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# Thermal spike effect on defect evolution in NaCl irradiated with light and heavy ions at 8 and 300 K

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**Abstract**

Single crystals of NaCl were irradiated at room temperature and at 8 K with energetic heavy ions ($^{12}$C, $^{50}$Ti, $^{58}$Ni, $^{74}$Kr, $^{152}$Sm, $^{197}$Au, $^{208}$Pb and $^{238}$U) of 50–2600 MeV providing mean electronic energy loss values from 0.7 to 19 keV/nm. The creation and evolution of color centers were investigated as a function of fluence and temperature by in situ absorption spectroscopy and thermo-stimulated luminescence, complemented by thermal annealing and optical bleaching. For irradiations at 8 K, primary hole centers are observed which typically annihilate at temperatures between 10 and 80 K. The efficiency of color center creation at 8 K strongly depends on the energy loss of the ions and is several times higher for U and Au ions than for C and Ti ions. Thermal spike estimations, taking into account the finite velocity of heat propagation, assign these effects to thermal stimulated separation of color centers in the genetic Frenkel pairs.


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## 1. Introduction

Under the irradiation of swift heavy ions, the formation of radiation damage in alkali halides is governed by the electronic energy loss of the projectiles and lattice defects are created by the decay of electronic excitations [1,2]. The damage is mainly induced in the anion sublattice where Frenkel pairs are formed, consisting of an F-center (an electron localized at an anion vacancy) and an H-center (a halogen molecule $X_2^-$ replacing a regular anion) or alternatively of an α-center (anion vacancy) and an I-center (interstitial anion). These defect pairs are created by the decay of self-trapped excitons or by recombination of delocalized electrons and holes typically within a time period between $10^{-11}$ s up to $10^{-3}$ s [3–5]. In NaCl the main electron and hole centers as well as various defect aggregates absorb light in the spectral range of 200–900 nm and thus can be easily identified by VIS–UV optical spectroscopy [6].

The irradiation temperature has a significant influence on the mobility of color centers and therefore plays a critical role for creation of stable point defects as well as their aggregation and recombination. For low-temperature (14 K) irradiations of NaCl and KBr crystals with swift ions (Ne, Zn and Xe), the exciton luminescence and the creation of F-centers was reported to show similar effects as under X-ray excitation [7–9].

In the study presented here, we investigated defects induced by heavier ions and analyzed them by VIS–UV spectroscopy and thermo-stimulated luminescence (TSL) in combination with thermal annealing and optical bleaching. Defects produced during 8-K irradiations are compared with room temperature (RT) results. Follow-up processes such as defect annihilation and aggregation are

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discussed in combination with a transient thermal spike which considers the finite velocity of heat transfer at cryogenic temperatures.

## 2. Experiments

Thin platelets of NaCl crystals (thickness ~ 1 mm) were irradiated at the UNILAC linear accelerator of GSI with different heavy ions ($^{12}$C, $^{50}$Ti, $^{58}$Ni, $^{84}$Kr, $^{152}$Sm, $^{197}$Au, $^{208}$Pb or $^{238}$U) of 50–2600 MeV providing mean electronic stopping values between 0.7 and 19 keV/nm. The fluences applied ranged from $10^8$ to $10^{12}$ ions/cm$^2$ and the beam flux was around $10^8$–$10^9$ ions/s cm$^2$. The irradiations were performed at RT and in a cryostat at 8 K (we note that the sample temperature could not be measured during irradiation; however, peaks at 10 and 12 K in the TSL spectra indicate a sample temperature below 10 K). During beam interruptions, the set-up allowed us to measure in situ the optical absorption and thermo-stimulated luminescence (up to 70 K with a heating rate of 4 K/min).

Crystals irradiated at nominally 8 K exhibit a pronounced TSL signal. Fig. 1 shows a typical spectrum measured during annealing between 8 and 70 K. The spectra of samples irradiated with different ions are rather similar, showing one group of peaks between 10 and 20 K and another group of lower intensity between 25 and 55 K. Each TSL peak corresponds to the recombination of a certain electron and hole center at the given temperature. The observation of various TSL peaks thus tells us that the ion irradiation induces defects of different mobility, annealing at different low temperatures.

The optical absorption spectra of all samples irradiated at 8 K exhibit a prominent band at 450 nm corresponding to F-centers. Additional bands appear at 330 and 245 nm which are mainly ascribed to H- and $H_A$-centers (an H-centers modified by impurities), respectively (Fig. 2). At 8 K the formation of $F_2$ centers (two neighboring F-centers with band maximum at 730 nm) is strongly suppressed, and the ratio of the concentration of $F_2$- to F-centers is more than two times lower than at RT. Optical spectroscopy of crystals warmed up after irradiation gave clear evidence that the H-centers in NaCl crystals are not stable above 80 K but recombine with electron centers or are converted into more complex hole centers. At RT the stable color centers are F-, $F_2$- and $V_3$-centers (absorption maximum at 220 nm) (see Fig. 2(b) and (d)). For a sample irradiated with Au ions at 8 K annealing to RT caused a conversion of about 20% of the H-centers into $V_3$-centers demonstrating that recombination of H- and F-centers is dominant.

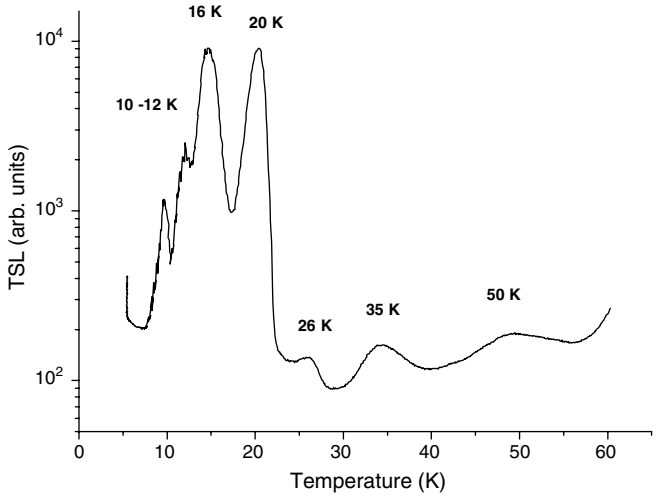


Fig. 1. Thermo-stimulated luminescence of NaCl irradiated at 8 K with 1400-MeV U ions at a fluence of $5 \times 10^8$ ions/cm$^2$. The different peaks illustrate the variety of defects responsible for recombination luminescence as well as their thermal stability.

A comparison of the evolution of the concentrations of F-centers as a function of fluence for irradiations at 8 K and RT is presented in Fig. 3. At 8 K, Au ions induce in the initial stage a linear growth of the F-center concentration, which finally saturates at high fluence. In the case of the RT irradiation, saturation requires higher fluences and the F-center production in the initial stage is lower than at 8 K (more details in [10]). This is in contrast to light ions (C, Ti) which produce in the initial irradiation phase less F-centers at 8 K than at 300 K.

Optical bleaching and thermal annealing of NaCl samples irradiated at 8 K mainly lead to a decrease of absorption bands of the F- and H-centers and not to an increase of the $F_2$-center band, indicating annihilation processes by recombination rather than aggregation.

## 3. Discussion

Color centers created at 8 K and their evolution as a function of temperature in comparison with RT observations, can shed light on the defect formation process in tracks of swift heavy ions (SHI) including the influence of ion induced heating (thermal spike).

### 3.1. Color centers and TSL at 8-K irradiation

The TSL spectra clearly demonstrate the large variety of induced defects. The thermal stability of these defects is limited to temperatures between 10 and 60 K, which is even lower than the annealing temperature of H-centers (~80 K) [1,6]. The TSL peaks at 10, 16 and 20 K are assigned to the recombination luminescence of I-centers and the peaks in the temperature range 26–50 K correspond to recombination luminescence of H-centers with electron color centers [3,11]. The mobility of these different hole centers is determined by various lattice microstructures (impurities or vacancies in the neighbor position). They modify I- and H-centers into $I_A$- and $H_A$-centers, respectively with different activation energies for diffusion. Such centers are known from TSL measurements which are much more sensitive to the defect microstructure than absorption spectroscopy [11]. The interstitial ions such as I-centers have the highest mobility and the lowest activation energy