

DEFECT PRODUCTION IN QUARTZ CRYSTALS BY CHARGED PARTICLES WITH VARIOUS ENERGY AND CURRENT DENSITIES

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INTRODUCTION

In the recent years much attention is paid to the investigation of the mechanism of the formation of point defects, structural transformation and other radiation stimulated phenomena in the crystal and amorphous SiO₂ having different types and concentrations of primary structure defects under influence of over- and sub-threshold energies. It is of scientific and practical interest since crystal and amorphous SiO₂ is widely used in the various fields of modern science and technology. Analysis of the results [1,2] on this matter shows that the nature of physical processes leading to the formation stable point defects, structure transformations and other radiation stimulated phenomena in the quartz crystals having complete and defect structure irradiated by different types of radiation with over and sub-threshold energies is not yet established. On the basis of above given account the aim of this work is determined. It will be an investigation of the view of the dependences of formed defects on the current density beam of the protons, deuterons, α -particles with over-threshold energy and electrons with sub-threshold energy, influence of the variation type and concentration increase of the primary defects at the time of radiation on the following process of the defect formation in the quartz crystals. In this connection spectrum of absorption (SA), photo- and gamma-luminescence (PL, GL) nominally pure quartz and quartz crystals with Fe after irradiation was investigated.

EXSPERIMENTAL RESULTS

Investigation of spectrum of absorption was shown that in the SA of the non-radiated samples SiO_2 : Fe absorption bands (AB) are observed with maximums at 220 and 260 nm. Investigation of difference SA of non-radiated and radiated samples has shown that the band 220 nm is superposition of two AB with maximums at 215 and 230 nm. It is established that in the SiO_2 intensity of the bands 220, 248, 260, 350, 540 nm and bands of GL at 470 nm linearly increased as current density beam (CDB) j of the electrons ($E=100, 120$ keV), protons ($E=18$ MeV), deuterons ($E=16$ MeV) and α -particles ($E=18$ MeV) increased (fig. 1). Absorption bands at 215, 248 and 260 nm are induced also in the spectra of nominally pure quartz crystals irradiated.

Investigation of the spectra of PL crystals irradiated by electrons having different j and energy showed that up to $j=30$ mcA/cm² bands of PL are absent. Band of luminescence at 420 nm is appeared beginning with 40 mcA/cm² and bands of PL with maximums at 550 and 660 nm are appeared at 60 and 80 mcA/cm² (fig.2). Comparison spectra of PL crystals irradiated at $j=60$ mcA/cm² shows that the bands of 550 and 660 nm are appeared only at the energies 100, 120 keV. Intensity of bands of PL at 550 and 660 nm in the samples irradiated with the energy 120 keV more than in the samples irradiated with energy 100 keV. Investigation spectra of PL excited at 315 and 337 nm show that at 315 nm of band intensity of 550 nm is more than band intensity of 660 nm. Reverse scene is observed at excitation of PL with 337 nm.

The bands at 550 and 660 nm are observed also in crystals, irradiated by fluencies of protons, deuterons and α -particles $4 \cdot 10^{14} - 10^{15}$ cm⁻². In the crystals, irradiated with the fluencies under the different CDB, the increasing of density of current brings about the growing of intensities of bands 550 and 660 nm.

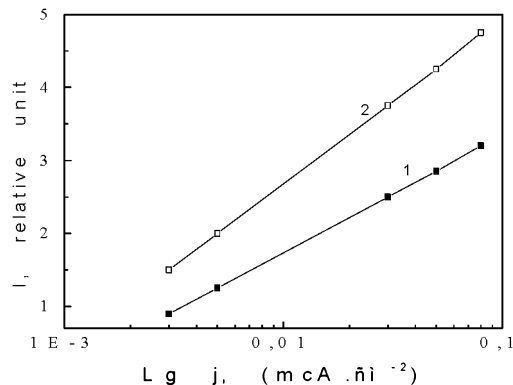


Fig.1. Dependence of GL band at 470 nm in SiO_2 : Fe irradiated by proton fluences of 10^{14} (1) and 10^{15} cm⁻² (2) on the beam current density.

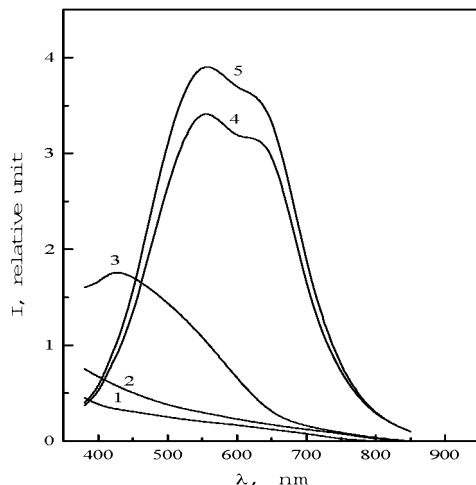


Fig 2. Spectra of PL ($\lambda_B=315$ nm) non-radiated (1) and irradiated by electrons with energy 100 keV at $j= 30$ (2), 40 (3), 60 (4) and 80 mcA/cm² (5) crystals SiO_2 .

DISCUSSION OF RESULTS

It is shown in [3] that in the quartz crystals irradiated by fluency of neutrons $>10^{18}$ n. cm⁻², band of luminescence with maximum at 660 nm, which is excited in the interval 270-280 and 620 nm, is appeared. This band of PL is observed as well as in the quartz glass at the time of excitation in the bands 260 and 620 nm. It is assumed that appearance of the band of 660 nm is caused by luminescence of the nonbridging atoms (NBA) of oxygen stably existing only in the amorphous region. NBA is responsible for the absorption bands at 260 and 620 nm. Concentration increase of NBA of oxygen under neutron irradiation of α -SiO₂ characterizes the degree of amorphization of a sample.

It is established in [1] that the band with the maximum at 660 nm appears also in the spectrum of luminescence of quartz crystals irradiated by fluency of neutrons $5 \cdot 10^{16}$ n.cm⁻². It is assumed that centers of luminescence of the bands are nonbridging oxygen atoms situated in the volume of amorphous phase formed around nucleus of β - phase. Structure of amorphous phase differs from that of quartz glass. Investigation of PL neutron irradiated quartz crystals at different wavelength shows that the intensity of the band of 660nm reaches maximal value at excitation in 315 nm. In this case the band of 550 nm is also excited.

We have detected luminescence band of 550 nm in the γ - irradiated quartz glasses with a big deficient of oxygen which in these experiments regulated by adding silicon powder at the time of glass synthesis in the synthetic raw material vacuum. In this case a brown color appears, which is connected with the inclusions of silicon [4].

It is shown in [1] that in the spectrum of PL of the neutron-irradiated crystals appears bands with maximums at 550 and 840 nm, simultaneously with the band of 660 nm. It is assumed that appearance of the band of 550 nm is connected with the formation of nucleus of β -phase. Centers of luminescence in the band of 550 nm are peroxy radicals being in the volume of the amorphous volume formed around nucleus of β - phase. Occurrence of the band of 840 nm is connected with the formation of silicon nanocrystals (NC).

The band of 550 nm occurs also in the spectrum of luminescence of the different modification SiO₂ irradiated by neutrons [2] and electrons [5]. At the time of investigation luminescence spectra of cathode-luminescence (KL) it is shown in [5] that centers of luminescence in the 550 nm appears after definite values of current density beam of electrons. It is shown that the band of 550 nm is connected by formation of silicon NC. It is assumed that the centers of luminescence are in the border of division of the silicon NC and amorphous SiO₂. The nature centers of luminescence are not discussed.

It is established at present that amorphization of the structure of α - quartz crystals takes place under influence of neutrons [1, 6, 7], ions and electrons with over and sub-threshold energies [see in 8]. It is predominated opinion that vacancy of oxygen is responsible for the formation of nucleus of amorphous phase. It is shown in [9] that the silicon NC is formed in quartz glasses with oxygen deficient under influence of γ -rays. Efficiency of the formation of silicon NC is increased as the degree of the oxygen deficient is increased. In [5] it is established that the value of the current density beam of electrons j , at which the band of 550 nm occurs, is decreased as the density of the irradiated modifications SiO₂. Based on this it can be considered that there exists critical value of the oxygen vacancy concentrations reaching on which formation of the silicon NC takes place.

Investigation was shown that in the crystals spectrum irradiated by electrons with the energy 100 and 120 keV by fluence 10^{16} cm⁻² at $j < 60$ mcA.cm⁻² absorption band of 215 nm and at $j \geq 60$ mcA.cm⁻² absorption bands with 215, 248, 260 nm and luminescence band with 550, 660 are appeared. It is seen that band of the 660 nm is excited at 315 nm. In this case as in case of the neutron-irradiated crystals [1], the band 550 nm is also excited. Absorption band at 248 nm is attributed to neutral oxygen vacancies in the amorphous phase of quartz [7]. This shows that at the time of influence on the quartz crystals by the same fluency of electrons, which are accumulated from j to 60 mcA.cm⁻², E_1' - centers are appeared basically and at $j \geq 60$ mcA.cm⁻²

E_1' - centers and nucleus of the amorphous phase of the quartz containing neutral vacancies and nonbridging oxygen atoms as well as silicon NC are formed.

Perhaps this takes place following way. It is well known that at the beginning of irradiation (for example, at the first seconds of the influence-first stage), when the fluence of electrons 10^{16} cm^{-2} accumulated at $j = 30, 40, 60$ and 80 mcA.cm^{-2} , falling electrons interact with crystal which have complete structure. It is shown in [1] that E_1' -centers are formed in the nominally pure crystals irradiated by fluency of electrons 10^{16} cm^{-2} at different j electrons with energies 100 keV, beginning from $j = 30 \text{ mcA.cm}^{-2}$. Increase of j leads to the increase of concentration of the E_1' -centers formed in the time unit. It is assumed that formation of the E_1' -centers takes place because of the collective decay of two or more interacting electron excitations. In this case according [10] as complementary pair to the E_1' -centers peroxy radicals appears. Therefore, can be accepted that the increase of j leads the increase of the number of peroxide radicals. So, in the first stage of the influence of the fluency of 10^{16} cm^{-2} electrons accumulated at $j=30, 40, 60$ and 80 mcA.cm^{-2} different concentration of defect structure are formed, i.e. crystals have different degree of the defectiveness of the structure. At the second and the next stages of irradiation electrons interacts with defect states of the crystals formed on the previous stages.

Based on the results of investigation influence of the electrons fluency of 10^{16} cm^{-2} accumulated at the same value of j , on the crystals with different structure defectiveness, it is shown in [1] that the concentration of the additional intruded E_1' -centres increase as the structure defectiveness is increased. In the crystals with the same structure defectiveness under fluency of electrons of 10^{16} cm^{-2} with the energy 100 keV but at different values of j concentration of the additional formed E_1' -centres increased as j increased. Based on this we consider that for the each value of j efficiency of formation, defects will be increased as the time of irradiation is increased. Obviously, in each stage of efficiency of formation E_1' -centers and other types of defects are increased as j increases. Therefore, at the time of influence on the crystals of the same fluence 10^{16} cm^{-2} but accumulated at different values of j , concentration of formed E_1' - centers is increased as j of electrons increased. This leads to the increase of the intensity of absorption bands with maximum of at 215 nm.

In the crystals irradiated by fluence of 10^{16} cm^{-2} at $j=60 \text{ mcA.cm}^{-2}$ electrons with the energies of 100 and 120 keV critic concentration of oxygen vacancy necessary for the formation of nucleus of amorphous phase and silicon NC are appeared. This leads to the appearance of the absorption band of 248 and 260 nm and luminescence of 550 and 660 nm. At $j=80 \text{ mcA.cm}^{-2}$ efficiency of the formation of oxygen vacancy in each stage of irradiation is more than 60 mcA.cm^{-2} . Therefore, critic concentration of oxygen necessary for the formation of nucleus of amorphous phase and the silicon crystals appears on the earlier stage of irradiation. In the next stages of irradiation efficiency of formation nucleus of amorphous phase and NC will be increased. Hence, concentration of nucleus of amorphous phase and NC will be more than at $j=60 \text{ mcA.cm}^{-2}$. This causes the increase of the bands luminescence of 550, 660 nm at $j=80 \text{ mcA.cm}^{-2}$.

On the basis of results [1] and the detection of the bands of luminescence at 550 and 660 nm which are caused by peroxy radicals and nonbridging atoms of oxygen, as well as AB of 248 nm attached to neutral vacancy of oxygen being in the regions of amorphous structure of quartz, we consider that in the growing in $\text{SiO}_2\text{:Fe}$ defects of structure are appeared which are in the nucleus of the amorphous phase of quartz. Nucleus of amorphous phase consist of E_1' - centers, neutral vacancy and nonbridging atoms of oxygen. Additional defects are created under influence of electrons. Efficiency of the creation of additional defects is increased as the concentration of preliminary disturbances of structure. It seems formation of amorphous regions are taken place at the expense of thermal peaks.

CONCLUSIONS

1. It is established that after influence fluence 10^{16} cm^{-2} of electrons with energy 100 and 120 keV accumulated at 60 mcA.cm^{-2} in the spectra of absorption and PL of quartz crystals bands connected with the formation nucleus of amorphous phase and silicon NC are appeared. At 80 mcA.cm^{-2} concentration of nucleus of amorphous phase and silicon NC will be more that more than 60 mcA.cm^{-2} .
2. It is assumed that the increase of defectiveness of structures relieves formation of amorphous nuclei in quartz. Oxygen vacancies are responsible for the formation of precipitates of amorphous phase and silicon NC. The critic value for the concentration of oxygen vacancies, when the nuclei form, decreases as the degree of crystal structure defectiveness increases.

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