## ORIGINAL PAPER

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# Electrons in feldspar I: on the wavefunction of electrons trapped at simple lattice defects

**Abstract** The purpose of this article is to make an initial consideration of the physical properties of electrons trapped at classic hydrogenic lattice defects in feldspar. We are particularly interested to determine the radial extent of the electron wavefunctions in the ground and excited states. It is shown that for $NaAlSi_3O_8$, the ground-state wavefunction is expected to be confined well within a single lattice unit cell, but the first excited state is far more extensive, being spread over several unit cells. This aspect is of direct relevance to understanding the nature of various luminescence processes in the materials. Under low-energy optical stimulation ($hv \sim 1.4$ eV), luminescence can be a competitive process between direct electron-hole tunnelling recombination (with the charge still trapped at the defect sites), and free-to-bound recombination (after the excited state electron accesses the conduction band). We show that analysis of the thermal behaviour of the luminescence can be used to separate the two processes.

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## Foreword

Despite being the most ubiquitous group of minerals in the Earth's crust, very little is known about the fundamental electronic properties of the feldspars. For example, the simplest parameters, such as the optical band-gap energies ($E_g$), have never been determined, although from the luminescence excitation results of Jaek et al. (1997a, b) we do know that $E_g$ must be greater than 7 eV for alkali feldspars. The situation is in stark contrast to the cases of associated minerals such as quartz and zeolitic sodalites, where there is considerable detailed theoretical and empirical information available (e.g. Xu and Ching 1991; Demkov and Sankey 1997; Sankey et al. 1998). These studies have mainly arisen because of the technological importance of the materials. Obtaining knowledge about how electrons behave in the feldspars is, however, not entirely academic, since these materials are widely used in luminescence dating of sediments (e.g. Aitken 1998). Considering that there are major problems associated with feldspar dating (e.g. Huntley and Lamoth 2001; Wallinga et al. 2001), trying to understand the physical properties of electrons in more detail is a worthwhile endeavour, since the luminescence involved is dependent on both the electron transport properties, and how the electrons are spread in the lattice.

In this respect, a partial breakthrough was recently made by Poolton et al. (2001) who were able, for the first time, to directly measure the effective electron mass $m_e^*$ in $NaAlSi_3O_8$ feldspar using cyclotron resonance methods. They found that the mass was basically isotropic, and had a value of $m_e^* = 0.79m_e$. This measurement is

highly significant on two counts. Firstly, the isotropic nature of the mass is strongly indicative that, unlike quartz, the electronic band gap is direct (i.e. no phonons are required per se for electronic transitions within or across the gap, making luminescence excitation and emission processes very efficient). Secondly, it means that it is possible to start building theoretical models of electronic processes in feldspar, many of which require knowledge of the value $m_e^*$.

The aim of this article, and the associated paper that directly follows (Electrons in feldspar II: band-tail states and their effects on luminescence processes) is to start to explore some of the electronic properties of feldspar, with the help of these new insights. Whilst Part I is concerned with a quite specific problem (that of electrons trapped at a particular defect that is used in dating studies), Part II begins to explore, in a more general way, the nature of the electronic conduction band. We wish to emphasise that the main source of published empirical data that can be called upon to compare with the ideas comes from the luminescence properties of feldspar. Insights provided by the work are therefore tested in the luminescence arena.

## Introduction

### Luminescence processes and trapped charge in feldspar

Many impurities or native lattice defects commonly present in natural feldspars result in the appearance of isolated electronic levels within the band gap, located between the conduction and valence bands. Depending on their nature, these can trap electrons or holes (generated by, for example, exposure to $\beta$-particles, X- or $\gamma$-rays). The two hole-trapping centres that have been most widely investigated and understood are $Fe^{3+}$ and $Mn^{2+}$ (Manning 1970; Telfer and Walker 1978; White et al. 1986). If the defects are deep enough, they can trap and store the charge for considerable periods of time (many thousands of years), making them useful for dating or radiation dose applications (e.g. Aitken 1998).

Although there are many ways in which defects can participate in luminescence emission, the processes of particular interest to this work are where trapped electrons can be stimulated to recombine with trapped holes. In dosimetry and dating, this provides a simple way of assessing the relative amount of trapped charge in the material. Simply put, the brighter the luminescence emitted during stimulation, the greater the amount of trapped charge (and hence, the older the material, or the greater the radiation exposure).

Consider the specific case where such luminescence arises from optical stimulation (OSL: optically stimulated luminescence). Hütt et al. (1988) found that in alkali feldspars, OSL is dominated by electron transfer from just a single (unidentified) defect type, possessing a distinctive resonant transition in the near infrared (IR)

at around 1.4 eV. The resonance was thought to arise from the ground-to-excited state transition of electrons trapped at the centre and, once in the excited state, electrons could escape (with thermal assistance) to recombine with holes trapped elsewhere in the material. Such recombination yields anti-Stokes shifted luminescence, which decays with time as the quantity of trapped charge becomes depleted. The proposed model for this type of emission is shown schematically in Fig. 1a. More recently, luminescence *emission* arising from relaxation between the excited and ground states has been monitored during irradiation, and the dominance of this particular defect has been confirmed (e.g. Krbetschek et al. 2000; Trautmann et al. 2000). The physical nature of the defect has, however, not yet been identified.

### Why we need to know about the extent of the trapped-charge wavefunctions

Whilst the above description gives an indication of the general properties of infrared stimulated luminescence (IRSL), there are many anomalies which are poorly understood. For example, from the original work of Hütt et al. (1988) onwards, it has been known that the IR transition is thermally assisted. However, the amount of thermal assistance is strongly dependent upon the optical excitation energy chosen and, in some instances, is much lower than expected. Poolton et al. (1995a) tentatively suggested that this may be due to the occurrence of tunnelling from the excited state which would, in principle, require no thermal assistance. Here, the thermal release to the conduction band would be short-circuited, and the electrons and holes would directly recombine whilst still trapped at their respective sites: the process is shown schematically in Fig. 1b.

In order to compare the probability of tunnelling in the ground and excited states, it is necessary to compare the extent of the electron wavefunctions in the two states.
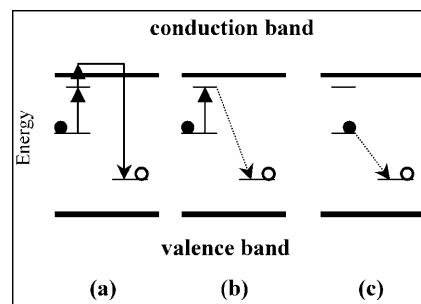


**Fig. 1** Schematic of the luminescence processes considered, involving an electron trap (with a well-defined excited state) and a hole trap. *a* Low-energy optical excitation (~1.4 eV) raises the electron to the excited state; thermal energy available from the lattice allows ionization, followed by recombination elsewhere in the lattice; *b* if the defects are closer, direct (tunnelling) recombination is possible from the excited state, because the extended electron wavefunction overlaps the hole; *c* for very close pairs, even the compact ground-state wavefunctions overlap, allowing direct tunnelling recombination

In turn, this requires a basic working electronic model of the defect in question, so that we can apply simple quantum mechanical treatments to solve the problem.

## A simple electronic model for the IRSL electron trap in feldspar

The physical nature of the dominant OSL electron trap in feldspar is unknown: building a detailed crystal-field model for the centre is thus not possible (as has been done, for example, in the case of the recombination centre $Mn^{2+}$, e.g. Telfer and Walker 1978). Nevertheless, progress can be made. Consider the simplest possible electron-trapping defect in a solid. This would be one in which a single electron is trapped by a central, single positive charge at the defect's core: such a defect is analogous to the simple hydrogen atom. The electron would move in a potential of the form

$$V(r) = -\frac{e^2}{(4\pi\varepsilon_0\varepsilon_r)r} \quad , \tag{1}$$

where $r$ is the distance from the nucleus, and $\varepsilon_0, \varepsilon_r$ are, respectively, the permittivity of free space, and the relative permittivity of the material. The one-body, three-dimensional Schrödinger equation that describes the motion of the electron is (e.g. Bransden and Joachain 1989):

$$\left[ -\frac{h^2}{8\pi^2 m_e^*}\nabla^2 - \frac{e^2}{(4\pi\varepsilon_0\varepsilon_r)r} \right]\psi(\mathbf{r}) = E\psi(\mathbf{r}) \quad , \tag{2}$$

yielding the bound state eigenvalues for the $n$th level of the system as

$$E_n = -\frac{2\pi^2 m_e^*}{h^2}\left[ \frac{e^2}{4\pi\varepsilon_0\varepsilon_r} \right]^2\left( \frac{1}{n^2} \right) \quad . \tag{3}$$

As described in the Foreword, the effective electron mass has been measured only very recently (Poolton et al. 2001). For $NaAlSi_3O_8$, an isotropic value of $m_e^* = (0.79 \pm 0.02)m_e$ was determined. Taking the published data for the relative permittivity of the same material to be $\varepsilon_r = 2.33$ (Keller 1966) then, for the simple defect described above, the energy of the 1s–2p ($n = 1 \rightarrow 2$) optical transition would be expected to occur at $1.48 \pm 0.04$ eV.

Remarkably, this is very close to the experimentally observed value for the IR transition in alkali feldspars, which have been reported to occur in the range 1.41–1.47 eV (Bailiff and Barnett 1994; Clark and Sanderson 1994; Poolton et al. 1995b; Godfrey-Smith and Cada 1996; Barnett and Bailiff 1997). Using Eq. (3), the ionisation energy of the defect in Na-feldspar would be expected to occur at 1.97 eV. Whilst the IR resonant transition is easily observable and well defined experimentally (see Fig. 3b), the ionisation energy is more difficult to determine, although 1.97 eV does correspond to the switch-on of a rising continuum in the excitation spectrum, as electrons are excited well away from the centre.

What is special about this result, is that by taking all known parameters ($m_e^*, \varepsilon_r$), the simple hydrogenic model adequately describes the observed optical excitation properties of the IRSL electron trap. Previous consideration of the problem has been only conjectural and limited in scope: Poolton et al. (1995b) imagined that if the defect was hydrogenic in nature, this would imply that the effective electron mass in the material would have to be around $0.76m_e$. (This conjecture is thus borne out.) They went on to show that variations in the relative permittivity within the Ca–Na–K feldspar group could well explain the variations in the IR transition energy experimentally found: this is very small in the K–Na series (varying between 1.44 and 1.42 eV), but much larger in the Na–Ca series (1.42–1.27 eV).

## Determining the extent of the electron wavefunction

The Schrödinger equation (2) is particularly useful, since it allows calculation of the radial extent of the trapped electron wavefunction, which is of direct interest to the present work. It allows us to determine where the recombination centres would need to be (in relation to the electron traps) if they are to participate in tunnelling recombination (from either the ground or excited states); this process would not be subject to thermal assistance (Fig. 1b, c).

The path to the solution of Eq. 2 is lengthy, but the solutions are standard, and can be found in most elementary quantum mechanics texts. Following Bransden and Joachain (1989) for example, the wavefunctions for the first two states $n = 1$ (1s), and $n = 2$ ($^2p_0$) are found to be:

$$\psi_{1s}(r) = C_1 \exp(-r/a^*) \tag{4}$$

$$\psi_{2p}(r) = C_2(r/a^*)\exp(-0.5\,r/a^*) \quad , \tag{5}$$

where $C_1$ and $C_2$ are constants, and $a^*$ is the modified, effective Bohr radius, such that

$$a^* = \frac{\varepsilon_0\varepsilon_r h^2}{\pi m_e^* e^2} \quad . \tag{6}$$

Note that we are not particularly interested in the angular part of the wavefunctions, $\psi(\theta, \phi)$, and these are not considered (the ground state is spherically symmetrical anyway): Eqs. (4) and (5) are valid only for the radial part of the wavefunctions, and these are plotted in Fig. 2. Here, comparison can be drawn with the physical Na-feldspar crystal structure, where a single unit cell is plotted as a projection onto the (1 0 0) crystal plane (after Papike and Cameron 1976: the $a$, $b$, $c$ cell parameters are taken as 8.1, 12.8 and 7.2 Å, respectively).

For luminescence, the extent of the electron wavefunction is only partly of interest: emission only occurs when there are holes to recombine with. For a completely ubiquitous distribution of recombination centres in the material, the number of defects located within a small volume $dr$ at $r$, increases as $r^2$. (This would lead
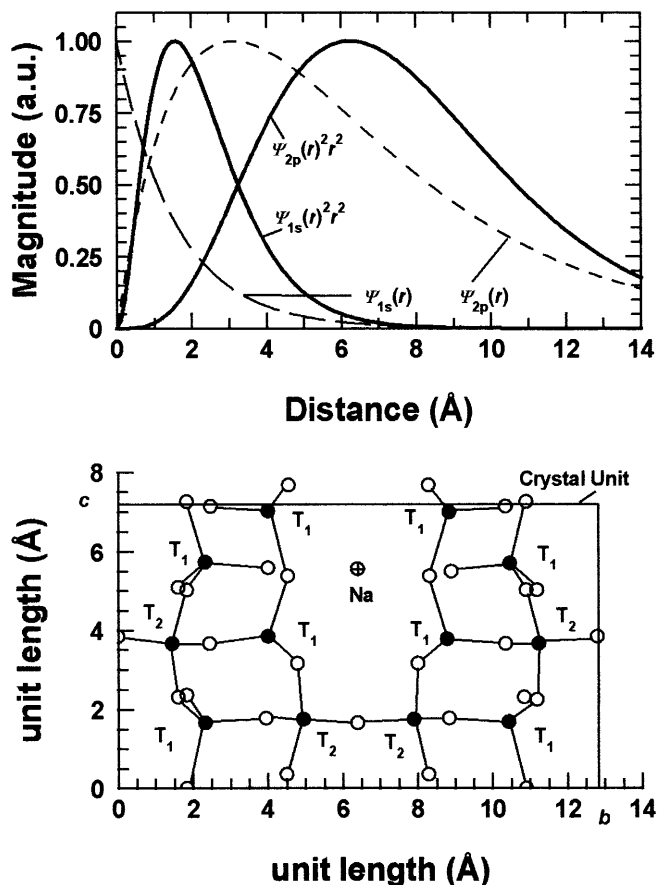
**Fig. 2** *Top* Radial extent of the ground and first excited state of an electron trapped at a simple donor defect in Na-feldspar, obtained from the solution of Schrödinger's equation. The probability of luminescence arising from tunnelling to acceptor defects at distance $r$ will be proportional to $|\Psi(r)|^2 r^2$. All *curves* have been normalized to their maximum value. (*Bottom* after Papike and Cameron 1976) Comparison can be drawn with the crystal structure of $NaAlSi_3O_8$ feldspar, here shown as a projection onto the (1 0 0) crystal plane. *Open circles* Oxygen; *filled circles* silicon lattice positions; interstitial *Na* as marked; $T_1$ and $T_2$ are the two crystallographically distinct silicon sites

to the total number contained within a sphere of radius $r$ being, as required, $\propto 4/3\pi r^3$). Since the actual probability of finding the electron at any position $r$, is $|\psi(r)|^2$, it follows that the probability that the electron will recombine with a trapped hole (and hence the intensity of the luminescence) at distance $r$, is proportional to $|\psi(r)|^2 r^2$. These tunnelling recombination probability functions are also plotted in Fig. 2 for both the ground and excited states.

(In the above argument, we have made two simplifications and assumptions. Firstly, that the ground-state wavefunction of the recombination centre is highly compact – as is the case for $Mn^{2+}$, for example (Manning 1970; Telfer and Walker 1978); we therefore regard it as a point defect – which it is, in respect to the excited state electron. Secondly, we ignore the possibility that the electron and hole traps may not be nearest pairs).

For tunnelling from the electron's ground state, the maximum recombination probability occurs at a distance of 1.6 Å. (Obviously, at very short distances, only discrete defect pair separations are allowed, and 1.6 Å would in reality be the minimum likely electron-hole trap separation, since this is the Si–O bond length in feldspar). The significant point about Fig. 2, is how far the function extends – reaching 1% of its maximum value at 8 Å (the $a$, $c$ unit-cell parameter) or 0.001% at 14 Å (the $b$ dimension): by $2b$, the figure drops to $10^{-10}$%. As far as radiation dosimetry is concerned, any hole trapped within one unit cell's distance from the electron trap will almost certainly have recombined through tunnelling from the ground state of the electron trap during geological times. It is hardly surprising, then, that significant tunnelling afterglow is observed in feldspars following laboratory irradiation (e.g. Spooner 1994; Visocekas et al. 1994, 1998; Visocekas and Zink 1995): this process can lead to age underestimations in luminescence dating (e.g. Wintle 1973; Huntley and Lamothe 2001). In contrast to the ground state, $|\psi_2(r)|^2 r^2$ for the excited state peaks at 6.2 Å and drops to 17 and 0.03% of its maximum value at the radial ($b$, $2b$) distances $r = 14$ and 28 Å, respectively.

We conclude, therefore, that any tunnelling recombination that might occur in IRSL arising from the excited state would principally be taking place at least one unit cell removed from the electron trap. The exception would be in freshly irradiated samples where recombination through tunnelling from the electron trap's ground state, with holes trapped within the unit cell, has not yet taken place.

## Evidence of tunnelling from the excited state of the electron trap

For feldspars, considerable evidence is available, showing that trapped electrons and holes can tunnel from their ground states (as discussed in the preceding section). In some cases, this can be simply monitored by detecting the luminescence that is emitted during recombination, of the type shown in Fig. 1c (e.g. Visocekas 2000).

Proving that tunnelling occurs from the electron excited state (and that it is a potentially significant source of IRSL emission) is somewhat more difficult, but worthwhile, because of its application to dating (see following section). Intuitively, the two luminescence processes shown in Fig. 1a and b will both provide a resonant peak if the photon excitation energy is scanned, of the type shown in Fig. 3b. The luminescence emission spectra from both cases may well be identical (if the recombination centres are $Mn^{2+}$ of $Fe^{3+}$, for example). However, the thermal dependence of the two processes is expected to be very different, the first requiring heat to access the conduction band prior to recombination, and the latter (tunnelling) emission requiring no thermal assistance.
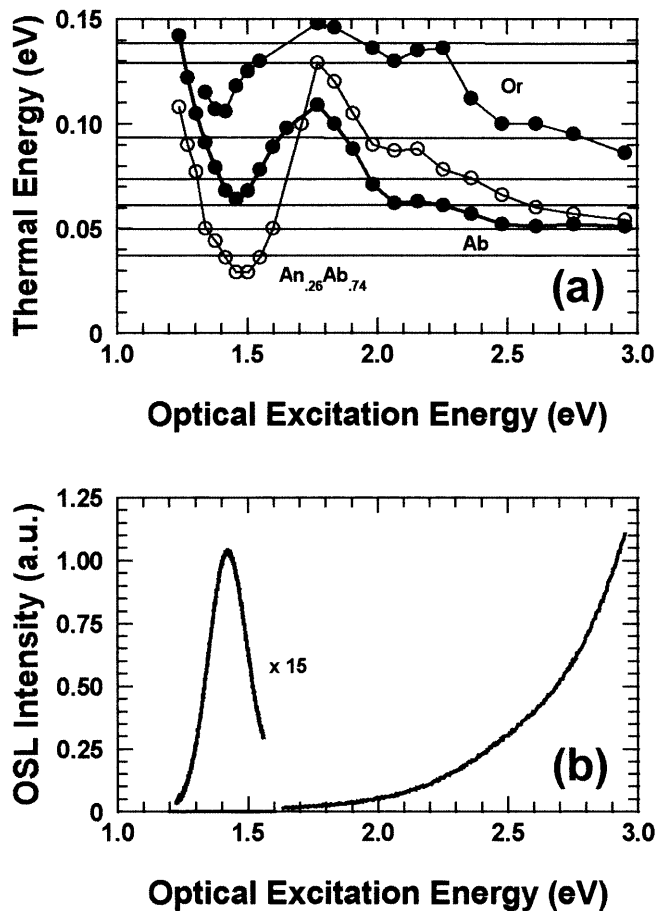
Fig. 3 a (After Poolton et al. 1995a). Thermal activation energy dependence on the optical excitation energy, determined for three different feldspars (where *Or* orthoclase; *Ab* albite; *An* anorthite). Comparison is drawn **b** with the OSL stimulation spectrum for albite. The *horizontal lines* in **a** are the main phonon vibrational energies in feldspars (see Salje 1993)
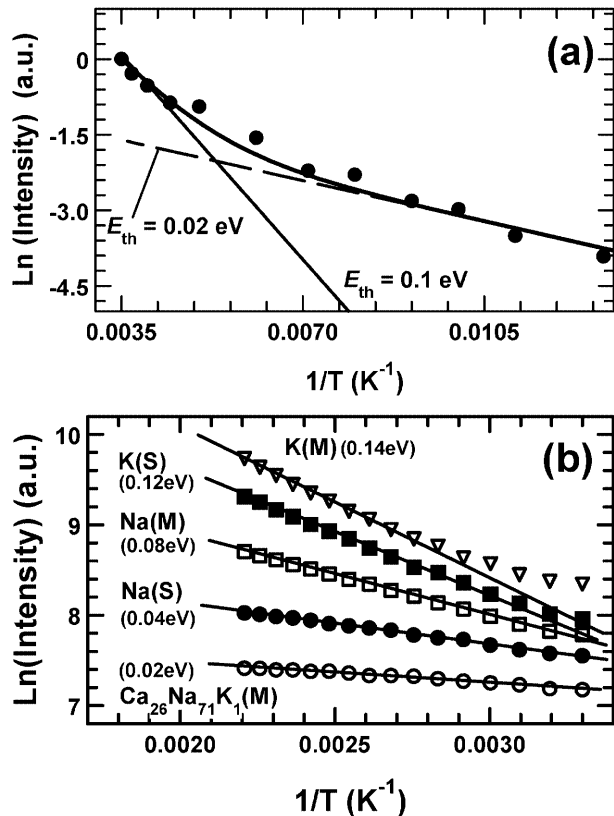


Fig. 4 a Low-energy Arrhenius components of IRSL are best analysed at low temperatures (e.g. 80–300 K). For optical stimulation at 1.41 eV, the data (*points*) of Bailiff and Barnett (1994) are consistent with two components, one with a thermal activation of 0.02 eV, the other with 0.1 eV. **b** Arrhenius plots of the thermal dependence of IRSL (when excited at 1.49 eV), for a range of typical sedimentary (*S*) and single-crystal mineral (*M*) feldspar samples. The temperature range is 300–480 K. Each sample has been dosed in the laboratory to 100 Gy. The thermal activation energies of the main components are as labelled

Simply measuring the thermal activation (TA) energy at the IRSL resonant position can present a confusing picture, since values anywhere between 0.02 and 0.2 eV have been reported (Hütt et al. 1988; Bailiff and Poolton 1991; Duller and Wintle 1991). Being more selective in the excitation energy chosen, however, reveals the source of this confusion, as shown in Fig. 3a. *Off* resonance, (where tunnelling is unlikely), the thermal activation energies are nearly always high – typically 0.14 eV. The anomalies occur at resonance, where there is a possibility of admixture of a tunnelling component. The meaning of the high TA values is discussed in Part II, since it is intricately linked to the nature of the conduction band.

In order to pull apart two thermal components with activation energies at ~0.14 and ~0 eV requires Arrhenius plots of the emission intensity to be made down to cryogenic temperatures. Two groups of workers have done this: Bailiff and Barnett (1994) working on K-rich feldspar and Rieser et al. (1997) working on microcline.

The data points of Bailiff and Barnett (1994) are reproduced in Fig. 4a. Whilst for high temperatures ($T > 225$ K) the data was consistent with a TA energy of 0.1 eV, below this temperature, the second component becomes clearly apparent. Although they do not evaluate this, we suggest that their data are not inconsistent with the second component having a TA of just 0.02 eV. Rieser et al. (1997) also found two components in their material, having activation energies of 0.15 and 0.05 eV.

We believe that the two components found in the IRSL transition are indicative that the tunnelling and non-tunnelling luminescence processes can coexist; which is dominant in any one material will be dependent on the particulars of the trapped charge distribution. [The fact that the low-energy tunnelling component is not exactly zero is explainable by a minor thermal positioning within the ground state of the electron trap: Visocekas (2000) for example, has established that ground-state to ground-state tunnelling can be thermally activated in some cases].

## Implications for luminescence dating

Where feldspars are to be used in radiation dosimetry applications (such as in dating), radiation-induced charge trapped at defects in the material would, ideally, remain trapped indefinitely. Unfortunately, the presence of close electron-hole trapping pairs gives rise to tunnelling recombination (from their ground states). As a consequence, the leakage of charge causes the measurable OSL to "fade" with time (e.g. Huntley and Lamothe 2001). Because of its tunnelling origin, such fading can continue over geological time periods. For dating, testing for the presence of fading is standard practice, since samples can either be rejected for measurement or appropriate compensation for the rate of charge loss can be made. If fading is not taken into consideration, it can lead to an underestimation of the palaeodose and, consequently, of the age of the sample. To detect the presence of fading, the OSL signal level usually needs to be monitored at regular intervals over a period of weeks or months (e.g. Spooner 1994), which is both time-consuming and inconvenient.

Is there a more rapid (even "instant") way to test for fading, without waiting months? Clearly, the pure quantum mechanical tunnelling process of the ground-state charge cannot be accelerated. However, Fig. 2 shows that the electron wavefunction can be greatly expanded, by stimulating electrons into the excited state of the defect. This method, then, can be used to probe for nearby recombination centres that would, in time contribute to the fading process. The simple hypothesis is thus: if IRSL shows a tunnelling component, then the sample may be subject to fading. Conversely: if there is no IRSL tunnelling component, there can be no nearby holes trapped at recombination centres and, by implication, the sample will not be subject to fading.

We have carried out initial experiments that show that this idea is sound. A range of pure mineral (M) and sedimentary (S) feldspar samples were selected for study, and the thermal activation characteristics determined in the temperature range 300–475 K, as shown in Fig. 4. (Ideally, the work would be done from 80–475 K). The samples can be grouped into three sets:

1. Those showing a single component with a low TA energy, around 0.02 eV, such as $Ca_{26}Na_{71}K_1(M)$. This component is assumed to be the excited-state tunnelling component.
2. Those showing predominantly a single component with high TA energy, such as K(M) and K(S). This component is assumed to be the non-tunnelling IRSL component.
3. Those samples with an intermediate TA value, such as Na(M) and Na(S).

Group (1) faded by typically 65% over a 7-day measurement period. (All samples were maintained at a temperature of 100 °C during this time). Group (2) showed no observable fading in 7 days. Group (3) showed some fading, typically around 10–15%. This is, we believe, a clear initial indication of the validity of our ideas, and points the way to developing a rapid method for the detection of fading, for use in dating.

## Conclusions

We have been able to consider and map the extent of the wavefunction of charge located at simple electron traps in feldspar, for the first time; the effects on luminescence arising from direct electron-hole recombination have been considered. Comparison of the extent of the ground and excited state wavefunctions suggests that any tunnelling recombination arising from the excited state in samples of geological age is likely to occur at least one lattice cell distant from the electron traps. In all cases, it is unlikely that any tunnelling recombination would occur at distances greater than about 30 Å.

Not all feldspars show an excited-state tunnelling component. Of a range of feldspar samples measured, we have found that those that do are prone to fading (i.e. the uncontrollable loss of charge from the defect's ground states). We have suggested that this is because the greater extent of the excited-state electron wave-function is able to probe for holes trapped near the electron centre, which would be liable to partake in the fading process. The work leads to possibilities for developing the method as a tool for the rapid assessment of the suitability of material for dating applications.

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