

Optical absorption and thermoluminescence in LiF TLD-100

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The thermoluminescent (TL) properties of many LiF samples have demonstrated a strong influence on trace impurities and impurity content on the TL process. Hence, the general validity of any particular model must be tested against its applicability in a standard material such as Harshaw LiF (TLD-100). In this paper the validity of the use of Harshaw LiF(54) by Mayhugh *et al.* in their model development for the TL process in LiF is demonstrated by comparing the TL and optical properties of LiF(54) with TLD-100. The specific properties of optical absorption bands at 310 and 380 nm, the *F* band near 250 nm, and the Z_3 band near 225 nm are intercompared with observed TL peaks in both materials. Both the gamma and uv-exposure behavior of the Z_3 center demonstrates a direct relationship between Z_3 and TL peak 10, while no direct conversion between Z_3 and the 310-nm band is observed in TLD-100. These results do not support recent models identifying Z_3 with TL peak 6 and Z_2 with the 310-nm band (and TL peak 5); this is probably due to differences in purity and defect state in the LiF samples used by other investigators, emphasizing the need for standardization of materials in this system.

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I. INTRODUCTION

Thermoluminescence (TL) in dosimetry grade LiF depends on at least two impurities, Mg and Ti.¹⁻⁴ Magnesium-related defects constitute the TL traps and during irradiation these capture electrons to form the TL centers. Thermally releasing the electrons from these trapping centers initiates the TL process for the major glow peaks numbered 2 to 5 and peaking between 100 and 220 °C. In this paper the prefix "TL" indicates the traps and centers active in this initiation, and a specific label like "5" similarly implies the specific trap or center which gives rise to peak 5, the main dosimetry peak. While intact, the TL centers cause optical absorption in the 310–380-nm region and they may be studied through this attendant absorption.⁵⁻⁸ Both 5 and 4 centers cause optical absorption bands centered near 310 nm while 2 and 3 centers are thought to have bands near 380 nm. As inferred from dielectric loss and ionic conductivity measurements, the structures of the TL traps are thought to involve Mg^{++} cation vacancy dipoles (2 centers) and dimer or trimer complexes of these (5 centers),^{4,9} although Nink and Kos have proposed an alternative *Z* center model to explain the observed optical and TL behavior.^{8,10,11} In addition, Mg and other defects also create deep traps whose centers are relatively stable at 250 °C and higher.

Titanium activates the luminescence process; that is, once electrons are thermally released from the TL centers, the radiative transitions causing photon emission occur at luminescence sites involving the Ti impurity.^{1,2} Further, Ti in some configurations causes optical absorption bands near 200 nm, present even before irradiation¹²; illumination into this band produces a photoluminescence whose emission

spectrum is the same as that of the TL. Further studies have indicated the importance of the hydroxyl impurity^{13,14} possibly transformed to oxygen impurities during irradiation^{15,16} in the luminescent process.

The complete TL mechanism represents the combination of the trapping and luminescence parts. For radiation exposures to about 0.1 C kg⁻¹, the TL output (peaks 2 to 5) increases linearly with exposure, reflecting a growing population of TL centers. Above about 0.1 C kg⁻¹, TL grows more rapidly, that is, supralinearly, until at about 25 °C kg⁻¹ the growth response begins to decrease (saturation).⁴ Heating a sample at 280 °C after irradiation into the supralinear region leaves the phosphor with an increased TL response to a subsequent exposure and the material is said to be sensitized.^{4,17} The sensitizing treatment leaves a high-temperature TL peak 10 near 400 °C and the Z_3 band (225 nm) in the optical absorption spectrum. Recent work demonstrates that the Z_3 center does not relate directly to the normal TL traps.¹⁸

A variety of TL work on many LiF samples has demonstrated that trace impurities and variations in impurity content can drastically affect the results obtained.^{1-4,13,16,18} Since the data used in developing the model often used to explain TL in LiF:Mg, Ti was obtained using 1954 vintage Harshaw LiF rather than commercial Harshaw LiF TLD-100, the first goal of this paper is to confirm that the optical and TL behavior of 1954 vintage Harshaw LiF and TLD-100 are indeed the same in terms of those parameters used in model development. A second goal is to demonstrate in a consistent fashion the relationship between the various impurity-related supralinearity and sensitization effects in

TLD-100. Finally, these data are utilized to comment on the recent Z -center model for TL trapping in LiF.

II. EXPERIMENT

Dosimetry grade LiF TLD-100 and vacuum ultraviolet grade ("pure") LiF, originating from Harshaw Chemical (Solon, Ohio) were used as single crystals and as powder (800-200 Tyler mesh). Irradiations were performed with ^{137}Cs γ rays or 50-kV peak x rays (effective energy estimated at 20 keV). Calibration of the exposure rate for the x-rays is correct absolutely to only about 15%, although reproducibility is 3% or better.

Thermoluminescence was measured using a Harshaw Model 2000 reader sometimes modified to use a Thermal Technology Associates temperature programmer for slow heating rates ($< 140^\circ\text{C}/\text{min}$) or high temperatures (to 500°C). So-called "permanent" planchets were employed. Reproducibility was about $\pm 3\%$. Optical absorption was measured with a Zeiss Model DMR-21 WZ spectrophotometer capable of scanning the region 2500 to 185 nm. The optical samples measured about $1 \times 1 \times 0.15$ cm and were mounted for measurement along a 1-cm optical path.

Light for optical treatments originated from a Bausch and Lomb SP-200 super-pressure mercury source sometimes coupled to a matching high-intensity monochromator. The intensity of monochromatic light was measured with an E.G.&G. Model 580 radiometer. Illumination of powdered samples occurred by way of a front-surfaced aluminum mirror.

III. RESULTS

A. Comparison of TLD-100 and LiF(54)

Early LiF thermoluminescent model development concentrated on old LiF samples purchased in 1954 rather than TLD-100.^{5,6,12} This old material, labeled LiF(54), accidentally displayed useful TL properties and has low enough impurity concentrations to be studied in the vacuum ultraviolet region while TLD-100 has concentrations that are too high for such studies. Studies on LiF(54) included optical and thermal treatments to identify charge carriers and correlate TL peaks with optical absorption bands. We have repeated many of these experiments in TLD-100 to be certain that results in the two materials can be compared, and find that the two behave similarly, a fact which others have also noted in specific cases.¹⁹

In both materials the glow curve is composed of five prominent peaks occurring at the same temperatures. The optical absorption spectra also appear to be similar. Before irradiation, overlapping impurity bands are seen near 200nm, one at 198 nm and the other at 205 nm. The 205 nm band can be reduced by thermal treatments or irradiation, as in LiF(54). As will be shown below, the sensitization procedure produces the Z_3 band near 225 nm in TLD-100, much as in LiF(54). The 310- and 380-nm bands are also present in TLD-100, as many others have also noted. These are thought to correlate with TL peaks 2 and 3 (380-nm band) and with peaks 4 and 5 (310-nm band).⁵⁻⁸

Following irradiation, an hour's 100°C anneal eliminates peaks 1, 2, and 3 from the TL glow curve and the 380 nm band from the optical absorption spectrum while peak 5 and the 310-nm band grow. Observing this growth in peak 5 depends on the heating rate. The F band (250 nm) falls during the 100°C anneal as does the nonradiation-induced 20-nm band. These facts are already well established in both TLD-100 and LiF(54); the natural course of this work reconfirmed them

After irradiation and a 100°C anneal the glow curve contains peaks 4 and 5, while the radiation-induced absorption spectrum is dominated by the 310-nm (and F) bands; these situations are shown in Fig. 1. The effect of a subsequent 310-nm optical bleach is also shown in this figure; TL peak 5 falls, peak 4 grows, and the 310-nm band decays. During this bleach the F band also grows. Growth of the F band (and decay of the V_3 band near 110 nm, which was not studied in TLD-100) is the main evidence that 5 centers release electrons during heating.

The details of the decay of the 310-nm band are shown

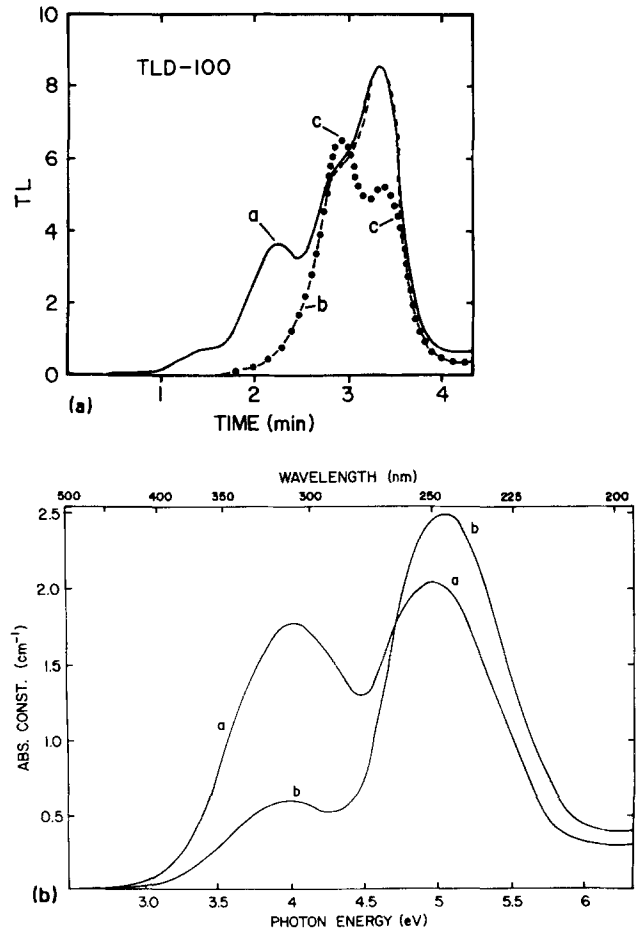


FIG. 1. (a) Thermoluminescence. Curve a shows the thermoluminescence induced by irradiation to 0.2 C kg^{-1} . The dominant peak is 5. Curve b shows the TL remaining when irradiation is followed by 60 min at 100°C before read out. Curve c results when irradiation and annealing are followed by 227 min of illumination with 310-nm light. (Heat at $60^\circ\text{C}/\text{min}$). (b) Optical absorption. Curve a is the optical absorption induced by irradiating TLD-100 to 7.2 C kg^{-1} then annealing for 60 min at 100°C . Curve b results when the sample has been exposed to 310-nm light for 300 min.

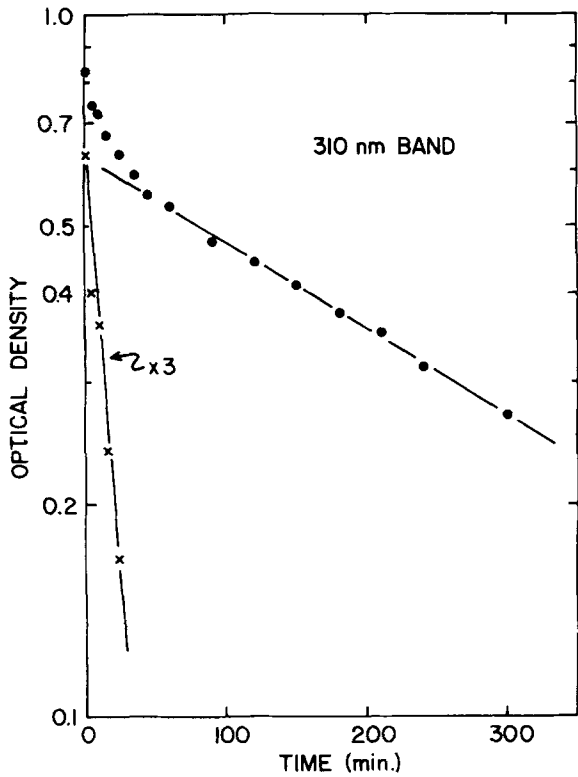


FIG. 2. Decay of 310-nm band during illumination with 310-nm light.

in Fig. 2. The band falls in a manner characteristic of two exponential components, similar to the results for LiF(54). The fast bleaching 310-nm centers have been correlated with peak 5 and the slow ones with peak 4. The decay rates for the 310-nm band (5 and 4 centers) cannot be compared since the lamp intensity was not accurately measured for the LiF(54) experiment. However, the ratio of the two exponential rates should be independent of lamp intensity; indeed this ratio is the same for LiF(54) and TLD-100.

After irradiation and a 100 °C anneal, irradiation with 250-nm light has the effects shown in Fig. 3. The TL (peak 5) begins falling at once, as does the *F* band, but the 310-nm band is initially unaffected. Eventually the 310-nm band begins to fall. After long bleaching times, the absorption is seen to consist of the 310-nm band, a possible absorption band in the 270–280-nm range, also reported earlier,^{6,7,20} and the *Z*₃ band near 225 nm. These results are similar to those observed in LiF(54).

B. Trap filling, supralinearity, and sensitization

In this section we discuss an experiment which confirms that supralinearity and sensitization for glow peak 5 do not result from an increase in the number of 5 centers produced during irradiation. The relative number of 5 centers present after irradiation is measured using the intensity of the 310-nm optical absorption band, which, prior to bleaching, has been shown to measure directly the concentration of 5 centers.

The circles in Fig. 4 show the growth of TL peak 5 (solid) and the 310-nm band (open) in a single crystal of TLD-

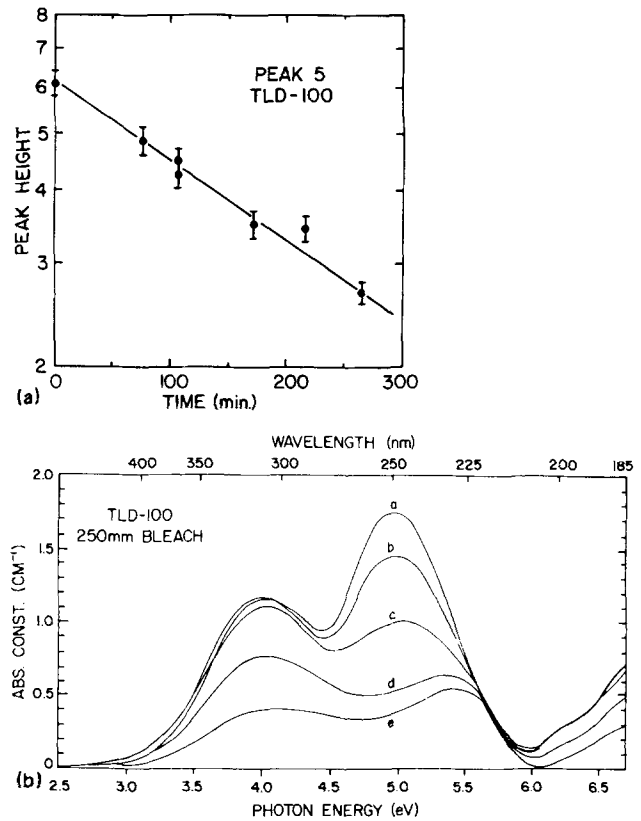


FIG. 3. (a) Decay of TL peak 5 during illumination with 250-nm light. Samples were irradiated to 8.8 C kg⁻¹ and annealed 30 min at 110 °C prior to illumination. (b) Changes in the optical absorption spectrum during 250-nm bleaching. Curve a results from 16 C kg⁻¹ irradiation plus 60 min at 100 °C. Curve b results after 15 min of 250-nm light, curve c after 60 min total, curve d after 210 min total, and e after 540 min total.

100. Clearly the TL grows supralinearly while the absorption grows linearly. After 7.7 C kg⁻¹ the samples were heated 15 min at 280 °C, then exposed again. Figure 4 also shows

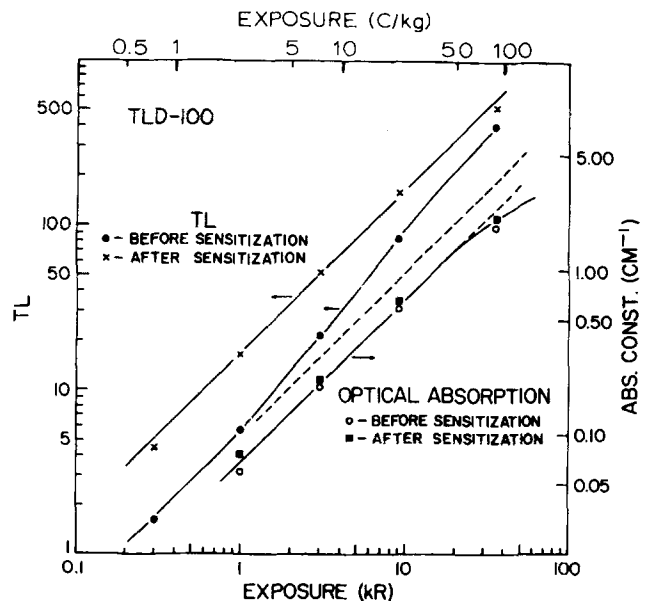


FIG. 4. Growth of radiation-induced TL, peak 5, and the 310-nm optical absorption band in a single crystal of TLD-100 before and after sensitization (see text).

growth of the TL and optical absorption after this sensitization. Now the TL growth is linear but sensitized while the optical absorption remains linear with no comparable sensitization. These results confirm the conclusion¹⁷ that supralinearity and sensitization result from changes in the luminescence efficiency (i.e., the number of photons produced per thermally freed electron) rather than from changes in the number of centers created during irradiation.

The details of this experiment are as follows. Twenty small crystals were cleaved from a larger optical crystal of TLD-100, then the growth of optical absorption in the large pieces and TL in the smaller ones were measured, all for increasing exposures to radiation. After each exposure the samples were annealed for 15 min at 100 °C, since this treatment is the one for which the 310-nm band is most strongly correlated with peak 5. The TL samples were treated together with the optical one until the TL was read. (For example, 0.25 C kg⁻¹ and 100 °C was administered to the optical sample and all the TL samples, readings were taken on the optical sample and two of the TL samples, then these two TL samples were discarded. The optical sample and the remaining TL ones were then given 0.5 C kg⁻¹ more plus the 100 °C, readings were repeated, etc.) Thus, the absorption and TL readings are for TLD-100 samples from the same crystal which have received identical treatments. After the exposure totalled 7.7 C kg⁻¹ the optical sample and the 10 remaining TL crystals were annealed for 15 min at 280 °C. This history of exposure and annealing sensitized the TL response. After this sensitization the growth versus exposure was measured again.

The absorptions at 250 nm (*F* band) and at 185 nm are plotted in Fig. 5 against exposure both before and after sensitization. The *F* band grows linearly before and after sensitization

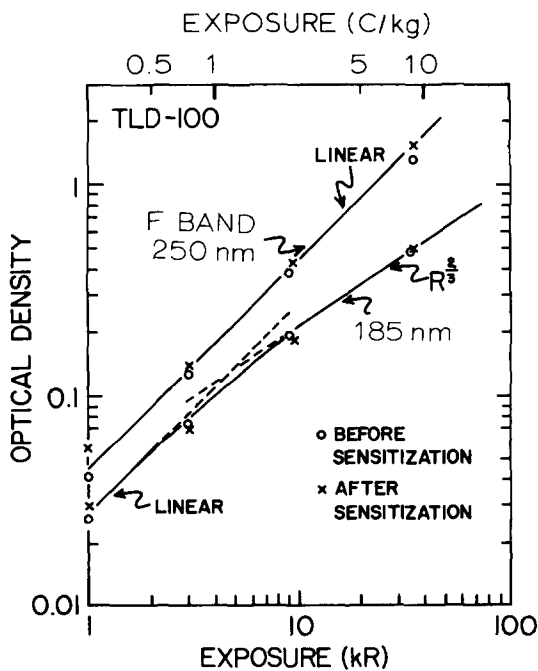


FIG. 5. Radiation-induced growth of the *F* band and bands at 185-nm, before and after sensitization.

just as the 310-nm band does. The absorption at 185 nm first grows linearly, then as the two-thirds power of the exposure; this latter behavior is similar to a broad band at 165 nm (7.5 eV) in pure LiF.⁶ The optical absorption at other wavelengths may also be of interest and for completeness Fig. 6 displays the entire absorption spectra for growth before and after sensitization. These data indicate a lack of supralinear response in the *F*, 310- or 185-nm bands, and that, to within 20%, the radiation-induced absorption curve after sensitization [Fig. 6(c)] is identical to that before the sensitization treatment [Fig. 6(a)]. The same treatment

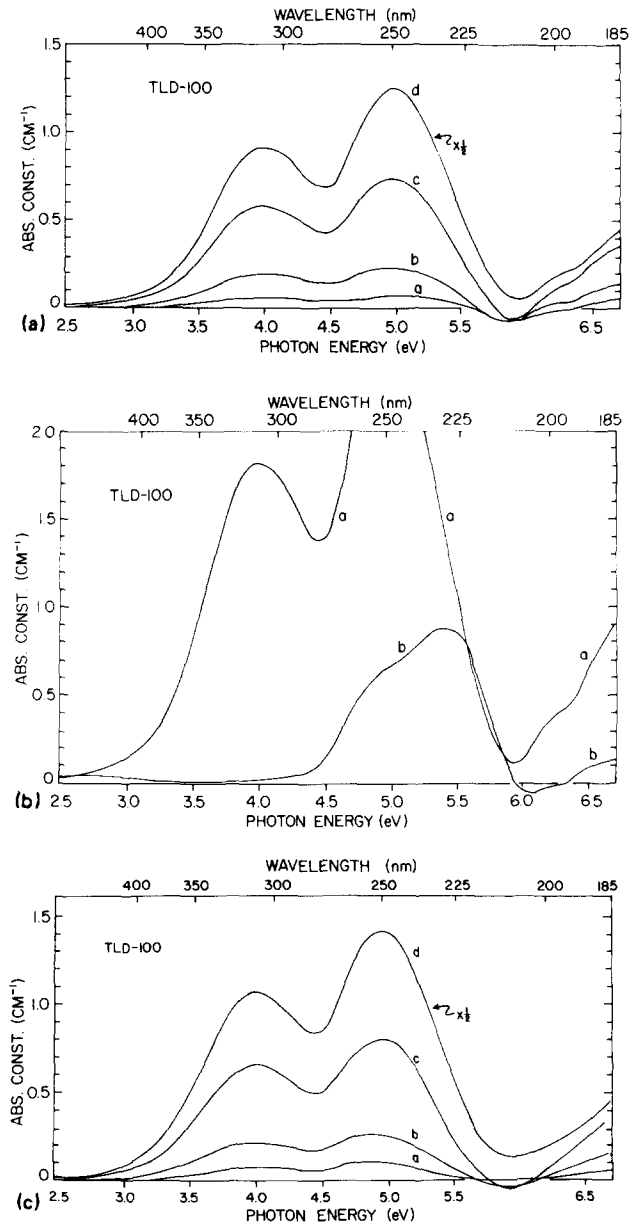


FIG. 6. The detailed growth of radiation-induced optical absorption before and after sensitization. All irradiations are followed by 15 min at 100 °C. (a) Unsensitized. Curve a, 0.26 C kg⁻¹; curve b, 0.77 C kg⁻¹; curve c, 2.3 C kg⁻¹; curve d, 8.8 C kg⁻¹ total exposure. (b) Effect of annealing. Curve a is the same as curve d in (a). Curve b shows the effect of annealing 15 min at 280 °C. (c) Sensitized. Curve a, 0.26 C kg⁻¹; curve b, 0.77 C kg⁻¹; curve c, 2.4 C kg⁻¹; curve d, 9.0 C kg⁻¹ total exposure. All curves in (c) are referenced to the base line of a sensitized sample, i.e., curve b in (b).

causes an increase in the TL response by a factor of 5. This indicates that sensitization and supralinearity are not associated with increases in any of these optical absorption bands or, by inference, with increases in the population of TL traps.

C. Deep centers

Sensitized TLD-100 displays the Z_3 (225-nm) band in the absorption spectrum [Fig. 6(b)] and a glow peak labeled No. 10 near 400 °C in the glow curve. Each of these has been suggested to be critical to sensitization,¹⁸ and in fact this absorption band and this glow peak may be different manifestations of the same centers.

To produce Z_3 , irradiation *plus* annealing (usually at or above 280 °C), is required. Once produced, this band is relatively stable at 280 °C, as indicated in Fig. 7(a). This stability puts in question the identification of the Z_3 center as the trap responsible for a TL peak near 285 °C, identified by Gartia,²¹ or for a TL peak near 250 °C reported as peak 6 by Kos and Nink.²² Our results, not illustrated, demonstrate that peaks 6 and 7 in the 250–285 °C region are thermally depopulated by annealing for 15 min. in this temperature range, ruling out peaks 6 and 7 as being associated with the more stable Z_3 center. The isothermal decay of the Z_3 band at higher temperatures is also shown in Figs. 7(a) and 7(b). At each temperature thermal destruction is exponential with time, except for a small portion of the band (20%) which decays

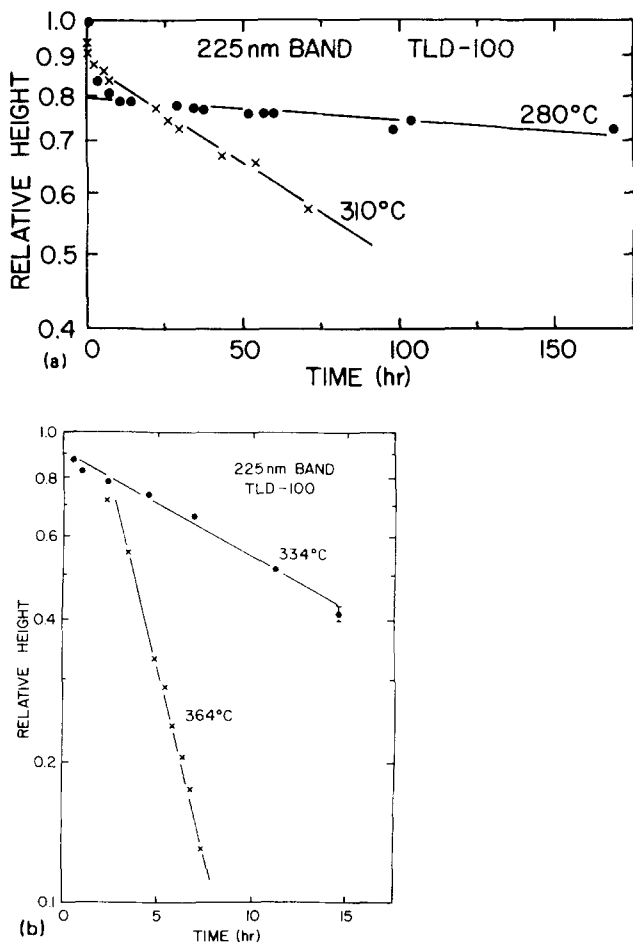


FIG. 7. Isothermal decay of the Z_3 band. (a) at 280 and 310 °C. (b) At 334 and 364 °C.

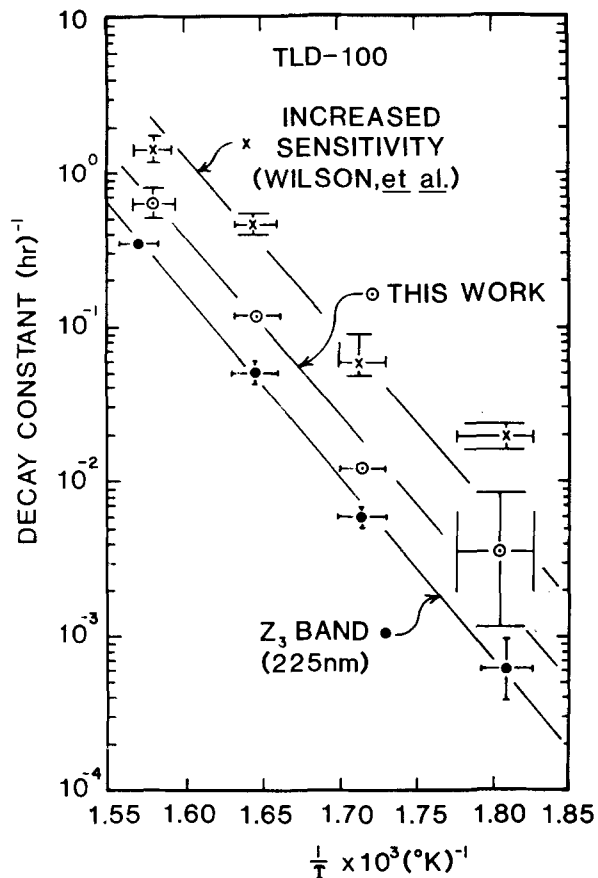


FIG. 8. Thermal decay constant plotted against reciprocal temperature for the Z_3 band (dots) and for the increased sensitivity (circles), this work; (×)'s, see Wilson *et al.*, Ref. 23.

faster than the characteristic rate. Sensitized TL samples also show an exponential decay of the increased sensitivity, but at each temperature the Z_3 band decays more slowly than the increased sensitivity.

The decay constant for the thermally stimulated decay of both the Z_3 band and of the TL sensitivity, plotted versus reciprocal temperature, is shown in Fig. 8. In this figure, the lower curves shows the decay constant α for the Z_3 band determined from Figs. 7(a) and 7(b) using the relationship $I = I_0 \exp(-\alpha t)$, where I_0 and I are the initial and actual Z_3 band height at times t_0 and t . The upper curve shows thermal decay of the increased sensitivity as measured by Wilson *et al.*,²³ while the middle curve is the same decay measurement for the TL sensitivity performed during this work. The observation that the slopes of these curves are nearly the same may indicate that a similar mechanism is operating for the thermal destruction of both Z_3 centers and the increased TL sensitivity. However, the rate of decay of the increased TL sensitivity is greater than that of the Z_3 centers at each temperature, indicating that this mechanism has a greater probability of removing the enhanced sensitivity than of removing the Z_3 center from the system. The apparent activation energy for the destruction mechanism, measured from the slope of Fig. 8, is 2.35 eV.

When a sample is exposed to 250-nm light while being held at approximately 280 °C, the Z_3 band is destroyed much

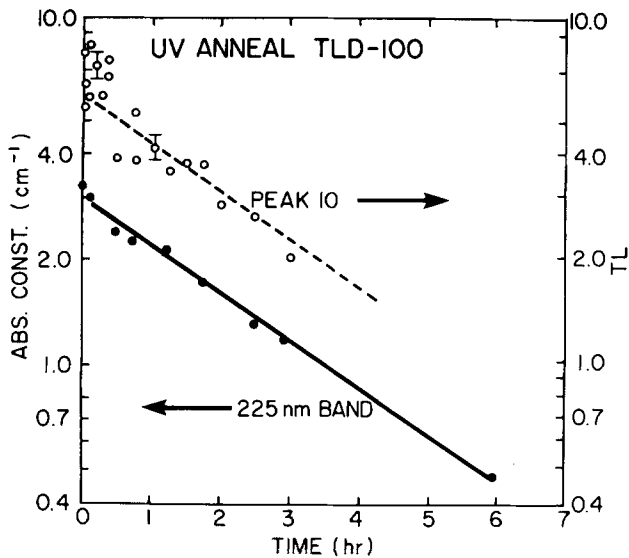


FIG. 9. Decay of the optical absorption of the Z_3 band (left scale) and of the TL glow peak 10 (right scale) caused by ultraviolet light irradiation at 280°C as a function of ultraviolet irradiation time.

more quickly than by the thermal treatment alone. Figure 9 shows the decay of the Z_3 band during such a uv anneal along with a similar decay for glow peak 10. Results not shown verify that this same treatment does not alter the increased sensitivity in peak 5, as also noted by others.^{24,25}

Finally, Fig. 10 shows the growth of the Z_3 band as a function of exposure. Notice that Z_3 growth is linear well beyond 25 C kg^{-1} where the TL (peak 5) saturates. Recall that most of Z_3 is not produced directly during irradiation, but rather the band arises from exposure plus annealing. To construct Fig. 10 five individual crystals were given the indicated exposures then all received a 280°C anneal for 15 min. The figure presents the resulting band height at 225 nm versus exposure. Parallel TL measurements show that peak 10

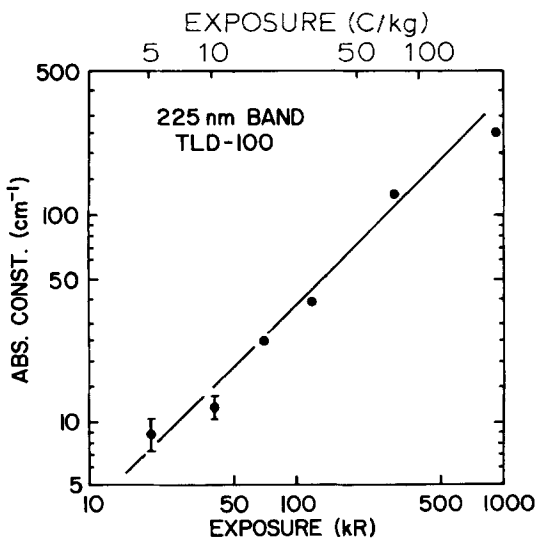


FIG. 10. Height of the Z_3 band in individual samples irradiated to the indicated exposures then annealed at 280°C for 15 min.

also is linear in its response to radiation exposure at least to 65 C kg^{-1} .

IV. DISCUSSION

The results of this investigation indicate clearly that the TL models developed using the optical and TL behavior of LiF(54) may be applied equally to LiF TLD-100. This is an important result because it is seen too often that minor discrepancies in specimen impurity content or distribution^{1-4,13} or in sample history¹⁴ can change the optical and TL behavior enough to confuse model development.

The relationships observed in Figs. 4, 5, and 10 indicate linear behavior with exposure for the optical absorption band at 310 nm, the F band near 250 nm, and the Z_3 band near 225 nm. This observation is contrary to the model of Kos, Takeuchi, and co-workers,^{26,27} who suggest that sensitization occurs by the conversion of Z_3 centers to Z_2 centers (identified in their model as the 310-nm band). In particular, the lack of a change in the rate of growth of the 310-nm band in sensitized versus unsensitized samples (Fig. 4), and the lack of significant changes in the shape of the absorption bands (Fig. 6), demonstrate that these absorption bands play no direct part in sensitization or in the supralinearity of the TL response.

As seen in Fig. 8, the activation energy of the decay process of Z_3 centers is identical to that for sensitization, a point also made by others.¹⁸ Z_3 grows linearly beyond the saturation point of peak 5, however, and its bleaching by uv light does not affect sensitization in proportion to the effect on the Z_3 band, indicating that any interrelation between Z_3 and sensitization is only indirect. The results show a definite relationship between Z_3 centers and peak 10, both in terms of their behavior with exposure and after uv bleaching.

The question of the identity of the TL trapping centers has been discussed widely.⁴ The recent work of Nink and Kos^{8,10,11} attempts to identify the 310-nm absorption band, known to be related to TL peak 5 as a Z_2 center. This identification is made on the basis of an observed increase in Z_3 centers ($Mg^{++} - F$ center pairs) as the 310-nm absorption decreases during postirradiation annealing.⁸ Nink and Kos argue that the logical center which would produce Z_3 centers after the electron liberation that takes place during TL is the Z_2 center ($Mg^{++} - F'$ center pair, where F' is an F center with an extra trapped electron).

While it is true that the above $Z_2 \rightarrow Z_3$ conversion is a logical and convenient model for the TL mechanism in LiF, Nink and Kos's results are in conflict with both current and previous optical studies in TLD-100^{6,7,18}. The results of Fig. 1, for example, show that bleaching irradiated TLD-100 with 310-nm light reduces the peak 5 TL but produces only a small and poorly resolved increase in absorption at 225 nm that could be interpreted as a Z_3 center absorption. Further, in the Z -center model, a 250-nm (F -band) bleach should increase the intensities of Z_2 and/or Z_3 if the appropriate empty traps are available. Experimentally the F -band bleach overlaps the Z_3 and 310-nm bands, so that the results observed, in which peak 5 decreases immediately, but the 310-nm band follows somewhat later, seen in Fig. 3, are more difficult to interpret. The 250-nm bleaching does cause Z_3

center formation (225 nm) either by providing electrons to Mg^{++} -anion vacancy pairs already present, or by promoting the formation of Mg^{++} -anion vacancy pairs. If Nink and Kos's mechanism was valid for TLD-100, further F bleaching should provide electrons that could be trapped at Z_3 centers, forming Z_2 centers. Also, in this case, Z_2 and Z_3 should saturate together. These effects are not observed.

Nink and Kos's arguments bring up the question of the location of the Z_1 , Z_2 , and Z_3 bands in LiF. A Mollwo-Ivey plot for these centers based on their positions in NaCl, KCl, and KBr (Ref. 28) indicates that in LiF, Z_1 and Z_2 centers should be observed in the range 265–285 nm. It is unlikely that enough error could exist in the Mollwo-Ivey relationship to allow a Z_2 center at 310 nm. This conclusion was reached early in the 1960's by several research groups working at both room temperature and at 77 °K (Refs. 29 and 30) and has been reconfirmed in this study.

In another experiment, Kos and Nink²⁰ report the identification of the band near 280 nm, also seen by others^{6,7} as the Z_1 band. Their experiment in this case, in which bleaching is done using broad band uv light, causes electron liberation from the observed centers at 380-nm, 310-nm, and the F band, and causes growth of the 280-nm and Z_3 bands. However, such bleaching could also cause growth of the Z_2 band. Since either Z_1 or Z_2 could occur in this range, the exact identification of this band is uncertain.

The above remarks concerning both trapping and sensitization indicate the failure of the Z -center model in explaining the TL process in LiF TLD-100. It should be noted, however, that the LiF samples utilized in the development of this model by Nink, Kos, and co-workers were prepared in their own laboratory. As noted earlier, a variety of workers in this area have seen variations in TL properties with purity and found it to be a virtual impossibility to match the TL properties of TLD-100. Hence, Nink and Kos's results may be valid for their own LiF material but not for TLD-100. This underscores the importance of the verification, presented in this paper, that the TL models developed for LiF(54) are indeed valid in LiF TLD-100.

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