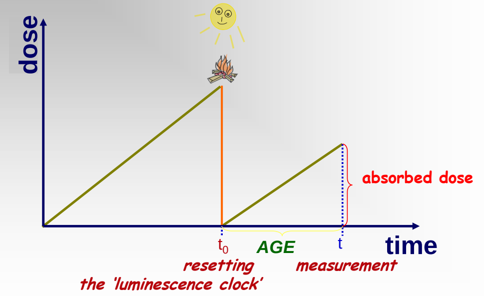
Growth and Resetting of the Luminescence Signal



Excitation and Luminescence Photon Energies Used in OSL Dating

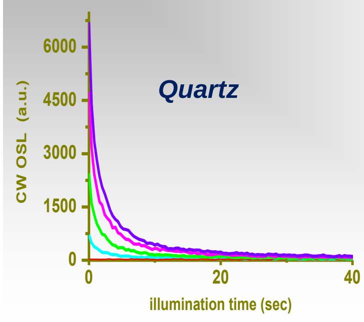
Mineral	*Energy (wavelength) of excitation photons*	*Energy (wavelength) of luminescence photons*
Quartz (SiO_2)	2.2 – 2.4 or 2.7 eV (510 – 560 or 470 nm) green-blue	3.35 eV (370 nm) ultraviolet
Potassium Feldspar $KAlSi_3O_8$	1.4 eV (880 nm) infrared	3.1 eV (400 nm) Violet

Appropriate detection filters required

Types of OSL

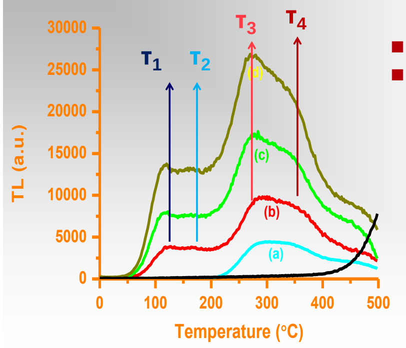
1. *Continuous Wave OSL* ***(CW-OSL)***: the stimulation intensity is kept constant throughout the duration of the experiment, with simultaneous monitoring of the signal.
2. *Linearly Modulated OSL* ***(LM-OSL):*** the stimulation intensity is linearly increased with time, with simultaneous monitoring of the signal.
3. *Non-linearly Modulated OSL* ***(NLM-OSL):*** the stimulation intensity is non-linearly increased (parabolically, hyperbolically, etc) with time, with simultaneous monitoring of the signal.
4. *Pulsed OSL* ***(P-OSL):*** the sample is exposed to stimulation pulses, while monitoring of the signal takes place when stimulation mode is off ***(NO FILTERS REQUIRED)***.

Experimental CW-OSL Curves



- Experimental parameters
  1. Illumination t (sec)
  2. $P_{stimulation}$ (mW/cm^2)
  3. $T_{measurement}$ (°C)
  4. $T_{preheat}$ (°C)
  5. Preheat t (sec)
- Dependent Variables
  1. I_{max} (a.u.)
  2. σ (cm^2)

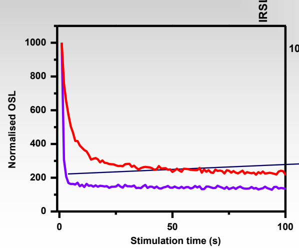
Why T_{max} and $T_{preheat}$

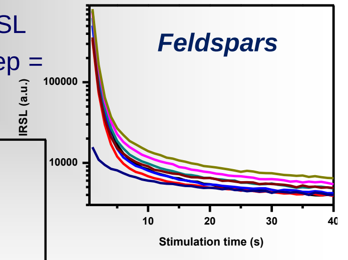


- $T_1 < T_2 < T_3 < T_4$
- **Assumption (which experimentally is turned to be correct: TL and OSL stimulate the exact same traps→the intention is to isolate signal from more stable traps.**

Examples of CW-OSL curves

Initial part of the OSL Signal should be steep = Fast decaying

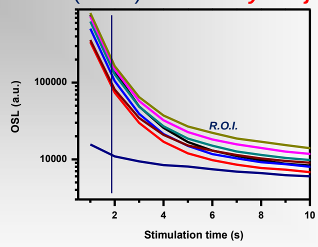




Dominated by fast OSL

Quantifying luminescence

➢Regions of interest (R.O.I.) → ***Arbitrary/Subjective limits***



➢This method alleviates the statistical fluctuations suffered by peak height methods but is still based on the overlap of several peaks. Very common in the case of OSL dating protocols such as SAR OSL.

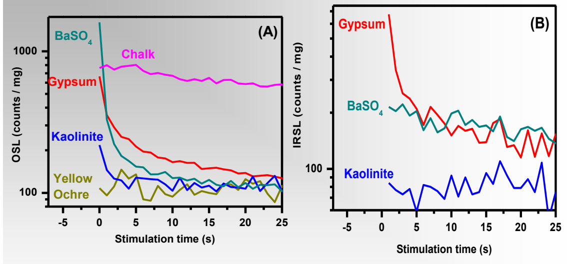
Fast OSL component

➢For quartz OSL, it is desirable to use a well-separated easily bleached (termed as ***fast***) OSL component in dating routines. The fast component is yielded at the beginning of stimulation. The age is determined by using the initial part of the CW-OSL signal minus a background based on the signal level at the end of the stimulation period. In some quartz samples the fast component dominates the initial CW-OSL signal. However, in others the contribution from other less light-sensitive signals can be significant.

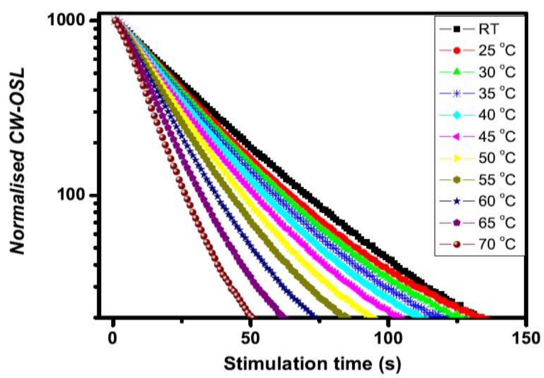
➢FAST OSL decays after roughly 50 seconds of stimulation!

➢Isolating and separating the most appropriate luminescence signal based on stability and bleach-ability criteria (mainly among others).

Examples of OSL, IRSL curves

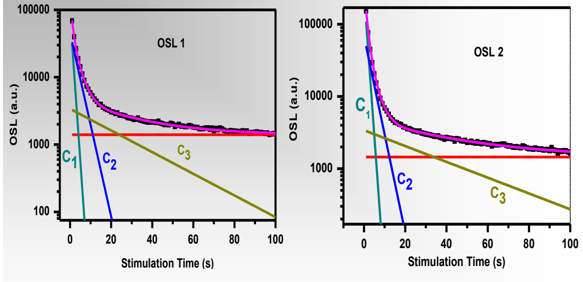


Signal isolation and separation is required!

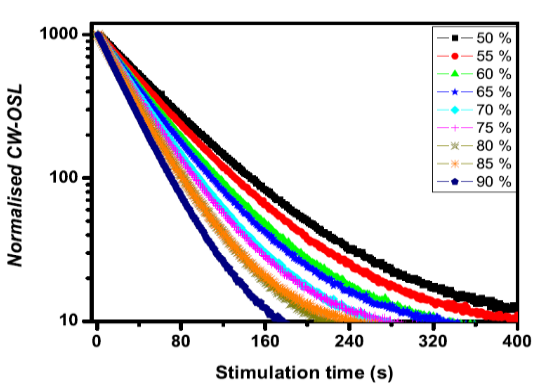


Isolation via high T measurement

Deco examples: Quartz

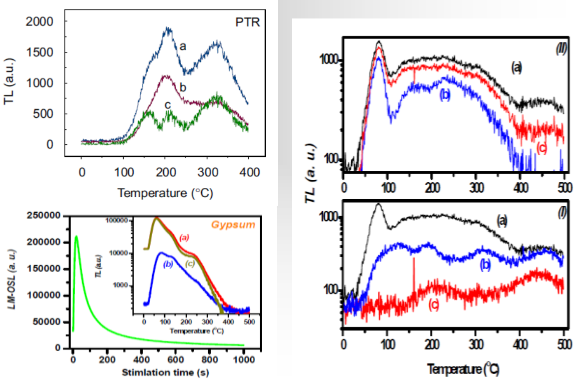


Isolation via de-convolution



Isolation via high stimulation intensity

Correlation between TL and OSL



$$I_{LUM}(t) = -\frac{dn}{dt} = pn$$

$$p = \sigma(\lambda)\,\phi(t)$$

$$I_{OSL}(t) = I_0 \exp\left(-\frac{t}{\tau}\right) = n_0\sigma(\lambda)\phi \cdot \exp\left(-\sigma(\lambda)\phi \cdot t\right)$$

Photo-ionization cross section

$$I_{LUM}(t) = -\frac{dn}{dt} = p\frac{p_r}{p_t}\frac{n^2}{N}$$

Stimulation intensity flux: Constant (CW-OSL)

$$I_{OSL} = \sigma(\lambda)\cdot\phi\cdot\frac{p_r}{p_t}\frac{n_0^2}{N}\cdot\left(1+\frac{p_T}{p_t}\frac{n_0}{N}\cdot\sigma(\lambda)\cdot\phi\cdot t\right)^{-2}$$

$$I_{LUM}(t) = -\frac{dn}{dt} = p\frac{p_r}{p_t}\frac{n^b}{N}$$

$$I_{OSL} = \sigma(\lambda)\cdot\phi\cdot\frac{p_r}{p_t}\frac{n_0^b}{N}\cdot\left(1+\frac{p_T}{p_t}\frac{n_0^{b-1}}{N}\cdot(b-1)\cdot\sigma(\lambda)\cdot\phi\cdot t\right)^{\frac{b}{b-1}}$$

$$I_{LUM}(t) = -\frac{dn}{dt} = pn$$

$$p = s\cdot\exp\left(-\frac{E}{kT}\right)$$

$$I_{TL}(T) = n_0\cdot s\cdot\exp\left(-\frac{E}{kT}\right)\exp\left[-\frac{s}{\beta}\cdot\int_{T_0}^{T}\exp\left(-\frac{E}{kT'}\right)dT'\right]$$

$$I_{LUM}(t) = -\frac{dn}{dt} = p\frac{p_r}{p_t}\frac{n^2}{N}$$

$$I_{TL}(T) = \frac{p_r}{p_t}\frac{s}{\beta}\exp\left(-\frac{E}{kT}\right)\frac{n_0^2}{N}\left(1+\frac{n_0}{N}\int_{T_0}^{T}\frac{p_r}{p_t}\frac{s}{\beta}\exp\left(-\frac{E}{kT'}\right)dT'\right)^{-2}$$

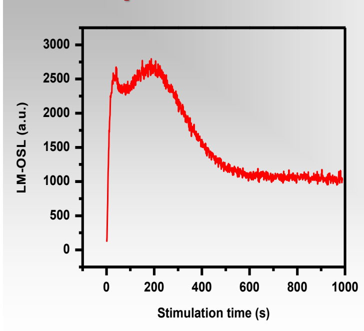
$$I_{LUM}(t) = -\frac{dn}{dt} = p\frac{p_r}{p_t}\frac{n^b}{N}$$

$$I_{TL}(T) = s\cdot\frac{n_0^b}{N}\frac{p_r}{p_t}\cdot\exp\left(-\frac{E}{kT}\right)\cdot\left(1+\frac{p_r}{p_t}\frac{s}{N}\frac{n_0^{b-1}}{\beta}\cdot(b-1)\cdot\int_{T_0}^{T}\exp\left(-\frac{E}{kT'}\right)dT'\right)^{\frac{b}{1-b}}$$

Photo-ionization cross section

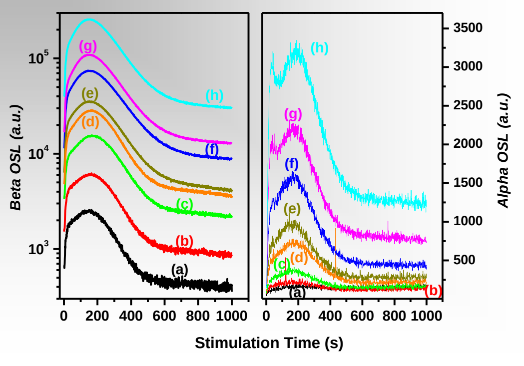
- A **cross section** is the effective area that governs the probability of some scattering or absorption event. Together with particle density and path length, it can be used to predict the total scattering probability via the Beer-Lambert Law.
- In **nuclear** and **particle physics**, the concept of a **cross section** is used to express the likelihood of interaction between particles.
- In ***OSL***, the concept of a **cross section** is used to express the likelihood of interaction between one photon of the excitation and one electron.

Experimental LM-OSL Curves

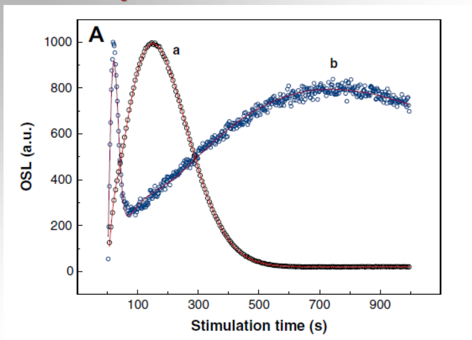


- Experimental parameters
  1. Illumination t (sec)
  2. $P_{stim.init}$ (mW/cm^2)
  3. $P_{stim.fin}$ (mW/cm^2)
  4. $T_{measurement}$ (°C)
  5. $T_{preheat}$ (°C)
  6. Preheat t (s)
- Dependent Variables
  1. I_{max} (a.u.)
  2. σ (cm^2)
  3. t_{max} (s)

Examples of LM-OSL curves



Examples of LM-OSL curves



$$I_{LUM}(t) = -\frac{dn}{dt} = pn$$

$$I_{OSL}(t) = n_0 \cdot \sigma(\lambda) \cdot q \cdot t \cdot \exp\left[-\frac{\sigma(\lambda) \cdot q \cdot t^2}{2}\right]$$

$$I_{LUM}(t) = -\frac{dn}{dt} = p\frac{p_r}{p_t}\frac{n^2}{N}$$

$$I_{OSL} = \sigma(\lambda) \cdot q \cdot t \cdot \frac{p_\tau}{p_t}\frac{n_0^2}{N} \cdot \left(1 + \frac{p_T}{p_t} \cdot \frac{n_0}{N} \cdot \frac{\sigma(\lambda) \cdot q \cdot t^2}{2}\right)^{-2}$$

$$I_{LUM}(t) = -\frac{dn}{dt} = p\frac{p_r}{p_t}\frac{n^b}{N}$$

$$I_{OSL} = \sigma(\lambda) \cdot q \cdot t \cdot \frac{p_\tau}{p_t}\frac{n_0^b}{N} \cdot \left(1 + \frac{p_T}{p_t} \cdot \frac{n_0^{b-1}}{N} \cdot (b-1) \cdot \frac{\sigma(\lambda) \cdot q \cdot t^2}{2}\right)^{\frac{b}{b=1}}$$

1-b

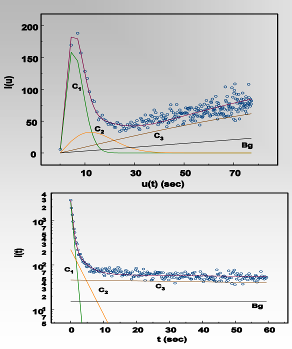
$$p = \sigma(\lambda)\,\phi(t)$$

Photo-ionization cross section

Stimulation intensity flux:

Linear (LM-OSL)

CW-OSL curves



$$u = \sqrt{2td}$$

$$y = I\frac{u}{d}$$

Pseudo LM-OSL (PS LM-OSL)

The shape of OSL curves

- All CW-OSL curves, decaying with time but not according to a single exponential, yield a featureless shape in the sense that it is very difficult to distinguish prominent components by their shape.
- LM-OSL method of measuring the OSL suggested by Bulur (*Radiat. Meas. 26, 701–709, 1996*) yields bell-shaped curves, similar to those of TL, with each bell-shaped peak corresponding to a unique component.
- Even in the case where there is only one, unique TL trap, the OSL signal yields at least 2 different components.
- Overlapping of various OSL components!!!
- All components start decaying at t=0 s.

De-convolution

- Quantitative isolation – separation of the luminescence signal of each component based on analytical models
- Model dependent procedure since various peak shape methods can be used
- Time-consuming
- Has the potential of delivering the greatest amount of information with great precision and accuracy
- The most frequently peak shape methods used include various combinations of first-order, second-order, mixed-order and ***general-order kinetics***

(*Horowitz and Moscovitch, Rad. Prot. Dos. 153, 1–22, 2013*)

Equation for OSL curves: CW-OSL

$$I(t) = I_0\left[1 + (b-1)\,\frac{t}{\tau}\right]^{-\frac{b}{b-1}}$$

- Fitting parameters

1. b = kinetic order (ranging between 1 and 2)
2. τ = decay lifetime of the OSL component
3. I_0 = maximum intensity of the OSL component

Independent variable: time t (s)

Equation for LM-OSL curves

$$I(u) = \frac{I_m}{u_m} \cdot u \cdot \left(\frac{\beta - 1}{2 \cdot \beta} \cdot \frac{u^2}{u_m^2} + \frac{\beta + 1}{2 \cdot \beta} \right)^{\frac{\beta}{1-\beta}}$$

- Fitting parameters

1. u_m = stimulation time where the signal gets its maximum value
2. β = kinetic order (ranging between 1 and 2)
3. I_m = maximum intensity of the peak

Independent variable: Stimulation time u (s)

(*Polymeris et al., PSSA 203, 578–590, 2006*)

Parameters - Constraints

➢The proposed general-order kinetic function has free parameters being the experimentally evaluated I_m, I_0 and T_m (u_m) as well as the kinetic order parameter *b* (as well as *E and σ* for the case of TL and OSL respectively) instead of intrinsic parameters of the material.

➢For n components/peaks, the model requires 4n+1 parameters for the case of TL while 3n+2 for the case of OSL, as well as great ambiguity in the selection of the range of allowed values.

➢An important aspect of successful de-convolution of complex glow curves consisting of multi-peak/overlapping components is not to let the program diverge; for such large number of parameters, and if common sense/previous knowledge is not used to fix as many parameters as possible within certain limits/constraints, unphysical results can be easily obtained due to local minima in the x^2 hyperspace.

Computer/Software specifications

➢The availability of powerful computers and multifunctional software packages has led to significant development and ease of application of de-convolution.

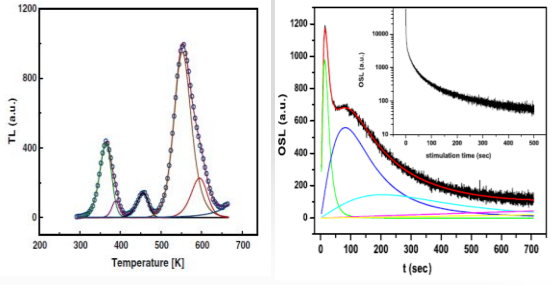
➢Many of the de-convolution programs are based on easily available, standard computer software, e.g. Microsoft/Excel/spreadsheets with the solver utility, Matlab or Mathcad, MatlabR2008b with curve fitting toolbox.

➢The solver is a general purpose optimisation package that uses the generalised reduced gradient non-linear optimisation code and is an Excel Add-in.

➢ For the majority of the examples bellow, the latter was applied.

(*Afouxenidis et al., Rad. Prot. Dos. 149, 363–370, 2012*)

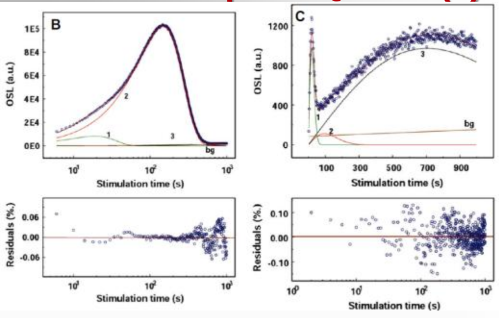
Deco examples: CaF_2:N



→ One-to-one correlation between TL peaks and OSL components

(*Polymeris et al., NIM B 251, 133–142, 2006*)

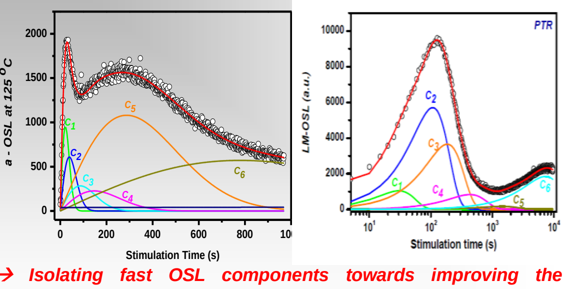
Deco examples: Quartz (1)



→ Correlation between 110°C TL peak and room temperature OSL

(*Polymeris et al., Rad. Meas. 44, 23–31, 2009*)

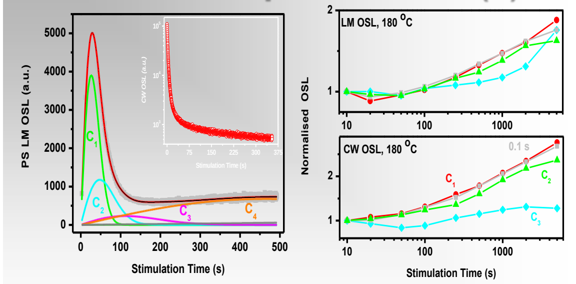
Deco examples: Quartz (2)



→ Isolating fast OSL components towards improving the luminescence age limits

(*Kiyak et al., Rad. Meas. 42, 144–155, 2007*)

Deco examples: Quartz (3)



→ Isolating fast OSL components towards improving the luminescence age limits

(*Kitis et al., Geochronometria 38(3), 209–216, 2011*)