Studies on F-band absorption of irradiated KCl single crystals excited with laser light

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The absorption in the F-band of KCl single crystals after X-ray irradiation and subsequent excitation with laser light is studied. Similar investigations have been carried out when these crystals are subjected to high a.c. electric fields prior to X-ray irradiation and also at elevated temperatures. The effect of laser excitation on γ-ray irradiated samples is studied. An attempt is made to interpret the results in terms of the disorder that sets-in in these crystals.

1. Introduction
The colour centre phenomenon in alkali halide crystals has been extensively studied and has paved the way for considerable insight into the defect processes taking place in them [1-3]. The influence of plastic deformation on the colour centre phenomenon in alkali halides is to enhance the F-centre yield [4]. If alkali halide crystals are subjected to an appreciable fraction of their electrical breakdown strength ($\approx 10^6$ V) and subsequently irradiated with X-rays, a significant increase in F-centre concentration is observed [5, 6]. It has been shown [7, 8] that a considerable concentration of F-centres is destroyed even at room temperature ($\approx 30^\circ$ C) by subjecting X-ray irradiated alkali halide crystals to high a.c. or d.c. electric fields. A similar decrease in F-centre concentration was observed in X-ray irradiated KCl and NaCl crystals when they were subsequently excited with laser light [9, 10]. A study of the influence of laser excitation on X-ray irradiated alkali halide crystals through controlled variation of defect concentration can yield valuable information in understanding the processes leading to the decrease in F-centre concentration in such crystals. It is the aim of this paper to report the results of such studies in KCl single crystals.

2. Experimental methods
The KCl crystals used in the present work are laboratory grown; measurements were also taken on samples obtained as a gift from the Crystal Physics Laboratory, Massachusetts Institute of Technology, USA. The samples were cleaved and polished, the final dimensions being 1 cm x 1 cm x 0.1 cm. Apparatus having a step-up transformer with proper ratings and a laboratory-made sample holder were used for a.c. field treatment.

X-ray irradiation was performed at room temperature ($\approx 30^\circ$ C) for 1 h (unless otherwise stated), with 35 kV and 10 mA, keeping the samples 2 cm away from the window of a Norelco Unit. γ-ray irradiation was carried out with a $^{60}$Co source of $10^5$ Ci strength giving out a dose of 78 krad h$^{-1}$, for 54 h. Excitation with laser light by a He-Ne laser, 2 mW power and 632.8 nm wavelength was undertaken, keeping the samples at a distance of 30 cm from the laser.

The F-band absorption measurements were taken using a Beckman 26 Spectrophotometer at room temperature. The accuracy in the measurement of the absorption coefficient ($\alpha$ cm$^{-1}$) was 0.03.

3. Results
The F-band absorption characteristic of X-ray irradiated KCl crystals showed an absorption peak at 560 nm. F-band absorption is found to decrease with the time of laser excitation (i.e. laser dose). Using Smakula's equation and assuming the F-band to be gaussian in shape, the F-centre concentration in these samples under various conditions is calculated and plotted as a function of time of laser excitation. Fig. 1 shows the F-centre concentration for KCl crystals which have been initially subjected to various high a.c. electric fields, then X-ray irradiated and subsequently excited with laser light. The F-centre concentration in these samples was found to decrease with time of laser excitation and this decrease was in two stages. The initial F-centre concentration in the field-treated samples was observed to be larger compared to as-cleaved and X-ray irradiated samples; also the initial F-centre concentration increased with increasing magnitude of a.c. field. The decrease in F-centre concentration with time of laser excitation was found to be lower and faster in KCl samples which were subjected to higher a.c. electric fields.

Fig. 2 shows the concentration of F-centres in γ-ray irradiated KCl samples subsequently excited with laser light for various times. Here also we found a two-stage decrease in the F-centre concentration with laser excitation. This figure also gives F-centre concentration in X-ray irradiated KCl crystals (X-ray irradiated for $\frac{3}{4}$ h when the F-centre concentration is practically the same as in the γ-ray irradiated samples) which were later excited with laser light. It can be seen...

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from this graph that the decrease in F-centre concentration in γ-ray irradiated samples was smaller and slower compared to X-ray irradiated samples for the same laser excitation times.

F-centre concentration as a function of laser excitation for KCl crystals which were X-ray irradiated at different temperatures and excited with laser light at that temperature, is shown in Fig. 3. We noticed that the initial concentration of F-centres in these crystals was smaller compared to that when these were irradiated at room temperature. Subsequent laser excitation of these irradiated samples decreased the F-centre concentration at a faster rate and more efficiently.

4. Discussion

If the irradiated alkali halide are excited with laser light a considerable interaction of laser light with the crystal lattice can occur, particularly in the major defect regions like dislocations, leading to a considerable increase in the disorder already produced due to irradiation. This increased disorder seems to facilitate the destruction of F-centres even at room temperature [9, 10]. It seems reasonable to assume that the disorder created by laser excitation would be larger in the regions where initially dislocations and vacancy clusters are present in the crystal (compared to the normal lattice regions). Therefore, we expect a larger and faster decrease in F-centre concentration with time of laser excitation in these perturbed regions of the crystal lattice (Stage I). The disorder in the vicinity of F-centres formed in the normal lattice of the crystal is comparatively less and we can ascribe a slower F-centre reduction rate for this region (Stage II). Also, the rate of decrease of F-centre concentration would be proportional to the concentration of F-centres present, and because F-centres normally tend to accumulate around major defect regions, the decrease in F-centre concentration in Stage I would be larger than that in Stage II. For the Stages I and II we can write

\[
\frac{dN_p}{N_p} = -K_1 \, dt \quad (1)
\]

and

\[
\frac{dN_n}{N_n} = -K_2 \, dt \quad (2)
\]

where \(N_p\) is the concentration of F-centres in the perturbed regions of the lattice and \(N_n\) is concentration in the normal regions, \(K_1\) and \(K_2\) are proportionality constants. By integration of the above equations, we get

\[
N_p = n_1 e^{-K_1 t} \quad (3)
\]

and

\[
N_n = n_2 e^{-K_2 t} \quad (4)
\]

where \(t\) is the time of laser excitation. The total number of F-centres, \(N\), would then be

\[
N = N_p + N_n = n_1 e^{-K_1 t} + n_2 e^{-K_2 t} \quad (5)
\]

where \(n_1\) is the initial concentration of F-centres and \(n_2\) relates to the possible initial number of F-centres that