

Ionoluminescence of silica bombarded by 420 keV molecular hydrogen ions

O.V.Kalantaryan, S.I.Kononenko, V.P.Zhurenko

V.Karazin Kharkiv National University,
4 Svobody Sq., 61001 Kharkiv, Ukraine

Received June 5, 2013

The paper presents experimental results for optical range silica luminescence induced by 420 keV molecular hydrogen ion bombardment. Ion beam from van der Graaf accelerator impinged on silica target, varying absorption dose from nonirradiated state up to $4.35 \cdot 10^{10}$ Gy. We focused upon evolution of spectrum shape and angular spectral characteristics (observation angle $0-70^\circ$) depending on absorption dose. The influence of absorption dose growth on luminescence indicatrix measured for wavelengths 456 and 644 nm was studied.

Представлены экспериментальные результаты исследования люминесценции кварца в оптическом диапазоне, возникающей при бомбардировке молекулярными ионами водорода с энергией 420 кэВ. Ионный пучок с ускорителя Ван де Граафа падал на кварцевую мишень с изменением поглощенной дозы кварца от слабооблученного состояния до $4,35 \cdot 10^{10}$ Гр. Акцентируется внимание на изменениях формы спектров и спектральных угловых характеристик (угол наблюдения $0-70^\circ$) в зависимости от поглощенной дозы. Изучалось влияние роста поглощенной дозы на индикатрисы люминесценции, измеренные для длин волн 456 и 644 нм.

Іонолюмінесценція кварцу під час бомбардування молекулярними іонами водню з енергією 420 кеВ. О.В.Калантар'ян, С.І.Кононенко, В.П.Журенко.

Представлено експериментальні результати дослідження люмінесценції кварцу в оптичному діапазоні, яка виникає під час бомбардування молекулярними іонами водню з енергією 420 кеВ. Іонний пучок з прискорювача Ван де Граафа вдаряв кварцову мішень, змінюючи поглинену дозу кварцу від слабо опроміненого стану до $4.35 \cdot 10^{10}$ Гр. Зосереджено увагу на змінах форми спектрів та спектральних кутових характеристик (кут спостереження $0-70^\circ$) в залежності від поглиненої дози. Досліджувався вплив зростання поглиненої дози на індикатрисы люмінесценції, що виміряні для довжин хвиль 456 и 644 нм.

1. Introduction

Construction of thermonuclear devices requires transparent materials such as glass and crystals. They can be mostly used as materials for optical elements. In a thermonuclear reactor such elements are windows designed for optical diagnostics. Silica glass is widely used in modern devices and facilities both as insulators and optical elements for diagnostics and optical radiation input-outputs. It is considered to be convenient as

window for UV and visible spectroscopy in thermonuclear facilities. One of the possible ways of silica monitoring under ion irradiation is our ionoluminescence technique [1, 2].

Earlier we proposed the new technique to monitor proton and molecular hydrogen absorbed dose up to $4.35 \cdot 10^{10}$ Gy in silica (see, for example, [3]). The technique is based on change of SiO_2 ionoluminescence spectra in process of ion bombardment and relation between the light intensity at some

defined wavelengths and absorbed dose. Hydrogen absorption dose can be determined using a ratio F of the luminescence intensity at 645 nm to the intensity at 456 nm, because these wavelengths correspond to maximum changes of the spectral shape [2]. We found absorption dose calibrating curve for H_2^+ irradiation of silica [3]. This curve is used by the technique proposed for H_2^+ absorption dose monitoring (up to $4.35 \cdot 10^{10}$ Gy) in silica.

The absorption dose calibrating curve is ambiguous at the beginning of absorption dose growth [3]: from nonirradiated state up to $2 \cdot 10^{10}$ Gy. We performed some additional experiments with angular spectral characteristics in order to remove the ambiguity and develop our monitoring technique.

2. Experimental

The experiments were carried out with Van der Graaf accelerators on the setup described in detail in our previous papers [3].

Molecular hydrogen ion beam with energy of 420 keV bombarded silica target at incidence angle $\alpha = 30^\circ$. Beam current density was varied from 0.3 to 30 $\mu A/cm^2$. The targets were prepared from 1 mm thick plane-parallel silica plate. We used flexible light guide for independent change of observation angle β in $0-70^\circ$ range. The radiation was detected from whole irradiated surface of the sample independently of β angle. Luminescence spectrum measurements including angular dependences were performed in the wavelength range of 400–700 nm by means of grating monochromator (1200 mm^{-1} , 1.3 nm per mm).

The optical channels were calibrated by means of incandescence spectrometric lamp. Luminescence spectra were corrected according to the spectral sensitivity. Residual gas pressure was less than 10^{-4} Pa. As luminescence light is generated along all of ion track in solid, the influence of surface contamination on optical spectra can be neglected.

3. Results and discussion

Typical luminescence spectrum induced by H_2^+ ions in visible wavelength range consists of two wide bands with maxima near 456 nm and 645 nm (Fig. 1). Such double-band shape (blue and red wavelength ranges) of luminescence spectrum is well known fact not only for ion bombardment by different species and energy [4, 5], but

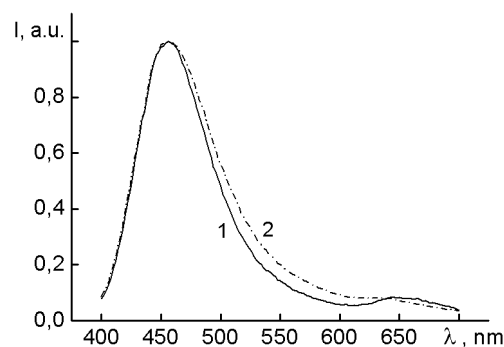


Fig. 1. Normalized ionoluminescence spectra ($\beta = 30^\circ$) for different absorption doses: 1 — at the beginning of irradiation; 2 — $2.5 \cdot 10^{10}$ Gy.

also for electron or neutron irradiation (see, for example, [6, 7]).

In order to perform some comparative study of the spectra for different absorption doses and observation angles we normalized each spectrum to the corresponding intensity at 456 nm maximum (measurement error at maximum intensity was minimal). After this procedure we found, that the most apparent changes occurred in the red band of spectra. Hereinafter we consider only normalized ionoluminescence spectra.

Fig. 2 shows red band luminescence spectra for absorption dose $1.1 \cdot 10^{10}$ Gy depending on observation angle. Minimal and maximum intensities of luminescence light were observed at $\beta = 0^\circ$ and $\beta = 60^\circ$, respectively. As it can be seen, angular changes of spectrum shape at 645 nm are more than ones at 606 nm. Thus, the F ratio depends on observation angle too.

Such angular dependences took place starting from small absorption doses and up to $1.1 \cdot 10^{10}$ Gy. But subsequent growth of absorption dose led to disappearance of apparent angular dependence (see, for example curves for absorption dose $2.5 \cdot 10^{10}$ Gy on Fig. 3). Thus, the F ratio does not depend on observation angle at large absorption doses more than $1.1 \cdot 10^{10}$ Gy.

As we mentioned above, the absorption dose calibrating curve [3] is ambiguous at the beginning of absorption dose growth. Namely, the same value of F ratio corresponds two different absorption doses. Therefore, it is not enough to measure ionoluminescence spectrum and calculate F ratio in order to determine absorption dose of silica. Ascertained angular dependence of ionoluminescence spectrum (red band region) for silica in a weakly irradiated state

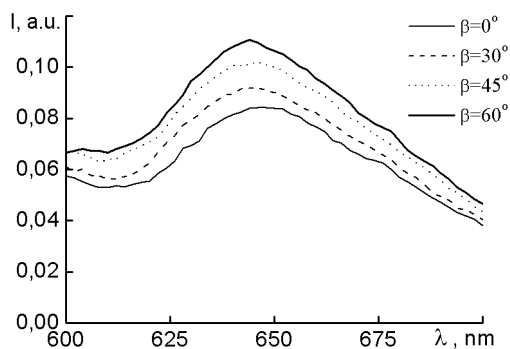


Fig. 2. Normalized ionoluminescence spectra for absorption dose $1.1 \cdot 10^{10}$ Gy.

and up to absorption doses $1.1 \cdot 10^{10}$ Gy removes the ambiguity of the calibrating curve.

In order to disambiguate absorption dose determination one should use the following procedure. The first step: angular dependences of the spectrum intensities at 645 nm and 606 nm wavelengths must be measured simultaneously. The second step: F ratio must be calculated for corresponding angles. The third step: if F ratio will have appreciable angular dependence than one should use the part of calibrating curve up to $1.1 \cdot 10^{10}$ Gy. Otherwise, the part of calibrating curve more than $1.1 \cdot 10^{10}$ Gy must be used. Thus, angular dependence data extend possibility of our remote monitoring technique for silica irradiated by H_2^+ .

We also paid attention to indicatrices measured for wavelengths 456 and 644 nm at different values of absorption doses (Fig. 4). For visual demonstration of indicatrix features they were normalized to $\cos\beta$ (the dependences measured must follow the Lambert law $I = I_0 \cdot \cos(\beta)$, where I is intensity corresponding to observation angle β , I_0 is indicatrix maximum corresponding to $\beta = 0^\circ$) and corresponding intensity values at observation angle $\beta = 0^\circ$.

As it can be seen the solid curves (absorption dose $7.64 \cdot 10^9$ Gy) show different dependences on observation angle. On the contrary, the dotted curves (absorption dose $3 \cdot 10^{10}$ Gy) converge.

Let's consider mechanism of luminescent light generation and possible reasons for indicatrix convergence with growth of absorption dose.

The most intensive blue band of spectra is usually connected with E' center intrinsic silica defects (Si with broken Si-O bond) by decay of self-trapped exciton [8]. Such de-

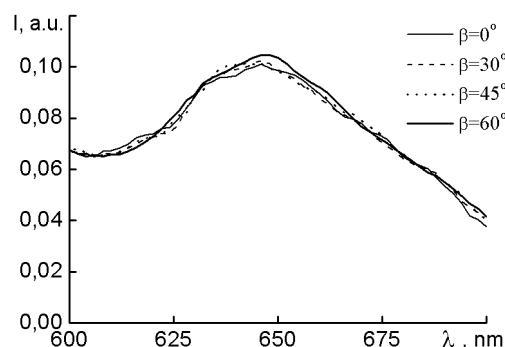


Fig. 3. Normalized ionoluminescence spectra for absorption dose $2.5 \cdot 10^{10}$ Gy.

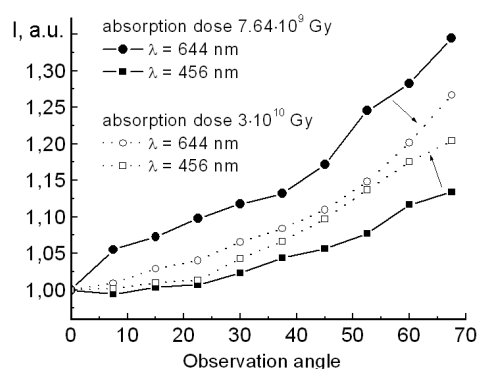


Fig. 4. Normalized indicatrices at $\lambda = 456$ nm and $\lambda = 644$ nm for two value of absorption dose.

fects as neutral oxygen vacancy (oxygen deficiency trapping two electrons) [6, 9, 10], twofold coordinated silicon [10], threefold coordinated silicon [11] can also be considered as additional sources of blue band radiation. Investigators attribute red band with 646 nm wavelength maximum to non-bridging oxygen centers (see, for example, [12]).

Concerning our case, intensity of the first band (456 nm maximum) is considerably larger than one for the second band. Luminescent light generation due to E' centers (intrinsic silica defects) is highly effective in the blue band range. Moreover, on account of large intensity of the first band its long wavelength wing affects the shape of spectra at the second band wavelength range [13]. It means, that in the case of the first absorption dose the indicatrix for $\lambda = 456$ nm is determined only by E' center intrinsic silica defects (solid curve with black circles), while the indicatrix for $\lambda = 644$ nm (solid curve with black squares) corresponds

to luminescence through non-bridging oxygen centers and to a small degree through E' centers. These solid curves display slightly different behavior, but general tendency is the same: luminescent intensity increases with growth of observation angle.

Notice that defect formation in silica irradiated by hydrogen ions is accompanied by process of hydrogen accumulation [13]. Implanted hydrogen particles are able to diffuse along the ion tracks in silica. It is known, that hydrogen accumulation can block intrinsic silica defects (especially non-bridging oxygen centers) resulting in luminescent intensity decrease. Perhaps, at certain absorption dose blocking of non-bridging oxygen centers by hydrogen begins to predominate over formation of non-bridging oxygen center defects. As a result, contribution of non-bridging oxygen centers to luminescence generation in the red band wavelength range reduces. While, E' centers influence light generation in this wavelength range to the greater extent. Fig. 4 shows this fact by means of convergence of the both dotted curves ($\lambda = 456$ nm and $\lambda = 644$ nm).

4. Conclusions

We performed the detailed experimental study of silica ionoluminescence induced by 420 keV H_2^+ at absorption dose up to $4.35 \cdot 10^{10}$ Gy. The ionoluminescence spectra changed with growth of absorption dose and accumulation of radiation defects. It was found that angular dependence of spectrum shape took place only at absorption dose less than $1.1 \cdot 10^{10}$ Gy. This result permitted to disambiguate the calibrating curve for absorption dose determination (up to $2 \cdot 10^{10}$ Gy). This makes it possible to use

our remote monitoring technique for silica irradiated by H_2^+ at absorption dose range from nonirradiated state up to $4.35 \cdot 10^{10}$ Gy. The indicatrices at $\lambda = 456$ nm and $\lambda = 644$ nm tended to coincide starting from absorption dose $3 \cdot 10^{10}$ Gy. Cleared-up facts can be explained by complicated dynamic processes of defect formation and modification (E' -centres and non-bridging oxygen centers) with growth of ion absorption dose.

References

1. S.I.Kononenko, O.V.Kalantaryan, V.I.Muratov, V.P.Zhurenko, *Nucl. Instr. Meth. Phys. Res. B.*, **246/2**, 340 (2006).
2. S.I.Kononenko, O.V.Kalantaryan, V.I.Muratov, V.P.Zhurenko, *Radiat. Meas.*, **42**, 751 (2007).
3. O.Vasylichenko, N.Zheltopiyatova, V.Zhurenko et al., *Functional Materials*, **17**, 67 (2010).
4. F.Jaque, P.Townsend, *Nucl. Instr. Meth. Phys. Res.*, **182–183**, 781 (1981).
5. T.Tanabe, A.Omori, M.Fujiwara, *J. Nucl. Mater.*, **258–263**, 1914 (1998).
6. A.Morono, E.R.Hodgson, *J. Nucl. Mater.*, **258–263**, 1889 (1998).
7. F.Sato, T.Iida, Y.Oyama et al., *J. Nucl. Mater.*, **258–263**, 1897 (1998).
8. F.Pio, M.Guzzi, M.Martini, *Phys. State Sol. (B)*, **159**, 577 (1990).
9. J.P.Duraud, F.Jollet, Y.Langevin, E.Dooryhee, *Nucl. Instr. Meth. Phys. Res. B*, **32**, 248 (1988).
10. L.N.Skuja, A.N.Streletsky, A.B.Pakovich, *Sol. State Commun.*, **50**, 1069 (1984).
11. N.Trukhin, *Nucl. Instr. Meth. Phys. Res. B.*, **91**, 334 (1994).
12. L.N.Skuja, A.R.Silin, *Phys. State Sol. (A)*, **70**, 43 (1982).
13. I.Yarova, N.Zheltopiyatova, V.Zhurenko et al., *Visnyk Lviv University. Ser. Physics*, **46**, 196 (2011).