

СООБЩЕНИЯ  
ОБЪЕДИНЕННОГО  
ИНСТИТУТА  
ЯДЕРНЫХ  
ИССЛЕДОВАНИЙ

Дубна

95-193

E14-95-193

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LUMINESCENCE SPECTRA FROM  $\alpha$ -Al<sub>2</sub>O<sub>3</sub>  
UNDER 1 MeV/amu ION IRRADIATION

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1995

# 1 Introduction

As known, the radiation defects in sapphire are formed just under irradiation causing the atomic displacements, this excludes the ionization as a possible source of primary structural damages. At the same time the production of electrons and holes simultaneously with the atomic displacements significantly affects the evolution of the defect structure of the irradiated crystal. An example of such effect is the radiation-induced conductivity (RIC) the value of which depends on the ratio of the ionizing and damaging component of the irradiation effect. Thus, investigations of the radiation damage of  $Al_2O_3$  under high-energy heavy ion irradiation when high rate of the defect formation and high level of the ionizing energy losses are involved are much currently problem. Experiments of Morono and Hodgson[1] have shown the importance of the optical absorption and luminescence spectra registration for the RIC studies as an indicator of the presence of the certain type of radiation defects. It stimulates the "in-situ" optical investigations of sapphire under irradiation. In the present work experimental results of studying the luminescence spectra of  $Al_2O_3$  excited by the ions B, Ne and Ar with the energies 13.6, 26.7 and 46.3 MeV, respectively, are being discussed.

# 2 Experimental

The uv grade  $Al_2O_3$  samples used in our study were obtained from the Institute of Crystallography(Moscow) in the form of discs (the diameter- 10 and 20 mm, the thickness 0.34 and 2 mm) and were oriented so that the crystallographic axis lay approximately perpendicular to the surface. Under irradiation the crystals were oriented at  $45^\circ$  to the ion beam and monochromator directions.

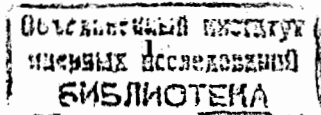
Experiments have been carried out with the IC-100 cyclotron beams. The ion beam induced luminescence (IL) spectra have been registered in the wavelength range of 225-550 nm in the temperature range 80 - 350 K and corrected for system response. The flux density of B, Ne and Ar ions was  $2 \times 10^9$ ,  $1.2 \times 10^9$  and  $5.8 \times 10^8 \text{ cm}^{-2}\text{s}^{-1}$ , respectively. Ion beam pulse duration was 1.2 - 1.5 ms.

# 3 Results and Discussion

The luminescence generation by high-energy ions occurs together with radiation damage formation, that, naturally, suggests the spectra dependence on its concentration. Due to this reason the most correct comparison of the IL spectra is possible at the same damage level. This value has been calculated as:

$$D = \int_0^{R_p} \sigma_D(x) dx / R_p$$

where  $\sigma_D(x)$  - damage cross-section as a function of the ion penetration depth and  $R_p$  - ion projective range. The  $\sigma_D(x)$  and  $R_p$  values have been calculated by using TRIM - 90 computer code (for example [2]). So,  $D$  equals total dpa value, divided per projective range. Luminescence spectra excited by B, Ne and Ar ions at 80 K, normalized on ion beam intensity are given in Fig. 1. for  $D \approx 1.5 \times 10^{-5}$  dpa. One can see, that spectra composed of three broad bands peaked at 3.0, 3.8 and 4.35 eV. The luminescence yield in these bands depends on ion type, irradiation temperature and dose. As an example, the dose dependence of the IL intensity  $N(D)$  for Ne and Ar ions at 80 and 293 K is presented in Fig. 2. As our results show, there are, at least, two stages of dose dependence which differ by spectral decomposition. The



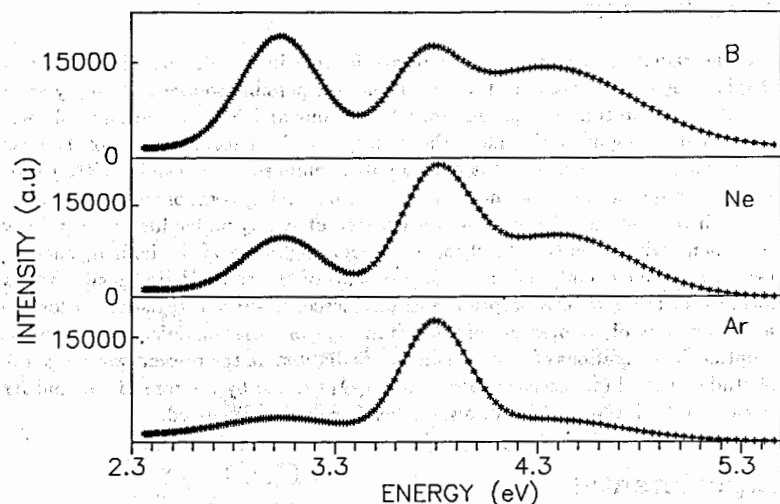


Fig. 1. Luminescence spectra excited by 1 MeV/amu heavy ions at 80 K.

initial time of the irradiation, three bands peaked at 3.0, 3.8 and 4.35 eV are registered. The next stage is characterized by the presence of two bands 3.0 and 3.8 eV only. The irradiation by argon ions is characterized by low radiation efficiency in bands 4.35 eV and 3.0 eV the reason of which will be discussed in a particular work.

A considerable body of works is devoted to the sapphire luminescences in a spectral range studied by us [1, 3-19]. Thus, first of all, the comparison of parameters of the measured spectra with the available literary data is of interest. Information on spectra involving the excitation type, the temperature of a sample during measurements, the position of the band maxima, the lifetime of the excited state, the full width on half maximum (FWHM) are summarized in Table 1.

So, the band 3.0 eV, as seen from the Table, is in the luminescence spectra at irradiation by all types of the ionizing radiation and at excitation by the light in the absorption band 6.1 eV and attributes to the F-centers (two electrons in the field of the anion vacancy). The position of the maximum of this band does not depend on crystal temperature in the range from 10-300 K this is also true in the case of high-energy ion irradiation. The luminescence band at 3.8 eV (one electron in the field of the anion vacancy) is also observed under the effect of all kinds of radiations, however in contrast to the band 3.0 eV it is a complex one and involves up to three elementary bands [18]. These bands have the maximum at the same energy but differ by polarization characteristics, the FWHM, excitation spectrum and temperature dependence as well. The structure of the band 3.8 eV is analyzed in the work [18] in the most detail, where the spectral structure and kinetics of the cathodoluminescence, the photoexcitation and photoluminescence spectra, kinetics and the spectral structure of thermally stimulated luminescence of the nominal pure samples and samples with different concentration of the intrinsic defects have been studied.

Table 1. The luminescence characteristics of sapphire for different types of the excitation (here PL-photoluminescence, CL-cathodoluminescence, STE - self-trapped excitons, STH - self-trapped holes).

Type of the excitation	$h\nu_{max}$ , eV	FWHM, eV	Lifetime	Nature of the band	Ref.
PL 4.1, 4.8 5.4 eV	3.75-3.8	0.351	<50 ns	F <sup>+</sup>	3, 4, 10, 11, 12, 14
	3.82	0.34	<7 ns	F <sup>+</sup>	-
PL 6.1 eV	2.99-3.00	0.36	10-36 ms	F	3, 5, 9, 15
X-ray	3.8	0.35(300K)	<4 ns	F <sup>+</sup>	18
		0.26(80K)		F	-
CL	3.0			F	-
	4.3			STH	-
	3.8			F <sup>+</sup>	18, 19
	3.0			F	-
e <sup>-</sup> 0.430 - 2 MeV	3.76-3.81			F <sup>+</sup>	1, 3
	2.98-3.008			F	-
e <sup>-</sup> 0.48 MeV	3.81			F <sup>+</sup>	13, 17
	4.1			F	-
n <E>1.3 MeV	3.87			F <sup>+</sup>	6
	2.95			F	-
He <sup>+</sup> , D <sup>+</sup> 6.5, 25 keV	3.80-3.87			F <sup>+</sup>	6
	2.95-3.00			F	-
H <sup>+</sup> , He <sup>+</sup> 2-3.8 MeV	3.68			F <sup>+</sup>	7, 8
	3.17			F <sub>2</sub>	-
	2.99			F	-
Ar <sup>+</sup> 2.4 MeV	3.68			F <sup>+</sup>	-
	3.17			F <sub>2</sub>	-

First of all, in samples irradiated by neutrons the main contribution in emission at 3.8 eV is conditioned by the radiative transitions in the F<sup>+</sup>-centers. The maxima of the typical excitation bands of these intercenter transitions are at 3.8 and 5.35 eV. The halfwidth - 0.35 eV at 300 K (.26 eV at 80 K), the polarization ratio  $I_{||}/I_{\perp} = 1.3$  and the lifetime  $\tau < 4$  ns are the spectral passport of the F<sup>+</sup> band. The band with the maximum 3.8 eV, FWHM = 0.25 eV at 300 K and the polarization ratio  $I_{||}/I_{\perp} = 6$  was observed in the neutron irradiated samples under photoexcitation in the band of intercenter transitions with the maximum at 4.1 eV and at excitation in the region of the fundamental absorption [18, 19]. This band in the author's opinion is caused by the interstitial atoms of aluminum [15].

And at last, the band with the maximum 3.8 eV, halfwidth 0.36 eV and  $\tau \leq 28$  ns at

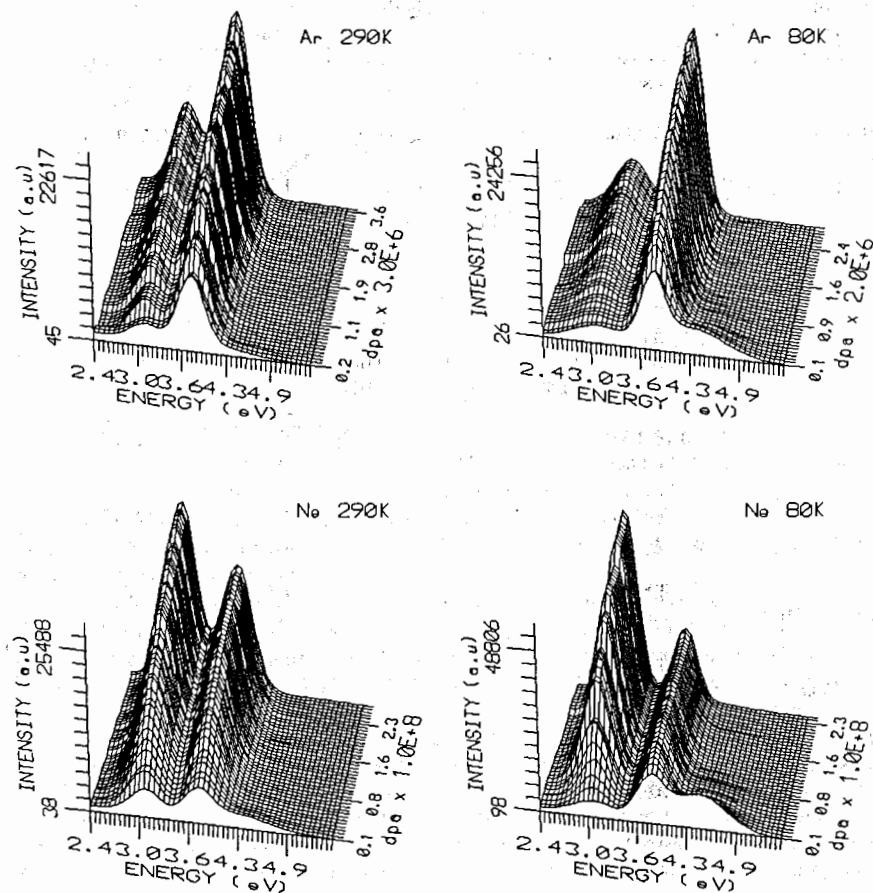


Fig. 2 The dose dependence of the IL intensity for Ne and Ar ions at 80 and 290 K.

300 K (.35 eV and 280 ns at 80 K), the polarization ratio  $I_{\parallel}/I_{\perp} = 0.8$  has been detected in samples with the undamaged anion sublattice. The emission in this band (E-luminescence by the author's terminology) is connected with the known intrinsic luminescence characterized by the band with the maximum at 7.5 eV (A-luminescence)[16]. Due to the analysis of all possible mechanisms of the E-luminescence the authors came to the conclusion that the experimental data obtained by them can be interpreted from the common point of view just in the framework of conceptions on the exciton nature of the both related luminescence bands. So, in this case the exciton has two stable configurations of the relaxed state which are separated by the potential barrier. The low-energy configuration of the relaxed exciton involves self-trapped hole as a hole component and responds for the E-luminescence. Thus, the E-luminescence can be interpreted as a luminescence of the self-trapped exciton.

Besides the STE emission in sapphire crystals at the luminescence excitation by the X-rays [18] there has been detected spontaneous interdefect radiative recombination independent on the temperature of observation below 200 K. It is a so-called tunnel luminescence (TL) the spectrum of which is presented by the band with the maximum at 4.3 eV. According to [18] the TL mechanism involves the diffusion-controlled tunnel recombination of the electrons and holes, i.e. the reactions of the tunnel transfer of the electron between the electron and hole centers which is limited in the velocity of their diffusive approach. Thus, the emission in the band of 4.3 eV is due to the reaction which includes the thermostimulated migration of the self-trapped holes at low temperature (<200 K). Another interpretation of 4.3 eV band nature is as a radiative relaxation of the 1p-like state of the F-centers[17].

From the data of the works [7,8] the spectra of the sapphire luminescence measured during low-energy ion implantation except for the F and F<sup>+</sup> bands involve also the band at 390 nm (3.13 eV) associated with the F<sub>2</sub>-centers. The relative IL yield in this band increases with the increase of the mass of incident ions. Approximation with the Gaussians of the spectra measured by us showed that the inclusion of the band 3.13 eV gives better agreement with the experimental data. Therefore, one can assume that while irradiation by the high-energy ions the band 3.13 eV is in the IL spectra. However, the obtained data are deficient in the studied regions of fluences so that one could with certainty to determine the contribution of this emission in the total luminescence intensity.

So, summarizing the given information one can make a conclusion that the IL yield in the spectral range considered at high-energy ion irradiation is mainly determined by the intercenter transitions in the F and F<sup>+</sup> centers and also by the radiative recombination of the self-trapped electrons and holes. As it was mentioned before, the contribution of these processes depends on the level of the damage of the anion crystal sublattice. Thus, to determine the damage level of the which is peculiar to the above-listed processes is of interest. As seen from the Fig. 2 the intensity of the bands 3.8 and 3.0 eV increases with the irradiation dose and reaches the saturation for the Ar ions or the level close to the saturation for the Ne ions. At the same time the intensity of the band 4.35 eV decreases up to the undetected level proportional to D<sup>-n</sup>, where n is the parameter equal to  $0.55 \pm 0.05$  for both types of the ions, as seen from the Fig. 3. Two possible processes causing the irradiation in the band 4.3 eV has been discussed above, they are - the radiative relaxation of 1p-like state of the F-centers and the tunnel recombination of the self-trapped holes with the electrons. As in our case there is no correlation in the dose dependencies of the band 4.3 eV and F band, then there are no grounds to believe that the luminescence in them is caused by the same centers. It is also known, that the tunnel STH recombination involves a stage of the diffusive approach of the self-trapped holes and electrons (electron centers).

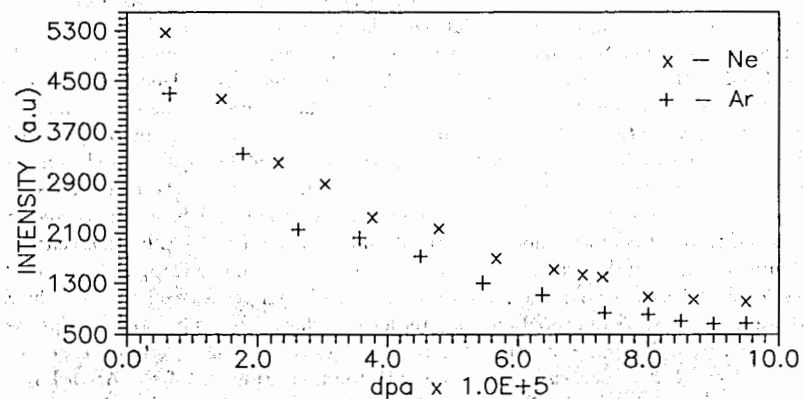


Fig. 3. The dose dependence of the IL intensity in the band 4.35 eV at 80 K.

The availability of this stage in any radioluminescence processes means so-called migration losses due to the charge carrier captured by structural defects and therefore, a certain dependence of the luminescence yield on the concentration of defects similar to the observed one for the band 4.35 eV. Among such defects in our case are first and foremost the oxygen vacancies which are the effective traps for the electrons and as a result the F-centers are formed. One should note that the STH radiation is suppressed under irradiation dose common to the Ar and Ne ions and corresponding to the dpa value just  $10^{-4}$ . Taking into account the character of the band 3.8 eV, the IL dose dependence in the most general form the dependence  $N(D)$  should be presented as a sum:

$$N(D) = N_{F^+} + N_{STE}(D)$$

where  $N_{F^+}$  and  $N_{STE}(D)$  relate to the luminescence of the  $F^+$ -centers and the STE, respectively. The formation of all  $F^+$ -centers and the STE are the competition processes. In this case the first one preferable as the anion vacancy disturbs the crystal sublattice much more than the hole. Therefore, the value  $N_{STE}(D)$  as well as in the case with the band 4.35 eV will decrease with increasing the irradiation dose. However, taking into account the dispersion of experimental data to separate the contribution of the  $F^+$ -centers and STE luminescence from dose dependence is impossible. As a rough estimation of the upper boundary of damage dose range where one should take into account the presence of the exciton luminescence in the band 3.8 eV one take value obtained for 4.3 eV band -  $10^{-4}$  dpa.

#### 4 Conclusion

Thus, the spectral structure of the luminescence of  $Al_2O_3$  produced by high-energy heavy ions has been studied. It is shown that in the spectral interval of 2.3-5.6 eV the luminescence is caused by the intercenter transitions in the  $F^+$  and F-centers, the radiative STE recombination and the tunnel STH recombination. It has been found that the damage level whereby the STH emission is suppressed corresponds to  $10^{-4}$  dpa.

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Received by Publishing Department  
on April 25, 1995.