Thermal Inactivation of Defect Formation Induced by Exciton Self-Trapping in Rare-Gas Solids

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Selective excitation of excitons in rare-gas solids by photons with energies $h\nu < E_g$ results in accumulation of Frenkel-pairs by intrinsic excited-state mechanism of defect formation via self-trapping of excitons [1]. Recently the simple kinetic model was proposed, which allows fitting the experimental dose dependences of "defect" luminescence subbands and obtaining the particular kinetic parameters [2]. Application of this model provided a way of qualitative and quantitative analysis of rare-gas crystals, which is indispensable at any attempt of comparison of luminescence spectra from different samples. At the same time it is well known that there is a strong thermal quenching of the defect formation processes, which was initially explained by temperature dependence of lifetime of emitting states [3]. In the same temperature range the electron traps become active and charge recombination processes result in rich spectra of thermoluminescence.

In the present study we apply the Eyring's transition state concept [4] to the processes of thermal activation-inactivation of exciton trapping states resulting in luminescence spectra evolution under selective synchrotron irradiation. The experiments were carried out at the SUPERLUMI-station at HASYLAB, DESY, Hamburg. The selective photon excitation was performed with spectral resolution $\Delta\lambda = 0.2$ nm. The VUV-luminescence analysis was performed with $\Delta\lambda = 2$ nm, Pouey high-flux monochromator equipped with a multisphere plate detector. The dose dependences of self-trapped exciton luminescence at different temperatures under irradiation by photons with energies $E < E_g$ were measured. These curves are saturated at long time of irradiation therefore we used the slopes of the initial linear parts of the dose curves at $t = 0$ as the defect formation rates w.

Following Eyring assumption we can fit the temperature dependence of the defect formation rate as $w(T) = \beta \cdot T \cdot \exp(-E/kT) \cdot (1 + \exp(\Delta S/k) \cdot \exp(\Delta H/kT))^{-1}$, where k – the Boltzmann's constant, E – Arrhenius activation energy, β – scaling factor. The values of activation energy E and enthalpy of inactivation ΔH may be determined from the upper and lower tangents of the $w(T)$ in the coordinates $(\ln[w(T)])$ vs. (T^{-1}). The value of entropy of inactivation ΔS may be obtained from the equilibration condition $d(\ln[w(T_{max})])/dT = 0$. The best fit of the data for solid Xe was obtained with $T_{max} = 30$ K, $E = 2.8$ meV, $\Delta H = 28$ meV, $\Delta S = 0.8$ meV$\cdot$K^{-1} For solid Kr correspondent values are $T_{max} = 27$ K, $E = 4$ meV, $\Delta H = 30$ meV, $\Delta S = 1$ meV$\cdot$K^{-1}.

References:
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