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V. G. ZUBOV and L. P. OSIPOVA

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V. G. ZUBOV and L. P. OSIPOVA

RAMAN SCATTERING IN α-QUARTZ IRRADIATED WITH FAST NEUTRONS

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1. In the present work we set forth the results of an investigation of the Raman-scattering spectrum of a quartz single crystal irradiated with fast neutrons. We have not been able to find other works devoted to this question.

The specimen studied had the form of a cube $30 \times 30 \times 30$ mm, oriented along the principal axes and cut from a single block of collection Brazilian quartz. The results of an investigation of the Raman-scattering spectrum of this specimen before irradiation are given in ([1]). After irradiation with an integral dose of fast neutrons of $1 \cdot 10$ n/cm^2, the density of the quartz decreased from 2.65 to 2.49 g/cm^3. The specimen retained residual γ-activity, acquired a light-violet coloration, and began to absorb approximately 50% of the light incident on it in the interval 5800–4000 Å. A strong fluorescence arose in the irradiated specimen, the maximum of which corresponded to λ 5750 Å. In the region 5750 –4000 Å the fluorescence intensity decreased monotonically and produced a rather significant continuous background.

2. The Raman-scattering spectrum of irradiated quartz was studied by the photographic method with an ISP-51 spectrograph and by the photoelectric method with a reconstructed DFS-4 spectrometer. A description of the latter apparatus is given in ([1]). The spectra obtained were compared with the corresponding spectra of unirradiated crystalline and fused quartz.
3. Photographically it proved possible to determine only the frequencies of the strongest diffuse maxima (see Table 1). It was not possible to estimate the intensity and width of these maxima from the spectrograms because of the strong general background.

Table 1

Frequencies of the Raman-scattering spectra (in cm^{-1})

Unirradiated α-quartz	Irradiated α-quartz, photographic method	Irradiated α-quartz, photoelectric method	Fused quartz	Unirradiated α-quartz	Irradiated α-quartz, photographic method	Irradiated α-quartz, photoelectric method	Fused quartz
–	–	–	–	–	750	740	760
128	–	–	–	795–805	–	–	–
–	170	–	–	–	830	820	–
206	–	–	–	–	930	950	970
–	240	230	–	–	–	1010	–
266	–	–	–	–	1050	–	1070
–	290	300	–	1063	–	–	–
357	–	–	–	1081	–	–	–
–	400	380	–	–	1100	1100	–
403	–	–	–	1159	1180	1170	1170
466	480	470	430	1228	1260	–	1260
–	540	550	600	–	1350	1320	–
696	–	–	700				

The photoelectric method made it possible to record, with greater reliability, the entire Raman-scattering spectrum of the irradiated quartz up to frequencies of 1500 cm^{-1}. Figure 1 shows the spectra we obtained for irradiated crystalline quartz and for unirradiated fused quartz. The Raman lines of the spectrum of unirradiated α-quartz are also plotted there.

It is seen from Fig. 1 that the Raman scattering in irradiated quartz has the character of a continuous spectrum extending approximately to 1500 cm^{-1} and containing a large number of different maxima (see Table 1). Comparison with the spectrum of unirradiated crystalline quartz shows that a large number of these maxima can be identified with the lines of unirradiated quartz, which, owing to destruction and distortion of the original crystalline structure under the action of irradiation, have somewhat changed their frequency and become strongly broadened. The maxima at 540, 930, 1050, and 1350 cm^{-1} are new, and their origin requires special explanation.

Fig. 1. Raman-scattering spectra of α-quartz irradiated by fast neutrons (1) and of unirradiated fused quartz (2). The vertical strokes indicate the Raman-scattering lines of unirradiated α-quartz, the intensities of which are expressed relative to the line 1159 cm^{-1}. The intensity scale for the lines 128, 206, 266, 357, 403, and 466 cm^{-1} in the plot is reduced by a factor of 20 in comparison with the intensity scale for the remaining lines.

The intensity in the broadened maxima of the spectrum of irradiated quartz is considerably lower than the intensity of the corresponding lines in the spectrum

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Figure 1: Fig. 1. Raman-scattering spectra of α-quartz irradiated by fast neutrons (1) and of unirradiated fused quartz (2). The vertical strokes indicate the Raman-scattering lines of unirradiated α-quartz, the intensities of which are expressed relative to the line at 1159 cm^{-1}. The intensity scale for the lines 128, 206, 266, 357, 403, and 466 cm^{-1} in the plot is reduced by a factor of 20 in comparison with the intensity scale for the other lines.

of the unirradiated crystal. The greatest changes in intensity occur in the region 100—500 cm^{-1}. Thus, the intensity in the maximum corresponding to the line 466 cm^{-1} is approximately 20-30 times smaller than the intensity of this line in the spectrum of the unirradiated specimen. Comparison of the spectra of irradiated and fused quartz shows that in the region 900—1500 cm^{-1} the character of the scattering in these specimens is practically the same, whereas in the region 100—500 cm^{-1} these spectra differ sharply both in their general appearance and in the number and positions of the maxima. The spectrum of irradiated quartz in this region is more intense and richer in its structure.

4. Comparison of the Raman-scattering spectra in measurements along directions coinciding with the optical axis Z of unirradiated quartz and along the second-order axes showed that the scattering intensity depends very strongly on the orientation of the irradiated specimen. In the region from 100 to 700 cm^{-1}, the intensity of light scattered along the direction coinciding with the Z axis of the unirradiated crystal proved to be almost two times smaller than the scattering intensity along the second-order axes X and Y. In the region 700—1300 cm^{-1}, the influence of the orientation of the irradiated crystal on the scattering intensity is practically not manifested. Thus, the dependence of the Raman-scattering intensity of light on the orientation of the irradiated specimen proved to be fundamentally different from that for natural crystalline α-quartz ([1,2]).

5. A comparison of Raman scattering in the Stokes and anti-Stokes regions showed that, in the interval from 200 to 500 cm^{-1}, the intensity of Raman scattering in the anti-Stokes region is anomalously large in comparison with the intensity in the Stokes region. Table 2 gives the experimental values of I_{ast}/I_{st} and the theoretical values of this ratio calculated from (3).

Table 2

$\Delta\nu$, cm^{-1}	220	300	400	466	500
I_{ast}/I_{st}, exp.	0.5	0.45	0.30	0.15	0.09
I_{ast}/I_{st}, theor., $T =$ 300°K	0.35	0.24	0.15	0.11	0.09

For control, the ratio I_{ast}/I_{st} was measured for the 466 cm^{-1} line in the spectrum of unirradiated quartz. Within the limits of measurement error, the experimental value coincided with the theoretically calculated one.

6. The fundamentally different dependence found for the scattering intensity on the orientation of the irradiated quartz single crystal indicates the existence of a certain anisotropic state, substantially different in its properties both from crystalline and from fused quartz.

The anomalous increase in scattering intensity in the anti-Stokes region is possibly connected with the fact that, after irradiation, the sample began to absorb approximately 50% of the light incident upon it.

Moscow State University
named after M. V. Lomonosov

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CITED LITERATURE

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