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Thermoluminescence in LiF crystals and the role of impurities

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Abstract. Eight nominally pure lithium fluoride crystals, obtained from a single LiF crystal containing less than 100 ppm impurities, were irradiated by gamma-rays from a ⁶⁰Co source at room temperature, with doses from 8.4 to 2.5 x 10⁵ Gy, but one at -60 °C. Optical density measurements were performed to investigate the radiation-induced color centres (CCs) and to evaluate their concentrations. Thermoluminescence (TL) glow curves were collected and simulated by a first-order kinetics approach, and from the best-fit procedure ten glow peaks (GPs) spanning from 100 to 450 °C were highlighted. A comparative analysis of GP intensities and CC concentrations as a function of the irradiation dose has questioned their association as obtained in previous measurements, showing the impurities, less than 10¹⁸ cm⁻³, still playing a predominant role in TL spectra. New measurements on LiF crystals more pure, at least an order of magnitude, are required to establish for sure the association of GPs to CCs.

1. Introduction

Thermally stimulated light emission from an insulator or a semiconductor crystal, following the previous absorption of energy from ionizing radiation, results in the typical thermoluminescence (TL) glow curves, from which information on the electron traps present in the crystal band gap can be extracted [1]. TL is one of the most frequently used techniques in radiation dosimetry for personal and environmental use, with outstanding success [1, 2]. Among the various crystalline materials utilized as TL dosimeters, lithium fluoride (LiF) is the most common because of its almost biological tissue equivalence and its chemical-physical properties. LiF crystals are relatively hard and almost not hygroscopic. Irradiation of LiF crystals by ionizing radiation (X-rays, gamma-rays, protons, electrons, etc.) induces the stable formation of primary and aggregate color centers (CCs), whose optical and spectroscopic properties are rather well known [3]. The primary electronic defect is the F center, which consists of an anionic vacancy occupied by an electron. Its absorption band, called F band, is located at about 248 nm. Up to now, photoluminescence originating from the F center in LiF has not



been detected unambiguously. The F_2 and F_3^+ aggregate centers (two electrons bound to two and three anion vacancies, respectively) possess almost overlapping absorption bands at about 450 nm, which together form what is generally called M band [3]; under optical pumping in this spectral region they emit broad photoluminescence bands peaked at 678 nm and 541 nm for F_2 and F_3^+ centers, respectively [4]. The F_3 aggregate centers (three F centers, located at the vertices of an equilateral triangle in the lattice plane $\{1,1,1\}$) give rise to two principal absorption bands, $F_3(R_1)$ and $F_3(R_2)$, located at around 3.92 eV (316 nm) and 3.31 eV (374 nm), respectively [5].

Even though the TL properties of LiF have been extensively investigated since long times, the role of impurities and radiation-induced CCs in TL spectra [6] has still to be clarified. Indeed, the mechanism of TL is not known in detail yet, although a general description has been attempted with some success. This situation has been addressed lately having in mind the role of impurities versus the properties of pure LiF material [7, 8].

During the last decade systematic studies by TL technique have been carried out to establish the role of radiation-induced CCs in TL spectra, with promising results [6, 9]. In particular, four LiF crystals, irradiated with the same dose and properly treated in order to get a wide variation of CCs content, have been studied with interesting results [7]. In order to extend that latest investigation and clarify the role of CCs and impurities in TL glow curves, we started a new campaign of TL measurements on a larger number of LiF crystals and with a much wider range of irradiation doses. In this work some preliminary results are reported and discussed.

2. Materials and Methods

Eight nominally pure LiF samples, obtained from a single crystal, containing less than $10^{18}/\text{cm}^3$ impurity ions in total, were irradiated at increasing doses by gamma-rays from a ^{60}Co source. Seven samples were irradiated at room temperature (RT) with doses from 8.4 and 2.5×10^5 Gy, and one with a dose of 8.4×10^4 Gy while keeping its temperature at -60 °C (213 K).

Optical density measurements were performed at normal incidence and at RT, by using a double-monochromator Perkin-Elmer Lambda 950 spectrophotometer, in the 190-1400 nm spectral range with 1 nm of resolution.

TL was measured in the range 50-450 °C (323-721 K) with a heating rate of 0.25 °C/s (15 °C/min) by using a commercial automated RISØ TL/OSL-DA-15 reader, also equipped with a $^{90}\text{Sr}/^{90}\text{Y}$ beta (β) radiation source (0.565 MeV mean β energy, dose rate 0.119 Gy/s). The glow curves were measured from small fractions of powder obtained by pounding small pieces of the LiF crystals under a nitrogen flux. Five pieces of each irradiated LiF crystal, of about equal weight, were pounded and measured for statistical purposes, and the final TL glow curves were averaged by taking in account the weight of each powder sampling.

3. Results and Discussion

Figure 1 shows the optical density spectrum and its best fit of sample S15, which was irradiated at RT at a dose of 2.5×10^3 Gy. The most visible spectral features are the F band, peaked at 5 eV (248 nm), and the M band located at about 2.76 eV (450 nm). The absorption band contributions of more complex CCs, such as $F_3(R_1)$, $F_3(R_2)$, $F_4(N_1)$ and $F_4(N_2)$ located at about 3.92 eV (316 nm), 3.31 eV (374 nm), 2.40 eV (517 nm) and 2.26 eV (547 nm), are also highlighted, although not observed in this sample.

Figure 2 shows an example of a TL glow curve (sample S15) and its best fit achieved by using the GlowFit free-downloadable software, which was especially designed to simulate TL glow curves resulting from up to ten single or superimposed glow peaks (GPs) following first order kinetics [10]. Comparing the results of the present investigation [11] with previous ones [6, 9], it appears that the GP temperatures differ in the two measurements by about 19 °C in average. This is due in part to unavoidable instrumental systematic errors and mostly to an improved best-fit procedure in the present case.

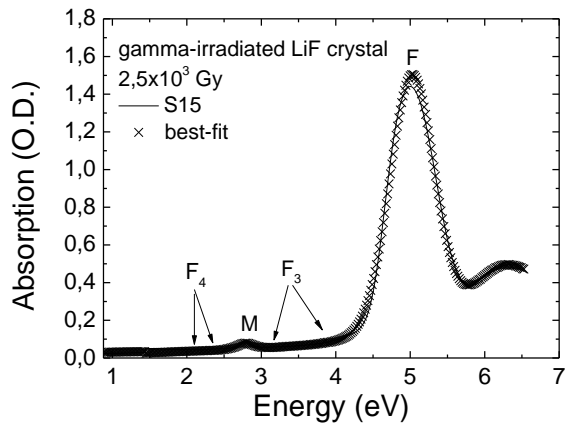


Figure 1. RT optical density spectrum of the gamma colored LiF crystal at a dose of 2.5×10^3 Gy, S15 sample, (black full line) and its best fit (x symbol). The thickness of the crystal is 2 mm.

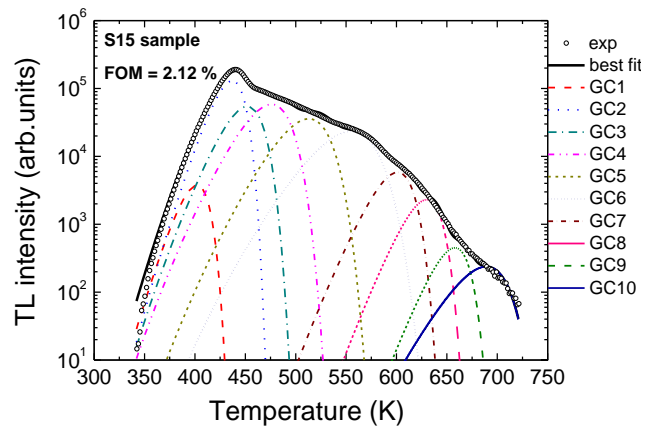


Figure 2. TL spectrum of the gamma colored LiF crystal at a dose of 2.5×10^3 Gy, S15 sample, (circle), its best fit (black full line) the ten glow peaks (colored lines). The Figure Of Merit (FOM) is indicated.

From optical density measurements it is possible to approximately calculate the amount of CCs in the irradiated LiF crystals [12]. Figures 3 and 4 report in a log-log scale the concentrations of F centers, and $F_3(R_1)$ and $F_3(R_2)$ centers, respectively, as functions of the irradiation dose. The experimental points have been interpolated with a line, except for sample S18 which was treated differently (dose of 8.4×10^4 Gy at -60°C).

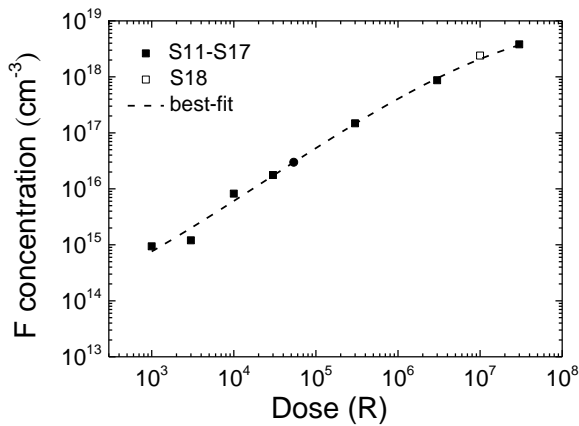


Figure 3. Concentration of F centers as a function of gamma ray dose for the LiF crystals colored at RT and utilized in the present study ($1 \text{ R} \cong 8.3 \cdot 10^{-3} \text{ Gy}$).

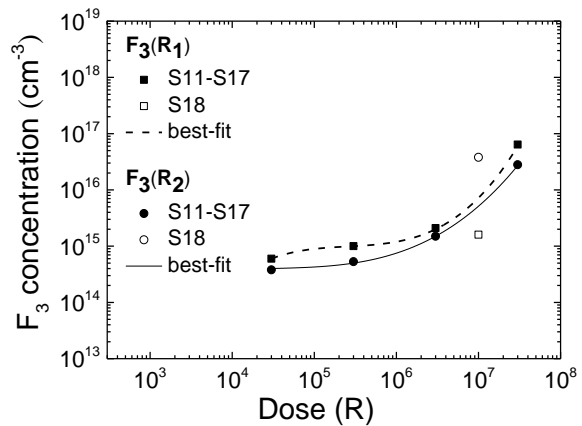


Figure 4. Concentration of $F_3(R_1)$ and $F_3(R_2)$ centers as functions of gamma ray dose for the eighth samples utilized in the present study. The three samples irradiated at the lowest doses do not give appreciable numerical values ($1 \text{ R} \cong 8.3 \cdot 10^{-3} \text{ Gy}$).

Figures 5 and 6 report the intensities of GP7, and GP3 and GP4, respectively, as functions of the irradiation dose. GP7 intensity shows a linear behavior with dose, thus it can be related to F centers, whose concentration is also linear with the dose, see figure 3, and moreover the sample S18 result is compatible with linear behavior in both graphs. In the previous analysis it was associated with F centers bound to impurities contained in LiF crystals [9], which cannot be excluded by current investigation.

In the previous analysis [9], GP3 and GP4, figure 6, were associated with F_3 centers, but examining the behavior of the F_3 concentrations as functions of dose, figure 4, this assignment cannot be confirmed. Indeed, the peak intensities of GP3 and GP4 grow together with the concentrations of $F_3(R_1)$ and $F_3(R_2)$, albeit with a different behavior, but, while the concentrations continue to grow, GP3 and GP4 intensities saturate. Moreover, also the values of the F_3 concentrations and the GP3 and GP4 intensities of sample S18 do not support this association. The observed saturation can be explained with the hypothesis that these GPs are due to the presence of CCs bound to impurities contained in the LiF crystals. Indeed, at a certain dose, there are not “enough impurities” to be bounded to CCs, and the related GP intensities cannot grow anymore. Similar behavior has been found in the other samples [11].

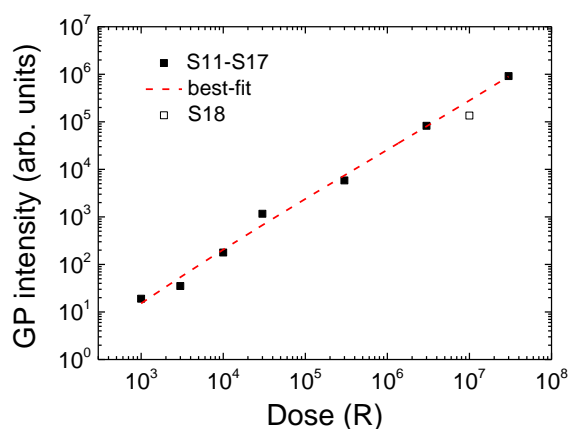


Figure 5. Intensity of GP7 as a function of gamma ray dose for the eighth samples utilized in the present study ($1 R \cong 8.3 \cdot 10^{-3} \text{ Gy}$).

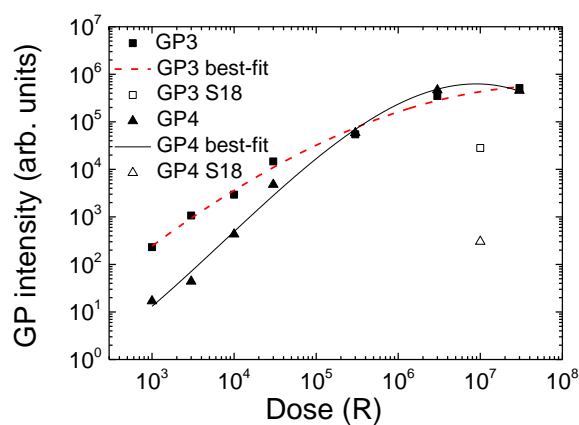


Figure 6. Intensity of GP3 and GP4 as a function of gamma ray dose for the eighth samples utilized in the present study ($1 R \cong 8.3 \cdot 10^{-3} \text{ Gy}$).

4. Conclusions

The association between GPs and CCs performed a few years ago [4, 7] was based on the knowledge of the physics of CCs and accurate annealing measurements on a restricted set (only four samples) of LiF crystals that had been gamma-irradiated at the same dose, and so not completely satisfactory.

This new campaign of measurements, performed on a larger number of LiF crystals and with a much wider range of irradiation doses, has shown, in the framework of the first-order kinetics theory, that the previous association between CCs and GPs is strongly questionable and that impurities could be responsible for certain GPs that had been earlier assigned to CCs. These results highlight the paramount influence of residual impurities in LiF crystals, even if apparently negligible, in TL measurements. New measurements on LiF crystals, containing much less impurities, are required to establish on more sound bases the association of GPs to CCs.

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