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# Crystal perfection of $HgI_2$ studied by neutron and gamma-ray diffraction

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# Crystal perfection of $HgI_2$ studied by neutron and gamma-ray diffraction [a)]

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The crystalline perfection of wire sawn pieces of vapor grown single crystals of mercuric iodide was compared with the perfection of (00*l*) cleaved sections of the same crystal from which nuclear radiation detectors have been fabricated. The crystalline perfection was studied using neutron and gamma-ray diffraction rocking curves. Most of the gamma-ray data were obtained using a high intensity source of $^{153}Sm$ gamma rays with a wavelength of $\lambda = 0.12$ Å. Some of the data were obtained using highly penetrating $^{198}Au$ gamma rays with a shorter wavelength of $\lambda = 0.03$ Å. The neutrons had a wavelength of $\lambda = 1.07$ Å. It was found that, in terms of the mosaic spread of the crystals, the cleaved detector plates have a much lower crystalline perfection than the thicker uncleaved detector plates. At the same time, the results show that for detectors cut from the same crystal, the one with the lower spectral resolution for radiation detection will also have a lower perfection and larger width of the gamma-ray rocking curve. These results suggest consideration should be given to alternative fabrication procedures for $HgI_2$ nuclear radiation detectors.

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## INTRODUCTION

Mercuric iodide ($HgI_2$) single crystals are used as room-temperature gamma radiation and x-ray semiconductor detectors.[1–3] The charge transport properties of $HgI_2$ have been thoroughly studied and the literature mentions some excellent[4,5] and some very poor[6] charge transport properties. The trapping mechanism of $HgI_2$ is also discussed in the literature[7] and a more recent study[8] reports an updated list of the trapping levels present in $HgI_2$ as determined by thermally stimulated current (TSC), photoconductivity, and photoluminescence measurements.

Poor charge transport properties may be caused by either unwanted traps (particularly hole traps) present in the bulk of the crystal after completion of the growth process, or by traps that stem from a faulty fabrication process.[9] The method by which nuclear radiation detectors of $HgI_2$ are produced can be summarized as follows: The crystals are usually grown from the vapor phase[10,11] but sometimes from solutions.[12] The present paper describes the perfection of only vapor grown crystals grown either in a vertical[10] or horizontal[11] furnace. The crystals are manufactured into nuclear detectors by solution wire sawing and cutting into blocks with a square section of about $10 \times 10$ mm$^2$. The blocks are handcleaved or sawn into 0.5-mm-thick plates which are partially coated with a carbon layer made from Aquadag.[9] Alternatively, the plates are covered with evaporated palladium or platinum contacts.[6] Thin palladium wires are then attached to the contact layer and the detector is subsequently encapsulated with an acrylic protective layer.

$HgI_2$ is a tetragonal layer-like compound with relatively strong coupling within the layers and quite weak coupling between the layers. If one therefore assumes that the degree of crystal perfection within the crystalline layers is quite high, there are two significantly different types of crystalline imperfection which may exist in $HgI_2$. One type of disorder corresponds to random rotations of successive layers about the direction perpendicular to the plane, i.e., observed for crystal rotations about the [001] direction (referred to as twists). The second form of disorder corresponds to a random tilting of the normal to the planes (referred to as layer tilts). Layer tilts are measured by studying the 00*l* reflection or other reflections by rotating about any direction perpendicular to [001]. If the tilting is not random, but preferential about some axis, then the mosiac about different rotation axes perpendicular to [001] will be different. This may be related to anisotropy in the growth process or to mechanical treatment.

Both neutron diffraction and gamma-ray diffraction rocking curves have been used in the past to study the mosaic spread of crystals which are used as monochromators for neutrons.[13–17] The present paper describes the results of neutron diffraction and 0.03-Å gamma-ray diffraction on one

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cleaved piece of $HgI_2$ and the results of extensive experiments on a variety of cleaved, uncleaved and solution sawn crystals with 0.12-Å gamma-rays at the University of Missouri Research Reactor.

## DESCRIPTION OF INVESTIGATED SAMPLES

A cleaved section from a crystal S-4-19, $8 \times 10 \times 1$ mm grown in a vertical furnace was investigated by both neutron diffraction at the research reactor RISO, and gamma diffraction at the Hahn-Meitner Institut, Berlin. Two crystals, one unsawn (H-3-2) and one wire sawn (S-2-17) were studied by gamma-ray diffraction at the University of Missouri (MURR). Since it was seen that cleaved detectors had lower perfection than the crystals from which they were cleaved, two sections of the S-2-17 crystal were cleaved and studied. Seven fabricated detectors were studied at MURR by gamma diffraction. Five of these were cleaved from large crystals including the H-3-2 and S-2-17 crystals while two were solution sawed. The spectral resolution of these detectors varied from grade B to grade D where grades B, C, and D mean that the spectral resolution, expressed as the full width half maximum (FWHM) of the $HgI_2$ detector to the 662-keV photopeak of $^{137}Cs$, is ~8% (B), ~12% (C), or greater than 15% (D).

## EXPERIMENTAL

Characterization of crystal perfection with neutron diffraction was done at the RISO nuclear reactor in Denmark. The experimental setup is shown in Fig. 1.

The mosaic spread of the monochromator crystal $\eta_m$ and the collimators $\alpha_0, \alpha_1, \alpha_2$ (which may depend on sample size), determine both the monochromaticity and the divergence of the incident beam. The width of the neutron rocking curve depends on these parameters, the crystal mosaic, and the Bragg angle $\theta$ in a complicated fashion[18] and only if the sample mosaic is large can it be unfolded reliably from these other effects. A computer program written by R. G. Hazell of Aarhus University has been used to fold the mosaic with the other (known) parameters to approach the observed curves.

Characterization with gamma-ray diffraction was carried out at both the Hahn-Meitner Institute (Berlin) at 0.03 Å and at the University of Missouri Research Reactor (MURR) with 0.12-Å gamma rays. Since the gamma-ray beam is initially highly monochromatic,the resolution for rocking curves depends only on the collimation which can be adjusted to match the sample characteristics.

The Hahn-Meitner instrument uses the 0.03-Å gamma-rays emitted from an $^{198}Au$ source. This wavelength is advantageous for minimizing attenuation due to absorption, and in fact x-ray diffraction would only probe the surface of the $HgI_2$ samples. The small scattering power for this radiation makes it less than ideal for studying thin samples such as the ~500-$\mu$m-thick $HgI_2$ crystals fabricated into nuclear radiation detectors.

The design of the MURR gamma-ray instrument is shown in Fig. 2. This uses a $^{153}Sm$ source, which while less penetrating than the $^{198}Au$ radiation, give greater intensity for thin samples, and the intrinsic Ge solid-state detector gives an improved signal-to-noise ratio.[19,20] The source-sample distance is variable as is the source collimation. For high-resolution studies, the source-sample distance was approximately 2 m and the source collimation had a 0.25-mm exit slit width for an angular divergence of 0.007°. For relatively imperfect samples the resolution was relaxed by opening the exit slit width to 2.5 mm which resulted in higher diffracted intensities. The samples were mounted on the full circle goniometer so that a series of reflections corresponding to rota-

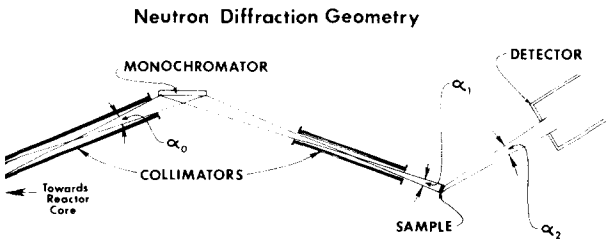


FIG. 1. Schematic representation of the neutron diffractometer in RISO, Denmark. The wavelength used for this study was $\lambda = 1.07$ Å.

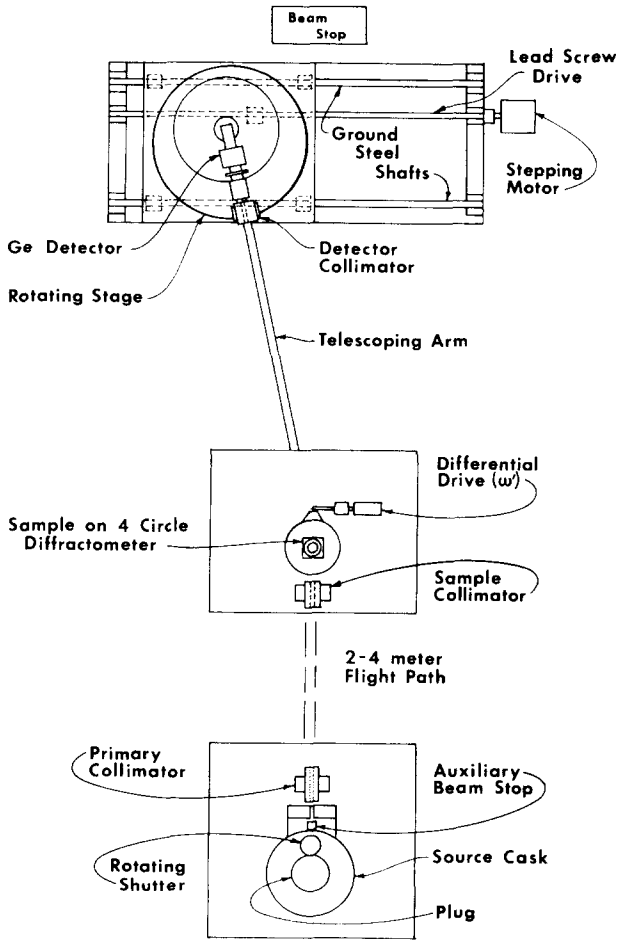


FIG. 2. Schematic representation of the gamma-ray diffractometer used at the University of Missouri Research Reactor (MURR) which uses a $^{153}Sm$ source ($\lambda = 0.12$ Å).

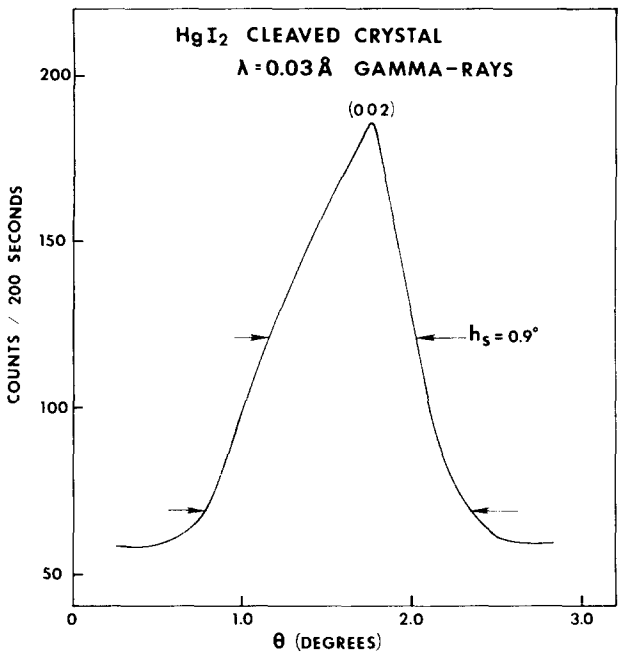


FIG. 3. Typical gamma-ray diffraction rocking curve performed in Berlin on the 002 reflection taken with a $^{198}$Au source, $\lambda = 0.03$ Å. The mosaic for layer tilt is $h_s = 0.9°$ compared to $h_s = 1.5$ obtained for neutron diffraction on the same sample.

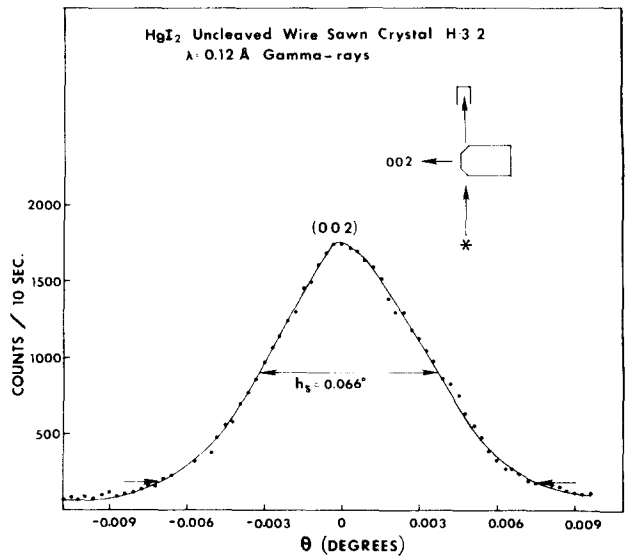


FIG. 4. High-resolution rocking curve for the 002 reflection at the top of the H-3-2 crystal with $\lambda = 0.12$ Å showing the layer tilt mosaic.

tions about different axes could be investigated. In addition, the samples were mounted on a gonimeter with translations which enabled us to examine different locations on the sample in order to determine the sample homogeneity.

For the most perfect samples with sharp mosaic structure and the highest instrumental resolution, count rates up to 200 cps were achieved. Less perfect crystals, even with relaxed collimation, gave as little as 20 cps at the maximum.

The anisotropic structure of $HgI_2$ was found to result in distinctly different mosaic spreads for the tilt and twist mo-



TABLE I. Summary of results of crystal perfection for samples studied with 0.12-Å gamma rays.

| Sample characteristics | | | | | Layer tilt | | | Twist Width | | |
|---|---|---|---|---|---|---|---|---|---|---|
| Crystal | Detector | Thickness (μm) | Grade | Reflections | Number of peaks | Total width at 10% max | Width-main peak at 50% max | Number of peaks | Total width at 10% max | Main peak at 50% max |
| H-3-2 | CN4 | 645 | C | 200 | 2 | 1.5 | 0.75 | 1 | 0.14 | 0.09 |
| | | | | 020 | 3 | 0.56 | 0.12 | | | |
| H-3-2 | CN4 | 565 | C | 200[a] | 4 | 1.2 | 0.24 | | | |
| | | | | 200[a] | 10 | 3.0 | | | | |
| | | | | 020[a] | 3 | 0.56 | 0.10 | | | |
| | | | | 020[a] | 4 | 1.5 | 0.24 | | | |
| H-3-2 | Unsawn | crystal | | 200 | | | | 1 | 0.14 | 0.05 |
| | | | | 220 | | | | 1 | 0.13 | 0.05 |
| | | | | 002 | 1 | 0.15 | 0.06 | | | |
| S-2-17 | CN4 | 535 | B | 200[a] | 3 | 0.80 | 0.24 | 1 | 0.18 | 0.13 |
| | | | | 200[a] | 2 | 0.40 | 0.14 | | | |
| | | | | 020[a] | 6 | 1.5 | 0.32 | | | |
| | | | | 020[a] | 2 | 0.48 | 0.10 | | | |
| S-2-17 | CN10 | 453 | D | 200 | 15 | 5.5 | 4.5 | 2 | 0.73 | 0.22 |
| | | | | 020 | 6 | 4.5 | 1.7 | | | |
| S-2-17 | Sawn | crystal | | 200 | | | | 1 | 0.12 | 0.08 |
| | | | | 020 | | | | 1 | 0.12 | 0.09 |
| S-2-17 | Thin | cleavage | | 200 | 3 | 0.80 | 0.16 | | | |
| S-2-17 | Thick | cleavage | | 200 | 7 | 5.0 | 0.24 | | | |
| S-1-14 | CD5 | 490 | D | 200 | 15 | 3.0 | 1.5 | | | |
| | | | | 020 | 12 | 3.8 | 2.0 | | | |
| S-8-10 | | 650 | C+ | 200[a] | 1 | 0.02 | 0.035 | | | |
| | | | | 200[a] | 1 | 0.033 | 0.05 | | | |
| | | | | 020[a] | 1 | 0.04 | 0.09 | | | |
| | | | | 020[a] | 1 | 0.04 | 0.065 | | | |
| S-8-10 | | 670 | C | 200 | 3 | 0.055 | 01.4 | | | |
| | | | | 020 | 1 | 0.035 | 0.085 | | | |

[a]Measurement of two positions for the same reflection, i.e., crystal translation.

saics discussed in the Introduction. However, the geometry of the samples did not allow us to characterize the tilt and twist mosaics for all samples investigated. The trends seen for one type appear, however, to be reflected by the other.

## RESULTS

A cleaved section of the S-4-19 crystal was studied both with neutrons and with gamma rays. Two neutron measurements for tilt (116,118) and two for twists (020,220) were performed. Since the observed widths were large the mosaic spread could be reported with some reliablity as follows:

$$\left.\begin{matrix}006\\008\end{matrix}\right\} \quad h_s = 1.5°, \qquad \left.\begin{matrix}020\\220\end{matrix}\right\} \quad h_s = 2.9°,$$

where $h_s$ is the full width at half maximum. Since the neutron beam bathes the entire sample, these values are global averages. These results are quite surprising in that all our other measurements give tilt mosaics larger than twists, and the reason for this difference is not yet understood.

The same sample, studied with 0.03-Å gamma rays gave a smaller value for $h_s$ than seen with neutrons (Fig. 3),

$$002: \quad h_s = 0.86°, \quad 102: \quad h_s = 0.9°.$$

The difference between the two techniques is most likely due to the fact that the gamma-ray beam irradiates only a small volume of the sample while the neutron beam envelopes the entire speciment. Any sample curvature would give a broader line in the neutron case.

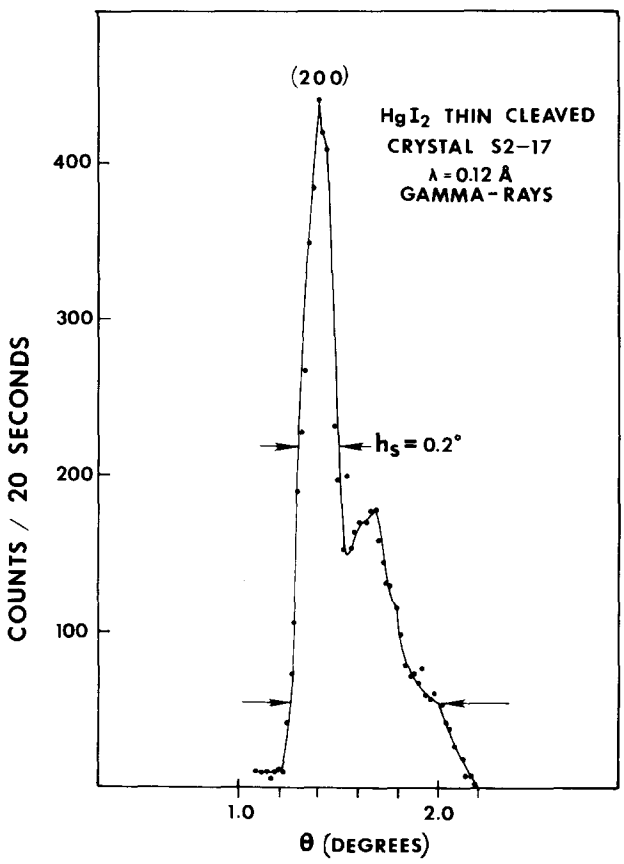


FIG. 5. Rocking curve of a cleaved section of the S-2-17 sawn crystal showing the layer tilt mosaic for the 200 reflection.

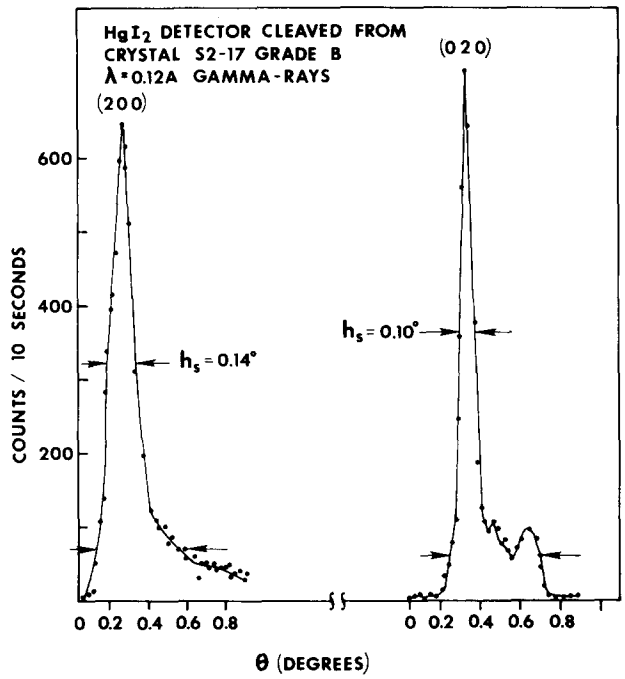


FIG. 6. Rocking curves for a Grade B detector fabricated from the S-2-17 crystal, showing layer tilt mosaic for the 200 and 020 reflections.

The results of the remaining studies with the 0.12-Å gamma rays are summarized in Table I. Certain systematic effects can be observed in these results.

(1) The two crystals studied, H-3-2 and S-2-17, show relatively high degree of perfection; $h_s = 0.05°$ and 0.08° respectively for twist. The tilt mosaic for H-3-2 (Fig. 4) is slightly broader than the twist, a trend which is common through these measurements. The perfection of the S-2-17 crystal which is sawn appears to be less than that of the unsawn H-3-2 crystal.

(2) The perfection of the two pieces cleaved from the S-2-17 crystal for tilts is much lower than the uncleaved H-3-2 crystal. We have reason to believe that during the cleaving, hard dislocations are encountered which create damage and reduce the crystalline perfection (Fig. 5).

(3) There is a clear correlation between crystal perfection and detector performance for detector fabricated in the same manner. For example, two detectors fabricated from S-2-17, with B grade and D grade spectral resolution (Figs. 6 and 7) have vastly different mosaic spreads, with the better resolution corresponding to higher perfection.

(4) The possibility of traps other than those due to defects must not be neglected. Detectors fabricated from the S-8-10 crystal (Fig. 8) show the highest degree of perfection and were fabricated by solution wire sawing rather then by cleaving to eliminate the effects of damage during the cleaving process. Apparently, surface traps have been introduced which reduce the performance of the detectors. These traps can possibly be eliminated through the use of a different solution. Nevertheless between the two detectors fabricated from S-8-10 the correlation between perfection and performance is observed.

(5) On the basis of these studies, it appears likely that all

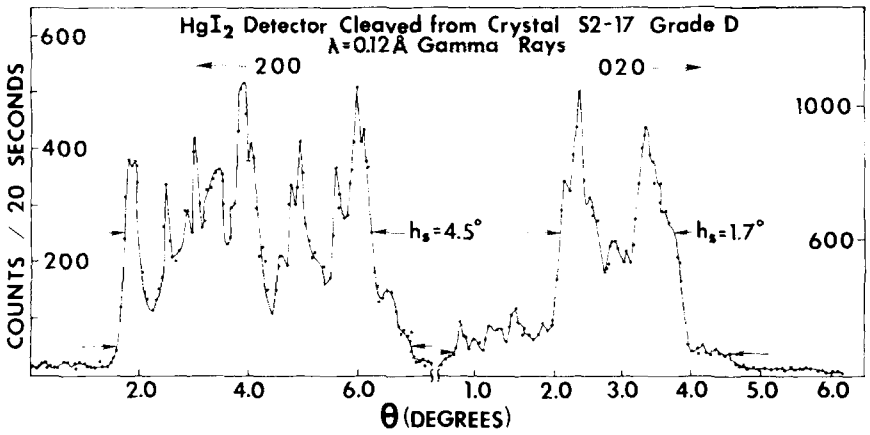


FIG. 7. Rocking curves for a Grade D detector fabricated from the same crystal (S-2-17) showing the much broader layer tilt mosaic for the 200 and 020 reflections.

A grade detectors will show a high degree of perfection. However, all perfect crystals may not give A grade detectors due to intrinsic traps. This technique will, at least, permit us to separate the intrinsic and extrinsic effects and ultimately to determine the optimum conditions for both crystal growth and detector fabrication.

## CONCLUSION

Gamma-ray diffraction rocking curves are a powerful method to characterize the perfection of single crystals of $HgI_2$, which, due to their high $Z$ cannot easily be studied by x-ray rocking curves. This study shows that the present fabrication procedure used to prepare detectors from the grown

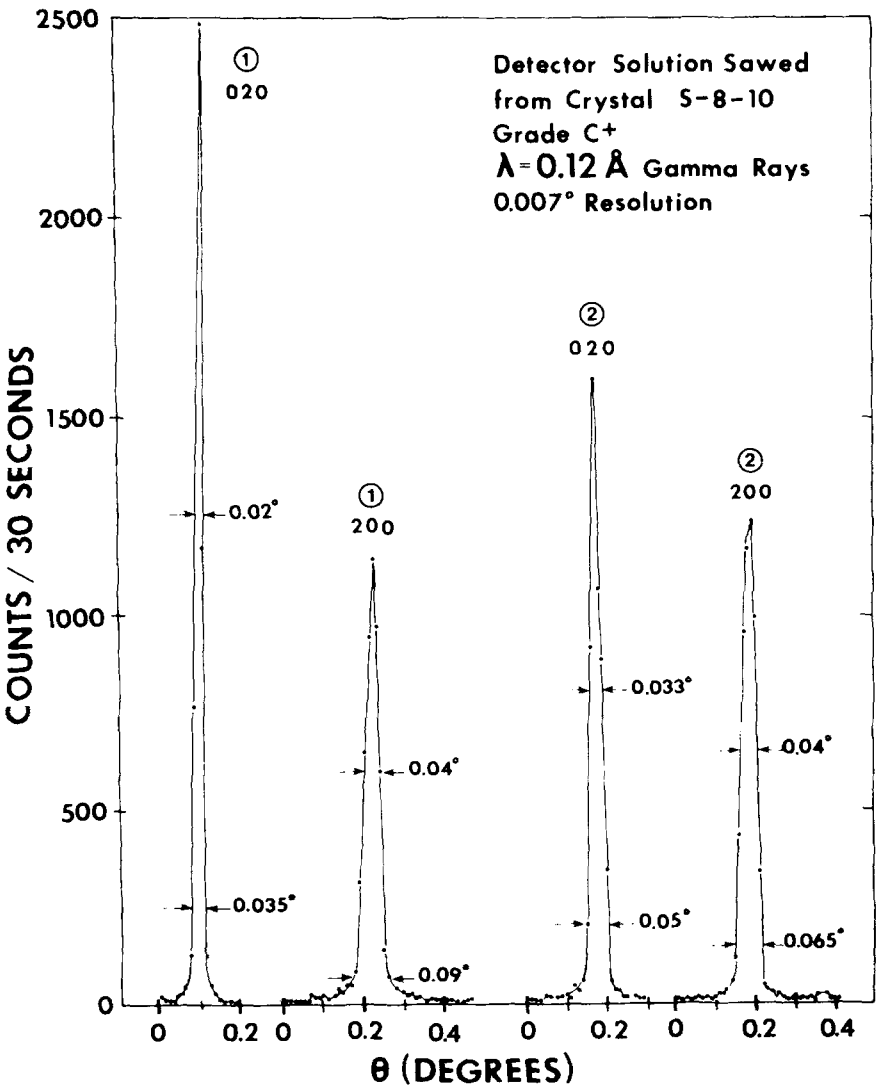


FIG. 8. High resolution rocking curves of the solution sawn S-8-10 detector, $C^+$ grade for two positions on the sample showing the layer tilt mosaic for the 020 and 200 reflections.

crystals should be reevaluated, since it was found that the cleavage process deteriorates the perfection of the single crystals while solution sawing preserves the quality to a considerable degree. It was also shown that there is a correlation between the nuclear spectroscopic resolution of the detector and the width of the gamma-ray rocking curve, i.e., the lower the perfection, the poorer the spectral response of the gamma radiation detector.

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