A Geometrical Correction for Precise Lattice Constant Determination

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Dedicated to Professor G. Hildebrandt on the occasion of his 60th birthday

A correction term for high precision lattice constant investigations has been derived, which is caused by geometrical and absorption effects. Surprisingly there is no dependence on absorption and extinction in the case of high absorption and/or "infinite" crystal thickness. Only the half width of the rocking curve has to be considered. In the case of low absorption and/or thin crystal, there are terms of higher order which still depend on μt but can be neglected compared to the main effect.

The main reason for the correction is that the observed rocking curve is the result of a "true" rocking curve multiplied by a geometrical function which is not symmetrical with respect to the rocking angle. For high precision measurements ($< 10^{-5}$) the application of the correction term is recommended.

Introduction

Lattice constant measurements of high precision ($< 10^{-5}$) have recently found new interest in investigations of the quality of basis materials for integrated circuits, solar cells and other semiconductors.

Mainly the Bond-method [1] has been used, in which symmetric reflections are considered in order to avoid uncertainties in zero point determination and other systematical and instrumental errors.

Quite a few papers deal with the correction of additional errors, and serious efforts have been made to push up the accuracy to a level of 10^{-6}.

One kind of error in the Bond-method — the effect of asymmetrical absorption — has not been considered yet, although this method doubles the effect. Surprisingly the correction term is independent of the absorption coefficient in the case of infinite crystal thickness. It mainly depends on the width of the measured "rocking curve", whatever the reason of its broadening may be.

Theory

Let $I_0(x)$ be the intensity profile of the primary beam and let the geometry perpendicular to the goniometer plane be constant (line focus). The coordinate x is perpendicular to the primary beam direction and y perpendicular to that of the reflected beam (Figure 1).

Here the primary beam is considered to be parallel; the case of divergent radiation will be discussed later.

The primary beam enters the crystal and is weakened by absorption and extinction. This and the total path length of the beam within the crystal determines the profile of the reflected beam which is collected by the wide open detector. This model neglects other (e.g. dynamical) effects and describes the beam behaviour merely geometrically.

In the Bond-method detectors are put in the positions 2θ and -2θ (Fig. 2) and small oscillations β of the crystal around the positions θ and $-\theta$ are required to register a full response curve with a half width w. The following considerations are not in principle affected if we limit ourselves to the case

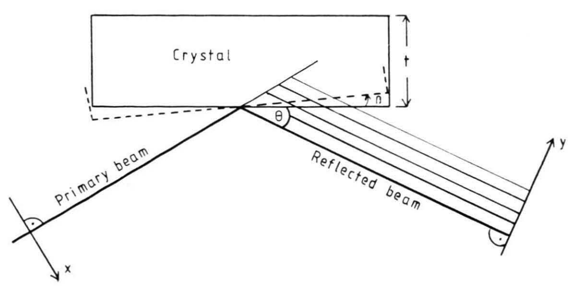

Fig. 1. Experimental arrangement for the Bond-method of precision lattice constant measurement.

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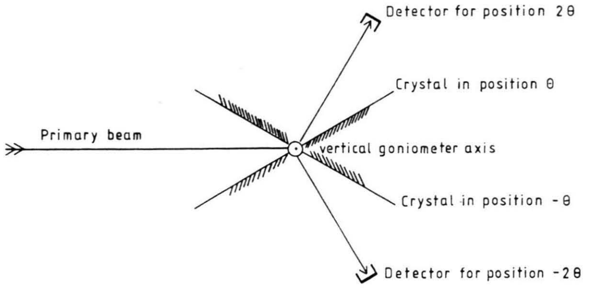

Fig. 2. Definition of quantities used.

that this halfwidth is caused by the natural line width alone, whereas the crystal is regarded as perfect and the necessary convolution with the dynamical diffraction pattern [2] is being neglected.

For the symmetrical case $\beta = 0$ the geometrical relations have been calculated earlier [3] and the profile of the reflected beam was found to be the one-dimensional convolution product

$$I(y) = I_0(x)\, S(x, y)\,, \tag{1}$$

where $S(x, y)$ is the absorption function

$$S(x, y) = \exp(-\mu r) \tag{2}$$

and $r = (y - x) \cdot 2/\sin 2\theta$ is the total path length.

For the oscillation case $\beta \neq 0$ we have

$$r = a_1 y - a_2 x$$

with

$$a_{1,2} = \frac{2}{\sin 2\theta} \pm \frac{2 \sin\beta}{\cos\beta - \cos(2\theta \pm \beta)}\,. \tag{3}$$

Note that r is not symmetrical in β.

For further simplification we now take a primary beam without lateral extension i.e. $I_0(x) = I\,\delta(x)$. Let $V(\beta)$ be the angular intensity distribution in the reflected beam without geometrical correction, where $\beta = 0$ corresponds to the exakt reflection angle θ which is derived from the Bragg-angle by adding all the well known corrections such as refraction, Lorentz-polarisation etc. Then we have for the y-profile of the reflected beam in a given crystal position β

$$I(y, \beta) = I\,V(\beta)\exp(-\mu\, r(y))\,. \tag{4}$$

The open detector integrates this reflection profile and one gets the response curve profile

$$E(\beta) = \int I(y, \beta)\,\mathrm{d}y\,. \tag{5}$$

The lower limit is $y = 0$ for the first possible beam, the upper limit can be set to infinite if the crystal is sufficiently thick and/or the absorption coefficient is high. If not, this limit is calculated to be

$$y_{\max} = t \sin 2\theta / \sin(\theta - \beta)\,, \tag{6}$$

where t is the crystal thickness. With (3) and (4), integration of (5) yields for infinite $y_{\max}$

$$E(\beta) = I\,V(\beta)\,(1 + \tan\beta \cdot \cot\theta)/2\mu\,, \tag{7}$$

which means that $V(\beta)$, taken as a symmetrical Cauchy or Gauss distribution with center of gravity and peak at $\beta = 0$ and with the half width w, is distorted by a slowly varying geometrical function $G(\beta) = I(1 + \tan\beta \cdot \cot\theta)/2\mu$ in a similar way as by the Lorentz-polarisation factor (see [1], eq. (10)).

For further evaluation either the displacement of the center of gravity

$$\beta' = \int \beta E(\beta)\,\mathrm{d}\beta \Big/ \int E(\beta)\,\mathrm{d}\beta$$

can be calculated or the peak displacement β'' by the condition that

$$\mathrm{d}E/\mathrm{d}\beta = V\,\mathrm{d}G/\mathrm{d}\beta + G\,\mathrm{d}V/\mathrm{d}\beta = 0\,. \tag{8}$$

As can be seen from Fig. 3, no considerable difference between the two results can be expected, and with the second way we obtain exactly

$$\beta'' = w^2\, \frac{(1 + \tan^2\beta'')\cot\theta}{1 + \tan\beta'' \cdot \cot\theta} \tag{9}$$

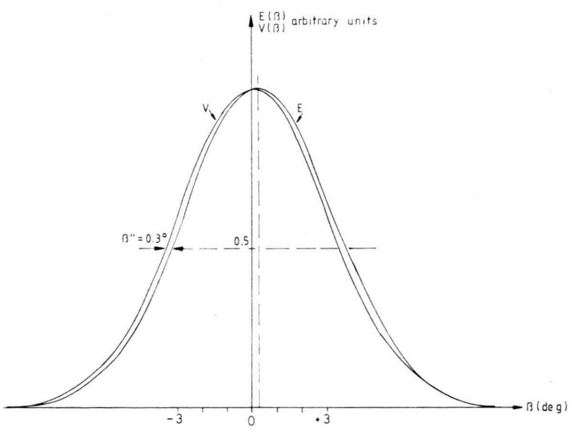

Fig. 3. Example for the angular shift due to the asymmetrical geometry function $G(\beta)$, where E is the observed rocking curve centered around $\theta_{\mathrm{obs.}}$ and V is the corrected distribution with peak position $\theta = 30°$.

or, as a first but very close approximation

$$\beta'' = w^2 \cot\theta\,, \tag{10}$$

where w can now be taken as half width of the observed response profile $E(\beta)$, see also Figure 3.

It must be noted that in this case of high absorption the absorption coefficient μ cancels out in deriving (9) and (10)! In the case of low absorption or thin crystal, (6) has to be taken into account in integrating (5) with the result that for $E(\beta)$ we have the much more complicated expression

$$E(\beta) = V(\beta)\, I\,\{1 - \exp[-\mu t a_1(\beta) \cdot \sin 2\theta/\sin(\theta-\beta)]\}/\mu a_1(\beta)\sin 2\theta\,. \tag{11}$$

Following the same way of treatment we obtain, ignoring all irrelevant terms,

$$\beta'' = w^2 \cot\theta + w^4 A(\theta, \mu t) + w^6 B(\theta, \mu t) \tag{12}$$

with $|A|$ and $|B| < \cot^3\theta$, which is dependent on μt. The value of β'' thus calculated has to be subtracted from the observed value of θ to which the well known corrections have been applied before. The order in which corrections are applied should not make much difference.

Discussion

Here, the final equations (10) for infinite μt and (12) for finite μt have been derived for the case of an incident beam, which is a) strictly parallel, b) has practically no lateral extension, c) contains the natural wave length distribution of say the CuKa1 emission line and is d) reflected by a perfect crystal. Then $V(\beta)$ is the result of c) only.

Certainly these conditions are not general. But if we keep a) and b), take highly monochromatic radiation together with a mosaic crystal, then $V(\beta)$ is the result of this mosaicity. If $V(\beta)$ can also be described by a Cauchy or Gauss function, the whole calculation is unaltered and yields the same results with w being again the half width of the observed response curve profile.

If we have both mosaicity and natural radiation, $V(\beta)$ is the convolution product of the two symmetrical distributions and therefore similarly symmetrical, and again the calculation is not affected. The same considerations hold for divergent and for laterally extended primary beams.

For the most general case we can conclude that (10) and (12) are valid wherever the observed response curve profile stems from.

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[1] W. L. Bond, Acta Cryst. **13**, 814 (1960).
[2] A. H. Compton and S. K. Allison, X-Rays in Theory and Experiment, Princeton, J.J., USA. See Eq. (9.58), p. 719.
[3] H. Bradaczek and R. Hosemann, Acta Cryst. **A 24**, 568 (1968).