Effect of laser excitation on F-band absorption and thermoluminescence of X-ray irradiated KBr single crystals

G. PRASAD, B. K. ROUL, R. N. P. CHAUDHARY, K. V. RAO
Department of Physics, Indian Institute of Technology, Kharagpur – 721 302, India

The F-band absorption and thermoluminescence (TL) of as-cleaved or quenched KBr single crystals after X-ray irradiation and later exciting with a laser beam have been studied. The F-centre concentration at room temperature is considerably reduced with laser excitation as is the TL output. These experiments indicate that a considerable concentration of F-centres in X-ray irradiated KBr crystals can be destroyed even at room temperature by laser excitation. An attempt is made to interpret these results.

1. Introduction

A study of colour centre phenomena in alkali halide crystals (e.g. KCl and KBr) irradiated with ionizing radiations such as X-rays, yielded valuable information regarding the defect processes taking place in them [1, 2]. The F-centre, which is a negative ion vacancy that trapped an electron, has been extensively studied and is well understood. The thermoluminescence (TL) of X-ray irradiated KBr exhibits two peaks: these lie at 90 and 152° C, and are shown to be due mainly to the destruction of F-centres [3, 4].

It has been reported [5, 6] that if these alkali halide crystals are subjected to high electric fields and later irradiated with X-rays, interesting results have been obtained. For example, the field-treated samples, on X-ray irradiation, exhibited an appreciable increase in the F-centre concentration; the TL light output also increased considerably. During subsequent studies on the colour centre phenomena in these crystals when X-ray irradiated under high electric fields, it has been shown that a considerable concentration of F-centres is destroyed even at room temperature ($\sim 30°$ C) by subjecting the X-rayed crystals to a high a.c. or d.c. field [7, 8]. It appears that if X-ray irradiated alkali halide crystals such as KBr are excited with laser light at room temperature, a similar effect seems to be exhibited. It is the aim of this paper to report the results of our measurements of the decrease in F-centre concentration in KBr crystals X-ray irradiated at 30° C and later excited with laser light for different times.

2. Experimental methods

The KBr single crystals used in the present work are laboratory grown; measurements were also taken on some samples of KBr crystals obtained as a gift from Crystal Physics Laboratory, Massachusetts Institute of Technology, USA. Samples are cleaved from large boules, ground and optically polished, the final dimensions of the samples being $1 \times 1 \times 0.1\,\mathrm{cm}^3$. These crystals are of high quality with a low defect concentration as indicated by a low value for the absorption coefficient measured at 200 nm (data not shown in the figures) [9].

X-ray irradiation of the samples was carried out at 30° C for 2 h (half the time on either side of the sample) with 35 kV, 10 mA, keeping the samples 2 cm from the window of a Norelco unit. Excitation with laser light was done using a He–Ne laser of 2 mW power and 632.8 nm wavelength, keeping the samples at a distance of about 30 cm from the laser source.

The F-centre absorption measurements were taken at 30° C using a Beckman 26 spectrophotometer. The accuracy in the measurement of absorption coefficient, α, is $0.05\,\mathrm{cm}^{-1}$. The TL light output was recorded on an Esterline-Angus recorder using conventional TL apparatus.

3. Results

Fig. 1 shows the F-band absorption characteristics of as-cleaved KBr crystals before and after X-ray irradiation and also when they are later excited with laser light. The absorption in the F-band (with a peak at 625 nm) is found to decrease with time of laser excitation. A similar result is obtained in KBr crystals which are initially quenched from 600° C, though the initial concentration of F-centres in the quenched crystals is larger (hence the figure is not presented). Using Smakula's equation [2] and assuming the F-band to be Gaussian in shape, the F-centre concentration in these samples under different conditions is calculated and plotted as a function of time of laser excitation (Figs 2a and b). We find that the F-centre concentration decreases in as-cleaved crystals by 33% for up to 2 min laser excitation and by 37% (i.e. from $1.2 \times 10^{16}\,\mathrm{cm}^{-3}$ to $0.8 \times 10^{16}\,\mathrm{cm}^{-3}$) in the next 3 min excitation; the total number of F-centres destroyed up to 5 min is nearly 58% (the total reduction in F-centre concentration for the same time of laser excitation in quenched and X-ray irradiated KBr samples is 65%). The TL output in X-ray irradiated KBr crystals which are later excited with laser light is also found to

0022–2461/86 $03.00 + .12

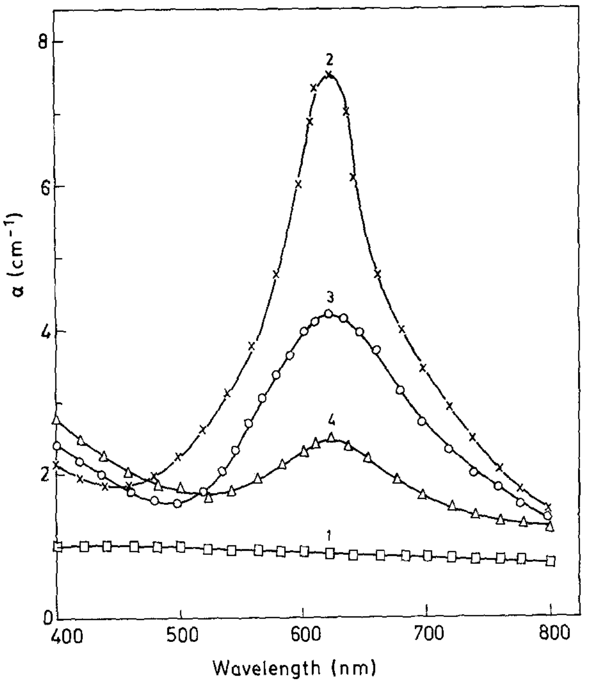

Figure 1 F-band absorption characteristics of KBr single crystals: (1) as-cleaved; (2) as-cleaved and X-ray irradiated; (3) as-cleaved plus 2 min laser excitation; (4) as-cleaved plus 5 min laser excitation.

decrease with time of laser excitation and this decrease is considerable up to 2 min laser excitation, beyond which the reduction in TL output with time of laser excitation is comparatively less.

The F-band absorption characterstics of KBr samples under the conditions (i) initially excited with laser light for 5 min, (ii) later irradiated with X-rays for 2 h, and then (iii) later excited with laser light for different times, are given in Fig. 3. We notice that the absorption in the as-cleaved KBr increases, particularly in the low wavelength region, on laser excitation of the samples; in addition the F-band absorption in the KBr crystals initially laser-excited and later X-ray irradiated, is larger (compared to the as-cleaved crystals which are X-ray irradiated only). Later, if these samples are laser excited, the F-centre concentration is found to decrease. Similar behaviour is exhibited by TL output.

4. Discussion

If alkali halide crystals containing F-centres are excited with light having wavelength corresponding approximately to the F-band peak, then F′-centres (i.e. an F-centre trapping one more electron) are reported to be formed. However, this conversion is shown to be efficient at very low temperatures and the F′-centres are reported [2] to exhibit an absorption

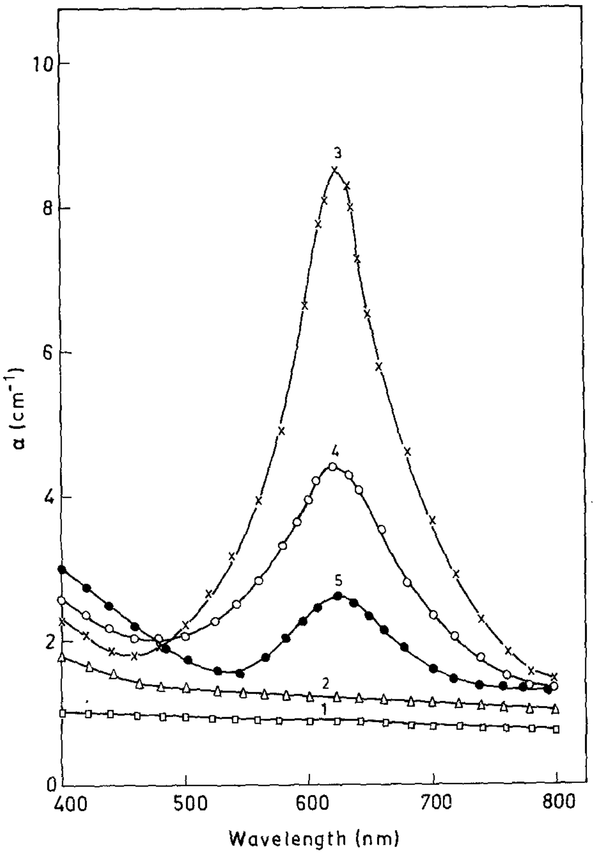

Figure 3 F-band absorption characteristics of KBr single crystals: (1) as-cleaved; (2) as-cleaved plus 5 min laser excitation; (3) as (2) plus X-rayed; (4) as (3) plus 2 min laser excitation; (5) as (3) plus 5 min laser excitation.

band in the larger wavelength region to the F-band, or at least overlapping the F-band, on the larger wavelength side. In the present work, which was carried out at room temperature, no absorption band is found either beyond the F-band or even overlapping it. When the X-ray irradiated KBr crystals are being excited with laser light they exhibit luminescence which is known to occur when electron-trapped centres (i.e. F-centres) recombine with hole-trapped centres. This analysis indicates that F-centres in the present work are not destroyed by being converted into F′-centres but mainly by recombining with hole-trapped centres only. Because of the high intensity of the laser beam, when a KBr crystal is excited with the laser light, thermal spikes may be generated which lead to the generation of defects (like vacancies) in the crystal; in addition, a laser radiation (electromagnetic in nature) associates itself with a strong electric field vector. When the KBr crystal is subjected to this strong electric field (during laser excitation), there can be considerable interaction of this electric field with the

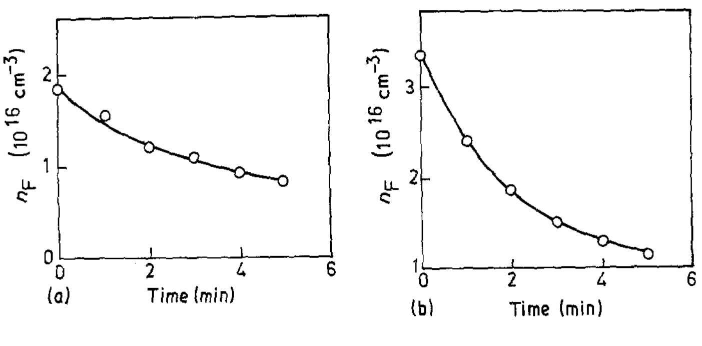

Figure 2 Change in F-centre concentration with time of laser excitation for: (a) as-cleaved, and (b) quenched KBr single crystals irradiated with X-rays.

crystal lattice, particularly at major defect regions like dislocations, and this interaction is known to lead to the generation of vacancies. The increase in optical absorption on laser irradiation, particularly in the low wavelength region, exhibited by as-cleaved KBr crystals supports this view [9]. In other words, excitation with laser light leads to considerable disorder in the lattice. The present results suggest that this disorder facilitates destruction of F-centres even at room temperature.

The decrease in F-centre concentration with time of laser excitation can be represented by the empirical equation [7]:

$$n = n_1 e^{-K_1 t} + n_2 e^{-K_2 t}$$

where n_1, n_2, K_1 and K_2 are constants and t is time of laser excitation. The values for K_1 and K_2 for as-cleaved KBr crystals X-ray irradiated for 2 h and later excited with laser light are 0.15×10^{16} and 0.096×10^{16} F-centres per minute. These data seem to indicate that considerable disorder sets in when the irradiated crystal is subjected to laser excitation for about 2 min resulting in the destruction of a comparatively large concentration of F-centres, but for larger excitation times the disorder is not appreciably further increased.

The total TL output in the irradiated KBr crystals in the as-cleaved condition or when they are later excited with laser light, is expected to be proportional to the initial concentration of F-centres present in them under the corresponding conditions. As such the TL output has to decrease with the time of laser excitation reflecting the behaviour of F-centre reduction with time of laser excitation, as observed in the present measurements.

The data on quenched KBr crystals or KBr crystals initially excited with laser light can be understood in a similar way. Quenching (or laser excitation) is known to produce a large concentration of defects like dislocations and "frozen-in" point defects [6, 10]. Hence the initial concentration of F-centres in these samples is expected to be larger.

We should mention here that the studies reported in this paper have been carried out on samples obtained from both the sources mentioned and similar results have been obtained. Several auxiliary experiments were also carried out (e.g. exciting the X-ray irradiated samples when they were kept in oil) to establish the fact that the reduction in F-centre concentration and TL output is due to laser excitation only.

In summary, the present work suggests that a considerable number of F-centres formed in KBr crystals with X-ray irradiation can be destroyed even at room temperature when these samples are later excited with laser light.

Acknowledgement
The authors are grateful to Professor A. Smakula, former Director, Crystal Physics Laboratory, Massachusetts Institute of Technology, Cambridge, USA, for kindly sparing some of the samples used in this work.

References
1. J. H. CRAWFORD Jr and L. M. SLIFKIN, "Point defects in Solids", Vol. I (Plenum New York, 1972).
2. J. H. SCHULMAN and W. D. COMPTON, "Colour centres in Solids" (Pergamon, London, 1963).
3. P. K. CHAKRABORTI and K. V. RAO, *Phys. Status Solidi (b)* **120** (1983) K107.
4. G. T. HAGESETH, *Phys. Rev.* **B5** (1971) 4060.
5. S. GOVINDA and K. V. RAO, *Solid State Commun.* **16** (1975) 297.
6. S. GOVINDA, PhD dissertation, Indian Institute of Technology, Kharagpur (1975).
7. D. ENAKSHI and K. V. RAO, *Physica Scripta* **31** (1985) 219.
8. *Idem, Nuclear Tracks* **10** (1985) 71.
9. A. K. GUPTA and K. V. RAO, *Phys. Status Solidi (a)* **59** (1979) 277.
10. P. K. CHAKRABORTI and K. V. RAO, *Cryst. Latt. Def. Amorph. Mater.* **10** (1983) 67.

Received 28 October 1985
and accepted 14 March 1986