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V.A.Skuratov, A.Yu.Didyk, S.M.Abu AlAzm*

HIGH-ENERGY
IONOLUMINESCENCE OF LiF

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*On leave from the Nuclear Research Center, AEA, P.C. 13759,
Cairo, Egypt

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Исследована люминесценция монокристаллов фтористого лития в процессе облучения ионами В (13,6 МэВ), Ne (26,7 МэВ), Ar (46,3 МэВ), Kr (210 МэВ) и Xe (158 МэВ). Спектры регистрировались при температуре облучения 80-300 К в диапазоне длин волн 250-700 нм. Установлено, что спектральное распределение люминесценции характеризуется набором широких полос с максимумами при 670, 525, 330 и 296 нм. Обсуждаются возможные причины зависимости выхода люминесценции в ультрафиолетовой полосе 296 нм от типа и энергии ионов в контексте радиационного повреждения LiF.

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The luminescence of pure crystals of lithium fluoride during irradiation by the ions В (13.6 MeV), Ne (26.7 MeV), Ar (46.3 MeV), Kr (210 MeV) and Xe (158 MeV) is investigated. Spectra were registered at the temperature of 80—300 K in the wavelength range of 250—700 nm. Spectral distribution is characterized by a set of wide bands with maxima of 670, 525, 330 and 296 nm. Possible reasons of the luminescence yield of ultraviolet band of 296 nm dependence on ion type and energy at liquid nitrogen temperature with regard to radiation damaging of LiF are discussed.

The investigation has been performed at the Flerov Laboratory of Nuclear Reactions, JINR.

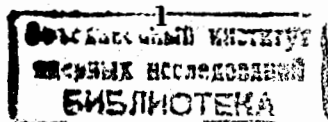
1. Introduction

The luminescence spectroscopy: photo-, cathode-, X-ray-, thermoluminescence is widely used to study a structure of ionimplanted crystals after irradiation and different stages of postradiation treatment, that is methods practically which do not lead to new defects forming during their use. In contrast to these classical methods the luminescence excitation by means of heavy charged particle beams allows one to study material structure just during formation and evolution of radiation damages. Ion energy in such experiments, as a rule, is of tens and hundreds keV. At the same time, in order to exclude the influence of surface sputtering processes one should use the light ions such as protons, deuterons, α -particles. Increasing of ion energy up to 1 MeV/amu allows one to decrease sputtering and due to increasing of ionizing energy losses to increase the luminescence yield. It is known that energy losses at the electron subsystem excitation are the maximum ones at the energy of bombarding ions of $\approx 1\text{MeV/a.m.u.}$. Thus, high-energy ionoluminescence (IL) can be one of few sources of a structural information while studying solid state properties on heavy ion beams.

In our previous work[1] we studied the luminescence of LiF under 46.3 MeV argon ion irradiation. In the present work the results of experiments of investigation of luminescence spectra of the LiF crystals generated by the ions B(13.6 MeV), Ne(26.7 MeV), Ar(46.3 MeV), Kr(210 MeV) and Xe(158 MeV) are discussed.

2. Experimental

Experiments have been carried out on the IC-100 (B, Ne, Ar) and U-400 (Kr, Xe) cyclotrons beams. The IL spectra have been registered in the wavelength range of 250-700 nm at the temperature from 80 to 300K and corrected for system response. Ion flux density B, Ne, Ar, Kr and Xe was $2.5 \cdot 10^9$, $2.2 \cdot 10^9$, $1.1 \cdot 10^9$, $8 \cdot 10^8$ and $1.3 \cdot 10^8 \text{ cm}^{-2} \text{ s}^{-1}$ respectively. Ion beam pulse duration was equal 1.2 - 1.5 ms at pulse period-to-pulse duration ratio of about three. Total impurity concentration in investigated samples did not exceed 0.001 molecular percent.



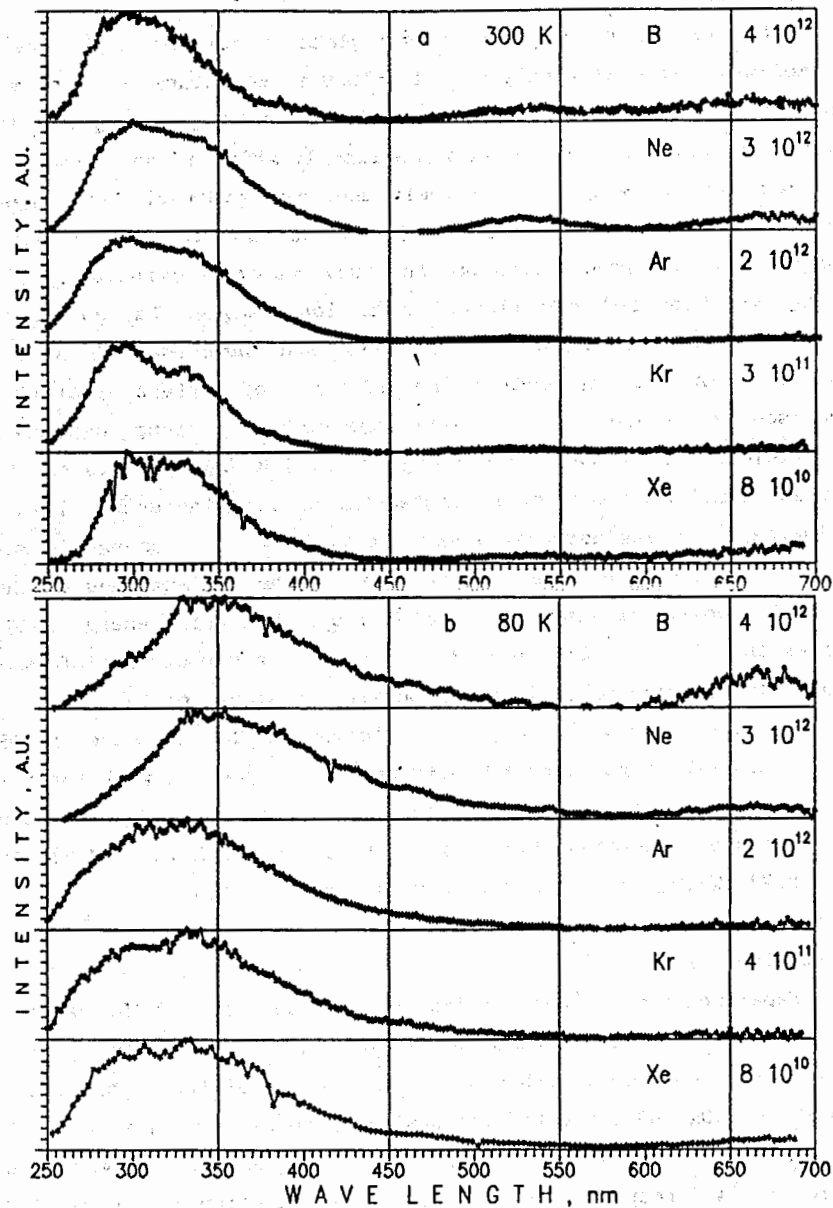


Fig. 1. Luminescence spectra of LiF during ion irradiation at room temperature and liquid nitrogen temperature.

3. Results

In fig. 1 a,b there are given the LiF IL spectra produced by heavy ions at 300(RT) and 80(LNT) K. All spectra have been normalized to the maximum value. As seen from the figures, at RT spectra represent a set of wide bands with $\lambda_{\max} = 670, 525, 330, 296$ nm. According to the work [2] where the CL of the LiF pure crystals has been studied, spectral distribution in the region of 300 - 500 nm besides the band of 330 nm includes the overlapping bands with $\lambda_{\max} = 350, 370, 410$ and 470 nm. Probably, the IL spectra also include the indicated bands, especially at low temperature, however, it is difficult to separate them. It should be noted that presented spectra are given at different ion fluences and are therefore compared only qualitatively in longwavelength bands strongly depended on irradiation dose.

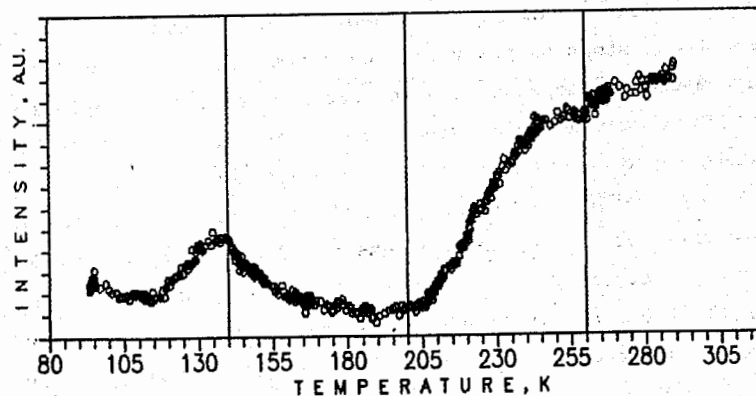


Fig. 2. The temperature dependence of IL intensity of band 296 nm under Ar ion excitation (heating).

The nature of bands 670, 525 and 330 nm is well known studied and described elsewhere [2,3]. Ultraviolet band of 296 nm in luminescence spectra besides 1 MeV/amu Ar ion irradiation was registered also under X-ray excitation after previous irradiation at 80 K and

following heating. We believe, that an emission in this band is associated with reaction $V_F + e^-$, where V_F denotes the V_k -hole trapped by cation vacancy. This suggestion is confirmed by temperature dependence of 296 nm band, presented in fig.2 for Ar ions. So, increasing of the IL intensity at 210-220 K is in accordance with beginning of thermal mobility of V_F -centers.

4. Discussion

As known, the IL is a particular case of radioluminescence effect (RDL). The RDL process assumes some stages which proceed in chronological order in alkali halides as follows: 1) production of thermalized electrons and holes as a result of energy dissipation of bombarding ions; 2) migration of these electron excitations to the luminescence centers; 3) electrons and holes recombination on the luminescence centers transforming these centres into excitation state and the next radiative transition into the ground state.

Electron excitation density evidently is determined by the level of electron stopping power. The carriers migration besides irradiation temperature depends also on number of radiation defects being traps. To a considerable extent the number and spectrum of radiation damages are determined by energy losses for elastic scattering. Therefore, before to analyze the differences in the IL spectra let present specific losses of energy B, Ne, Ar, Kr and Xe ions for electron stopping and elastic collisions defining the character of the two first IL stages. These data obtained by using TRIM-90 program are presented in fig. 3a,b versus ion penetration depth.

The results of present measurements given in fig. 1 a,b lead to two significant observations of IL spectra generated by high energy heavy ions:

1. Luminescence yield decreases in the longwavelength bands of 525 and 670 nm with the increase of atomic number and, hence, density of ionization and nuclear energy losses with respect to exciton band of 330 nm.

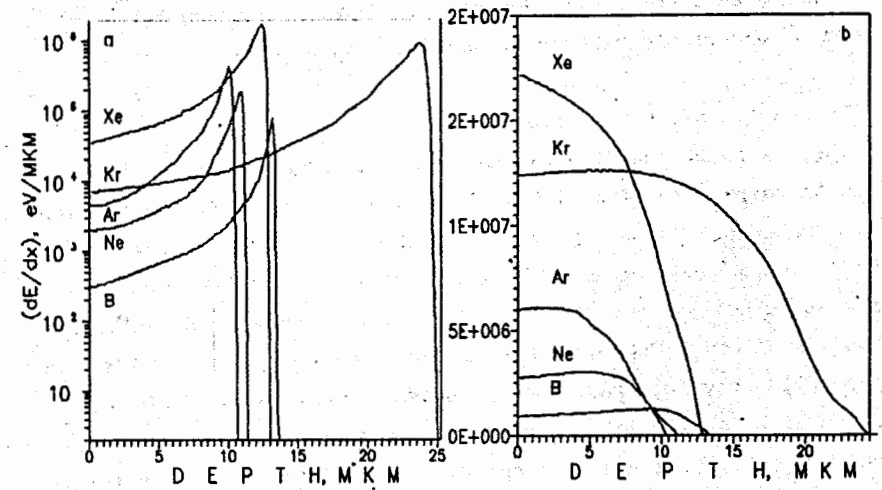


Fig. 3. Specific energy losses of ions for elastic scattering (a) and electronic excitation (b) in LiF.

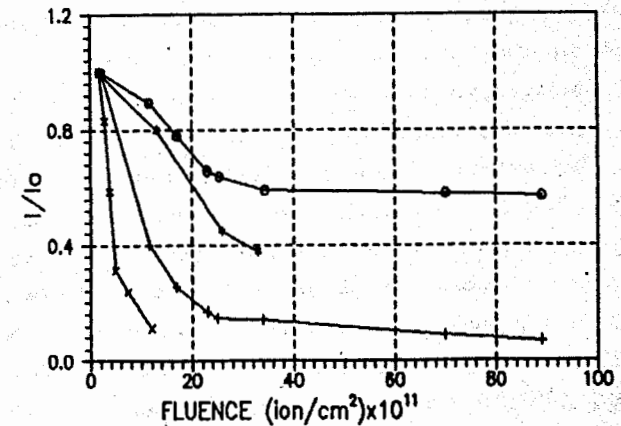


Fig. 4. Dose dependence of luminescence yield with respect to initial value at RT. o - Ne(330nm), * - Ar(330 nm), + - Ne(525 nm), x - Ar(525 nm).

2. One observes a complicate dependence of the IL intensity in 296 nm band at the liquid nitrogen temperature on excitation density.

4.1 The 525 and 670 nm emission bands

The reason of the IL yield decrease in the bands of 525 and 670 nm in comparison with 330 nm band with the increase of ion atomic number one should, probably, search in more high efficiency of radiation damage production. It is seen fig 3b that energy losses from elastic collision for Kr ions 4 times more exceed the analogous values for B ions. One should also take into account that in alkali-halide crystals radiation defects can be produced also due to electron stopping of high energy ions. So, in the work [4] there was shown that distribution of F_2 -centers in LiF crystals irradiated by C(110 MeV) and Cu(180 MeV) ions correlates with the distribution of ionization energy losses along these ions range. The consequence of this is a great amount of radiation defects in ion tracks on which electrons and holes can be localized during migration process, that finally decreases the probability of radiative recombination. At the same time at exciton annihilation there is excluded migration stage as the electron and hole exciton component are combined. Therefore, the absence of carriers migration being the necessary condition for luminescence appearance in 525 and 670 nm bands explains a weak dependence of luminescence in exciton band on radiation damages of crystal lattice. This is quite well seen on example of dose dependence of luminescence intensity in considered bands at RT presented in fig. 4. As one can be see, the IL yield in 525 and bands decreases within ion fluence much stronger than in 330 nm band. One can also observe the essential difference in the IL intensities for the Ar and Ne ions in the longwavelength bands at the same fluences, while for exciton band the corresponding values are more close.

4.2 The 296 nm emission band

Let consider possible reasons of the observable IL intensity dependencies in 296 nm on bombarding ion type at liquid nitrogen temperature.

Thus, the emission in 296 nm band is, most probable, attributed with recombination of V_F -centers on electrons. As follows from fig.1 the IL yield in this band at LNT with respect to exciton band equals to 0.3 for B, and Ne ions, 0.6 - for Ar ions and 0.75 for Kr and Xe ions. At the same time, at RT this ratio for B, Ne and Ar is ≈ 0.9 and approximately equally and for Kr it is 1.2.

The 296 nm band is registered at 80 K, therefore, the formation of V_F -centers takes place already at this temperature. The formation of V_F -centers one usually connects with migration of V_K -holes and their capture by cation vacancies. However, the 525 nm band, due to holes migration and recombination practically is not observed at LNT, although it is difficult to separate it because of overlapping with other bands. Therefore, one could suppose that the formation of V_F -centers occurs just in bombarding ion tracks in cascades of atomic collisions. By the Ar, Kr and Xe irradiation the concentration of cation vacancies is higher than by the B and Ne irradiation, that leads to form a great number of V_F -centers and to increase the probability of their radiative recombination.

Discussing possible factors which influence upon an emission in the band of 296 nm one should take into account crystal heating under ion irradiation. One can neglect the increase of average temperature of sample as a maximum power inserted by the ion beam did not exceed 0.1 Wt. However, taking into account the low thermoconductivity of LiF, one could assume that the temperature in irradiated layer exceeds the average target temperature. Moreover, the experimental techniques having been used did not allow to measure temperature for time comparable with ion beam pulse duration. It is impossible also to experimentally determine temperature distribution by thickness of irradiated sample. Therefore, the temperature field $T(x,t)$ (the x axis coincides with beam

direction) was determined from the thermoconductivity equation by assumption that the target is essentially extended in other directions.

Boundary and initial conditions were chosen in the form of $T(x=1, t) = T_0$, $T(x, t=0) = T_0$, (l is the sample thickness, T_0 - initial temperature, i.e. the target is cooled on the side opposite to the beam input, that corresponds to samples irradiation on cooled holder in the vacuum.

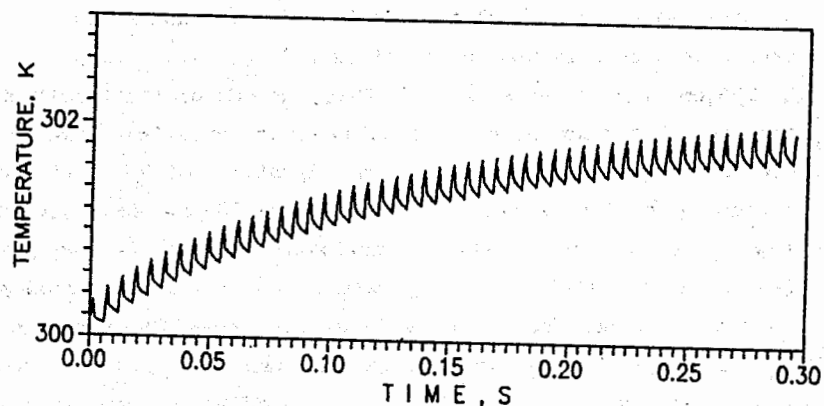


Fig. 5. The temperature of LiF sample (1 micrometer from the surface of irradiated layer) during 210 MeV Kr ion bombardment. $T_0 = 300$ K, $l = 1$ mm., $\Phi = 2.4 \cdot 10^9 \text{ cm}^{-2} \cdot \text{s}^{-1}$.

In fig.5 the dependence $T(t)$ on the LiF sample surface irradiated by the ions Kr with the energies of 210 MeV at RT is presented. As seen from the figure, the temperature mode of irradiation can be considered as a quasi-stationary one.

In fig 6a,b there are given the IL spectra measured for three values of ion flux at 80 and 300 K. The increase of ion flux density three times did not lead to variation of spectral distribution at LNT and at RT.

Temperature increase in the region of high-energy ion track much more influences on defect structure forming and also on the IL.

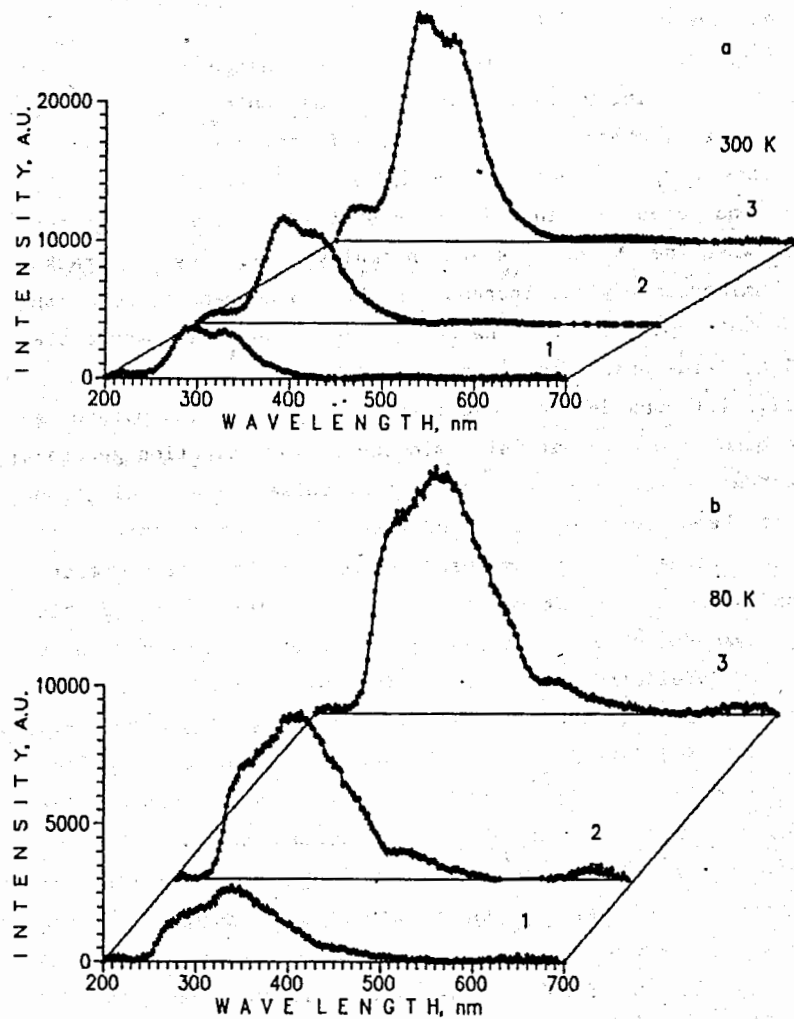


Fig. 6. Luminescence spectra of LiF during Kr ion irradiation for various ion beam intensities at RT(a) and LNT(b). 1- $8 \cdot 10^8$, 2- $1.6 \cdot 10^9$, 3- $2.4 \cdot 10^9 \text{ cm}^{-2} \cdot \text{s}^{-1}$.

migration stage. Experimentally this effect manifests itself in the annealing of radiation damage in materials irradiated by heavy ions with the energies more than 0.5 MeV/a.m.u..

Therefore, one can assume that mobility of charge carriers and radiation defects including V_F -centers is determined not only by the mean crystal temperature but also by effective temperature in the ion track T_{eff} . It means that radiation recombination of the V_F -centers and electrons in the region of track is possible at the temperature when the V_F -centers are thermally fixed beyond track. Thus, the luminescence yield increase in the 296 nm band at LNT with the increase of specific ionizing energy losses one can associate with the T_{eff} value proportional to $(dE/dx)_{ion}$.

Finally, let consider one more interpretation of the nature of the 296 nm band. Recently, at CsI luminescence investigation generated by synchrotron radiation [5] and also at pulse X-ray excitation [6] there has been revealed the band with the maximum shifted to shortwavelength range against luminescence band of triplet selftrapped exciton and equals to 296 nm as in our case. The origin of this band, which was not observed at stationary irradiation the authors [5, 6] explain by electron excitations interaction with fast defects the result of which is a recombination of close located correlated selftrapped hole and fast defect with production of an exciton-like state and the following emission in 296 nm band. The analogous situation is possible also in high energy ion tracks when in anion and cation sublattices of a crystal there are produced both stable and unstable Frenkel pairs (fast defects by authors terminology [6]). So, more detailed than in [1] investigations of luminescence yield in the 296 nm band with ion fluence showed that this dependence is close to dose one of exciton band 330 nm. Therefore, the emission in the band 296 nm can be caused by annihilation of the exciton like state $V_k \cdot e^-$ where V_k is trapped by cation vacancy during the existence of unstable Frenkel pair at the same time forming the unstable V_F -center. On this case the IL total yield in this band combines radiative recombination of stable and unstable V_F -centers. At LNT,

when V_k - and V_F -centers are fixed, the luminescence is caused by recombination of unstable centers as the mechanism of their production assumes weak dependence on temperature. With increasing of temperature and defects mobility the contribution of stable V_F -centers increases in accordance with data given in fig. 2.

Conclusions

Spectral distribution of the LiF crystals luminescence produced by high-energy ions B, Ne, Ar, Kr and Xe versus irradiation temperature and ion fluence has been investigated. There has been determined that in spectra is a band with $\lambda_{max} = 296$ nm irrespective of irradiation temperature in contrast to other types of the excitation. The luminescence yield in this band depends on exciting density and is associated with radiative recombination stable and unstable V_F -centers on electrons.

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