Self-trapping features of excitons in elastically deformed alkali halides

S. Tulepbergenov [1], K. Shunkeev, I. Spivak-Lavrov

Aktobe State University, 463000 Aktobe, Kazakhstan

Abstract

The self-trapping (ST) features of excitons in alkali halides (AH) are theoretically studied under both uniaxial and hydrostatical compressions. The variational calculations of the ground state energy of interacting exciton-phonon system in elastically deformed AH are carried out within the adiabatic approximation and the continuum model of ionic crystals. The results are discussed in terms of the stress-induced change of the exciton ST barrier. It is shown that the small hydrostatic and uniaxial compressions (which almost do not change the force constants) of AH lead to the decreasing of the exciton ST barrier. But at large hydrostatic pressure the force constants of AH, the bandwidth of excitons and the exciton ST barrier are noticeably increased.

Key words: Self-trapping of exciton; Alkali halides; Elastic deformation

1. Introduction

The lowering of the crystal lattice symmetry induced by uniaxial stress has been expected to affect the efficiency of the different stages of electronic excitations (EE) relaxation in AH (e.g. their migration along the crystal lattice, self-trapping, radiative and non-radiative decay), as well as the structure and the parameters of the self-trapped exciton (STE) states.

The influence of the uniaxial stress on the radiative decay of the STE of various configurations, the non-radiative decay of EE resulting in defects formation, their migration along the crystal lattice and consequent ST have been studied by the methods of luminescence and absorption spectroscopy [1, 2]. The following effects have been found in uniaxial elastic stress field: (i) enhancement of the self-trapping efficiency of free excitons with radiative annihilation and (ii) suppression of the nonradiative decay of excitons into primary radiation defects - F, H pairs. For explaining the effect of intrinsic luminescence enhancement of STE it is necessary to take into account the influence of uniaxial compression on potential barrier, existing between free and self-trapped states of exciton.

The specific features of ST of EE in various materials are essentially different. In particular, the ST of excitons in some AH (i.e. alkali iodides and bromides) occurs with overcoming of the potential barrier and other AH such a barrier is absent [3].

2. The influence of uniaxial stress on potential barrier in the continuum model

In the present paper we develop the continuum theory of self-trapping of EE within the adiabatic approximation elastically stressed AH. In the continuum model of solids the functional of the total energy of interacting exciton-phonon system in the uniaxially stressed ionic crystal just as in the undeformed crystal [4] depends on the dilation $\Delta(r)$ described by the deformation potential of acoustic phonon, the electrostatic potential $\phi(r)$ due to the lattice polarization at optical lattice vibrations and the wave function of ex-

[1] Corresponding author. E-mail: sagyn@rambler.ru

citon chosen for uniaxially stressed crystals in the form

$$\psi_u(\boldsymbol{r}) = A\varepsilon^{-\frac{1}{2}}\exp\left\{-\pi\mu^2\left[\left(\frac{\rho}{a_0}\right)^2+\left(\frac{z}{\varepsilon a_0}\right)^2\right]\right\}, \quad (1)$$

where $A = \left(\mu\sqrt{2}/a_0\right)^{\frac{3}{2}}$, $\rho^2 = x^2+y^2$, ε is the degree of relative uniaxial compression, a_0 is the lattice constant and μ is the variational parameter characterizing the degree of exciton localization.

For the calculations of the ground state energy of exciton (hole)-crystal lattice system the functional of its total energy after minimizing with respect to lattice deformation and polarization has the form

$$E(\psi) = \frac{\hbar^2}{2m^*}\int(\nabla\psi)^2\,d\boldsymbol{r} - \frac{E_d^2}{2\beta}\int\psi^4(\boldsymbol{r})\,d\boldsymbol{r} - $$

$$-\frac{e^2}{2\epsilon^*}\iint\frac{\psi^2(\boldsymbol{r})\psi^2(\boldsymbol{r}')}{|\boldsymbol{r}-\boldsymbol{r}'|}\,d\boldsymbol{r}d\boldsymbol{r}', \qquad (2)$$

where $m^* = m_h^* + m_e^*$ is effective mass of the exciton, β is an elastic constant, $\epsilon^* = \epsilon_\infty/(1-\epsilon_\infty/\epsilon_0)$, E_d is the deformation potential. E_d is estimated as $E_d = \frac{2}{3}E_F$, where E_F is Fermi energy.

Considering $\varepsilon = 1-\delta$ in Eq. (1) and taking as the limit the first power related to small value δ we have the following expression

$$\psi_u(\boldsymbol{r}) = A\exp\left[-\pi\left(\frac{\mu r}{a_0}\right)^2\right]\left(1+\frac{\delta}{2}\left(1-4\pi\frac{\mu^2}{a_0^2}z^2\right)\right).$$

Using this trial wave function, the integral in third term in (2) can be presented in the form

$$\iint\frac{\psi^2(\boldsymbol{r})\psi^2(\boldsymbol{r}')}{|\boldsymbol{r}-\boldsymbol{r}'|}\,d\boldsymbol{r}d\boldsymbol{r}' = J_1(1+2\delta) - 2J_2\delta$$

Here through J_1 and J_2 the following integrals are mentioned

$$J_1 = 32\pi\left(\frac{\mu}{a_0}\right)^6\iint\frac{\exp\left(-2\pi\frac{\mu^2}{a_0^2}\left(r^2+r'^2\right)\right)}{|\boldsymbol{r}-\boldsymbol{r}'|}\,d\boldsymbol{r}d\boldsymbol{r}',$$

$$J_2 = 32\pi\left(\frac{\mu}{a_0}\right)^8\iint\frac{\exp\left(-2\pi\frac{\mu^2}{a_0^2}\left(r^2+r'^2\right)\right)}{|\boldsymbol{r}-\boldsymbol{r}'|}z^2 d\boldsymbol{r}d\boldsymbol{r}'.$$

After integrating by radial variables the following results are obtained $J_1 = 2\frac{\mu}{a_0}$ and $J_2 = \frac{7}{3}\frac{\mu}{a_0}$.

Thus, calculation of ground state energy gives

$$E_u(\mu) = A_u\mu^2 - B_u\mu^3 - C_u\mu, \qquad (3)$$

where $A_u = \frac{\left(2+\frac{1}{\varepsilon^2}\right)\pi\hbar^2}{2m^* a_0^2}$, $B_u = \frac{E_d^2}{2\beta a_0^3\varepsilon}$, $C_u = \frac{e^2(2+\varepsilon)}{3\epsilon^* a_0}$.

The dependence of the potential barrier between free (F) and self-trapped (S) states on the degree of relative uniaxial compression is given by

$$E_{au} = \frac{4A_u^3}{27B_u^2}\left(1-3\frac{B_uC_u}{A_u^2}\right)^{\frac{3}{2}}. \qquad (4)$$

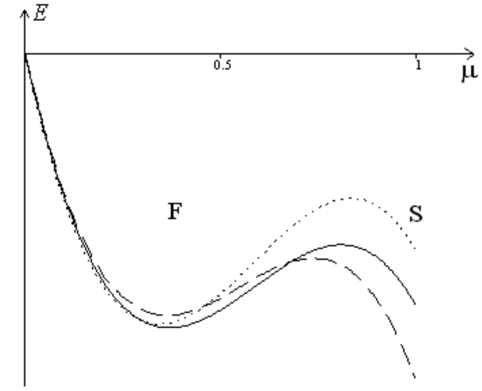

Fig. 1. The dependence of the ground state energy of exciton in nondeformed (solid line), elastically uniaxial (dashed line) and hydrostatical (dotted line) compressed AH on parameter μ.

From the analysis of Eq. (4) and how it is seen on the Fig. 1 as uniaxial compression increases, reduction of potential barrier with the comparison of nondeformed crystal potential barrier occurs.

3. Conclusion

Obtained results and analogous speculations for hydrostatically compressed crystal case have led us to the following conclusion.

Under uniaxial elastic and small hydrostatic deformation, i.e. when Hooke's law can be applied, potential barrier between free and self-trapped states of excitons decreases, which must lead to the increasing of the STE luminescence.

While, according to our estimates, at the nonlinear (high degree of pressure) deformation elastic constant increases - $\beta \sim (1/r^n)$, where $n > 4$, r is the distance between ions, hence exciton bandwidth expansion predominates above other terms in Eq. (3) which leads to the increasing of the self-trapping barrier for excitons and to the decreasing of the exciton self-trapping efficiency.

References

[1] V. Babin, A. Bekeshev, A. Elango et al., J. Lumin. **76**&**77** (1998) 502.

[2] V. Babin, A. Bekeshev, A. Elango et al., J. Phys.: Condens. Matter **11** (1999) 2303.

[3] Ch.B. Lushchik, A.Ch. Lushchik, Decay of Electronic Excitations With Defect Formation in Solids, Nauka, Moscow, 1989 (in Russian).

[4] Y. Toyozawa, Physica B **116** (1983) 7.