Optical radiation research establishment and annealing nanosized defects in the crystal NaCl

Galym Zhaksygeldinovich Alseitov, Almukhan Baimakhanuly, Erbol Anarbekovich Ospanbekov

Republican State Enterprise on the Right of Waging a «Kazakh National Pedagogical University named after Abai» Ministry of Education and Science of the Republic of Kazakhstan, Dostyk Avenue 13, Almaty, 050010, Kazakhstan

Abstract. Creation of radiation defects was investigated in NaCl crystals irradiated at 300 K by heavy Au ions, α particles, X-rays as well as XeCl or ArF laser radiation selectively forming in two-photon regime anion excitons or hot electron-hole pairs. The emission of self-trapped excitons and tunnel recombination of electrons (staring from two states of an F centre) with spatially close self-trapped holes were separated in cathodoluminescence and phosphorescence spectra at 6 K. The hopping diffusion of I and H interstitials is accompanied by thermally stimulated luminescence at 15-27 and 30-45 K, respectively.

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Introduction

Use of underground mines and caves, where in the past was mined rock salt as dry cells for long storage of metal containers with radioactive waste nuclear power demanded resumption of experimental and theoretical studies in NaCl radioative processes of creation structural defects resulting in long-term exposure to the material destruction [1]. We have continued contacts with the staff of Institute of Physics of Tartu University (IFTU) on studying of features of creation and annealing of nanoscale defects in crystals KCl, KBr, RbCl, RbBr [2, 3]. Our goal is to obtain new data on the characteristics of radiation defects in single crystals of NaCl (radius of Na⁺ half than Cl⁻) irradiated at 300 K X-rays (50 keV), α -particles (5.1 MeV), swift heavy ions Au (2.18 GeV) and electron beams (8 keV, T = 6 K). Particular attention is paid to radiation crystals XeCl-laser radiation (in the mode of two-photon absorption $4.02 \times 2 = 8.04$ eV), selectively creating anionic excitons in NaCl as well as ArF-laser (6.42 \times 2 = 12.84 eV), to create a split p-holes in the valence hot zone and p-electrons in the conduction band (in NaCl energy gap $E_{\sigma} = 8.7 \text{ eV}$).

Unlike the KCl, KBr and RbBr, the effectiveness of radiation in NaCl of creation stable charged anionic and neutral pairs of Frenkel defects (FD) at 6 K is low (for example, see [2]), although highly sensitive luminescence methods defects can not register on their annealing region $10 \div 60$ K. Effectiveness of F, H-pairs when irradiated NaCl at T = $100 \div 200$ K increases sharply due to the thermal removal of F-centers neutral interstitial H-centers, having the structure of the molecule Cl₂, taking one anion site and shifted along the (111) to tetrahedral voids in the packaging of anions. Charged defects

interstitial ions Cl⁻ and anion vacancies (I, α - pair) are effectively created at 200 ÷ 300 K.

The experimental technique used in the study of the processes of defect electrons at 6 K and their annealing in $6 \div 420$ K, is described in [4]. Applied constant heating rate $\beta = 0.17$ K / s. The absorption spectra were measured at 300K on a double-beam spectrophotometer JASKO V-550, allows measurement of the optical density up to 0.01. Annealing defects created by different types of radiation at 300 K, in the region of 300 K ÷ 750 implemented with $\beta = 2.86$ K / s at the installation SYSTEM 310 TLDF - Reader in a nitrogen atmosphere. XeCl-laser irradiation was carried out pulses of 10 ns and an energy of 50 mJ, 10 Hz [3]. ArF-laser irradiation was performed 5 ns pulses at 20 Hz. We investigated specially selected purest natural crystals NaCl, and crystals grown by «Korth» (Germany) and IFTU Stockbarger technique after special cleaning and 50-fold zone melting [5-7].

Figure 1 shows the absorption spectra of crystals NaCl, irradiated at 300 K with heavy ions Au, α -particles, X-rays, and XeCl (308 nm) and ArF (193 nm) lasers in two-photon mode. Irradiation is carried out perpendicular to the plane (100) of the crystal. The thickness of the crystals at the laser irradiation was \sim 4 mm, in other cases, $0.6 \div 0.7$ mm. Au⁺ ions and α particles penetrate into the crystal at a depth of about 80 microns and about 15 microns, accordingly. [8] Measurements have shown that in two-photon mode laser radiation penetrated the entire depth of the crystal, easing to 4 mm thickness only twice. All kinds of irradiation created F-centers (absorption band maximum of about 2.7 eV at 295K), F₂-centers (1.72 eV), and nonelementary absorption at $4.8 \div 6.45$ eV. The ratio of the absorption maxima F-and F₂-centers after irradiation of X-rays and lasers close to 20, using α -particles (fluens 16 days was 4 • 1011 α / cm²) F/ F₂-ratio down to 6. When irradiated NaCl ArF-laser to create a hot electron-hole (e-h) pairs, this ratio was 11. Anomalously high value F/ F₂-ratios were to create excitons XeCl-laser. NaCl crystal irradiated as a linear accelerator LINAC (GSI, Darmstadt) ions Au¹⁹⁸ (2.18 GeV fluence Au/cm²) at 300 K. The absorption spectrum of the crystal (the average distance between the tracks of ions ~ 36 nm) except for F and F₂ visible centers F₃ (or R)-centers (2.27 eV), non-elementary strip 6 ÷ ~ 5 eV (Cl₃ - centers) and continuous absorption at 3 ÷ 1.6 eV corresponding to the NaCl formed in the metal colloid.

As is well known (see, eg, [8]), in LiF and NaCl heavy ions U^{238} and Au^{198} almost all its energy (> 99.8%) passed the electronic subsystem of the crystal, creating extremely high density of electronic excitations . Decay of anion excitons (energy creating them $8 \div 8.4$ eV), cationic excitons (60 ÷ 62 eV), and recombination of the conduction electrons (e) selftrapped holes (VK - centers) in NaCl lead to the creation of anion Frenkel defects (FD) - F, H- and α , I- Pr. At 300 K, F-, F^2 - and F^3 - centers in NaCl are stable, but interstitial anion (H- and I- centers), interstitial cation and anion and cation vacancies (v_a) and v_c) it is easy to move through the crystal lattice. Localization and interstitial stabilization is carried out as in the other alkali-halide crystals of [2, 4], as a result of association of several point defects. For example, the union of two H-centers with bivacancies $(v_a v_c)$ lead to the creation of a stable linear to 500 K (Cl3)aca -molecule located in two anionic and one cationic sites of the crystal. The same molecule is formed also when the association H-center, V_K -center and a single cation vacancy. Absorption of molecules (Cl₃-) _{aca} lies in the $5 \div 6$ eV (see Figure 1).



Fig. 1. Absorption spectra of NaCl, irradiated at 300 K X-rays (1); α -particles (4·10¹² α / cm² - 2), gold ions (1011 Au / cm² - 3); ArF-(4) and XeCl-laser (5)

As shown in Figure 2, for X-irradiated at 300 K crystals annealing F-absorption at 2.7 eV is carried out at 400 \div 550 K. In the same temperature annealed and a significant portion of the absorption ~ 5.5 eV. Process is caused the thermal dissociation of molecules (Cl₃⁻)_{aca}. Formed while moving H-interstitial nonradiatively recombine with F- centers.

In the dissociation of the molecules are also formed three-holoid mobile V_K or V_F (v_ch) [9], which partly radiatively recombine with F-centers (see Figure 2). Irradiation of NaCl crystals of high purity X-rays at 5 K also leads to the creation of F-centers (see [10] and references therein). However, at 5 K the efficiency of production of long-lived for many minutes of Fcenters is ten times less than at 300 K. Creation and annealing of low concentrations of anionic FD can not register highly sensitive luminescence methods. Figure 2 shows the curve of the thermally stimulated luminescence (TSL) of natural crystal NaCl, irradiated at 6 K electrons with energy of 8 keV. Within 30 minutes studied cathodoluminescence crystal, after which the electron beam is disconnected, recorded rapid weakening phosphorescence within $1 \div 300$, and then measured the spectra is slightly decreasing over time phosphorescence. 15 minutes after switching off the electron beam to heat the irradiated crystal to 420 K with $\beta = 0.17$ K / s, TSL recorded through a monochromator. Figure 2 shows one of the many curves TSL measured for emission 2.4 ± 0.15 eV. The emission is dominated by tunneling luminescence created by the electron beam $\{F \dots V_K\}$ pairs are the F-center electron radiatively recombine with localized within a few interionic periods of self-trapped hole.



Fig. 2. Thermoluminescence crystals NaCl, irradiated with electrons (8 keV, 200 for 40 min) with 6 K (1,1', glow 2.4 eV) or X-ray (50 keV, 18 mA, 1 hour) at 300 K (2 glow $1.8 \div 3.7$ eV). In the insets annealing EPR signal H-centers (3) [9], and the absorption of F-centers (4).

In the glow of 2.4 and 3.4 eV are easily detected by TSL narrow peaks at 16, 21 and 26 K, and peaks at 30 \div 45 K. X-irradiated at 8 K NaCl in 10 \div 27 K [9] measured annealing 2.9 eV luminescence centers α -stimulated photons 7.2 eV (Cl⁻ions absorption surrounding υ_a). Weak TSL at 30 \div 45 K (inset in Fig. 2) is associated with annealing H-centers established in NaCl electrons at 6 K [10].

Figure 3a shows the cathodoluminescence spectra of natural crystal NaCl, measured at 6 K under the influence of the electron beam 8 keV. In the spectra of all the amendments introduced. Similarly to the case of synchrotron radiation exposure [11], the spectrum is dominated by cathodoluminescence stationary luminescence bands with maxima at 3.4 and 5.4 eV, corresponding to the radiative decay of the two states of self-trapped exciton (STE) of the lowest primary and excited (see [10]). Recombination of the electron with the VK- center passes state of the STE.



Fig. 3. (a) cathodoluminescence spectra (8 keV, 200 nA, 6 K) NaCl (1) and the range of the TF NaCl, X-irradiated (50 keV, 4 mA) at 6 K (2). through the excited (b) HF spectra NaCl at 6 K (1) and in the TSL spectra of 261 K (2) and 400 K (3) for NaCl, irradiated with electrons

Figure 3a shows a greatly enlarged scale also shows the spectrum of the tunneling phosphorescence (FT) X- irradiated for 15 min at 6 K crystal NaCl, measured after 300 s after the cessation of exposure. The spectrum is dominated by the emission band TF STE. In Figure 3b shows the spectra of TF measured after cessation of exposure NaCl electrons at 6 K. At 6 K the spectrum is the band TF STE 3.4 eV, visible glow ~ 2, 2.4, and 2.9 eV, and the faint glow ~ 4 eV. During the measurement of TSL this crystal twominute stop in the 270 K TSL possible to measure the spectrum in this temperature range (Figure 3b). Subsequent heating to 362 K leads to the almost complete disappearance of 3.4 eV luminescence.

The study showed that F, H-and α , I-DF pairs in NaCl created even after irradiation at 6 K 8 keV with electrons. In the glow in the I-and H-TSL peaks where carried annealing charged and neutral interstitial anion, there is a significant enhancement of TF involving close $\{F \dots V_K\}$ -pairs. These pairs are annealed at $150 \div 180$ K after the V_K-centers begin hopping diffusion through the crystal lattice and recombine with electrons localized on the anion vacancies (F-centers) or other lattice defects. Confirmed that the glow of ~ 2.4 eV corresponds to the recombination of electrons tunneling from the lowest state of the F-center with holes in pairs {F ... V_{K} . Glow 3.4 and 5.4 eV correspond to the recombination of electrons with VK -centers across the ground and excited states of STE. STE lowest state of energy $\sim 8 \text{ eV}$ [12], a two-photon absorption mode Xe Cl-laser. Excited state STE effectively created not only by 8.4 eV photons [12], but also in the ArF- laser

two-photon mode at the interband transition with the formation of p- holes in the valence band and the pconduction electron. This excited state decays STE is particularly effective with the creation of spatially separated few F- and H- centers , which is consistent with the creation of the first spectrum of F-centers VUV radiation for NaCl in the single-photon regime [10,13]. A detailed discussion of the features of the radiation effects in NaCl will be published in collaboration with the Estonian physicists.

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Corresponding Author:

Dr. Alseitov Galym Zhaksygeldinovich

Republican State Enterprise on the Right of Waging a «Kazakh National Pedagogical University named after Abai» Ministry of Education and Science of the Republic of Kazakhstan

Dostyk Avenue 13, Almaty, 050010, Kazakhstan

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