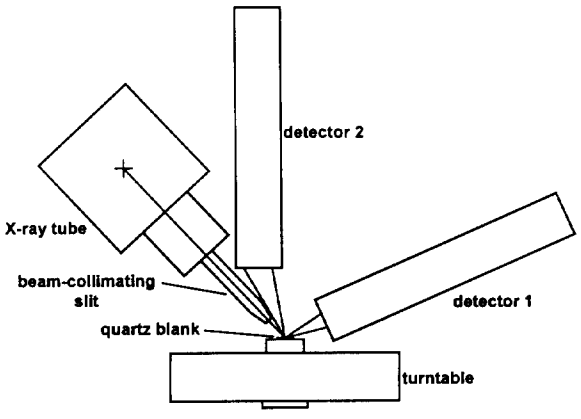

Fig. 1. Scheme of an Ω-Scan measuring arrangement with two detectors.

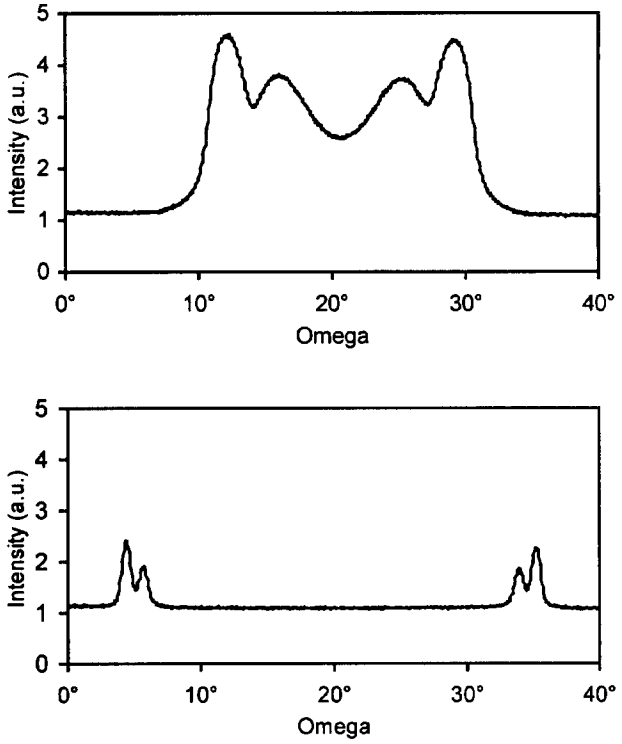

Fig. 2. Parts of the Ω-Scan measuring diagram (intensity vs. Ω). SC-cut quartz, CuKα radiation, reflections 3 3 1 (at the top) and 3-1 6 (at the bottom; same intensity scale for both reflections).

TABLE II
LATTICE PARAMETERS OF SC-CUT QUARTZ MEASURED AT DIFFERENT POINTS OF THE SURFACE
CuKα radiation; reflections 2 1 1, 2 1 3, 3 3 1, 3-1 6; uncorrected; 25°C

Measuring point	a(Å)	c(Å)	c/a
1	4.91350	5.40499	1.10003
2	4.91352	5.40492	1.10001
3	4.91347	5.40489	1.10002
4	4.91351	5.40495	1.10002

a reliable correction procedure is indispensible.

This procedure requires the exact simulation of the reflection curves. For this simulation, the spectral distribution as well as the dimensions of the collimator and the X-ray tube focus have been taken into account. Real intensities including measured values and dead-time losses of the detecting system have to be considered. For highest accuracy, the simulation can be performed individually for all the peaks of each measurement by fitting of the calculated to the measured curves, varying slightly the geometrical parameters. The peak positions used for the calculation of the best fitted curves are considered as the real positions.

In the case of the reflection 3–1 6 the measured intensity profile can be simulated properly in the upper part of the $K\alpha_1$ curve, which is used for the peak determination, as shown in Fig. 3. For the reflection 3 3 1, the Kα doublet is not so clearly resolved. Therefore, a reliable correction of the peak positions could not be performed for these preliminary measurements yet. However, it was shown by simulation calculations that a slightly changed incidence angle and a modified slit geometry for the incident beam would lead to a sufficient peak resolution, so that in this case the full correction procedure will be possible. The reliability of the corrected values should then be better than 5×10^{-5} Å (10 ppm).

IV. Applicability to other orientations

It has been estimated by simulation of the geometrical conditions that the measurement and evaluation will be possible in the whole SC range as well as for IT and FC cuts using the same reflections and a suitable incidence angle of the X-ray beam. In all cases the reproducibility should be of the same order of magnitude. Similarly, the lattice-parameter determination by means of the Ω-Scan Method can be also performed for AT-cut blanks using suitable additional reflections. This is valid for the geometrical conditions used in the conventional Cutting-Angle Sorting Machine as well as for the recently developed machine, the so-called "Carousel" [8]. The concerned reflections and the diffraction angles of the additional reflections are summarized in Table III for all cases. The additional reflections for AT-cut blanks occur twofold (e.g., 3 3 0 and 6-3 0) because a symmetry plane with regard to the X-ray reflections exists. The measuring arrangements have to be equipped with additional detectors, and the slit geometry for the incident beam has to be modified, too, in order to be able to correct reliably the peak-position error. The reproducibility is estimated to be in the same order as given for SC.

A similar procedure should be possible for any relevant orientation of quartz or of other single crystals. At least four reflections have to be selected, in each case two being especially sensitive for the orientation and the lattice parameters, respectively.

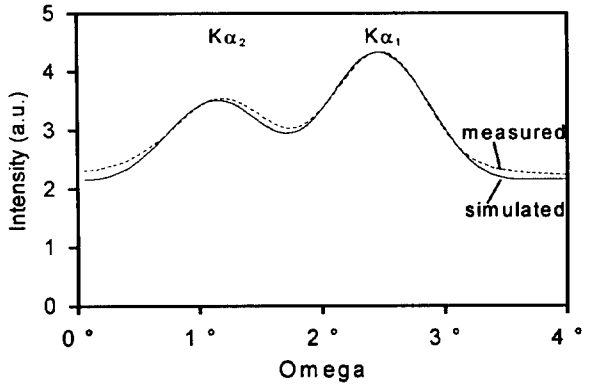

Fig. 3. Comparison of measured and simulated Ω-Scan reflection curves (intensity vs. Ω).
SC-cut quartz, CuKα radiation, reflection 3-1 6.

V. Conclusions

As shown by the simulation calculations as well as by the results of the preliminary measurements on SC-cut quartz blanks, the lattice parameters of quartz can be determined automatically by means of the Ω-Scan Method with total errors in a and c of smaller than 5×10^{-5} Å or 10 ppm taking a measuring time of a few minutes. For this purpose, the Cutting-Angle Sorting Machines have to be equipped with an additional X-ray detector in connection with slightly smaller beam-limiting slits in order to produce fully resolved peaks of the Kα doublet of the X radiation. The lattice-parameter determination need to be done only once at the beginning of the measurements of any charge.

TABLE III
REFLECTIONS TO BE MEASURED FOR THE LATTICE-PARAMETER DETERMINATION OF QUARTZ BY MEANS OF THE Ω-SCAN METHOD
$CuK\alpha_1$ radiation

Orientation	main reflections *h k l*	additional reflections *h k l*	diffraction angle
SC, IT, FC	2 1 1	3 3 1	72.05°
	2 1 3	3-1 6	78.53°
AT	2 0 2	3 3 0 / 6-3 0	70.15°
(conventional)	2 0 3	-1 3 6 / 2-3 6	78.53°
AT	1 0 1	-1 5 3 / 4-5 3	68.94°
("Carousel")	2 0 1	-1 2 6 / 1-2 6	65.60°

Using the correct lattice parameters, the cutting-angle sorting based on the measured orientation coordinates can be performed. However, also the lattice parameters themselves can be stored as they characterize the quartz quality, especially the impurity content.

Moreover, the arrangement as described above can also be realized as a special machine for the automatic lattice-parameter determination of quartz or of other relevant single crystals. The advantage of such a device compared with a conventional diffractometer is the simplicity of the measuring procedure. The sample, whose orientation must be roughly known, need not to be adjusted before the measurement, and all the necessary reflections are measured during one run utilizing only the continuous rotation of one measuring circle.

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