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MECHANISM AND MODEL OF IMPULSE LUMINESCENCE IN AHC BY THE ACTION OF β – PARTICLES STATIONARY FLOW

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Abstract. We studied the impulse luminescence of AH crystals in the visible and ultra-short wavelengths under stationary flow beta-particles. We propose a mechanism and model of the luminescence.

1. Introduction

The most common and promising ionic crystals include fluorides and chlorides of alkali metals, which is explained by the simplicity of their structure and nature of chemical bonds by which they are often used in scientific research as a model system to elucidate the nature of physical phenomena. Practical application of the crystals is most often caused by the peculiarities of their structure-sensitive properties. To change the properties in the right direction various methods can be used, including such method as intentional introduction of impurities, which leads to changes in the properties by changing the chemical composition of the crystal, and method of exposure of ionizing radiation. In the later case, changes in the properties associated with structural abnormalities that occur in the crystal under the action of radiation – the so-called electron and hole color centers. As noted in some studies, in ionic crystals the main mechanism responsible for the creation of color centers is the decay of electronic excitations. Local excitations can decay with the creation of free-electrons scattering, free-holes scattering and excitons. This reverse process, well-studied in AH crystals, is called the delocalization of the impurity electronic excitations. In ionic crystals, this process appears as a crystalline effect associated with the transfer of energy from the charge of the impurity center to the surrounding crystal lattice. Nowadays, such effects as the crystalline electronic hole conductivity, the creation of electron and hole color centers and recombination luminescence have been found in the AHC. Such effects may arise as a result of the detachment of free-electrons, and free-holes scattering, excitons and electron-hole pairs from the excited impurity centers.

The objective of this investigation is to determine the nature of the impulse luminescence, elementary mechanisms of energy transfer to luminescence centers and radiation processes in AH crystals during irradiation by the steady flow of beta particles with energies in the range 50- 75 keV; as well as expanding existing model concepts to explain the new and previously obtained experimental results [1].

2. Experimental method

In the experiments, we used the optically transparent single crystals of lithium fluoride having the impurity content 10-3 wt%, Ca+2, Mg+2, Ba+2. Cathodoluminescent and photoluminescent methods were used to study the fluorescent properties of the objects.

To excite the fluorescent light in a crystal, the electron flow having the following characteristics: particle fluence $1.2 * 10^{15}$ m^{-2}; energy 50 ÷ 75 keV, was applied, The whole experiment has been pursued in a vacuum with pressure below $1.3 * 10^{-2}$ Pa. For radiation detection of crystals, antimony-cesium photocells "F-4" and "F-13" were used, the waverange of which covered the entire visible region and capturing part of the ultraviolet and infrared light. The photoluminescence spectra were recorded at a fixed excitation energy, scanning the required spectral range by spectrometer "*spektra*$_1$".

3. Experiment results

To investigate the optical and temporal characteristics of lithium fluoride excited by beta particles with energies of 50-75 keV (at 300 K) an experimental plant was used; the flowchart is shown schematically in Fig. 1. The principle the experiment was as follows. Crystal (1) was irradiated with β-particles after the screen had been opened (4). Irradiation of the crystals was accompanied by luminescence, which was recorded by photocell (3) and spectrometer (8) established in the immediate vicinity of the crystal. A signal from the photo detector was fed to analog-to-digital converter (5), and then to a computer. Simultaneously with the luminescence registration, the potential of ionic crystal was measured with the use of electrodes embedded in insulating substrate (2), which was also recorded by the computer.

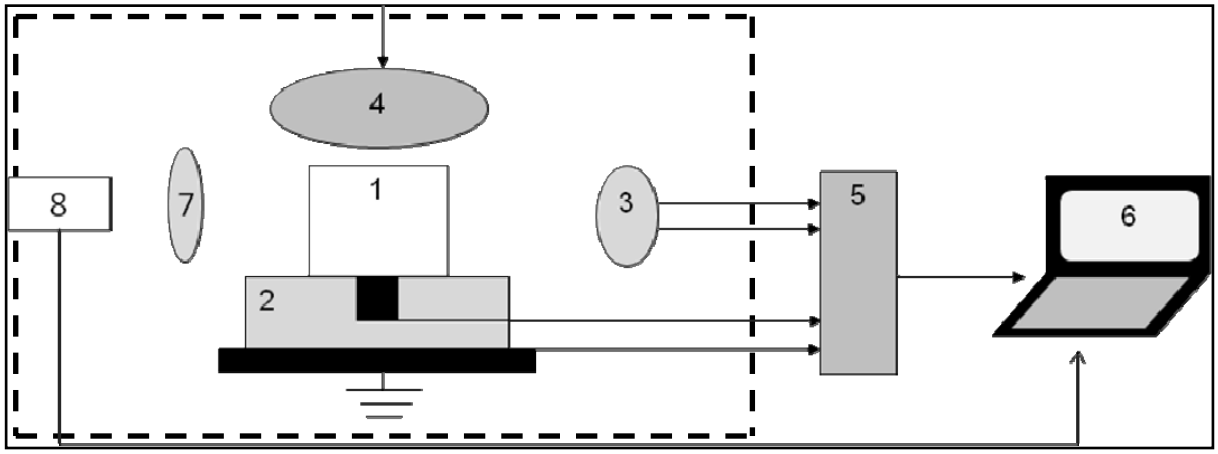

Fig. 1. Block diagram of the apparatus for investigation of luminescence in the visible spectrum.
1) ionic crystal; 2) insulator with built-in electrode; 3) solar cell;
4) screen, overlapping β-particle flux; 5) A to D converter;
6) personal computer; 7) lens; 8) spectrometer "spektra$_1$".

During the experiment it was found that along with cathodoluminescence of lithium fluoride crystals, light flashes appeared in the visible range. After one hour irradiation the frequency of outbreaks reduced by half [2]. In addition, it was found that the buildup and luminescence quenching was accompanied by accumulation and discharge capacities of the crystal (Fig. 2).

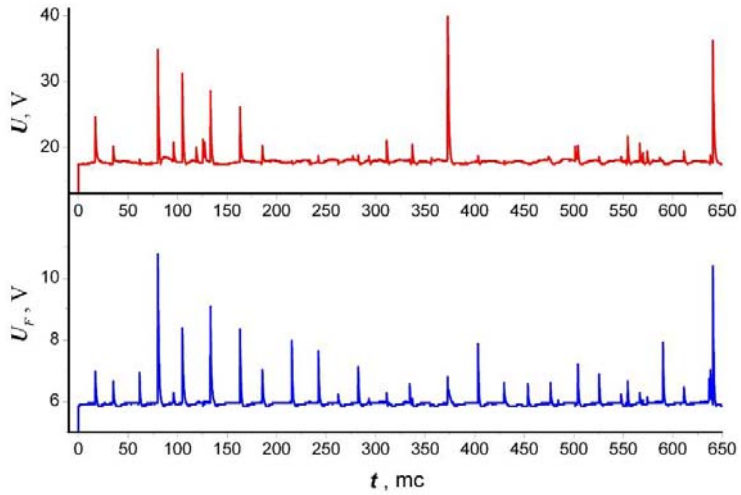

Fig. 2. Volt-brightness characteristic of luminescence; here the ionic crystal potential (above), the yield of luminescence (bottom).

However, as seen from Fig. 2. not all of the radiation peaks are associated with the crystal potential. At the time of outbreaks, the spectrometer registered the following (Tables 1 and 2).

Table 1. Spectral composition of a luminescence peak at the beginning of exposure.

λ, nm	380	410	480	520	570	630	740	780	810	860	910
I, %		0,15			0,1	0,4	0,35	0,3		0,1	0,17
						0,3	0,37	0,22		0,17	0,18
	0,12	0,16			0,26		0,28	0,24		0,28	0,24

Table 2. Spectral composition of a luminescence peak at the end of exposure.

λ, nm	380	410	480	530	560	650	740	790	820	830	920
I, %		0,35	0,23	0,31	0,28	0,31	0,28	0,23	0,26	0,45	
	0,24	0,32			0,2	0,27	0,25	0,32	0,25	0,2	0,27
	0,32	0,32	0,2	0,27	0,27	0,22	0,18	0,32	0,27	0,34	0,27

The tables show the characteristic values of the spectral composition for the flashes at the beginning and end of exposure. The average time interval of a registration is about 10 ms.

4. Discussion

Ionizing irradiation of the AH crystals produces high-energy electron-hole pairs. Later on these pairs transform to low-energy electronic excitations. During this process some energy is released, a part being transmitted to impurities and a part going to crystal heating. The latter causes delocalization of the defects, and then their recombination with complementary defects. The energy released during the relaxation and recombination of electron excitations goes not only into the electron-phonon interaction but also for generation of point defects and formation of several types of color centers in the selfsame crystal. Almost the whole of the wavelengths (Tables 1 and 2) can be associated with the emission of color-centers in the normal or ionized states, including also the impurity color-centers, except for the single peak with a wavelength of 380 nm (3.3 eV).

Table 3. Wavelength of absorption and luminescence of color centers [3].

Color-center	Absorption bands nm	Luminescence bands nm
F	240, 250	–
F^{2-}	960	670, 1120, 1150
F^{2}	440, 445	–
F^{2+}	645	900
F^{3-}	790	–
F^{3}	295, 306, 333	–
F^{3+}	–	530, 540
V	340, 348	–

It is known that at high excitation densities, induced by an electron beam, the recombination radiation associated with induced defects is an order of magnitude greater than the intrinsic emission [4]. In ionic crystals auto localization of excitons is mainly determined by the electron-phonon interaction estimated by Uhrbach rule. According the rule, the coefficient "α" of the fundamental absorption of matter depends on a temperature and photon energy. An important parameter is the steepness of the long-wavelength edge, σ_0 , of the fundamental absorption. If $\sigma_0>1$, we have a weak exciton-phonon interaction and free excitons; while $\sigma_0<1$, there is a strong exciton-phonon interaction and auto localization of excitons in a crystal.

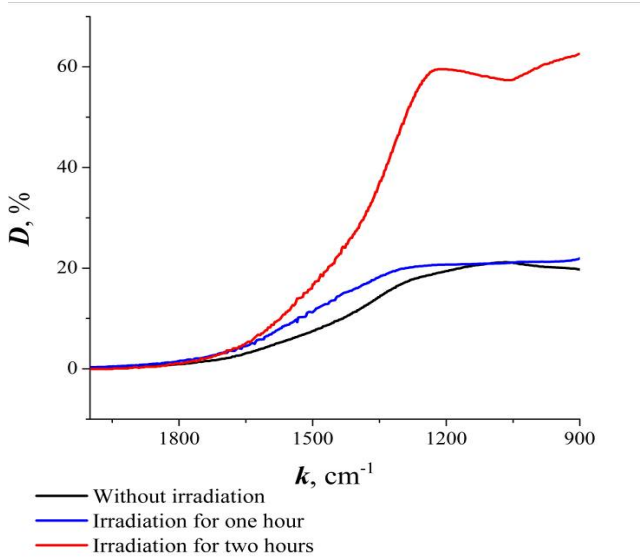

Fig. 4. Transmission spectrum of lithium fluoride; here *D* is the optical density, *k* is the wave number.

In the AHC, luminescence occurs according to the pure exciton mechanism and also according to the recombination of localized charge carriers [5]. Relaxation of the electron excitation, which forms Uhrbach's edge, proceeds through two channels and leads to the formation of various centers of the intrinsic luminescence. Figure 4 shows that the long-wave absorption edge in the control specimen and the specimen irradiated for one hour is markedly different from the sample irradiated for 2 hours, so it can be assumed that the type of dominant interaction has changed.

5. Model of electroluminescence

Experiment shows [1, 2] that the electroluminescence centers are initiated due to impact ionization. The impact ionization threshold is above the photon energy emitted by electroluminescence (3.3 eV). It follows that the luminescence of a band at 3.3 eV may be due to recombination of the excited electrons during the impact ionization. One would assume the existence of recombination step, which includes radiative transition 3.3 eV, implies the existence of levels in the forbidden zone, at a distance of 3.3 eV either from the valence band top or from the conduction band bottom. However, in reality, the excitation energy level is possibly above this value, and then the recombination must take place in several stages, one of which is the radiation in the band 3.3 eV.

We can now define a model of electroluminescence. At the beginning, formation of luminescence center and appearance of the levels in the band gap is associated with impurities or color centers, since neither monoenergetic beam of particles, no all the electrons can cause impact ionization, but only those having energies equal to or greater than the threshold. In this case, there is the excitation of the luminescence center, and then the emission of a photon 3.3 eV takes place. Initiated luminescence center can be excited both with the absorption of the electron excitation and without. Recombination is accompanied by the emission of a photon with an energy of 3.3 eV [6, 7].

The model uses the theory of electroluminescence by impact ionization [8]. However, if the levels are created in the forbidden zone of a crystal, the luminescence is also possible. Electroluminescence as a mechanism that reduces the average energy and number of carriers has a significant influence on the dynamics of a current, which is determined by the balance between processes that increase the number of carriers and compete with them. The main and the only significant process increasing the number of carriers is the impact ionization. In the case of a positive balance between impact ionization and competing processes, structure will be destroyed. Avalanche increase of the carrier number due to impact ionization is regarded as the main cause of dielectric breakdown in the Keldysh model [9]. Competing with impact ionization processes are divided into two groups. The first group includes processes that reduce the number of carriers (hole – electron recombination). The second group incorporates the processes with energy release due to scattering which is not accompanied by recombination (Fig. 2). The more acts of the impact ionization occurs, the more luminescent centers is initiated. The luminescence centers increase entails an increase in acts of electroluminescence which are not accompanied by recombination. In this case hot charge carriers, which can caused the glow, drop energy and cannot cause impact ionization.

6. Model of recombination luminescence

The basic model of the fracture of radiation coloring is considered to be recombination of electrons with their own holes producing color centers [10]. In addition, recombination of electrons with the impurity centers overcharged under the action of ionizing radiation is possible. If the concentration of rare-earth centers is low, they do not interact with each other. The absorption and emission will occur at each site independently of other centers. When after the light absorption, the optical center moves to the state with a greater energy, further transformation of this energy depends on the ratio between the probability of radiative and non radiative transitions from a state. After exciting light absorption the center usually falls into a state for which the non radiative transition to a low-lying level is the most probable. The non radiative transition occurs as long as the optical center is in no position for which the probability of non radiation transition is less than for a radiative transition. The radiative transition in the visible optical range is possible with energy which is several times more than the energy of vibrational quanta. Therefore due to non radiation transition, several vibrational quanta are produced.

In the case of low concentration of optical centers, there are only two ways of deactivation of the excited states: the emission of light and transformation of the vibration excitation in a center of the matrix. The situation changes with increasing a center concentration. In this case, the interaction of centers with each other occurs, during which there is the transfer of excitation. The excitation transfer reduces the quantum yield and time of luminescence decay of a donor. In this case, part of the donor excitation goes to an acceptor and is not transformed into radiation. If the donor gets excited to the metastable level, the excitation transfer is the cause of acceptor luminescence. The mechanism of excitation transfer between optical centers explains the microscopic model proposed and developed in the work of Forster and Dexter [11], where it is assume that there is electrostatic interaction between the centers. Sometimes, during experimental studies there is a problem of the radiative and radiation transfer mechanisms. To solve this problem, an extremely strong dependence of the non radiative transfer on the distance between the donor and acceptor is used. The probability of radiative transfer does not depend on this distance. Radiative transfer can also take place and for samples with low concentration of optical centers.

7. Conclusion

From the results obtained it follows that under chosen conditions of influence, there are several types of luminescence; the first is responsible for the steady-state type of luminescence, cathodoluminescence, the second of a pulse-type is characterized by a low-intensity electroluminescence, the third type is characterized by increased intensity compared to the second related to the relaxation of electronic excitation and destruction of color centers.

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