The rate of the exciton self-trapping in KI and RbI at different temperatures

N Zhanturina and K Shunkeev
Aktobe State Pedagogical Institute, 34 Moldagulova avenue, Aktobe, Kazakhstan
E-mail: nzhanturina@mail.ru

Abstract. The article disclosed the theory of the kinetics of excitons self-trapping in alkali halide crystals. On the example of KI and RbI crystals the time of excitons self-trapping and the length of free path before self-trapping were calculated. Also, the theory of self-trapping rate was revealed. According to the Arrhenius law the dependence of excitons self-trapping rate on the temperature and the degree of depth and uniaxial compression was analyzed. The increase of the rate of excitons self-trapping by temperature increasing was shown; while increasing the degree of compression at the full rate of strain localization decreases under uniaxial - is increasing. These data are good agreed with the experimental fact of luminescence increasing under uniaxial compression and allow to make a conclusion about weakening of the luminescence by applying hydrostatic pressure

1. Introduction
The progress in the development of modern ideas about physics of radiation-induced processes in ionic crystals is made by the development of rapid methods of registration. It is established that the formation of primary radiation defects occurs mainly in nonradiative decay of electronic excitations in the intact lattice. Despite that subpikosecond-range of measurements has been already mastered, such kinetic parameters as relaxation rate of self-trapped excitons, the luminescence time, the time of defects formation at different temperatures are still studied very poorly [1].

2. The height of excitons self trapping barrier
In some crystals quasiparticles (excitons) may exist in a free and in a self-trapped state. Excitons are produced by light in a free state, but they have self-trapped the time to decay, breaking barrier of self-trapping. These states are separated by a self-trapping barrier. Excitons are produced by light in a free state, but they have self-trapped the time to decay, breaking barrier of self-trapping. Kinetics of excitons self-trapping in alkali halide crystals plays an important role, since the formation of defects passes directly through the stage of localization, and the time, the rate of electronic excitations relaxation are directly dependent on the magnitude of the potential barrier of the excitons self-trapping [4]. The potential barrier height is determined by the formula

$$E = \frac{4A^3}{27B^2}(1 - 3\frac{BC}{A^2})^{3/2},$$

where A, B, C - a kinetic energy of the electron, an energy of the lattice relaxation, a contribution of optical phonons to the lattice relaxation energy, respectively [2].
Table 1 shows the calculated potential barrier of excitons self-trapping in alkali halide crystals and corresponding values of the parameters A, B, C.

| Crystal | A(eV) | B(eV) | C(eV) | E(eV) |
|---------|-------|-------|-------|-------|
| KBr     | 0.77  | 0.79  | 0.44  | 0.001 |
| NaI     | 0.82  | 0.63  | 0.24  | 0.033 |
| KI      | 0.66  | 0.316 | 0.376 | 0.026 |
| Rbl     | 0.52  | 0.239 | 0.348 | 0.016 |
| CsI     | 1.25  | 0.34  | 0.56  | 0.009 |

3. The kinetics of excitons self-trapping in alkali halide crystals

Mechanism of overcoming of the self-trapped barrier by excitons in AHC is quite different at high and low temperatures. In the first area it is activated, which corresponds to the Arrhenius law, in the second one - the tunnel mechanism operates, the temperature dependence of the rate is determined by the dependence of optimal tunneling path in configuration space. Value which depends on the temperature and determines the distribution of channel is the rate of excitons self-trapping [3].

According to [1] the excitons self-trapping's rate is the value $\omega = 1/\tau$ where the $\tau$ is the lifetime of the free exciton, which is defined by the parameters of the lattice and the temperature. At the same time, the exciton in alkali relaxes through two channels and the annihilation rate is defined as

$$\tau = \left(\tau^{-1} + \omega\right)^{-1},$$

where $\tau^{-1}$ is the rate of radiative annihilation, which does not depend on temperature, and $\omega$ is the rate of radiative annihilation determined by the Arrhenius law.

$$\omega = \omega_0 e^{-E/T},$$

where $\omega_0$ is defined as the rate of self-trapping at low temperatures, e.i. the value is inversely proportional to the time of excitons self-trapping at low temperatures.

From theoretical calculations, the calculation of the Hamiltonian system of exciton-lattice given in [2], the lifetime of the free exciton is

$$\tau \leq \frac{\hbar}{A},$$

where $A$ is the half-width of the exciton band.

The length of a free path is defined as

$$l = \frac{\hbar k}{m} \tau,$$

where $k = 10^6$ is the wave number of alkali halide crystals and the $m$ – is the effective mass of exciton in alkali halide crystals [4].
In the crystal RbI the scientists Song and Tanimura were able to calculate the time of luminescence 200±50 ps, from the time they took the time of exciton self-trapping 2±0.5 ns. In the crystal KI the corresponding times were calculated as 100±50 ps and 160±180ps. [3]

Following the experimental and theoretical data we have analyzed that the lifetime of free excitons is decreasing in the sequence RbI, KCl, KI, KBr, NaBr, NaI, CsI. Experimentally calculated lifetime of free excitons in the crystal KI is an interesting fact in connection with the fact that in this crystal the potential barrier separating the free and self-trapped exciton state is one of the largest. As the free-exciton lifetime depends on the width of the exciton band A in KI. The exciton band width is more than in RbI, so self-trapping of excitons occurs faster.

From the graph we can see that in the crystal KI excitons self-trapping rate changes faster while increasing temperature, which is associated with a significant width of the exciton band in this crystal. Using (5) and (6) and some experimental values of these parameters we have been able to calculate the value of the lifetime of free excitons and exciton length of free path before self-trapping in some alkali halide crystals [5].

The theoretical values of the lifetime of free excitons in alkali halide crystals which are calculated by us is similar to the experimental data that the length of free path of excitons in KI is several tens of lattice constants, and in KBr is one of the lowest values of the potential barrier of excitons self-trapping.

Table 2. Mechanical parameters of relaxation of excitons in alkali halide crystals

| Crystal | Time of the free exciton (ns) | The length of the free path (Å) | General exciton’s self-trapping rate ($10^7$ s$^{-1}$) |
|---------|-------------------------------|---------------------------------|---------------------------------|
| KI      | 1.6                           | 17000                           | 380                             |
| RbI     | 2                             | 191000                          | 45                              |
| KBr     | 0.0018                        | 2                               | -                               |

Uniaxial compression reduces the potential barrier of self-trapping of excitons in a crystal. The coefficients A, B, C which determine the potential barrier of self-trapping of excitons depends on the degree of compression:

$$A_u = \frac{2 + 1/\varepsilon^2}{3}, \quad B_u = \frac{B}{\varepsilon^{7/3}}, \quad C_u = \frac{2 + \varepsilon}{3} C.$$ (6)

There are interesting effects at the hydrostatic pressure of the crystals. In this case the lattice constant decreases smoothly with the increasing of a compression degree the potential barrier in alkali halide crystals have to grow [8].

In the case of hydrostatic compression coefficients take the form

$$A_u = \frac{A}{\varepsilon^2}, \quad B_u = \frac{B}{\varepsilon^7}, \quad C_u = \frac{C}{\varepsilon}.$$ (7)

Using the calculations presented above and experimental data on the excitons self-trapping time we have analyzed the dynamics of the self-trapping barrier at different temperatures and the function of hydrostatic compression and uniaxial deformation. The excitons self-trapping rate decreases at hydrostatic pressure with increasing compression, and in RbI the deformation significantly affects the dynamics of exciton’s self trapping barrier; at uniaxial one - increases.[6]

Upon analyzing the Arrhenius law it is not difficult to notice the increase of the rate of excitons self-trapping with increasing temperature. In the crystal RbI the rate of excitons self-trapping is changing faster than the crystal KI, which is associated with a smaller width of the exciton band. At low
temperatures the rate of excitons self-trapping increases significantly, further increasing the temperature increment of the function decreases [7].

![Figure 1](image1.png)

**Figure 1.** Dependence of the excitons self-trapping rate in crystals KI and RbI on the degree of hydrostatic compression at 77 K

![Figure 2](image2.png)

**Figure 2.** Dependence of the excitons self-trapping rate in crystals KI and RbI on the degree of uniaxial deformation at 77 K

4. Conclusion
With the help of theoretical calculations we calculated the lifetime of free excitons and free path length in the crystals KI, RbI, KBr; to investigate the dependence of the exciton self-trapping time on the temperature and the degree of compression [8]. The increase of excitons self-trapping rate by the action of uniaxial compression is confirmed by the experimental fact of amplification of the luminescence of self-trapped excitons in alkali halide crystals under uniaxial deformation [9]. Because of this, analyzed dependence of excitons self-trapping rate in crystals on the degree of compression allows to conclude about weakening of the luminescence under hydrostatic pressure in alkali halide crystals.

Our data are agreed well with the experimental results and allows a deeper study of the kinetics of luminescence decay in the AHC.

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