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Spectral and Angular Characteristics of Fast Proton-Induced Luminescence of Quartz

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Abstract: Experimental studies of the quartz luminescence during high-energy hydrogen ions bombardment are presented. The influence of observation angle and ion energy on shape of light-spectrum was investigated. Two experimental series were carried out as with rigid connection between incident and observation angles as without this one (true indicatrix). It was shown that the optical specter shape depended from ion energy and observation angle. The possibility of application of these results for distant monitoring quartz irradiation processes was proposed.

Key Words: quartz, proton, ion energy, ionoluminescence, spectra, observation angle, indicatrix.

Introduction

An increasingly keen interest in the behavior of some dielectrics under irradiation with charged particles has been shown in recent years. This is of particular urgency for the materials widely used as insulators and windows in thermonuclear facilities [1]. In the process of irradiation many physical properties of materials considerably change, and this may even lead to a functional inadequacy of the element. For example, the windows for input and output of optical radiation become opaque under a long-time irradiation with charged particles [1]. The main reason for the change in the properties of materials under irradiation lies in the formation and dynamics of radiation defects of various types. The defect formation occurs due to the kinetic particle energy dissipated in the substance.

In operation of a thermonuclear reactor, a great many high-energy ions are produced. These are mainly the ions of light elements. The flow of these charged particles exerts an efficient action on the materials being in an immediate contact with thermonuclear plasma. Therefore, the investigation of processes occurring in the substance under irradiation with these particles presents a great interest.

As a fast ion is moving in a solid, there occur the processes of defect formation (single points and cascades), annealing, recombination, etc. Besides, the process of defect formation is accompanied by various attendant phenomena (sputtering of material, electron emission, generation of photons of different energies, including the ones in the optical range).

Luminescence is also an accompaniment of defect formation. The optical radiation arises during the rearrangement of the system under the action of a particle moving in the substance. This rearrangement may manifest itself in an immediate formation of defects or in a generation of quasi-particles that may propagate in the solid. The decay of the quasi-particle may also result in the defect formation. For both channels of particle energy dissipation, the relaxation processes may lead to the electromagnetic wave generation. In both cases, it is the particle that serves as an energy source.

The process of electromagnetic radiation generation is much influenced by the dynamics of defect formation in the medium, particularly, by the spatial distribution of defects of various types and the existing dynamic equilibrium between them. Consequently, the characteristics of luminescence (intensity, spectral distribution, angular distribution, etc.) should depend on the sort and energy of projectiles, on the value of the absorbed dose of radiation, on the distribution of energy losses by the charged particle in the substance.

One of the materials most widely employed for both optical and insulating elements in thermonuclear facilities is quartz. It is also widely used in microelectronics (e.g., as one of the components of MOS), in space vehicles, etc. [7, 8]. Despite all the importance of the issue, the investigations dealing with ionoluminescence of quartz are obviously scarce and they have a fragmentary character (e.g., see refs. [2-5]). Of particular interest is the irradiation of materials with fast protons, the flux of which in a thermonuclear facility is rather substantial [6]. Therefore, a further investigation of optical properties of SiO_2 as it is exposed to ion flows appears to be rather urgent. This paper is concerned with the spectral and angular characteristics of quartz irradiated with protons of MeV energies.

Experiment

In experiments to investigate ionoluminescence we have used a combined setup, the constituents of which have been described in detail in refs. [10,11]. The targets were prepared from a plane-parallel quartz glass plate, 1 mm in thickness. The samples could be rotated around the axis perpendicular to the ion beam. The experiments were carried out with the use of a proton beam of energies ranging from 0.8 MeV to 2.4 MeV, the current density being up to 50 $\mu A/cm^2$. The residual gas pressure did not exceed 10^{-4} Pa and was maintained in the vacuum chamber by means of a magnetic-discharge pump. The optical radiation from the sample surface was transferred to the input slit of the grating monochromator using the optical channel. The direction of observation, the beam axis and the normal to the target surface were lying in the same plane. In experiments, we have used the channels of two types.

An optical channel No.1 has been described in detail in ref. [10]. When it was in use, a quartz condenser onto the input slit of the monochromator projected the luminescent radiation from the sample. The angle between the ion beam axis and the condenser axis was 90° and it remained unchanged in the process of investigations. In this case, the angle of ion beam incidence on the target α, and the angle of sight of radiation from the target, β, were related as:

$$\alpha + \beta = 90^\circ \qquad (1).$$

The angle β could be varied by rotating the target through the range from 20° to 70°. In this case, in accordance with relation (1), the angle α was simultaneously changed. Consequently, the size of the irradiated area of the sample and the current density on its surface also changed. This way of varying the angle of sight was previously used in all known-to-us experimental facilities for measuring angular characteristics of ionoluminiscence (e.g., see ref.[9]). The occurrence of relationship (1) is an essential drawback of these facilities. It considerably restricts the possibilities of investigations and complicates the interpretation of experimental results. At the same time, this optical channel had the maximum capacity and provided the investigation of optical radiation in the spectral range from 250 nm to 700 nm.

With the optical channel No.2 (described in detail in ref.[11]), the light was radiated from the sample was received by the input end of a flexible light guide. The input end of the light guide could be rotated around the sample. A quartz condenser onto the input slit of the monochromator projected the light from the output end of the light guide. In this case, the angles α and β could be set irrespective of each other. This scheme of optical channel made it possible to accept the generated radiation from the entire sample surface under irradiation, irrespective of the angle of beam incidence on the target and the angle of observation. The use of the light guide permitted one to obtain the true indicatrix of luminescent radiation for the arbitrarily given angle α. The range of variation in the angles α and β each was between 0° and 70°. As in the case with the optical channel No.1, the luminescence was also detected with a photoelectric multiplier, but the wavelength ranged here from 400 to 700 nm.

The optical channels of the facility were calibrated against the tungsten spectrometric incandescent lamp. The ionoluminescence spectra were corrected against the spectral sensitivity and were normalized by the beam current.

Experimental results and discussion

The typical luminescence spectra of quartz under bombardment with protons of different energies, obtained with the use of optical channel No. 1, are shown in Fig. 1. Each of them consists of three wide non-symmetric bands, the maxima of which lie in the vicinity of wavelengths of 282 nm, 456 nm and 644 to 648 nm. The spectra of the type shown in the figure are, first of all, characteristic of ionoluminescence [5,11,17]. These bands have also been previously observed in the luminescence spectra of SiO_2 under both the ion bombardment [4-6] and other types of excitation, e.g., with electrons [12, 13], neutrons [14, 15], gamma-quanta [14] or at quartz failure [16]. It should be noted that both the presence of separate bands and the relationship between their intensities depended on the way of luminescence generation.

The scientific literature comprises the following commonly accepted explanations for the existence of these bands:

(i) The ultraviolet radiation with its maximum near 282 nm is considered to be due to the oxygen vacancy [18].

(ii) The radiation in the band with the maximum close to a wavelength of 455 nm is most commonly attributed to intrinsic defects of quartz, namely, to E′ centers [18] (similar optical characteristics are also typical of threefoldcoordinated atoms of silicon [19]). Another popular explanation for the cause of this generation occurrence is the decay of an autolocalized exciton [19].

(iii) The radiation band lying in the red part of the spectrum has the maximum near 644-648 nm. The appearance of this radiation is accounted for by the presence of such defects in the sample as the centers of non-bridged oxygen, those are inherent in quartz [19].

It has been previously shown that the ionoluminescence spectra vary in the process of proton bombardment of quartz. A relationship of radiation intensities has been found for two wavelengths in the spectrum that can serve as a suitable criterion for the control of dose absorbed by the sample [3].

The ionoluminescence spectra obtained in our experiments and in those by other investigators have shown the radiation intensity in the band with the maximum at ~ 456 nm to be substantially higher than in the other two bands (e.g., see [5,11,17]). Unlike other types of excitation, the irradiation with ions is characterized by considerably higher specific ionization losses [20]. In our opinion, in this case, it is the excitation of excitons with their further decay and light generation in the band that may be one of the most probable processes. As the autolocalized exciton is generated, the silicon-oxygen bond becomes destroyed and this gives rise to two quasi-defect centers: threefoldcoordinated silicon and non-bridged oxygen. In the decay of the autolocalized exciton followed by light generation, the transformation of exciton energy by the first quasi-defect into luminescent radiation with the maximum at ~ 456 nm is predominant here, while the efficiency of red band generation due to the second quasi-defect is many factors lower. With an increasing energy of incident ions the radiation intensity increased throughout the spectrum (see Fig. 1).

In the process, the shape of bands remained unchanged, while the intensity at the maxima changed variously. To analyze the spectra, the radiation intensities in the first and third bands were normalized to the maximum intensity that corresponded to a wavelength of 456 nm. This approach appears quite justified in view of the following:

(i) in the measurements of maximum intensity the error was minimum;

(ii) the luminescent radiation with the maximum at $\lambda=456$ nm was observed practically in all the investigations devoted to this problem and known to us.

Figures 2 and 3 show the normalized parts of the spectrum for the bands with the maxima at 282 nm and 648 nm, respectively. It is seen that with an increasing proton energy the variation in the relative light intensity for these bands occurs in different ways: in the ultraviolet region the relative light yield increases, while in the red region it decreases. This fact may evidently be used for remote monitoring of bombarding proton energy. The observed dependence of radiation intensities in different parts of the spectrum indicates that the generation of the major part of power in each of the luminescent band is contributed by different defect centers.

Most often, the dependence of the intensity of light emission from the solid surface on the observation angle is described by the well-known Lambert law $I + I_0 \cdot \cos\beta$, where I_0 is the radiation intensity in the direction perpendicular to the sample surface, β is the observation angle [21]. This law is of geometrical character, i.e., such a dependence is observed at distances substantially exceeding the dimensions of the emitting surface provided that the radiation near the surface is isotropic [22]. We have measured the angular characteristics of the radiation at the maximum of the light-blue ionoluminescent band using the optical channel No.2. The measured dependences are found to be close to the above-mentioned law. Fig. 4 shows these experimental dependences for two energies of bombarding ions; the same figure shows the straight line corresponding to the Lambert law.

The both experimental curves differ from line 1. It seems likely that the radiation received by our detector comes not only from the sample surface but also from the bulk of the solid. As it is obviously seen from the figure, the angular characteristics of ionoluminescence exhibit the dependence on the projectile energy (cf. curves 2 and 3): With an increasing energy the deviation from the Lambert law also increases.

The present results testify that both spectral and angular characteristics of ionoluminescence can be used for monitoring the incident proton energy. The information obtained with the use of the two channels is complementary. To obtain exact quantitative characteristics, further investigations are needed.

Conclusion

As quartz is exposed to a flux of megaelectrovolt protons, the luminescence centers show different efficiency in generating the radiation in the corresponding bands, depending on the particle energy and the prehistory of the sample [3].

The results, obtained previously and presently, demonstrate that the analysis of ionoluminescence spectra provides new possibilities for control over the processes occurring during ion bombardment of solids:

(i) with the use of the relationship between the radiation intensities at certain wavelengths it becomes possible to determine at a distance the dose absorbed by a quartz sample [3];

(ii) the information comprised in spectral and angular characteristics of ionoluminescent radiation that corresponds to the maxima of wide bands, makes it possible to draw conclusions on the energy value of incident protons.

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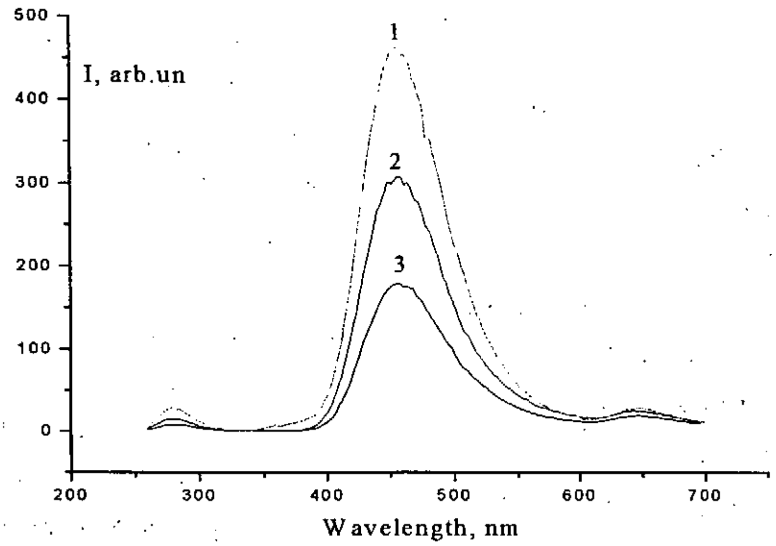

Fig. 1. Luminescence spectra of quartz under bombardment with 2.4 MeV, 1.6 MeV and 0.8 MeV protons (curves 1, 2 and 3, respectively).

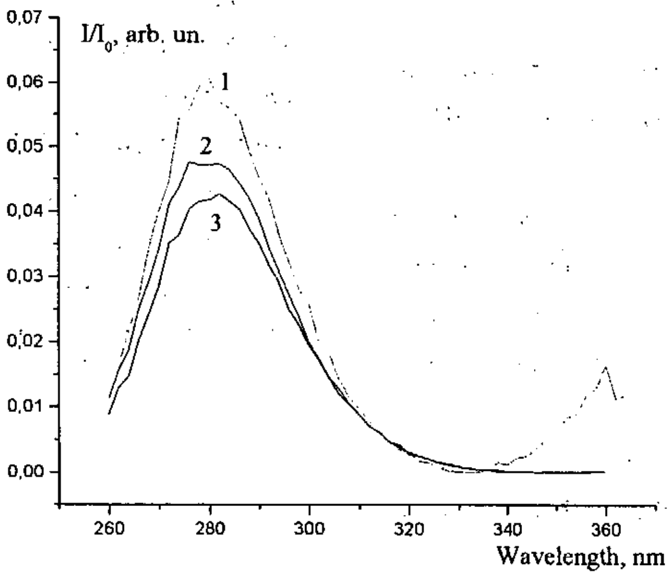

Fig. 2. Normalized spectrum of the band with the maximum at λ=282 nm at proton energies of 2.4 MeV, 1.6 MeV, 0.8 MeV (curves 1, 2 and 3, respectively).

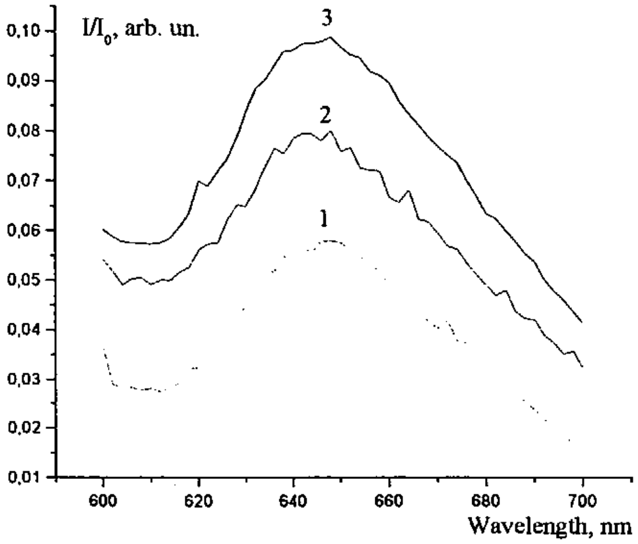

Fig. 3. Normalized spectrum of the band with the maximum at λ=648 nm at proton energies of 2.4 MeV, 1.6 MeV, 0.8 MeV (curves 1, 2 and 3, respectively).

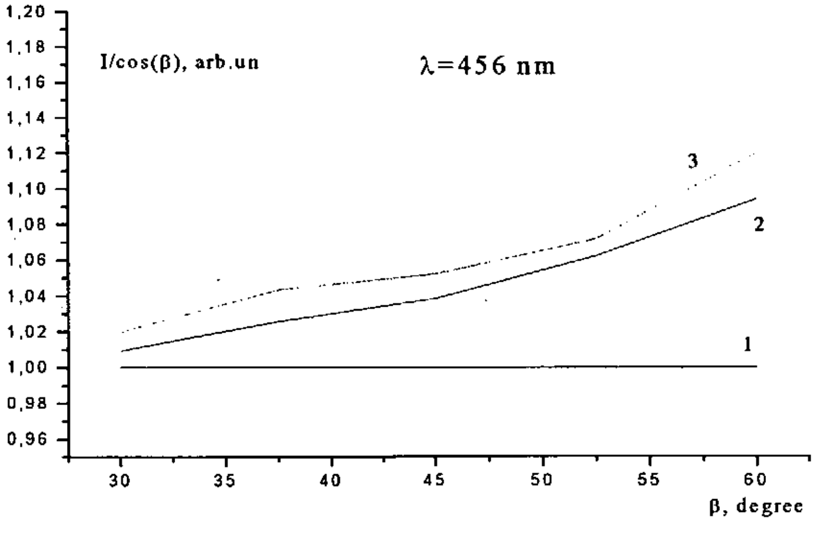

Fig. 4. Angular dependences of radiation: the Lambert law (curve 1) and luminescence of quartz under irradiation with 0.8 MeV and 1.6 MeV protons (curves 2 and 3, respectively).

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