Relaxation of electronic excitations in uniaxially stressed alkali halides at 4.2 and 80 K

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Abstract

Using the methods of absorption, luminescence and thermoactivation spectroscopy, we study the formation and relaxation processes of electronic excitations (EE) in alkali halides (AH) with the lowering lattice symmetry and at 4.2 and 80 K. The dependence of the efficiency of radiative relaxation of self-trapped exciton (STE) on the charge and size of point defects is studied. We have found that the emission efficiency of STE in uniaxially stressed AH is noticeably increased. Such an enhancement of the STE (e.g. σ, E_x, π) luminescences is very important for the elaboration of AH scintillators operating in the intrinsic STE emission without energy transfer to the impurities as in classic scintillators NaI-Tl and CsI-Tl.

Key words: Uniaxial stress; Alkali halides; Exciton luminescence

1. Introduction

The relaxation of the anion electronic excitations in AH is a process where excited anions transform into anisotropic centers, two-halogen STE's, with different final structures: a symmetric on-center, weak off-center and strong off-center STE configurations may be created [1]. The lowering of the crystal lattice symmetry and the compression effect induced by a uniaxial stress are expected to affect the efficiency of the different stages of EE relaxation in AH. The effect of the uniaxial stress on the luminescence and radiation defect creation in KCl, KBr, KI and RbI crystals has been studied earlier [2, 3].

2. Electronic excitations localized near vacancy defects of different sizes

In plastic deformed crystals ionizing radiation creates besides EE, self-trapped in regular lattice sites (e_r^0), also excitons, localized near single anion vacancies (e_α^0 or α-centers), divacancies (e_d^0) and quartets of vacancies (e_q^0). We have nominated divacancies and quartets of vacancies d- and q-centers, respectively, with the analogue of α-center.

We have distinguished excitation and radiation spectra of excitons localized near e_d^0 and e_q^0 created in KBr, KI, NaBr, and RbI by previous plastic deformation (see Table 1).

In Table 1, the relative Stokes shifts (S_R) and the energy distance ($\triangle$) between the maxima of the lowest energy absorption band of an exciton created in a regular lattice site and of the excitation band of the exciton created near corresponding defect are shown. One can see from Table 1 that the energy needed for the creation of the near-vacancy excitations (E_{exc}) decreases in sequence of the localized excitons e_q^0-e_d^0-e_α^0. The values of S_R are essential for the consideration of the relaxed excitons. As it is seen from Table 1, the S_R value increases reguarly in the sequence of the localized excitons e_q^0-e_d^0-e_α^0. The S_R value is connected with the electron-phonon interaction energy in the luminescence center. In the vicinity of a larger vacancy defect the possibility of the EE decay into phonons is limited,

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Table 1
Characteristics of the defects created by plastic deformation at 295 K and measured at 4.2 K.

Crystal	EE	E_{exc}(eV)	$\triangle$(eV)	E_{em}(eV)	S_R
KBr	e_r^0	6.82	0	4.42; 2.28	0.35; 0.66
	e_q^0	6.5	0.32	3.4	0.48
	e_d^0	6.4	0.42	2.88	0.51
	e_α^0	6.15	0.67	2.55	0.58
KI	e_r^0	5.83	0	4.15; 3.31	0.29; 0.43
	$e_r^0(E_x)$	5.83	0	3.04	0.48
	e_q^0	5.6	0.23	3.1	0.45
	e_d^0	5.4	0.43	2.42	0.52
	e_α^0	5.2	0.63	2.23	0.57
NaBr	e_r^0	6.73	0	4.58	0.32
	e_q^0	6.52	0.21	3.20	0.51
	e_d^0	6.36	0.37	2.7	0.57
	e_α^0	6.19	0.54	2.6	0.58
RbI	e_r^0	5.76	0	3.92; 2.26	0.32; 0.6
	e_q^0	5.52	0.24	3.03	0.45
	e_d^0	5.38	0.38	2.52	0.53
	e_α^0	5.23	0.53	2.22	0.58
	$e_r^0(E_x)$	5.76	0	3.10	0.46

E_{exc}-maximum of the corresponding excitation band; $\triangle$-the energy distance between the maxima of the lowest energy absorption band of an exciton created in a regular lattice site and of the excitation band of the exciton created near defect; E_{em}-maximum of the emission band; S_R-Stokes shift=$(E_{exc}-E_{em})/E_{exc}$.

and, therefore, the S_R value for e_q^0 is smaller than for e_α^0.

We want to emphasize that the charge of vacancy defects is the main perturbing factor for e_α^0, e_d^0, and e_q^0 creation and dimension of vacancy defects is the main perturbing factor for the radiative decay e_α^0, e_d^0, and e_q^0.

3. Uniaxial stress effect on the intensity of the intrinsic emission bands

Fundamentally different possibilities of investigating the relaxation of EE are produced when the analogous experiments are performed not on the postdeformed crystal, but while the sample is still under the stress produced by a definite mechanical action. In this case, effects due to changes of the parameters of the main lattice are added to the effects due to introduction of strain defects.

Cleaned-out crystal samples were deformed by uniaxial compression along one of the $\langle 100 \rangle$ directions at 80 K directly in a nitrogen cryostat. At this temperature deformation AH by about 1% is considered to be elastic.

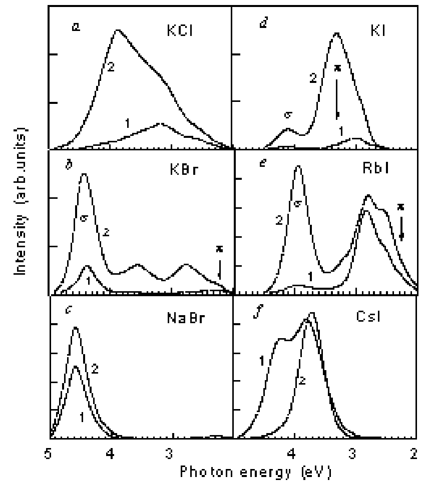

Fig. 1. Emission spectra of KCl (a), KBr (b), NaBr (c), KI (d), RbI (e), and CsI (f) crystals measured at 80 K under X-ray excitation for the unstressed samples (curves 1) and for the samples uniaxially stressed at 80 K by about 1% (curves 2).

X-ray luminescence spectra before (curves 1) and under (curves 2) uniaxial stress at 80 K are shown in Fig. 1 for KCl, KBr, NaBr, KI, RbI, and CsI crystals.

The main effect for all investigated crystals is concluded in redistribution intensity in favor of intrinsic σ- and π- emission bands of STE. It has been established (on the sample KI-Tl) that in uniaxial pressure stress field, exciton migration length before self-trapping considerably reduces due to enhancement of exciton-phonon interaction and it results increasing exciton self-trapping probability in regular lattice with consequent radiative decay.

Thus, the effect of radiative defect-formation decrease in stressed KI (also in RbI) is determined by unfavorableness of EE decay channel to F-H pairs in uniaxial elastic deformation field of crystal.

The method, which we have found, of direct influence on radiative channels of EE relaxation in AH plays an important role in search for new scintillation detectors, which action is based on intrinsic exciton luminescence without EE transfer to the impurities as in the case of traditional scintillators.

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