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Optical detrapping of charge from the 110°C quartz TL region

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Abstract : *A series of experiments has shown that the mechanism responsible for the enhanced rate of decay under illumination of PTTL (photo-transferred thermoluminescence) in the ~110°C region is most likely to be optical detrapping of charge. It has been demonstrated that an alternative explanation for the decay, related to changes in acceptor site concentrations, is unlikely to be the cause. An optical half-life of ~600s is deduced for the ~110°C PTTL emission for the conditions described (420-560nm, 12 mW.cm⁻² stimulation).*

Introduction

The aim of this work was to investigate the optical stability of electrons held in the quartz TL peak found at ~110°C. More specifically, the main concern was the issue of whether or not the decrease in PTTL as a function of OSL exposure time is due to optical bleaching (of the 110°C PTTL) or to a change in the concentration of available acceptor sites (such as the loss of luminescence centres or an increase in competition from non-radiative centres or electron traps) during optical exposure.

Basis of experiments

Following an exposure to the OSL stimulation light of 75s (at 20°C, stimulating with 2.2-2.9eV photons (420-560 nm) at ~12mW.cm⁻², within a standard automated TL-DA-12 Risø reader) the measurable OSL signal is generally depleted to a level of ≤1% of the initial intensity (I_0). During such an OSL shinedown a proportion of the mobile charge is transferred to the TL trap responsible for the ~110°C PTTL emission (Aitken and Smith, 1988). As the OSL signal has been reduced to such a low level, any further light exposure after this point (75s) will add only a minute fraction to the PTTL already present. If following 75s OSL the excitation light is turned off, the thermal decay of the PTTL (at 20°C) can be measured by making TL measurements of individual aliquots following time t . A plot of PTTL versus t both with and without illumination will show the effect of the light exposure on the PTTL.

Sensitivity change as a function of optical exposure time

It is necessary to quantify the degree to which the apparent loss of PTTL upon optical exposure (after

the initial 75s shinedown) is due to changes in the concentration of available acceptor sites. An attempt to indirectly measure such effects, as a function of OSL measurement time, was made using the ~110°C TL response. Individual aliquots of natural Chaperon Rouge (724g2) quartz (a sample chosen for its relatively strong TL / PTTL peaks at ~110°C), having a natural dose of ~12 Gy, were given ~0.1 Gy of beta dose and then immediately heated to 160°C (2°C.s⁻¹) to measure the TL at ~110°C (this procedure will be termed the *standard sensitivity test*). The aliquots were then exposed to the OSL excitation light for a range of times at 160°C. Following the OSL shine, the standard sensitivity test was performed again, the rationale being that during OSL shinedown a proportion of the de-trapped electrons recombine at the available L-centres (luminescence centres), hence the OSL, and the OSL traps are substantially depleted. Therefore, following the OSL measurement, the probability of any free electron moving around the lattice recombining at an L-centre is lessened, (both because of the reduced concentration of available L-centres and the increased competition for charge from the empty OSL traps), which should be expressed as a loss of sensitivity. This assumes that electrons released from the OSL and 110°C TL/PTTL traps recombine at the same L-centres - a view supported by the spectral emission work of Franklin *et al.* (1995).

The results (shown in Figure 1) are interpreted to mean that the loss of luminescence centres following OSL shinedown (to < 1% of I_0) is only a small fraction of the total, in the order of 9%. This is consistent with the findings of Bailey *et al.* (1997) who report an effectively constant charge transfer

rate throughout the OSL shine-down. There is a slight dose dependency in the results, showing less of a drop in sensitivity for aliquots dosed (see Figure 1 for details of administered doses) and preheated (220°C for 300s) prior to measurement. This can be tentatively interpreted in a number of ways (including thermal sensitisation via the mechanism described first by Zimmerman (Zimmerman, 1971) or there being an increased likelihood of trapping holes in the appropriate L-centres during dosing rather than trapping electrons at the OSL source traps). More work however is needed here and this is a relatively subsidiary point.

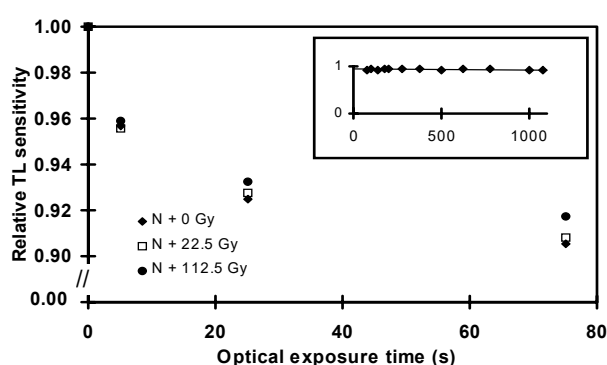


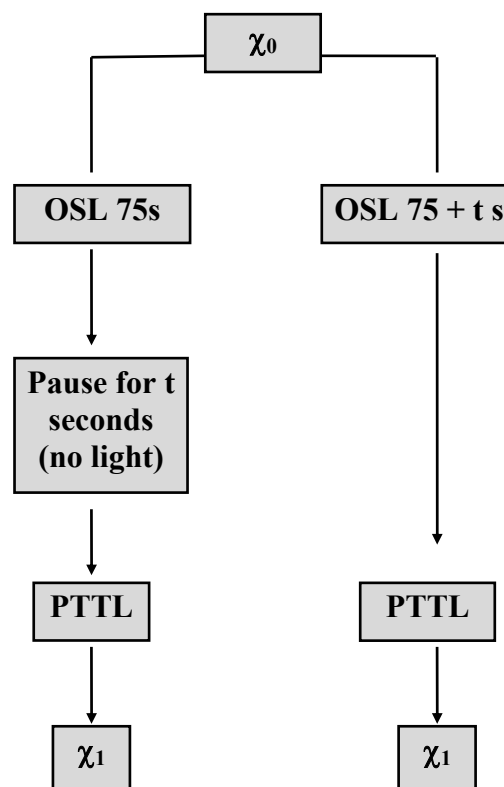
Figure 1: Relative sensitivity of the $\sim 110^\circ\text{C}$ TL emission as a function of OSL measurement time. Individual aliquots (of Chaperon Rouge quartz, 724g2) were β -irradiated with the doses shown and preheated (220°C for 300s). A standard sensitivity test was then made (see main text for description). The individual aliquots were then exposed to the OSL excitation light (as described in main text) whilst held at 160°C. Following light exposure a further standard sensitivity test was administered. The relative sensitivity is defined as the ratio of the two sensitivity measurements. Each data point is the mean of four individual aliquots. The standard errors are smaller than the size of the symbols. The inset box shows similar data for a natural sample of the same quartz with the optical exposure times extended to 1075s. Again each point is the average of at least two similar aliquots. A linear fit to the data is shown, which confirms the constancy of the relative sensitivity between 75 and 1075s of optical exposure (the gradient of the fit is less than 1 in 10^5).

The results from the present work show the same ca.9% drop in sensitivity following a shinedown to <1% for the same sample illuminated this time at

20°C. As expected the drop in sensitivity as a function of optical exposure appears to be proportional to the measured OSL signal. Consequently the change in sensitivity for OSL exposure times >75s (at either 20°C or 160°C), where the OSL signal has dropped to very low levels, are imperceptible, with no change in sensitivity seen for OSL exposure times between 75 and 1000s. Effectively all of the loss in sensitivity (as a function of optical exposure) has been completed by the 75s measurement time (i.e. where $t = 0$).

Procedure

A diagram for the experimental procedure is shown below. The same sample (natural Chaperon Rouge, 724g2) used for the experiment previously described was also used here. The characters χ_0 and χ_1 are the standard sensitivity tests, performed at the beginning and end of the procedure respectively. The TL measurements (of both PTTL and dose induced TL signals) were made at 2°C.s^{-1} , heating to 160°C. During illumination with the OSL excitation light, and for the subsequent hold for t seconds (where applicable), the samples were maintained at room temperature ($\sim 20^\circ\text{C}$).



Discussion of results

Figure 2 shows the integrated PTTL (20-150°C integral) remaining as a function of t , for both conditions (i.e. light on during t and light off during t). Each point represents the mean of at least two individual measurements, with the standard deviation shown. The data were normalised using the initial standard sensitivity measurement.

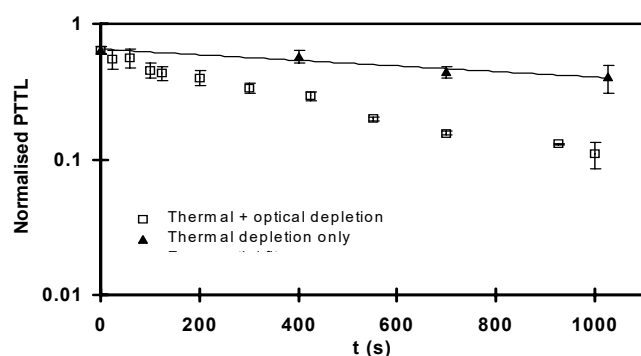


Figure 2: Comparison of thermal and combined thermal and optical decay of PTTL (see main text for details). Each data point is due to at least two replicate measurements. The thermal decay data has been fitted to a single exponential decay.

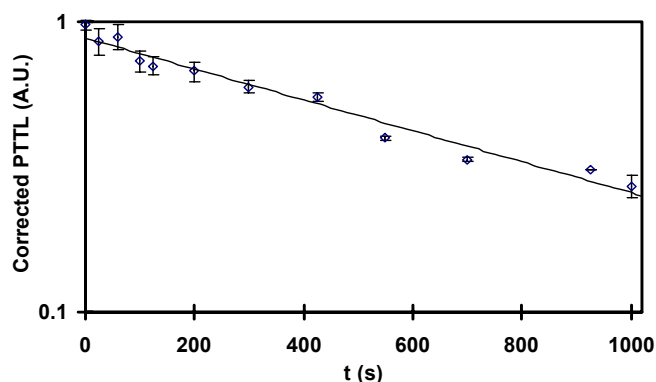


Figure 3: Optical decay of PTTL, corrected for thermal decay at 20°C and fitted to a single exponential decay. Data are normalised to the initial sensitivity value and are due to at least two replicate measurements (see main text for details).

There is a clear difference between the two sets of data shown in Figure 2. The increased rate of signal decay in samples exposed to the OSL excitation light during the hold for t seconds is easily apparent. As shown in the previous section, the sensitivity of the ~110°C TL peak is not subsequently affected by light

exposure past 75s and hence the subsequent changes in the availability of electron acceptors after this time are taken to be minimal. The drop in PTTL signal level as a function of illumination time therefore strongly suggests that optical bleaching of the PTTL is taking place.

An exponential fit to the thermal depletion data (see Figure 2) can be used to correct for the thermal loss of the PTTL signal during illumination, yielding the data shown in Figure 3. These data represent the true optical depletion of the ~110°C PTTL signal. The data cannot be particularly well fitted with a single exponential decay and there are a number of possible reasons for this. Smith and Rhodes (1994) have shown that the TL emission at ~110°C (for the same sample as that used here) comprises two components, with TL peak positions possibly corresponding to the 85°C and 110°C TL emissions (Smith *pers.comm.*). Other kinetic effects such as charge re-trapping (both to the traps responsible for the ~110°C PTTL and the OSL emissions) may also contribute to the overall form of optically induced decay. However if the data in Figure 3 are fitted to an exponential decay an optical half life for the ~110°C PTTL peak of ~600s is obtained (for the conditions described above). Wintle and Murray (1996) have also concluded that optical stimulation of electrons from the 110°C TL trap gave rise to a decrease in the PTTL signal under green light stimulation. They obtained a shorter optical decay lifetime of ~130s for the first 90% loss of signal decay for their sample of Australian quartz.

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Reviewer
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