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**Abstract**

**Full Text**

**Physics**

**E. V. Kolontsova, I. V. Telegina**

## **Radiation Disturbances in Quartz**

*(Presented by Academician G. V. Kurdyumov, 6 VI 1962)*

A change in the crystal structure of quartz, observed by the method of anomalous scattering of X-rays, is manifested first of all in a change in the ratio of intensities between the Laue and diffuse maxima: at a value of the integrated flux of about  $10^{19}$  n/cm<sup>2</sup>, an enhancement of the diffuse maxima becomes clearly noticeable (<sup>1</sup>); no other changes are found in quartz irradiated with such a dose. With an increase in the integrated flux to  $7 \cdot 10^{19}$  n/cm<sup>2</sup>, a substantial change in the diffraction pattern as a whole is observed (see Figs. 1 and 2). This is manifested: a) in a change in the symmetry of the diffraction pattern: instead of the third-order symmetry axis characteristic of  $\alpha$ -quartz, a sixth-order symmetry axis appears; b) in the appearance, in the angular interval from  $\vartheta = 6^\circ$  to  $\vartheta = 17^\circ$ , of diffuse "halo" rings characteristic of the scattering of X-rays by amorphous substances; c) in a sharp weakening of the Laue maxima (up to the complete disappearance of some of them) and an enhancement of the diffuse maxima; d) in a restriction of the interference field (cf. Figs. 1a and 1b); e) in a qualitative change in the pattern of diffuse scattering in accordance with the change in symmetry ( $C_3$  to  $C_6$ ) and in the appearance of certain features of a fundamental character, for example, a change in the contours of the "connecting bridges," i.e., a change in the distribution of the intensity of diffuse scattering in the intervals between diffuse maxima.

As is known, diffuse (thermal) scattering or, in the general case, anomalous scattering of X-rays is caused by deviations of atoms from positions of stable equilibrium (for example, as a result of thermal vibrations or as a result of some action on the crystal). The complete distribution of intensity in this case is determined by the crystal structure, the elastic characteristics, and the defect structure of the crystal. The increase in the intensity of diffuse maxima observed under the action of irradiation by neutrons (<sup>1,3-6</sup>) and X-rays (<sup>7</sup>), or upon deformation of crystals (<sup>8</sup>), is characteristic of the majority of metallic and ionic crystals. In the case where the shape of the diffuse maxima does not change, as occurs at the initial stage of irradiation of quartz by neutrons (<sup>1</sup>), the enhancement of diffuse scattering is associated with the appearance of point-type defects statistically disordered in the volume; by these are meant not only isolated vacancies and interstitial atoms, but also their clusters, which have small dimensions in three dimensions. Apparently, as applied to quartz, it is appropriate to relate the increase in the intensity of the diffuse pattern

Figure 1

Figure 1: Figure 1

Figure 2

Figure 2: Figure 2

to the destruction of the crystal structure, observed with obviousness at large irradiation doses. This is also manifested in the appearance of diffuse “halo” rings on radiographs irradiated with about  $7 \cdot 10^{19}$  n/cm<sup>2</sup> (see Figs. 1 and 2, and also Fig. 5 in work <sup>(3)</sup>), and in the sharp weakening of the interference maxima, and in the restriction of the interference field. At the same time, the change in symmetry, in our opinion, indicates a possible mode of rearrangement of the initial structure.

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Fig. 1. X-ray photographs obtained from quartz before (a) and after (b) irradiation with neutrons,  $7 \cdot 10^{19}$  n/cm<sup>2</sup>. Mixed Mo radiation. The beam is parallel to the [001] direction.

Fig. 2. Same as in Fig. 1 (monochromatic Mo radiation).

As is known, the presence of a sixth-order symmetry axis characterizes the structure of the high-temperature (above 575°) modification of quartz ( $\beta$ -quartz) and the structure of the so-called Dauphiné twin <sup>(9)</sup>. The latter can be obtained in  $\alpha$ -quartz at ordinary temperatures under the action of mechanical loading. On the basis of our data it is impossible to determine unambiguously what takes place under neutron irradiation of quartz—a  $\beta$ -transition or reorientation according to the twin law. However, the proposed detailed comparison of the intensity of the diffraction pattern in  $\beta$ -quartz, in the twin, and in  $\alpha$ -quartz irradiated at a lower dose will perhaps make it possible to make a more definite choice. This is of interest for checking the presence, in neutron-irradiated quartz, of regions with an amorphous, noncrystalline structure <sup>(10, 11)</sup>. If the transition proceeds according to the twin law, then, most probably, the conditions for this transition are created at the boundary of such amorphized regions because of the difference in density between crystalline and amorphous quartz. The transition  $\alpha \rightarrow \beta$  under the action of neutron irradiation at relatively low temperatures (about 100–200°) also appears possible to us, since, owing to the rupture and weakening of silicon-oxygen bonds <sup>(12, 13)</sup>, displacement and a change in the short-range order in the arrangement of Si–O tetrahedra\* may occur; all the more so because there are indications of a lowering of the transition temperature (by more than 100°) with an increase in the concentration of defects in the crystal—for example, the lowering of the temperature of the quartz  $\alpha \rightarrow \beta$  transition in the presence of impurity atoms of a definite type, noted in <sup>(14)</sup>. In this respect the complete destruction of the crystalline structure, observed in quartz at values of the total flux of about  $2 \cdot 10^{20}$  n/cm<sup>2</sup> <sup>(3, 15)</sup>, can be understood as the

rupture of a covalent bond with disturbance of the orientational correspondence in the arrangement of Si—O tetrahedra.

In analyzing changes in the bond in neutron-irradiated quartz crystals it is also advisable to take into account the qualitative change in diffuse scattering, recorded, as indicated above, at relatively large values of the total neutron flux. An attempt will be made subsequently to interpret this change.

The study of radiation-induced structural changes in quartz is being carried out in parallel with work, conducted under the direction of V. G. Zubov at the Department of General Physics of the Physics Faculty of Moscow State University, on the investigation of the physical properties of quartz.

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\* In such compounds as  $\text{BaTiO}_3$ ,  $\text{KNbO}_3$ , and  $\text{PbTiO}_3$ , a transition to high-temperature modifications was observed during neutron irradiation at a temperature of about  $85^\circ$  (<sup>5</sup>), whereas in unirradiated crystals the transition temperature is  $200\text{--}400^\circ$ .

*Note: Figure translations are in progress. See original paper for figures.*

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