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Quantitative analysis of time-resolved infrared stimulated luminescence in feldspars

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ABSTRACT

Time-resolved infrared-stimulated luminescence (TR-IRSL) from feldspar samples is of importance in the field of luminescence dating, since it provides information on the luminescence mechanism in these materials. In this paper we present new analytical equations which can be used to analyze TR-IRSL signals, both during and after short infrared stimulation pulses. The equations are developed using a recently proposed kinetic model, which describes localized electronic recombination via tunneling between trapped electrons and recombination centers in luminescent materials. Recombination is assumed to take place from the excited state of the trapped electron to the nearest-neighbor center within a random distribution of luminescence recombination centers. Different possibilities are examined within the model, depending on the relative importance of electron de-excitation and recombination. The equations are applied to experimental TR-IRSL data of natural feldspars, and good agreement is found between experimental and modeling results.

1. Introduction

When an insulating material is exposed to ionizing radiation, some of the free electrons resulting from the interaction will end up being trapped at defects in the crystal. The concentration of such trapped charges is a function of the radiation dose, and this relationship form the basis of trapped-charge dating methods. When the crystal is subsequently exposed to optical excitation, some of the trapped electrons can be released from the traps and recombine, radiatively, with holes trapped at appropriate centers to produce optically stimulated luminescence, which is widely used in sediment dating. In this paper we are concerned with feldspars, for which the excitation spectra of the optically stimulated luminescence have a prominent peak at ~1.44 eV [21]. The defects related to this resonance also exhibit tunneling effects associated with a variable tunneling distance. For a summary of previous work on feldspars the reader is referred to the books by Chen and Pagonis [7] and Bøtter-Jensen et al. [6].

During the past decade numerous experimental and modeling studies have established that quantum mechanical tunneling is the dominant mechanism for production of luminescence signals in feldspars [12–15,20,23–25,3,35,36,45–47,6]. While many of these studies are concerned with tunneling taking place from the ground state of the trap, research has also supported the existence of tunneling processes taking place from the excited state of the trap, as well as in combination with charge migration through the conduction band-tail states [1,18,2,21,29,42,43].

Jain et al. [17] developed a model in which localized electronic recombination of pairs of trapped electrons and recombination centers takes place via the excited state of the trapped electron, to the nearest center within a random distribution of them. They successfully simulated both thermally stimulated luminescence (TL) and optically/infrared stimulated luminescence (OSL, IRSL), and found that a power law behavior can occur for OSL. This model was examined by Kitis and Pagonis [19], to obtain analytical solutions for four different experimental modes of stimulation: TL, OSL, linearly modulated OSL (LM-OSL) and isothermal TL. These analytical solutions have been used in several recent experimental studies to fit luminescence from a variety of feldspars and apatites [28,31,32,34,39,40].

Time-resolved luminescence data provide a method of distinguishing between the different recombination routes in a variety of materials. Time-resolved optically stimulated and infrared

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stimulated luminescence (TR-OSL and TR-IRSL) from feldspars have been the subject of several recent experimental studies. Jain and Ankjærgaard [18] compared time resolved luminescence from feldspars using mainly IR, green and blue stimulations and examined the changes occurring as a function of photon energy, storage time after irradiation, and thermal or optical pre-treatments of the samples. These experimental results were interpreted within the framework of a model containing several possible pathways for charge movement, namely from the dosimetric trap via the ground and excited states of the trap, via the band tail states and through the conduction band. Jain and Ankjærgaard [18] concluded that the initial "fast signal" in their TR-IRSL data is most likely due to localized transitions, while the "slow signal" in their data originated in transitions through the band tail state.

Several experimental studies have identified five ranges of lifetimes in TR-IRSL signals, namely 30–50 ns, 300–500 ns, 1–2 μs, ~5 μs, and > 10 μs [10,11,38,44,8,9]. Some of these lifetimes were interpreted as due to internal transitions within the recombination centers [8]. In several of these earlier papers TR-IRSL curves were fitted using a sum of exponentials. Morthekai et al. [26] studied TR-IRSL from four feldspar mineral specimens, by assuming that de-trapped electrons undergo random walk in the band-tail states before recombining by tunneling. The hopping time for the random walk was derived from the OFF-time data of TR-IRSL experiments, and the extracted parameters were shown to be consistent with the variable range hopping mechanism of the Mott kind. Pagonis et al. [30] also analyzed TR-IRSL data from the same four natural feldspars samples, in terms of the sum of an exponential and a stretched exponential function.

The technology of measuring such time-resolved signals has been greatly improved during the past 10 years, and modeling advancements have contributed to a better understanding of the luminescence process. These recent developments in both experiments and models prompted us to re-examine the topic of the shape and mathematical characterization of such signals.

In this paper we use the model of Jain et al. [17] to describe the shape of TR-IRSL signals during and following short infrared pulses.

The goals of the present paper are:

(1) To investigate whether the model of Jain et al. [17] can be used to describe experimental TR-IRSL data from feldspars, in the microsecond time scale.
(2) To fit experimental TR-IRSL data from feldspars, both during and after short excitation pulses.
(3) To examine different outcomes from the model, depending on the values of the various kinetic parameters.

2. Samples and experimental setup

The TR-IRSL signals from two feldspar samples are analyzed in this paper. The first set of TR-IRSL data was previously published by Morthekai et al. [26] on sample FL1, a museum specimen of feldspar. X-ray diffraction analysis indicated that FL1 is a microcline also containing diopside and albite. The ratio of K:Na:Ca was calculated and these values place FL1 in the alkali feldspar series. The samples were crushed gently using an agate mortar and sieved to obtain the 90–150 mm size fraction, which was used without any further chemical treatment. The second sample is the potassium rich feldspar fraction (K-$AlSi_3O_8$) of a glacio-fluvial sediment from Jameson land, East Greenland with a grain size 106–180 μm (lab. code 951002FK). A few milligrams of the samples were mounted on stainless steel disks using Silkospray silicone oil.

Measurements were carried out on a Risø TL/OSL-20 reader equipped with an integrated pulsing option to control the IR LEDs, and a Photon Timer attachment to record the TR-IRSL [22]. The IR stimulation was performed with an 870 nm (FWHM 40 nm) LED array delivering 100 mW/cm^2. The stimulated luminescence emission was detected using a photomultiplier tube (EMI 9235QB; 30% QE at 395 nm), and the time-resolved signals for sample FL1 were detected using a combination of BG-39 (2 mm) and Corning 7–59 (4 mm) filters (transmitting photons at 395±50 nm). The TR-IRSL signals for the potassium rich feldspar fraction were detected in the UV emission (U340 filter, 7.5 mm).

For sample FL1 the TR-IRSL signals were measured for a stimulation temperature of 50 °C, after the sample was given a dose of 61.8 Gy, and preheated to 280 °C for 60 s, using an ON–time of 50 μs and an OFF-time 100 μs. For sample 951002FK the TR-IRSL curves were measured at a stimulation temperature of 50 °C, after the sample was given a dose of 70 Gy, and then preheated to 250 °C for 60 s, using an ON-time of 500 μs, and an OFF-time of 300 μs. The natural luminescence signals were erased by exposing the samples to IR at an elevated temperature of 250 °C, followed by heating the sample from room temperature up to 500 °C. For sample FL1, the total stimulation time was 100 s and hence 95 thousand pulses with a pulse period of 1050 μs were used. For sample 951002FK the total stimulation time was 10 s.

An important experimental consideration when fitting the TR-IRSL data is the switching-off speed (or the fall-time) of the LEDs. The diode intensity after the end of the excitation period reduces very quickly to ~5% of the initial intensity, but it takes an additional ~3 μs for the IR diodes to reach a light level of less than 1% of the stimulation light level during the pulse. Therefore, for the analysis of the experimental data in this paper, all data points for $t < 3$ μs are not used in the fitting process, since they may represent a mixture of the luminescence signal from the sample and the signal during the LED switching-off.

3. Analysis of time-resolved experiments using the model by Jain et al. [17]

In this section we use the model and notation of Jain et al. [17,16]. The model assumes a random distribution of trapped electrons and a random distribution of nearest-neighbor recombination centers, and that emission of luminescence is a result of tunneling recombination of a trapped electron from the excited state of the trap, excited by optical stimulation. Fig. 1 illustrates the different transitions considered within this model: Optical

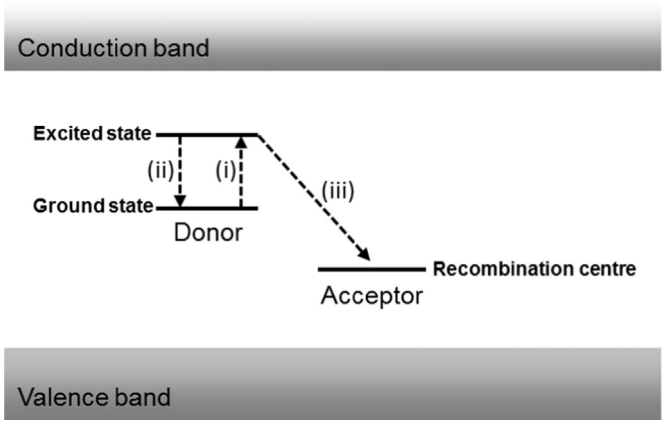

Fig. 1. Simplified band diagram showing the feldspar luminescence transitions considered in the model and discussed in the text: Transition (i) denotes the optical transition between the ground state of the trapped electron to the excited state, and transition (ii) is de-excitation back into the ground state. Transition (iii) is direct tunneling from the excited state of the trapped electron to the recombination center. The figure is adapted after Jain and Ankjærgaard [18] and Jain et al. [17,16].