Light scattering and singularities of the crystal structure of quartz at its phase-transition point

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Results are described of an investigation of a new phenomenon—the formation of a macroscopic phase structure at the α–β phase transition point of quartz crystals. The results are of importance for the elucidation of the mechanism of phase transitions in crystals.

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The anomalous light scattering at the α-β phase transition point of crystalline quartz was first observed in 1956 by I. A. Yakovlev, T. S. Velichkina, and L. F. Mikheeva [1]. This result was later confirmed in a number of studies.[2,3] However, the nature of the anomalous scattering of light at the phase-transition point is still under discussion.

To determine the character of the crystal inhomogeneities that are produced at its phase transition point and lead to so strong an increase in the intensity of the scattered light, we have undertaken an investigation of light scattering by quartz single crystals at small angles (several degrees). The experimental setup made it possible to measure simultaneously the intensity of the scattered light, to determine the pattern of the Fraunhofer diffraction, and to photograph the image of the investigated sample by the Toepler method.

Figure 1 shows the schematic diagram of the setup. A light beam from an He-Ne laser was focused by a long-focus lens O_1 ($f=80$ cm) into the investigated quartz sample Q. The scattered light was incident on objective O_2 ($f=9$ cm) in the front of which was placed a round screen S (2 mm diameter) which blocked the direct light beam. Part of the scattered light was reflected by the beam splitting plate SP_1 and the focal plane of the objective O_2 was photographed through lens O_3 on film in photographic camera PC_1. Camera PC_2 photographed the magnified image of the crystal.

The relative intensity of the scattered light was measured with a photomultiplier.

We used for the measurements samples of natural and synthetic quartz in the form of disks of 5 mm diameter and from 1 to 4 mm thick, cut either perpendicular or parallel to the optical axis of the crystal. The investigated samples were freely placed in an over whose construction is clear from Fig. 1 (*A*–ceramic, *B*–steel body of the over, *C*–platinum heating coil).

Our experiments have shown that in a narrow temperature region (0.5°) near the crystal phase transition point relatively large phase inhomogeneities are produced in the crystal and lead to strong scattering of the light, particularly at small angles. Figure 2 shows the Fraunhofer-diffraction pattern for a quartz sample 1 mm thick cut perpendicular to the optical axis of the crystal (the primary light beam propagated

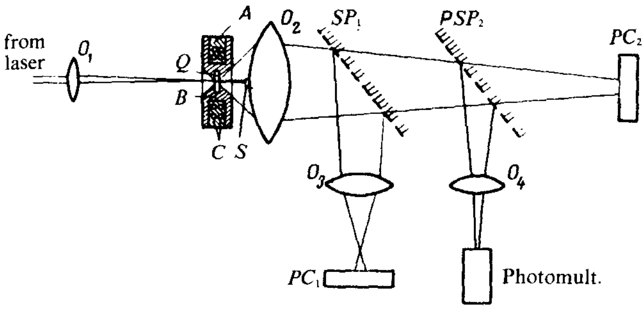

FIG. 1.

along the optical axis. The photograph shows the diffraction rings corresponding to diffraction by randomly disposed inhomogeneities. Figure 2b shows the picture of the phase inhomogeneities produced inside the crystal, obtained by the Toepler method. On the other hand, if the optical axis of the crystal makes a certain angle with the propagation direction of the primary light beam, then the diffraction pattern takes the form shown in Fig. 2c. This picture corresponds to diffraction by phase inhomogeneities that are elongated in the direction of the optical axis of the crystal. This conclusion is confirmed also by the photographs shown in Fig. 3 for a sample cut parallel to the optical axis of the crystal (the incident light is perpendicular to the optical axis). Figure 3a shows the Fraunhofer diffraction pattern, which consists of a system of diffraction peaks arranged along a straight line perpendicular to the optical axis of the crystal. In some cases the diffraction pattern is more or less periodic, and in others there is no periodicity. Figure 3b shows a photograph of the structure of the crystal for this case.

Measurements of the intensity of the scattered (diffracted) light have shown that when the incident light beam propagates perpendicular to the optical axis of the crystal the maximum intensity of the scattered light is 10^{-5} of the incident intensity (for a crystal 2 mm thick) in the scattering-angle interval from 0.7 to 8°. On the other hand, if the incident light beam propagates along the optical axis, then the ratio is much larger and reaches 10^{-3} for a sample 1 mm thick in the scattering-angle interval from 0.7 to 4.5°. It must be noted that the β-α transition is accompanied by the onset of more abrupt inhomogeneities of the crystal than the α-β transition, producing in the former case a much stronger increase of the intensity of the diffracted light than in the latter case. The data presented above pertain to the β-α transition.

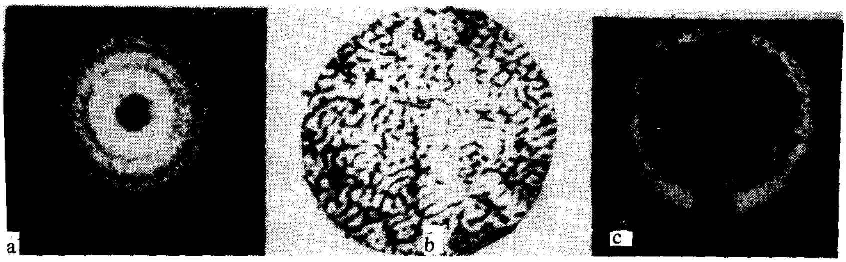

FIG. 2.

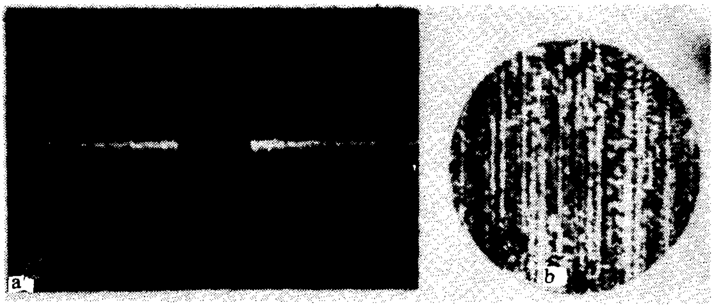

FIG. 3.

Thus, a crystalline state characterized by very abrupt stationary inhomogeneities of the refractive index exists in a narrow temperature interval near the phase-transition point of the quartz crystal. Under the conditions of our experiment this state extends over the entire crystal sample. The resultant inhomogeneities cause a strong scattering of the light, especially at small scattering angles. However, a structure of this type was observed already in 1977 prior to our experiments by T. S. Velichkina at a scattering angle 90°. The results of our diffraction measurements have shown that the inhomogeneities consitute small posts with average transverse dimension 0.03 mm, having a diffractive index that differs from the surrounding medium and elongated in the direction of the optical axis of the crystal.

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[2]S. M. Shapiro, The Johns Hopkins Univ., 1969 [sic!]
[3]The Theory of Light Scattering in Solids, Proc. First Soviet-Americ. Symp. Moscow, 1975, Nauka.