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Optically stimulated luminescence in strongly irradiated anion-deficient corundum and associated phototransfer effects

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Abstract. Optically stimulated luminescence (OSL), its mechanisms and the role of phototransfer effects in it have been comprehensively studied for strongly irradiated anion-deficient corundum with a high-temperature thermoluminescence (TL) peak at 830 K and without it. The fast component of the OSL kinetics in the samples was found to be due to the emptying of the main trap. The slow component was observed only in samples with the 830 K TL peak and was caused by the depletion of deep traps responsible for TL peaks in the range of 500 K≤T≤740 K.

1. Introduction
A new promising dosimetric information readout technology based on the phenomenon of optically stimulated luminescence (OSL), including TLD-500 detectors based on anion-deficient corundum (α-Al2O3-δ) has been under active development since the beginning of the 21st century. It was found in [1] that the upper limit of doses registered by α-Al2O3-δ is 30-300 Gy. Nevertheless, it was noted in [1] that the response and kinetics of the OSL decay curves depend on the degree of deep trap filling and emptying in the temperature region above 500 K. Thus, as the preliminary irradiation dose is increased from 1 to 1000 Gy, the OSL yields change dramatically, and the OSL decay kinetics slow down. As is well known [2], high-dose irradiation of α-Al2O3-δ induces deep trap filling causing a chromium 580 K thermoluminescence (TL) peak and a high-temperature 830 K TL peak with a 720 K shoulder. The light sum of these TL peaks depends on α-Al2O3-δ sample growing technology and impurity composition [3]. In particular, the TL peak at 830 K was found after high-dose irradiation only in a part of the detectors [3]. The proportion of such detectors in the tested batches was not constant, ranging from 40 to 60 %. The objective of this paper is therefore to carry out a comparative study of OSL and phototransfer effects in strongly irradiated α-Al2O3-δ crystals with an 830 K peak and without it.

2. Samples and experimental techniques
The objects of this study were samples of TLD-500 detectors on the basis of α-Al2O3-δ single crystals, some of which were discovered to lack the 830 K TL peak. X-ray irradiation of the samples was performed using an URS-55 X-ray unit (Cu-anode, Uf=55 kV, Imax=18 mA). OSL decay curves under continuous stimulation and TL curves at a heating rate of 2 K/s were measured using a dedicated automatic installation [4]. TL and OSL were recorded by a solar-blind FEU-142 PMT. The OSL signal
was separated from the stimulation light by an UFS-2 optical filter. Optical stimulation was conducted by means of blue (FYLP-1W-UBB, $h\nu_m = 2.6$ eV) LED. Wire mesh filters and current adjustments were used for both of them to provide a constant power density $P$ of 10 mW/cm$^2$ delivered to the sample. The samples were annealed at $T = 1200$ K for 10 s prior to each TL and OSL curve measurement for controlled filling of the main and deep traps.

Phototransfer TL (PTTL) was measured with the deep and less deep traps in $\alpha$-$\mathrm{Al}_2\mathrm{O}_3$ being filled by high-dose X-ray irradiation ($D = 2$ kGy). Then the shallow traps were emptied by annealing at 620 K. Carrier phototransfer was stimulated with filtered radiation from a 150 W xenon lamp at $h\nu_m = 4.0$ eV and $P = 6.67$ mW/cm$^2$ and an LGI-21 pulsed nitrogen laser at $h\nu_m = 3.68$ eV and $P_{\text{aver}} = 2.55$ mW/cm$^2$. The PTTL curves and residual TL in the 720 and 830 K peaks were measured during one heating cycle.

### 3. Results and discussion

The OSL curves of the $\alpha$-$\mathrm{Al}_2\mathrm{O}_3$ samples with an 830 K peak (a) and without it (b) irradiated with a dose of 0.2, 50 and 200 Gy are shown in figure 1. Whereas at $D = 0.2$ Gy the OSL curves of both sample types are not much different (figure 1a,b, curve 1), an increase in dose results in a substantial difference. In the sample without the peak, the OSL curves do not feature any considerable changes: with an increase in $D$ they demonstrate a growth in initial intensity and an insignificant slowing of the decay kinetics (figure 1b, curves 2 and 3). At the same time, the samples with the peak clearly show a complication of the OSL kinetics at the doses 50 and 200 Gy (figure 1a, curves 2 and 3). Firstly, they display a rise-up stage in the OSL kinetics (insert). Secondly, the fast and slow components of the decay kinetics are clearly distinguishable in the figure, the latter becoming even slower with a growth in $D$.

![Figure 1](image-url)

**Figure 1.** OSL curves of $\alpha$-$\mathrm{Al}_2\mathrm{O}_3$ samples with a 830 K peak (a) and without it (b) measured under stimulation by radiation with $h\nu_m = 2.6$ eV upon X-ray irradiation with doses $D_x$ respectively: 1 – 0.2 Gy, 2 – 50 Gy, 3 – 200 Gy. The insert shows the initial stages of the OSL kinetics in the sample with a peak at a dose of 50 Gy (2) and 200 Gy (3).

To ensure a more detailed quantitative description of the observed regularities, the OSL curves were approximated by exponential dependencies of the type $I_i \exp(-t/\tau_i)$ [5], where $\tau_i$ and $I_i$ are the decay time constant and pre-exponential factor (or initial intensity) of the $i$-component. The decomposition of the OSL curves into elementary exponents for the samples with 830 K peak and without it and estimation of the relative contributions from individual components $S_i$ were performed.
according to [5] using the relationships of the type \( S/S = I/\tau_i/\Sigma I/\tau_i \), where \( S \) is the light sum under the entire OSL curve. The values of \( S/S \) are given in table 1. It is also important to note that such decomposition of the OSL curves for the sample with the peak was carried out without allowing for the rise-up stage since it does not contribute in any way substantially to the total light yield.

**Table 1.** Parameters of the OSL signal decay in the \( \alpha-Al_2O_3:5 \) samples with the 830 K peak and without it as a function of the irradiation dose.

<table>
<thead>
<tr>
<th>Parameters of the OSL curves</th>
<th>( \alpha-Al_2O_3:5 ) with the 830 K peak</th>
<th>( \alpha-Al_2O_3:5 ) without the 830 K peak</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \tau_1 ) (s)</td>
<td>12.5\pm0.8</td>
<td>11.3\pm0.6</td>
</tr>
<tr>
<td>( I_1 ) (a.u.)</td>
<td>229\pm10</td>
<td>17451\pm50</td>
</tr>
<tr>
<td>( \tau_2 ) (s)</td>
<td>-</td>
<td>78.5\pm4.8</td>
</tr>
<tr>
<td>( I_2 ) (a.u.)</td>
<td>-</td>
<td>3081\pm11</td>
</tr>
<tr>
<td>( I_1\tau_1/(I_1\tau_1+I_2\tau_2) )</td>
<td>1</td>
<td>0.45\pm0.01</td>
</tr>
<tr>
<td>( I_2\tau_2/(I_1\tau_1+I_2\tau_2) )</td>
<td>-</td>
<td>0.55\pm0.01</td>
</tr>
</tbody>
</table>

A preliminary analysis of the quantitative data from table 1 shows the validity of the above purely qualitative reasoning based on the experimental results (see figure 1). Moreover, the quantitative values make it possible to accurately estimate the relative contributions of individual components to the total light sum and thus to proceed to describing the physical processes quantitatively.

It follows from table 1 that in the samples of both types the fast component time constant \( \tau_1 \) is approximately the same, being \( \sim10-13 \) s. In the 830 K peak sample irradiated with considerable doses \( (\geq50 \) Gy), the contributions of the fast and slow components do not differ significantly, which is shown in table 1 as the relationships \( S_i/S \) and \( S_i/S \). Nevertheless, we may note a certain tendency towards a reduction in \( S_i/S \) and increase in \( S_d/S \) with an increase in dose. Simultaneously, \( \tau_2 \) increases from 78 to 113 s, i.e. \( \sim1.4 \) times. In the sample without the peak, for \( D=50 \) Gy the relationships \( S_i/S \) and \( S_d/S \) are 0.72 and 0.28, respectively, while for \( D=200 \) Gy they are 0.64 and 0.36. This fact indicates that the contribution of the slow component with \( \tau_2=32 \) s is less substantial even for \( D=200 \) Gy.

By way of summarizing we may assume that the fast component in the samples of both types is due to the emptying of the main trap, which does not contradict the above data from [6], while the slow one, to the emptying of the deeper traps responsible for the TL peaks with \( T_c>500 \) K. Therefore we proceeded to investigating the residual TL in both types of sample after optical stimulation by radiation with \( h\nu_{\alpha}=2.6 \) eV (figure 2).

As can be seen from figure 2a,b (curves 1 and 1’), in the samples of both types irradiated with low doses \( (D\leq1 \) Gy), which corresponds to the emergence of one main peak only, stimulation by radiation with \( h\nu_{\alpha}=2.6 \) eV for 50 s \( (\geq3\tau_1) \) depleted the main trap completely. With an increase in dose, optical stimulation of the sample without the peak for \( 3\tau_2=200 \) s also depleted virtually completely not only the main but also the deeper traps (figure 2b, curves 2 and 2’).

A distinctly different picture is observed in the sample with the TL peak at 830 K (figure 2a, curves 2 and 2’) irradiated with large doses \( (D\geq50 \) Gy). In the residual TL curve measured after stimulation for 300 s, the peaks at 720 and 830 K remained unchanged while the lower temperature and main peaks decreased by no more than 5 times. According to the available data [6], the main trap under these stimulation conditions should get depleted. However, we observed the opposite, which may point to phototransfer of carriers into it from the deeper traps. As follows from a comparison of the residual TL curves for both types of sample (see figure 2a,b, curves 2 and 2’), the most probable source of phototransfer is the trap responsible for the TL peak at 660 K. Evidence of the fact that it was just phototransfer TL that was observed near 450 K is a shift in the TL peak maximum by 10 K
into the high-temperature region, which was found in [6] as well. An important result is that stimulation with $h\nu_m=2.6$ eV did not change the TL yields at 720 and 830 K peaks and, consequently, did not cause carrier phototransfer from the traps that caused them.

Thus, it may be stated that the OSL properties in the samples with the TL peak at 830 K and without it under stimulation with $h\nu_m=2.6$ eV are essentially different. In the samples with the peak irradiated with large doses ($D\geq50$ Gy), the OSL kinetics appear to be more complicated. At the initial stage, we observe a short rise-up stage in the OSL kinetics for 2–3 s followed by the stages of fast and sufficiently slow decay with $\tau_1=11-13$ s and $\tau_2\geq78$ s. The OSL kinetics in the samples without the 830 K peak measured under similar conditions contain predominantly fast decay components with $\tau_1=11-13$ s and are not different from the known ones. This fact is of practical importance for OSL dosimetry since such samples may allow a considerably shortened and simplified procedure of dosimetric information nulling. Moreover, OSL detector samples based on $\alpha$-$\text{Al}_2\text{O}_3$ without the TL peak at 830 K would enable the accuracy of absorbed dose measurements to be improved since they are subject to minimal phototransfer effects.

For separating the processes occurring at the depletion of traps responsible for the TL peaks at 720 and 830 K and for identifying their types we then investigated phototransfer thermoluminescence (PTTL). It was initiated by carrier phototransfer induced by UV irradiation with $h\nu>3.6$ eV [7] from the above-mentioned deep traps into empty shallower ones, including the main trap. The TL curves measured after UV stimulation with various energy densities $W$ applied to the sample are shown in figure 3. It may be seen (curves 1-5) that with an increase in $W$ from 0 to 6.4 J/cm$^2$, the residual TL yields at 720 and 830 K peaks decrease. The PTTL response in the main peak at 460 K increases only at increasing $W$ from 0 to 0.1 J/cm$^2$ (curves 1 and 2). Any further growth in $W$ to 6.4 J/cm$^2$ (curves 3-5) just reduces PTTL in the 460 K peak. At the same time we observe a slower decay of residual TL in the peaks at 720 and 830 K, which will be shown more explicitly below. Also note the shift in the PTTL peak by $\sim10$ K ($T_m=460$ K) into the high-temperature region and its broadening compared with the main TL peak observed near 450 K where the sample was irradiated with a low test dose of $D=0.3$ Gy (curve 6). This result provides evidence that under phototransfer the traps responsible for TL in the high-temperature part of the main peak get filled. According to [8], TL in the region of the main peak is not caused just by the release of holes and electrons in its low- and high-temperature parts respectively, but by the transformation of the peak-
inducing defect. So this feature of the PTTL peak at 460 K and its association with the predominant drop in residual TL at the 720 K peak may point to their common nature.

![Figure 3. PTTL and residual TL curves for an α-Al₂O₃ sample subjected consecutively to X-raying at D=2 kGy, annealing to 620 K and UV stimulation with hνₚ=4 eV at T=300 K for the following values of W: 0 (1), 0.1 J/cm² (2), 1.6 J/cm² (3), 3.2 J/cm² (4) and 6.4 J/cm² (5). For comparison, the figure shows a TL curve (6) for the same sample irradiated with a dose of D=0.3 Gy.](image)

A generalized picture of the pattern of changes in the PTTL peak at 460 K and residual TL in the peaks at 720 and 830 K depending on W is shown in figure 4a (curves 1–3, respectively). The light sums (S) achieved in the above peaks are normalized to their corresponding maximal values (Sₘₐₓ) for this series of experiments. Figure 4b compares changes in PTTL in the 460 K peak with derivatives of the functions S(W) with respect to W for residual TL in the 720 and 830 K peaks (curves 1–3, respectively). As is shown below, such derivatives are directly proportional to the speed of carrier phototransfer (dS/dt) from deep traps to less deep ones:

\[
\frac{d(S / S_{\text{max}})}{dW} = \frac{d(S / S_{\text{max}})}{d(P \cdot t)} = \frac{P \cdot S_{\text{max}} = \text{const}}{S_{\text{max}} \cdot P \cdot dt}, \quad (1)
\]

It is important that if an experiment had P=const, then Sₘₐₓ would also be a constant (Sₘₐₓ=const).

Such representation of data enabled us to compare relative changes in the PTTL response in the 460 K peak, residual TL in the 720 and 830 K peaks and their derivatives and suggests the following logical chain. From figure 4a it can be seen that the behavior of the functions S/W(Sₘₐₓ) with respect to the responses under consideration is clearly correlated. The maximum drop in the TL yields in the 720 and 830 K peaks at 0<W≤0.1 J/cm² has a corresponding extreme growth in the PTTL yield in the peak at 460 K. Comparison of this result (see figure 4b, curve 1) with the speed of carrier phototransfer from deep traps (ibid., curves 2 and 3 and the insert) suggests a preliminary conclusion that carrier phototransfer to the main trap occurs predominantly from one of them, namely from the trap responsible for the 720 K peak. This suggestion is also based on the fact that for 0<W≤0.1 J/cm² the speed of its depletion is ~10 times higher than one for the trap that gives rise to the TL peak at 830 K. It is also important that the speed of phototransfer under consideration is maximal at low values of W, and it decreases as W is increased.

It is no less important that UV stimulation with hνₚ=4 eV results not only in the depletion of the deep traps and filling of the main trap but also in the depletion of the latter. Moreover, the high probability of carrier release by light with hνₚ=4 eV from the main trap correlates well with its depletion spectrum [9]. Thus the stages of increase and decrease in the PTTL response in the peak at 460 K may be due to the competing processes of carrier phototransfer to the main trap and its
depletion. As W increases, the number of carriers phototransferred to the main trap decreases since the number of carriers released per unit time from the deep trap responsible for the TL peak at 720 K drops. Hence, as W increases, the probability of the main trap getting depleted will be increasing in relation to the probability of phototransfer while the PTTL signal will feature stages of growth and subsequent reduction.

Figure 4. (a) Changes in normalized light sums in the PTTL peak at 450 K (1) and residual TL in the peaks at 720 (2) and 830 K (3) as a function of W at $h\nu_m=4$ eV and $P=6.67$ mW/cm$^2$ (see figure 3); (b) Changes in normalized light sums in the PTTL peak at 450 K (1) and normalized speeds of residual TL decay in the peaks at 720 K (2) and 830 K (3) as a function of W. The insert shows part of the functions for $0<W\leq0.75$ J/cm$^2$.

Thus, the above data suggest that UV stimulation with $h\nu_m=4$ eV depletes simultaneously two deep traps (or converts defects) responsible for the TL peaks at 720 and 830 K. This approach renders the interpretation of the results more difficult. Therefore for selective depletion of one of them, namely a less deep one, we may probably use softer UV irradiation with $h\nu_m<4$ eV, for example from a laser with $h\nu_m=3.68$ eV.

The residual TL curves in the region of 630-950 K and PTTL in the region of 300-630 K as measured for $h\nu_m=3.68$ eV, $P_{av}=2.55$ mW/cm$^2$ and changes in W over the range of 0 to 4 J/cm$^2$ are shown in figure 5a (curves 1-4). As follows from it, as W grows from 0 to 4 J/cm$^2$, the residual TL yield in the peak at 830 K virtually does not change, falling in the peak at 720 K. At the same time the PTTL response is observed in the main peak at 460 K, which increases as W grows from 0 to 0.5 J/cm$^2$ and decreases with further increase in W to 4 J/cm$^2$. Figure 5b summarizes data on normalized PTTL light yields in the 460 K peak (curve 1) and residual TL in the 720 and 830 K peaks as a function of W (curves 2 and 3, respectively). Their analysis and comparison with the data obtained for UV-irradiation with $h\nu_m=4.0$ eV and those shown in figure 4b (curves 1–3) suggest that the PTTL response in the 460 K peak is caused predominantly by carrier phototransfer from the trap giving rise to the TL peak at 720 K. The second option, which cannot be ruled out either, is the photoconversion of the defect responsible for the TL peak at 720 K into one the thermal activation of which gives rise to the PTTL peak at 460 K. It should also be stated that the photodepletion of the trap responsible for the TL peak at 830 K does not cause any clearly detectable (noticeable) transfer of carriers to the main or other traps having thermal activation parameters which are close to those of the main trap (or defect photoconversion is not genetically associated with the defect giving rise to the main TL peak).
4. Conclusion

A combined investigation of OSL, phototransfer and residual TL in strongly irradiated $\alpha$-Al$_2$O$_3$-$\delta$ crystals has established that the fast component of OSL decay under continual stimulation by blue LED with $h\nu_m=2.6$ eV is associated with a depletion of the main trap. The slow component of OSL decay is recorded at $D \geq 50$ Gy, i.e. when the main and deeper traps are filled. It is caused by simultaneous release of carriers from the traps responsible for the TL peaks in the region of 500–680 K. It has also been shown that whereas photodepletion of a less deep trap (pre-peak at 720 K) causes its carriers to fill the main trap, in the case of the deep trap (830 K peak) no such phototransfer is observed.

References