# F-band absorption and thermoluminescence of NaCl single crystals X-ray irradiated at elevated temperatures and later subjected to high electric fields or laser excitation

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F-band absorption and thermoluminescence of NaCl single crystals X-ray irradiated at elevated temperatures (25, 40 and 90° C) and later subjected to high electric fields (up to 20 kV cm<sup>-1</sup> a.c. or d.c.) or laser excitation have been studied. Interesting results have been obtained and an attempt is made to understand them.

### 1. Introduction

Studies on colour centre phenomena in alkali halide crystals irradiated at room temperature with X-rays or y-rays have been carried out extensively giving valuable information about the defect processes taking place in them [1, 2]. It has been reported that if these crystals are subjected to high electric fields a.c. or d.c. (or excited with laser light) and later irradiated at room temperature with X-rays or y-rays, the F-centre concentration in these crystals increased considerably compared to those when they are only irradiated with these ionizing radiations. Even the thermoluminescence (TL) light output in the glow peaks of these crystals increased significantly [3, 4]. However, recently it has also been shown that if the high electric field (or laser excitation) is applied to the crystals after they are X-ray or y-ray irradiated, interesting changes in the colour centre phenomena are exhibited even at room temperature [5, 6].

It is known that if alkali halide crystals are irradiated with X-rays at slightly elevated temperatures, the colour centres formed, particularly F-centres, are more stable though their concentration is less; the TL peaks are pushed to higher temperatures, the light output being less. Attempts have been made to understand these results [1, 2].

It is felt worthwhile to study the effect of high electric fields (or laser excitation) on the F-centre concentration and thermoluminescence of NaCl single crystals X-ray irradiated at elevated temperatures. The results of such an investigation form the content of this research communication.

## 2. Experimental methods

NaCl single crystals used in the present work are laboratory grown; some crystals obtained commercially from Harshaw Chemical Company, Ohio and also as a gift from Professor A. Smakula (Director of Crystal Physics Laboratory, MIT, USA) have also been used to check the reproducibility of the nature of the data. The crystals are cut to desired size, ground and optically polished. The final dimensions of the samples are approximately  $1 \text{ cm} \times 1 \text{ cm} \times 0.1 \text{ cm}$ .

The sample was mounted on a holder which had a heating arrangement; the temperature of the sample could be measured by means of a chromel-alumel thermocouple attached very near to the sample.

X-ray irradiation of the sample at the desired temperature was carried out for  $1\frac{1}{2}$ h with a Norelco unit run at 35 kV, 10 mA, keeping the sample at about 1.5 cm from the window. Excitation with laser light was carried out at the corresponding elevated temperature with a He-Ne laser of 2 mW power and 632.8 nm wavelength.

For the application of high a.c. (or d.c.) field the samples carried thin aluminium foils on the large area faces to serve as electrodes. Then the crystals were subjected to the field at the elevated temperature for  $\frac{1}{2}$  h. The a.c. field was applied by a step-up transformer, the voltage of which can be varied from 0 to 2.5 kV. The d.c. field was applied by an electronically controlled conventional power supply the voltage of which can be varied from 0 to 2 kV. The value of the electric field was calculated. After the field application was over, the aluminium foils were removed.

The optical absorption measurements were carried out on a Beckmann 26 spectrophotometer in the wavelength region 300 to 600 nm always at room temperature ( $\sim 25^{\circ}$  C). The accuracy in the measurement of the absorption coefficient ( $\alpha$  cm<sup>-1</sup>) is 0.05. The thermoluminescence light output from the sample is recorded on an Esterline-Augus recorder using a conventional set-up [7].

## 3. Results

Figure 1 presents optical absorption characteristics of NaCl crystals X-ray irradiated at  $40^{\circ}$  C for  $1\frac{1}{2}$  h and when the crystals are subjected to different a.c. fields. The absorption in the band at 465 nm (which is the F-band) is found to decrease appreciably with the a.c.



Figure 1 Optical absorption characteristics of NaCl crystals X-ray irradiated  $(1\frac{1}{2}$  h) at 40° C and later subjected to different a.c. fields. (1) as-cut NaCl; (2) X-ray at 40° C; (3) (2) + 1 kV cm<sup>-1</sup>, (4) (2) + 2 kV cm<sup>-1</sup>; (5) (2) + 5 kV cm<sup>-1</sup>; (6) (2) + 10 kV cm<sup>-1</sup>; (7) (2) + 15 kV cm<sup>-1</sup>; (8) (2) + 20 kV cm<sup>-1</sup>.

field. Similar results are obtained for d.c. fields (data not presented).

Figure 2 gives the decrease in F-band absorption in NaCl crystal subjected to the same X-ray dosage at 40° C and later excited with laser light at that temperature for different times. The F-band absorption is again found to decrease with laser dosage.

Using Smakula's equation [2], the concentration of F-centres in NaCl crystals under different conditions is calculated. Figures 3 and 4 show the decrease in the F-centre concentration with the a.c. field or with laser dose for NaCl crystals which are X-ray irradiated at room temperature ( $\sim 25^{\circ}$  C), 40 and 90° C.

The thermoluminescence characteristics of these crystals X-ray irradiated at 25, 40 and 90°C are presented in Fig. 5. TL peaks are observed at 122 and 235°C for the samples irradiated at 25 and 40°C; however these peaks seem to be pushed to slightly



*Figure 2* Optical absorption characteristics of NaCl crystals X-ray irradiated  $(1\frac{1}{2}h)$  at 40°C and later excited with laser light for different times. (1) as-cut NaCl; (2) X-ray at 40°C; (3) (2) + 1 min laser; (4) (2) + 2 min laser; (5) (2) + 5 min laser; (6) (2) + 10 min laser; (7) (2) + 20 min laser.



Figure 3 (a) Concentration of F-centres  $(n_{\rm F})$  in NaCl crystals X-ray irradiated at different temperatures as a function of a.c. field; (b) log  $n_{\rm F}$  against a.c. field. (×) 25°C; ( $\bigcirc$ ) 40°C; ( $\triangle$ ) 90°C.

higher temperatures when X-ray irradiation is carried out at a sufficiently high temperature of 90° C.

#### 4. Discussion

It is known that the F-centre absorption band lies at about 465 nm in NaCl crystals irradiated with X-rays. When these crystals are heated, they exhibit thermo-



Figure 4 (a) Concentration of F-centres  $(n_{\rm F})$  in NaCl crystals X-ray irradiated at different temperatures as a function of time of laser excitation; (b) log  $n_{\rm F}$  against time of laser excitation. (×) 25° C; (O) 40° C; ( $\Delta$ ) 90° C.



*Figure 5* Thermoluminescence characteristics of NaCl crystals X-ray irradiated at different temperatures.

luminescence (TL) peaks at 122 and 235°C; these TL peaks are shown mainly to be due to the destruction of F-centres [8, 9].

It is reported that when NaCl crystals are subjected to high electric fields (a.c. or d.c.), there will be considerable interaction of the field with the crystal lattice particularly at major defect regions like dislocations which probably leads to the generation of vacancies and hence to more disorder in the lattice [3, 4].

As the electric field associated with the laser beam used in the present work is very small  $(2 \text{ V cm}^{-1})$ , there will be no defects produced in the crystal due to the field. It has been pointed out that excitation of alkali halide crystals with this laser beam is likely to generate thermal spikes in the crystals (particularly in the dislocation regions) leading to production of a large concentration of defects like vacancies and their clusters [5, 6]. Thus the influence of subjecting the X-ray irradiated NaCl crystals to either high electric fields or laser excitation is mainly to increase the disorder in the crystal lattice. The present results suggest that this disorder facilitates destruction of F-centres even at comparatively low temperatures.

The decrease in F-centre concentration with electric field (data presented only for a.c. fields) or laser excitation for NaCl crystals X-ray irradiated at room or elevated temperatures is found to fit the relation

$$n = n_1 \exp(-k_1 F) + n_2 \exp(-k_2 F)$$

where  $n_1$ ,  $n_2$ ,  $k_1$  and  $k_2$  are constants and F is the a.c. field (or can be considered as time of laser excitation

which is laser dose), Figs 3b and 4b. (The graphs in these figures have been drawn taking points at close intervals from the smoothly drawn curves of Figs 3a and 4a). The values of the constants  $k_1$  and  $k_2$  with some more pertinent data of the present work are given in Table I. These data seem to indicate (i) that the electric field (or laser dose) necessary to destroy a large concentration of F-centres in the first stage (represented by the first exponential term of the equation) is more for NaCl crystals X-ray irradiated at 90°C (compared to that when the irradiation is done at 25 or 40° C); and (ii) that in the second stage (represented by the second exponential term of the equation) the destruction of F-centres by field or laser excitation is not an efficient process particularly when the crystals are irradiated at 90° C. The percentage of F-centres destroyed at different stages and also the total percentage of F-centres destroyed from the initial condition of the crystal to that when  $20 \, kV \, cm^{-1}$  field is applied (or laser excitation done for 20 min) are presented in Table I. These experimental findings suggest that the F-centres formed in NaCl by X-ray irradiation at elevated temperatures (e.g. 90°C) are more stable (compared to the F-centres formed in these crystals when they are irradiated around room temperature) and this may be due to the differences in the surroundings of the F-centres under the two conditions.

As NaCl crystals X-ray irradiated at elevated temperatures contain a smaller concentration of F-centres, the thermoluminescence light output in the peaks is also smaller. The shift of TL peak positions to slightly higher temperatures, exhibited by NaCl crystals irradiated at 90° C, seems to support the earlier conclusion that the F-centres in these samples are comparatively more stable.

In summary, the present work indicates that (i) the F-centres produced in NaCl crystals by X-ray irradiation at elevated temperatures (90° C) are comparatively more stable and (ii) it is not necessary to heat the crystals to high temperatures (corresponding to TL peak positions) to destroy these F-centres; they can be destroyed to a large extent even at 90° C by subjecting the crystals to high electric fields or laser excitation.

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TABLE I Data on F-centre concentration, thermoluminescence and  $k_1$  and  $k_2$  values of NaCl single crystals X-ray irradiated at different temperatures and subsequently excited with laser light for different times\*

Sample condition X-ray irradiated at (°C)	F-centre concentration $n_{\rm F} \times 10^{16}  ({\rm cm}^{-3})$	TL peak positions (°C)	Laser excited samples				
			$\frac{k_{\rm t}}{(\times 10^{16})}$	% n <sub>F</sub> destroyed	$k_2$ (×10 <sup>16</sup> )	% n <sub>F</sub> destroyed	Total % destroyed
25	5.47	122, 235	0.31	56	0.024	34	70
40	4.21	122, 235	0.24	48	0.022	31	64
90	2.36	132, 250	0.16	38	0.014	21	52

\*Data for electric field treated samples are not presented because of their similar nature with those of laser excited samples.

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