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# Time-resolved luminescence from quartz: An overview of contemporary developments and applications

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#### ABSTRACT

Time-resolved optical stimulation of luminescence has become established as a key method for measurement of optically stimulated luminescence from quartz, feldspar and  $\alpha$ -Al<sub>2</sub>O<sub>3</sub>:C, all materials of interest in dosimetry. The aim of time-resolved optical stimulation is to separate in time the stimulation and emission of luminescence. The luminescence is stimulated from a sample using a brief light pulse and the emission monitored during stimulation in the presence of scattered stimulating light or after pulsing, over photomultiplier noise only. Although the use of the method in retrospective dosimetry has been somewhat limited, the technique has been successfully applied to study mechanisms in the processes leading up to luminescence emission. The main means for this has been the temperature dependence of the luminescence intensity as well as the luminescence lifetimes determined from timeresolved luminescence spectra. In this paper we review some key developments in theory and applications to quartz including methods of evaluating lifetimes, techniques of evaluating kinetic parameters using both the dependence of luminescence intensity and lifetime on measurement temperature, and of lifetimes on annealing temperature. We then provide an overview of some notable applications such as separation of quartz signals from a quartz-feldspar admixture and the utility of the dynamic throughput, a measure of luminescence measured as a function of the pulse width. The paper concludes with some suggestions of areas where further exploration would advance understanding of dynamics of luminescence in quartz and help address some outstanding problems in its application.

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#### 1. Introduction

Time-resolved optical stimulation (TR-OSL) is a key method for measurement of optically stimulated luminescence that relies on the use of repetitive light pulses of constant intensity to separate in time the stimulation and emission of luminescence. The luminescence is stimulated using a brief light pulse during which the measured signal comprises a monotonically increasing luminescence component and constant scattered stimulating light. After the pulse, the luminescence is measured over a background of photomultiplier noise only and decreases in intensity with time.

There are several advantages of time-resolved optical stimulation over steady-state stimulation. The method is capable of high signal-to-noise ratio over extended measurement times [1]. In addition, pulsing rather than continuous optical stimulation means that only a negligible proportion of the optically stimulable

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charge is sampled thereby enabling repeated monitoring of the luminescence without an appreciable loss in intensity. Time-resolved optical stimulation provides a means to measure luminescence lifetimes. The lifetimes are associated with charge transfer processes within the phosphor and as such help to better explain the luminescence emission process.

The first measurements of time-resolved luminescence from materials of interest in retrospective dosimetry were first reported by Sanderson and Clark [2] on feldspar stimulated with 470 nm light using a 10 ns pulse. The study showed evidence of luminescence lifetimes of the order of hundreds of nanoseconds. The method was then used to investigate luminescence processes from  $\alpha$ -Al<sub>2</sub>O<sub>3</sub>:C, a luminescence dosimeter [3–6]. Further studies on pulsed luminescence from feldspars were then reported by Clark et al. [7] and Clark and Bailiff [8] using an 850 nm pulsed laser. The first measurements of TR-OSL from quartz, a common natural dosimeter, independently reported by Bailiff [9] using a 470 nm pulsed laser system and by Chithambo and Galloway [1] using a pulsed 525 nm green LED system, showed a principal luminescence lifetime of about 40 µs from this material. Later work







showed the presence of shorter subsidiary lifetimes in quartz depending on certain combinations of annealing and measurement temperature [9–12] as well as on the thermal provenance of the quartz [13]. The method has also been widely used to study dynamics of luminescence in quartz e.g. [9–15], to develop a kinetic model of pulsed OSL in quartz [16] and applied in retrospective dosimetry [17, 18].

The aim of this paper, which is not meant to be exhaustive, is to present a review of some contemporary developments in the theory and applications of time-resolved optical stimulation of luminescence in quartz. The paper looks at instrumentation, discusses luminescence lifetimes and luminescence intensity and their use in evaluating kinetic parameters, presents an overview of models for time-resolved luminescence, summarises some notable applications of the method and concludes with an outlook of areas where further work would advance understanding of mechanisms of luminescence in quartz and help refine its applications.

#### 2. Measurement techniques

There are several methods for measurement of time-resolved or pulsed optically stimulated luminescence (POSL) that have been reported but in this section we reflect only on two, time-correlated photon counting and the other, a time-tag technique.

#### 2.1. Time-correlated photon counting

The stimulation of luminescence in this method may be discussed with reference to the schematic diagram of Fig. 1 which shows a light pulse of duration  $t_w$ . Luminescence is emitted both during and after the light pulse. Since the luminescence photons emitted during and after optical stimulation using a single light pulse are not sufficient for reliable statistical analysis, it is often necessary to build up a time-resolved luminescence spectrum by summing spectra from several scans or sweeps. In this case, the total measurement time known as the on-time  $t_{ON}$  is the product of the pulse width (i.e. the duration of the light pulse) and the number of sweeps. The data points displayed as a function of time in the time-resolved luminescence spectrum measured in this manner are not separated in time contiguously as in a conventional OSL decay curve. The data points are arranged depending on their correlation in time with the luminescence stimulating light pulse. The correlation in time between the light pulse and the resultant luminescence photon is determined by a multichannel scaler e.g. see Refs. [19, 20]. The multichannel scaler records the number of events (detection of luminescence photons) that occur during the time interval  $t_i$  to  $t_i + \Delta t$  as a function of time where  $\Delta t$ 



**Fig. 1.** A schematic diagram of a luminescence stimulating light pulse of duration  $t_w$  and period *P*. The dynamic range *t* is subdivided into an integral number of  $n\Delta t$  channels [20].



**Fig. 2.** A schematic diagram of the pulsing system showing the arrangement for detection and measurement of time-resolved luminescence spectra [20].

is the dwell time (Fig. 1). The measurement time per sweep or dynamic range t which need not be equal to the period P is quantized into an integral number of channels each of duration  $\Delta t$  by the relation  $t=n\Delta t$  where n is the channel or bin number.

Fig. 2 shows a schematic diagram of the luminescence detection assembly for a typical pulsing system [19]. The luminescence is detected by the photomultiplier tube (EMI 9635QA) whose signals are amplified by the timing filter amplifier (Ortec 474) and then counted by the constant fraction discriminator (Ortec 584). The multichannel scaler (EG & G Ortec MCS-plus<sup>™</sup>) triggers a set of light-emitting diodes as well as initiates a data-recording sweep in the computer. Once the sweep is started the multichannel scaler records photon counts sequentially in its memory with no deadtime either between channels or at the end of each sweep. The time-resolved luminescence spectrum measured in this way is a plot of cumulative photon counts against time for the dynamic range selected. An example of a time-resolved luminescence spectrum is shown in Fig. 3. The spectrum was measured from a



**Fig. 3.** Time-resolved luminescence from sedimentary quartz measured using a 60  $\mu$ s pulse over a dynamic range of 300  $\mu$ s and 10<sup>6</sup> sweeps following a beta dose of 7 Gy [21]. The parameters  $\tau_a$  and  $\tau_d$  are discussed later in the text.

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