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Zhanturina N.[*], Aimaganbetova Z., Sagimbaeva Sh.

K. Zhubanov Aktobe Regional State University,
Kazakhstan, Aktobe, [*]e-mail: nzhanturina@mail.ru

ABOUT THE ORIGIN OF THE EMISSION BANDS IN THE WAVELENGTH RANGE 320-600 NM IN KBR CRYSTAL AT LOW TEMPERATURES

In this paper the results of the registering of X-ray luminescence spectra of KBr crystal at the temperature range 10-325 K were presented. The spectrum of X-ray crystal KBr is composed of three bands – at ~ 280 nm (~ 4.44 eV) ~ 340 nm (~ 3.65 eV) and ~ 490 nm (~ 2.54 eV). The peak at 490 nm – ϖ-luminescence from the triplet state excitons. In article the temperature dependence of this band was explained by the theory of excitons self-trapping. The luminescence intensity of this band increases to 25 K then decreases. We have modeled by the continual theory that the potential barrier height of excitons self-trapping is minimal at a temperature of 25 K. So the theory confirms with experiment. The band at 280 nm eV – σ – luminescence from the singlet state exciton, reaches its maximum at low temperatures. The nature of the band at 340 nm eV is associated with the creation of excitons with more "symmetric" configuration than «strong-off».

Key words: exciton, KBr, band, luminescence, temperature dependence, configuration, self-trapping.

Жантурина Н.Н.[*], Аймаганбетова З.К., Сагимбаева Ш.Ж.

Қ. Жұбанов атындағы Ақтөбе өңірлік мемлекеттік университеті,
Қазақстан, Ақтөбе қ. [*]e-mail: nzhanturina@mail.ru

Төменгі температураларда KBr кристалында 320-600 нм интервалында жарқырау жолағының табиғаты туралы

Бұл мақалада KBr кристалының 10-325 K температуралық диапазонында рентгендік люминесценция спектрін тіркеуінің нәтижелері келтірілген. KBr кристалының рентгендік люминесценция спектрі үш жолақтан тұрады: ~ 280 нм (~ 4.44 эВ), ~ 340 нм (~ 3.65 эВ) және ~ 490 нм (~ 2,54 эВ). 490 нм-дегі шың – ϖ-люминесценцияға сәйкес. Мақалада осы жолақтың температуралық тәуелділігі экситондардың автолокализациясы теориясымен түсіндіріледі. Бұл жолақтың люминесценциясының интенсивтілігі 25 K-ге дейін артады, содан кейін азаяды. Экситондардың қармалу бөгетін әртүрлі температурада модельдеу нәтижесінде, биіктігі 25 K температура кезінде минималды болып келеді. Осылайша, теория экспериментпен расталады.

Түйін сөздер: экситон, KBr, жолақ, люминесценция, температуралық тәуелділік, конфигурация, автолокализация.

Жантурина Н.Н.[*], Аймаганбетова З.К., Сагимбаева Ш.Ж.

Актюбинский региональный государственный университет им. К. Жубанова,
Казахстан, г. Актобе, [*]e-mail: nzhanturina@mail.ru

О происхождении полосы излучения в интервале 320-600 нм в кристалле KBr при низких температурах

В настоящей работе представлены результаты регистрации спектров рентгенолюминесценции кристалла KBr в интервале температур 10-325 K. Спектр рентгенолюминесценции кристалла KBr состоит из трех полос – при ~ 280 нм (~ 4,44 эВ), ~ 340 нм (~ 3,65 эВ) и ~ 490 нм

(~ 2,54 эВ). Пик при 490 нм – ϖ-люминесценция экситонов из триплетного состояния. В статье температурная зависимость этой полосы объясняется теорией автолокализации экситонов. Интенсивность люминесценции этой полосы возрастает до 25 K, а затем уменьшается. В результате моделирования процесса автолокализации экситонов при различных температурах высота потенциального барьера экситонов минимальна при температуре 25 K. Таким образом, теория подтверждается экспериментом.

Ключевые слова: экситон, KBr, полоса, люминесценция, температурная зависимость, конфигурация, автолокализация.

Introduction

Alkali halide crystals (AHC) are members of a broad class of scintillation and dosimetric materials, in which the action of radiation, especially X-ray, is the emergence of electronic excitations relaxed, subsequently, on the radiative and nonradiative channels [1, 2]. The radiation relaxation of AHC is caused by the self-trapping of excitons in regular lattice sites due to the exciton-phonon interaction. It depends on different factors, among them are impurities, hydrostatic and uniaxial pressure, point defects [3]. The consequences of nonradiative relaxation are creation of F-centers, H-centers and other defects and their complexes. In AHC the bands of absorption characteristic for most of defects are widely researched [4]. The result of radiative annihilation during the exciton self-trapping in regular lattice sites is the luminesce. Studies of the luminescence of AHC under different conditions allows to determine the nature of the emission bands, as well as the annihilation of electron excitations and their interaction with defects [5]. Resently, besides alkali halide crystals are used as a scintillation materials, dosimetric materials, optical medium and simply model objects for many theoretical researches, their clasters became useful for the creation of batteries or as an ideal gas phase models for sea salt aerosols, which are essential components of atmospheric chemistry in marine regions. In this field the Infrared multiple photon dissociation is interesting method [6]. Recently, theoretical studies of the energy of electronic excitations in various states allow us to determine many characteristics of scintillation detectors, optical devices based on alkali halide crystals. For example, in the works, the binding energies of LiF clusters were calculated and this made it possible to determine the durability of lithium fluoride batteries [7,8].

It is known that the nature of the luminescence of AHC is radiation dihalogen self-trapped exciton (STE) by reducing the mean free path monohalogen exciton. With increasing temperature, the intensity of the luminescence is quenched by the exciton transition from a free state to a self-localized [9,10]. In the study of X-ray spectra of alkali halide crystals at low temperatures, special attention was paid to the nature and regularity of the main emission in the area of ~ 280 nm (~ 4.44 eV), which contribute to the integral luminescence of AHC, but has not yet been thoroughly studied spectra with low intensity ranges ~ wavelengths 320-360 nm (~ 3,88-3,45 eV) and ~ 450-560 nm (~ 2,76-2,22 eV) at temperatures from 10 to 85 K. A study changes in the intensity of these peaks can give information on the nature of the electronic excitations in AHC. Also, there was an interest to study the spectra of X-ray crystal KBr in connection with the discovery of the effect of the creation of excitons with "more symmetric" configuration in comparison with «strong-off» under the influence of uniaxial elastic compression up to 1.2% in AHC [11].

Samples

Samples of crystal KBr, used in the work were grown at the Institute of Physics of the University of Tartu in Estonia. A complex method comprising the steps of purification and cultivation of AHC:

1. Cleaning the original crystals from oxygen-containing impurities and OH;
2. Cleaning of the original crystals with impurities homologues zone melting method;
3. The multiple zone-refining of polyvalent metals in vacuum and subsequent cultivation in a vial at its passage of the last zone or again growth by Kyropoulos method in a helium atmosphere [12].

KBr crystals were obtained from the zone-refined raw materials by Kyropoulos in a helium atmosphere. This crystal was subjected to 60-fold zone melting. The impurity content of 10^{-6}-10^{-8} and atmospheric admixture of Rb in the amount of 10^{-5} [13].

Experiment

The experiment was done in the Nicolaus Copernicus University in Torun.

X-ray source series «Inel» with a copper anode allows for a maximum voltage of 60 kV. Operating voltage / current during the experiment was 45 kV / 10 mA. The X-ray generator has a water cooling system, his work is automated. With the help of cryogenic helium cooling apparatus with temperature controller 330 LakeShore possible to control the temperature from 10 to 325 K. The data are processed by a special program, the files are saved in ASCII [14,15].

As can be seen from Fig. 1, the spectrum of X-ray crystal KBr is composed of three bands – at ~ 280 nm (~ 4.44 eV) ~ 340 nm (~ 3.65 eV) and ~ 490 nm (~ 2.54 eV). It is known that the peak at 2.54 eV – π-luminescence from the triplet state excitons, at 4.44 eV – σ – luminescence from the singlet state exciton, reaches its maximum at low temperatures [16]. Intensity of σ – luminescence reaches its maximum at low temperatures and quenched starts at high temperature (Fig. 1). So, at 80 K, the intensity of the σ – emission is approximately 50 times weaker than that at 25 K (Fig. 1).

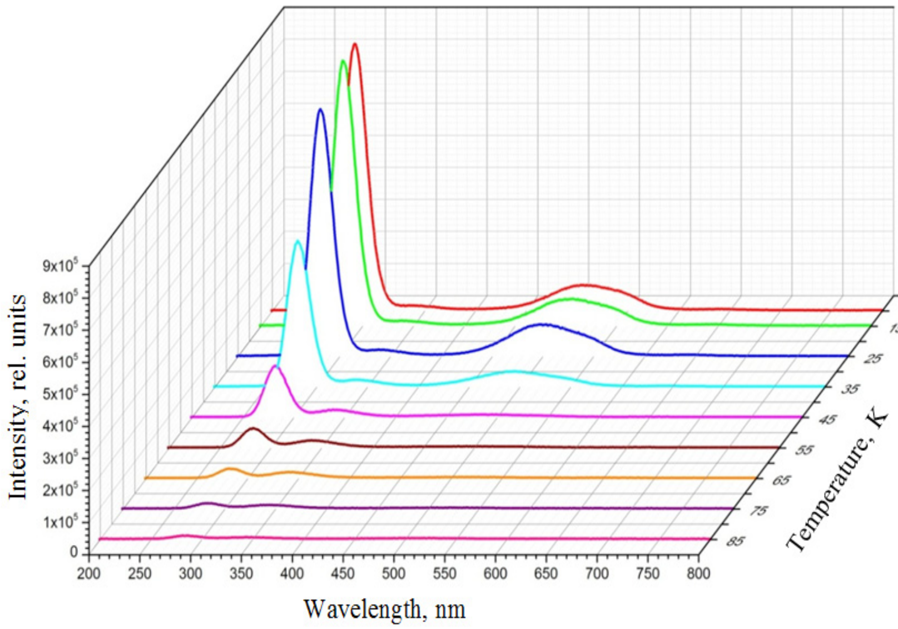

Figure 1 – The spectrum of X-ray crystal KBr in the temperature range 10-85 K

Temperature dependences of the intensities of the emission bands at ~ 340 nm (~ 3.65 eV) and ~ 490 nm (~ 2.54 eV) are shown in Fig. 2, 3.

The intensity of the luminescence of KBr at 340 nm is clearly observed at temperatures of 10-65 K and is in a high-energy part of the spectrum than the π-luminescence. Its intensity increases with temperature up to 50 K. Then, the intensity of this band begins to decrease.

Let's consider the nature of the band at 490 nm. There are passport details of this band that it corresponds to the radiation of self-trapped exciton [17]. And, for the π-emission of KBr released the following pattern: as the temperature increases from 10 K, the intensity starts to increase to a temperature near 30 K, and then decreases. This experimental result confirms our excellent model of self-trapping of excitons depending on the temperature [18].

Discussion

Analyzing the possible mechanisms of emission in KBr crystal at low temperatures and taking into account the results of studies of the luminescence of alkali halide crystals obtained under the influence of uniaxial strain, we formulate our arguments.

Apparently, the nature of the band at 340 nm is associated with the creation of excitons with more "symmetric" configuration than «strong-off», because:

– Radiation occurs in the high part of the spectrum than the π-emission;

– Under the influence of uniaxial stress in alkali halide crystals excitons with more "symmetric" configuration than the «strong-off» are created, and the impact of uniaxial strain is similar to the effects of low temperatures;

– In the literature is indicated recombination luminescence mechanism at low temperatures in crystals with a high yield of radiation defect formation [19];

– It is shown that at low temperatures the I – centers recombine with electron centers (F – centers) by the scheme:

$$F\left(\upsilon_a^+ e^-\right) + I\left(i_a^-\right) \rightarrow e^-$$

resulting in recombination luminescence of STE.

$$e^- + e_s^+ \rightarrow e_s^0 \rightarrow h\nu \,.$$

– Increase in the intensity of the luminescence to a temperature of approximately 50 K with its further decrease indicates the presence of a barrier for self-trapping of excitons to create STE of more "symmetric" configuration than «strong-off» [20].

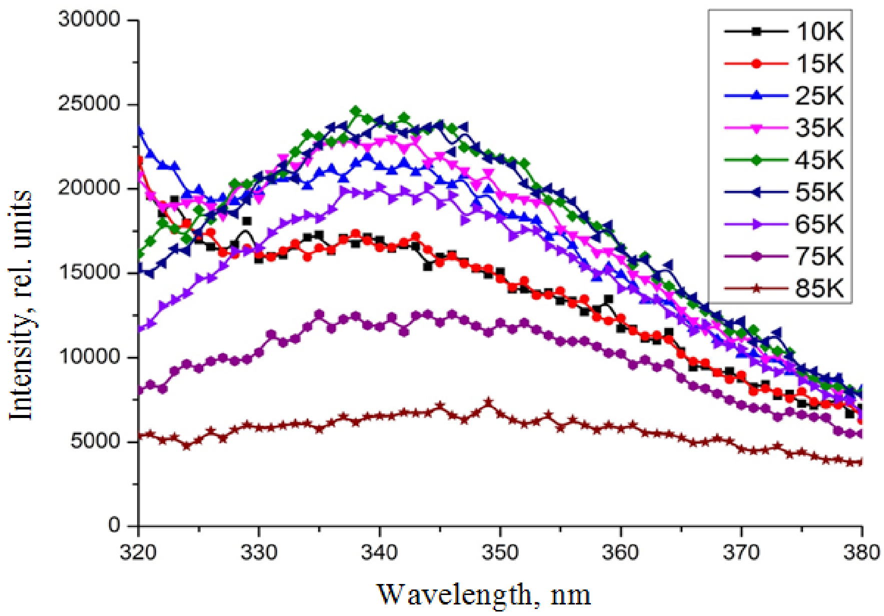

Figure 2 – Temperature dependence of the luminescence band at ~ 340 nm (3.65 eV) in KBr crystal

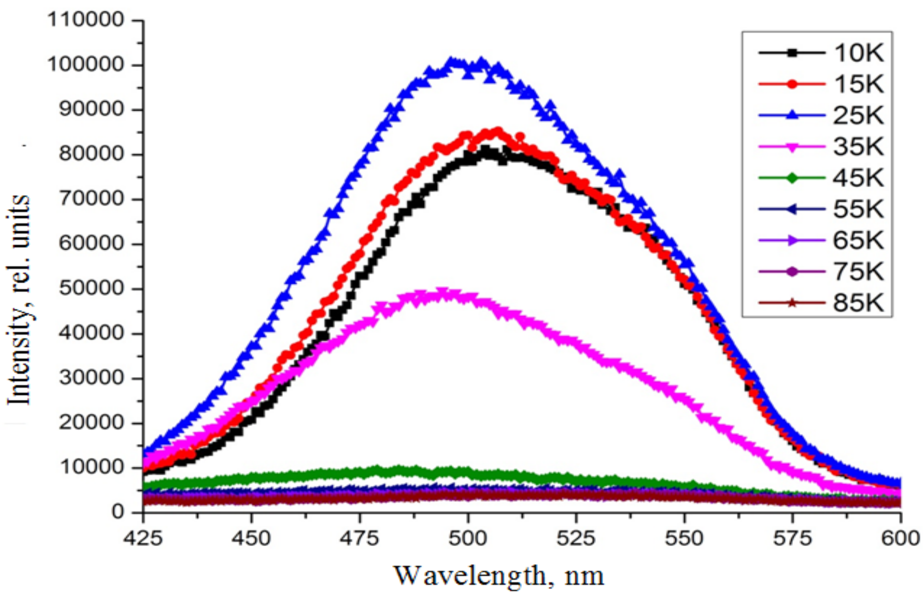

Figure 3 – Temperature dependence of the luminescence band at ~ 490 nm (~ 2.54 eV) in KBr crystal

However, since the radiation of STE occurs in KBr at 340 nm is apparently created STE with different configuration compared to «strong-off».

As for the patterns of change in the intensity of the luminescence band with the temperature at 490 nm, it can be argued that the luminescence intensity increases during the transition to self-trapped exciton state, this corresponds to a temperature of about 25 K, and then after the process of self-trapping of excitons the intensity of π- emission begins to fade.

In the simulation, the temperature dependence of the height of the potential barrier trapping of excitons in crystals KI, KBr based on the adiabatic potentials methods was found that the potential barrier has a minimum in KBr crystal at a temperature of about 25 K, in crystal KI – about 65 K. Here highlights of this theoretical investigation [21].

The nature of the changes the potential barrier height of self-trapping of excitons can be analyzed on the surfaces of the adiabatic potential (AP) excitons in alkali halide crystals:

$$E(\mu) = A\mu^2 - B\mu^3 - C\mu, \tag{1}$$

where μ is a variational parameter, varying from 0.1 to 1 and equal to the ratio of the lattice constant to the radius of the area of self-trapping of the exciton,

$$A = \frac{3\pi\hbar^2}{2m_h^* a_0^2}, \quad B = \frac{E_d^2}{2\beta a_0^3}, \quad C = \frac{e^2}{\tilde{\varepsilon} a_0}$$

determine respectively the kinetic energy of the exciton energy relaxation of the lattice, and the lattice polarization (the interaction of holes with optical lattice vibrations) [9]. The energy difference between the maximum and minimum of the expression is the height of the potential barrier of self-trapping of excitons [22, 23].

In the simulation, the temperature dependence of the height of the potential barrier of self-trapping of excitons we took into account two main factors – a lattice vibrations and changes in the lattice constant, which is part of every member in the functionality of the adiabatic potential. Lattice vibrations affect the kinetic energy of an exciton, resulting in a decrease of the quantity $D = \sqrt{2Bk_Б T}$,which is called the fluctuation potential [24].

The temperature dependence of the lattice constant in a first approximation, we find the following. At sufficiently low temperatures, when the energy of thermal motion $k_A T$ much smaller than the width of the exciton band A majority of excitons located in the wave vector space, with increasing temperature change of the wave vector of the exciton, and its deviation from the initial value χ can be found by the following formula:

$$k = k_0 - \chi\,. \tag{2}$$

Considering the non-degenerate exciton gas electron gas [25] applying the law of conservation of energy Boltzmann statistics, the deviation of the wave vector of the initial value can be written as the following expression:

$$\chi = \frac{j}{\hbar}\sqrt{3mk_Б T}\,, \tag{3}$$

where j is parameter depending on the thermal effects on or interatomic bond distances. For alkali crystals it ranges from 0.01 to 0.1.

Using expressions (2), (3) the temperature dependence of the lattice constant is written as

$$a = \frac{a_0\hbar\sqrt{\pi}}{\hbar\sqrt{\pi} - a_0 j\sqrt{mk_б T}}\,, \tag{4}$$

where $\dot{a}_0$ is lattice constant at 4,2 K.

So, by the expression $E(\mu) = (A - D)\mu^2 - B\mu^3 - C\mu$ we built the adiabatic potential surface excitons in KBr crystals at different temperatures.

As can be seen from this figure the potential barrier reaches a minimum at a temperature of about 25 K in the crystal KBr. Consequently, in KBr crystal near 25 K the luminescence intensity reaches its maximum with further quenching.

For crystal KI, for example, it is known x-shaped luminescence intensity of self-trapped excitons dependence on temperature, which confirms our theoretical results. For crystal KBr the intensity of the luminescence of self-trapped excitons on temperature has not been studied in detail experimentally due to the fact that this crystal is not effective scintillator.

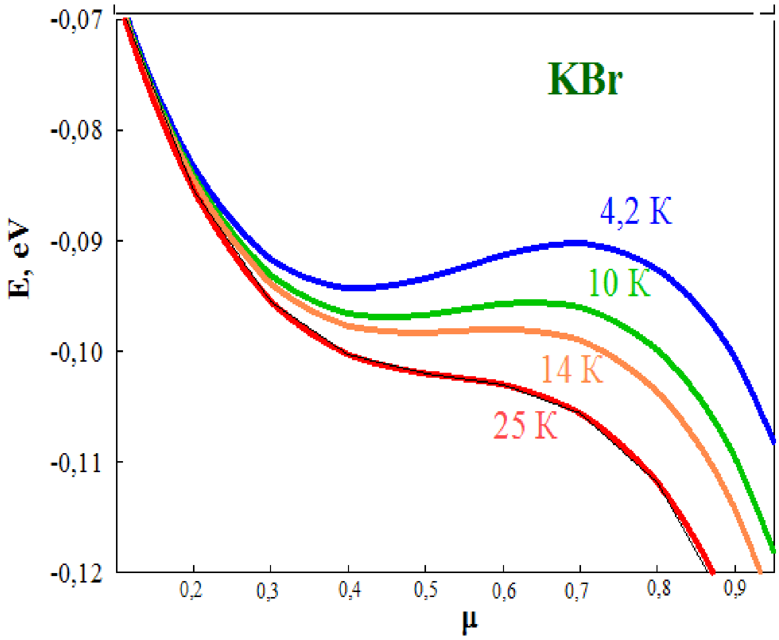

Figure 4 – Adiabatic potential surfaces of excitons in KBr at different temperatures

Conclusion

Thus, as a result of experimental study of X-ray luminescence spectra in KBr crystal in temperature range 10-90 K, we obtained the following findings:

1) The spectrum of X-ray luminescence of KBr crystal at low temperatures was studied and the nature of the luminescence band with a maximum at 340 nm was interpreted, which intensity increases to a temperature of about 50 K, the nature of which corresponds to the emission of self-trapped exciton with more “symmetric” configuration than «strong -off», created using the tunneling charge of radiation defects at low temperatures.

2) The effect of increasing the luminescence intensity of self-trapped excitons in KBr crystal (490 nm) to about 25 K, corresponding to the temperature at which the potential barrier of excitons self-trapping reaches a minimum value, has been experimentally discovered and theoretically interpreted.

The results obtained by measuring the X-ray spectra in alkali halide crystals at low temperatures allow us to explain the connection between the self-trapping of excitons and the intensity of the luminescence, efficiency of radiation defect formation as well as to explain the regularities of luminescence by the theory of electronic excitations.

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