



WIR SCHAFFEN WISSEN – HEUTE FÜR MORGEN

The basics of TSL and OSL

Eduardo G. Yukihara

*Head of the Radiation Metrology Section and Dosimetry Group, Paul Scherrer Institute
Adjunct Professor, Physics Department, Oklahoma State University*

Summer school on scintillation, dosimetric and phosphor materials
7-8 September 2018, Prague, Czech Republic

Where is TL/OSL used today?

Diagnostic imaging (computed radiography)

Personal/environmental monitoring



Luminescence dating

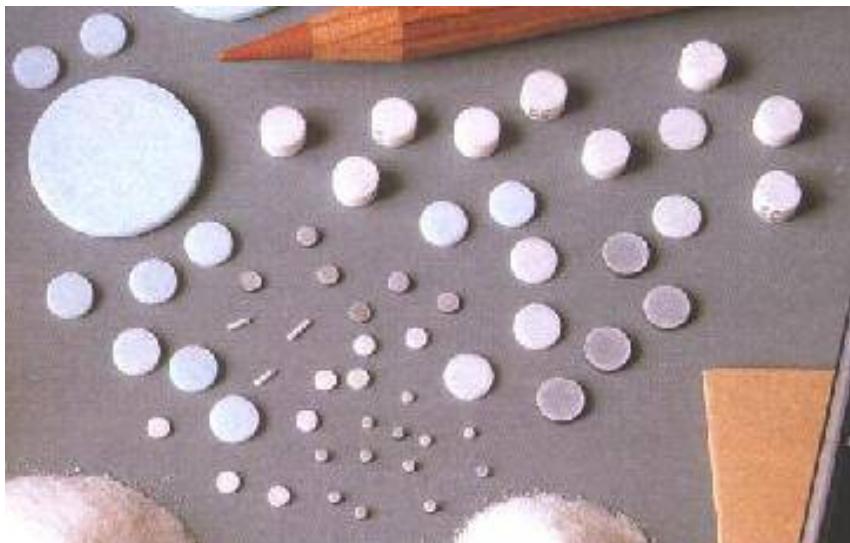


in-vivo measurements



TL/OSL detectors

LiF TLDs



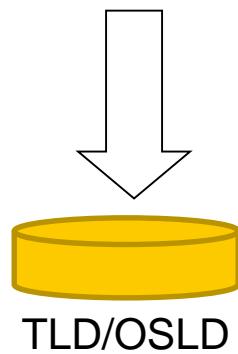
$\text{Al}_2\text{O}_3:\text{C}$ TLDs and OSLDs



Basic scheme

(a) excitation

X-rays, γ -rays, e-beam,
protons, etc.

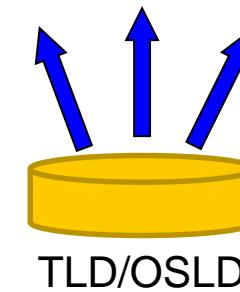


(b) storage



(c) stimulation

heating or
illumination



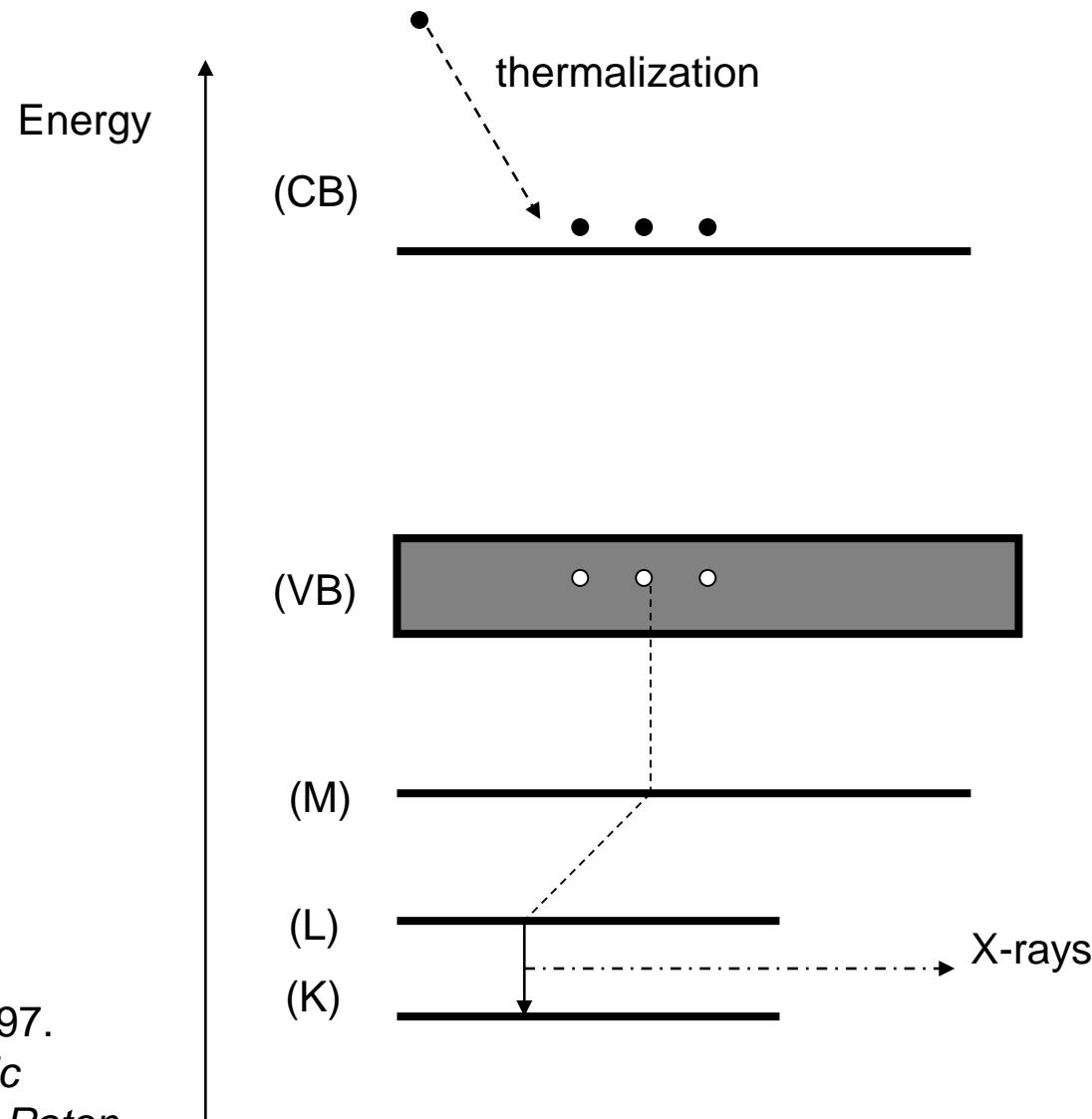
Outline

- Fundamentals
- Measurement techniques
- Main characteristics
- Advantages/disadvantages
- Materials R&D and trends

How to explain the TL/OSL?



Energy levels in a crystal

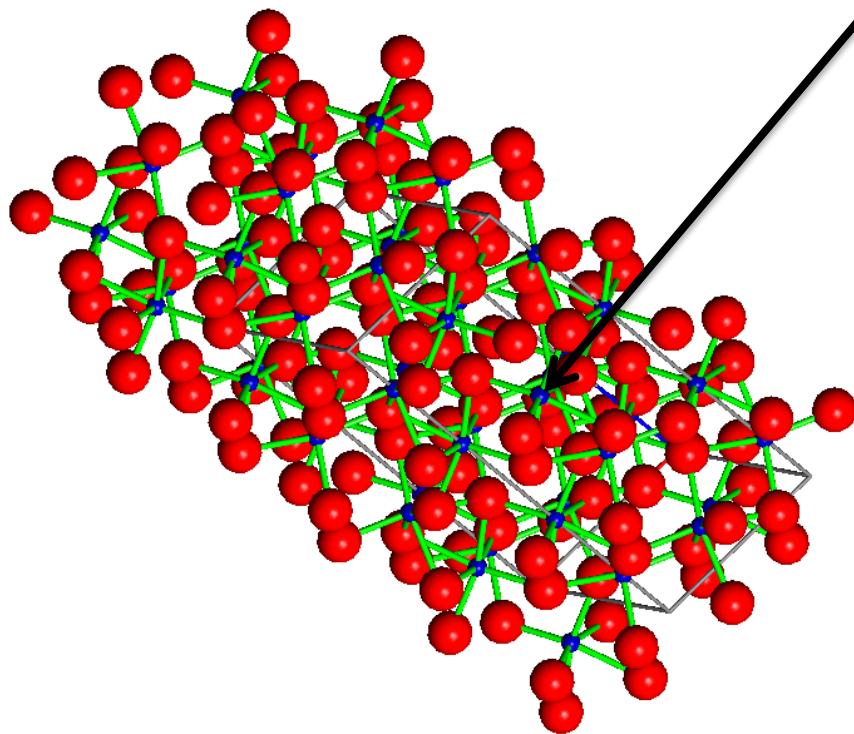


Adapted from Rodnyi, P.A., 1997.
Physical processes in inorganic scintillators. CRC Press, Boca Raton.

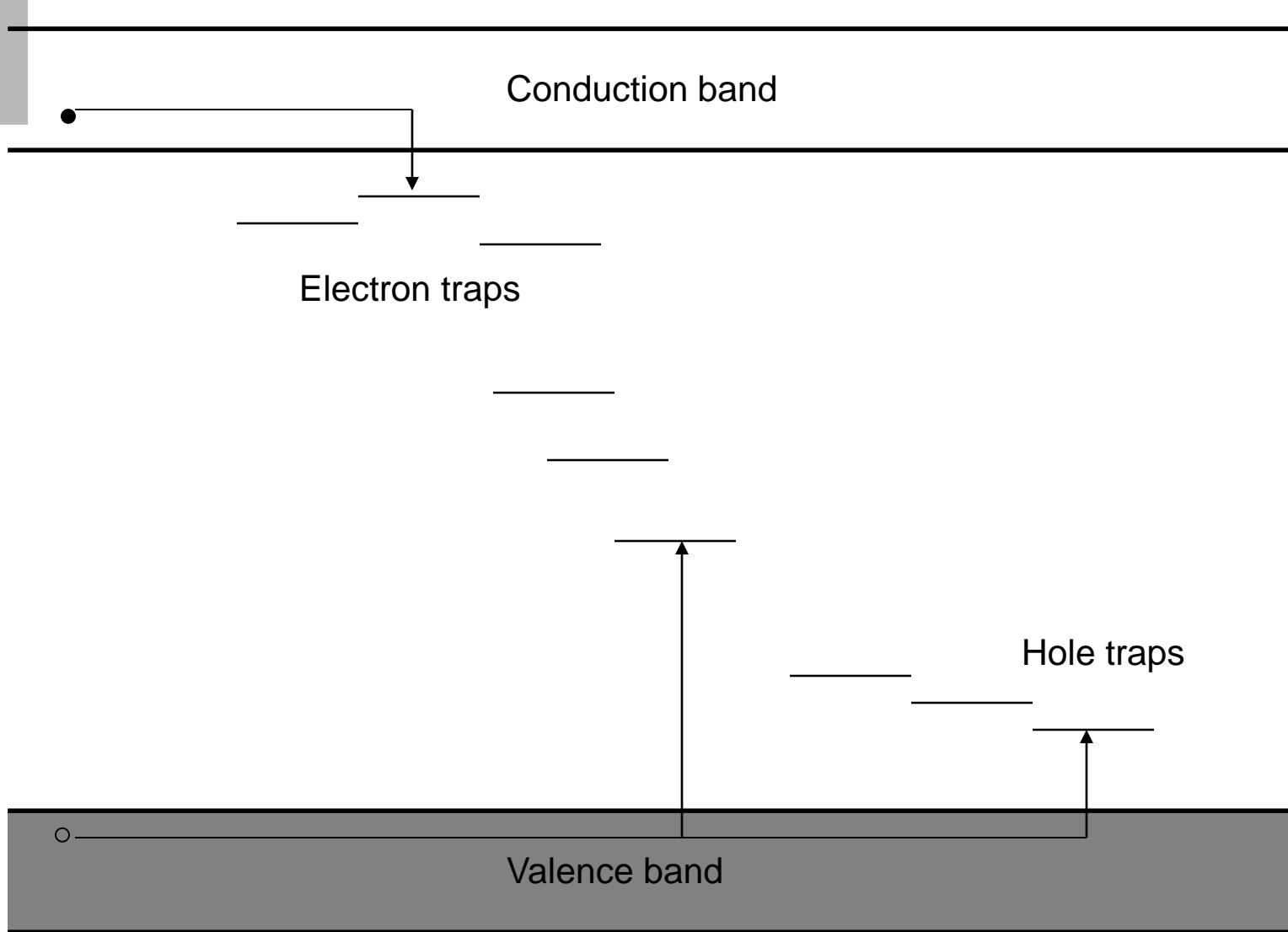
Defects in the crystalline lattice

Impurity (instead of Al^{3+}):

- Fe^{3+}
- Ce^{3+}
- Mg^{2+}

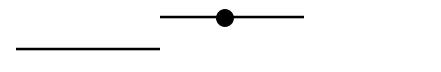


Defects in the crystalline lattice

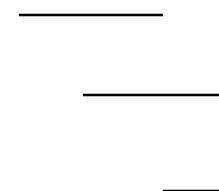


Defects in the crystalline lattice

Conduction band



Electron traps

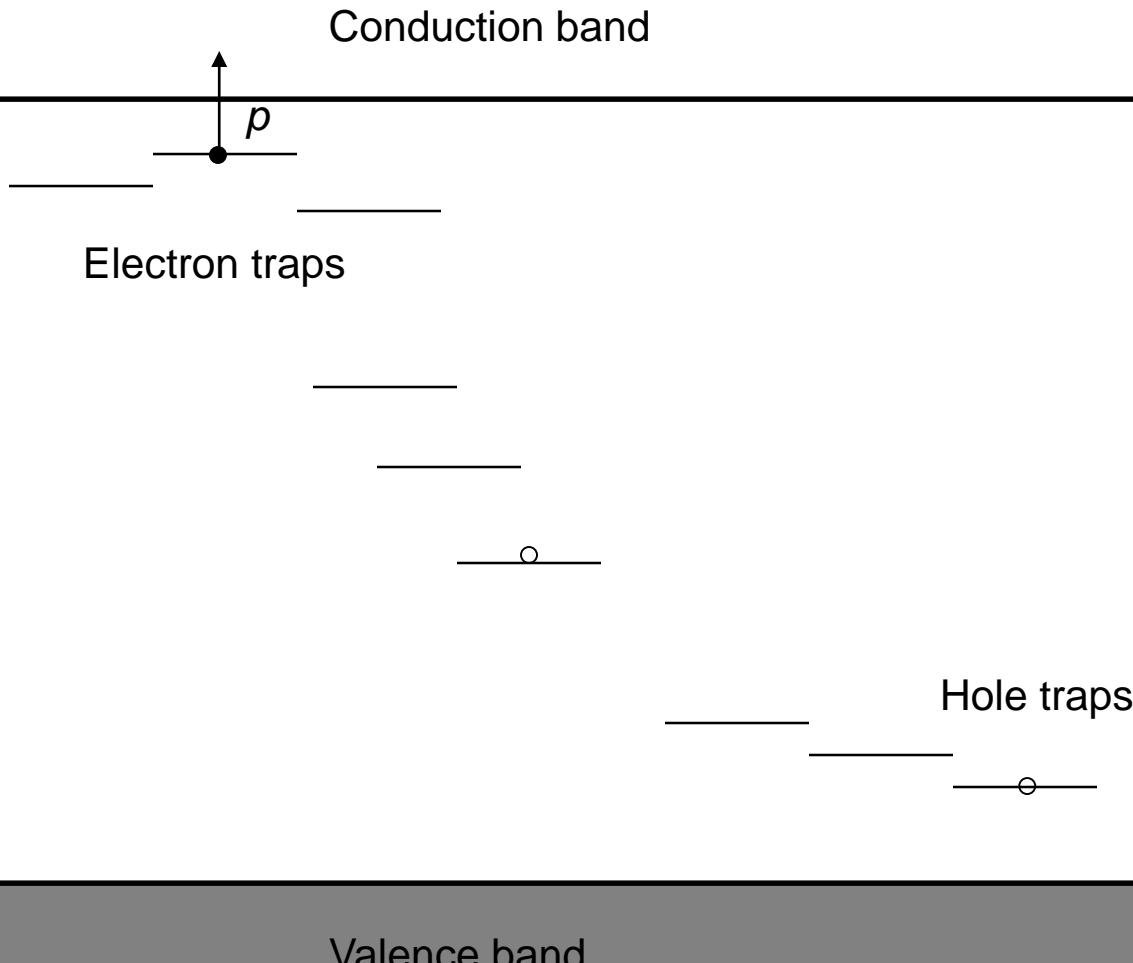


Hole traps

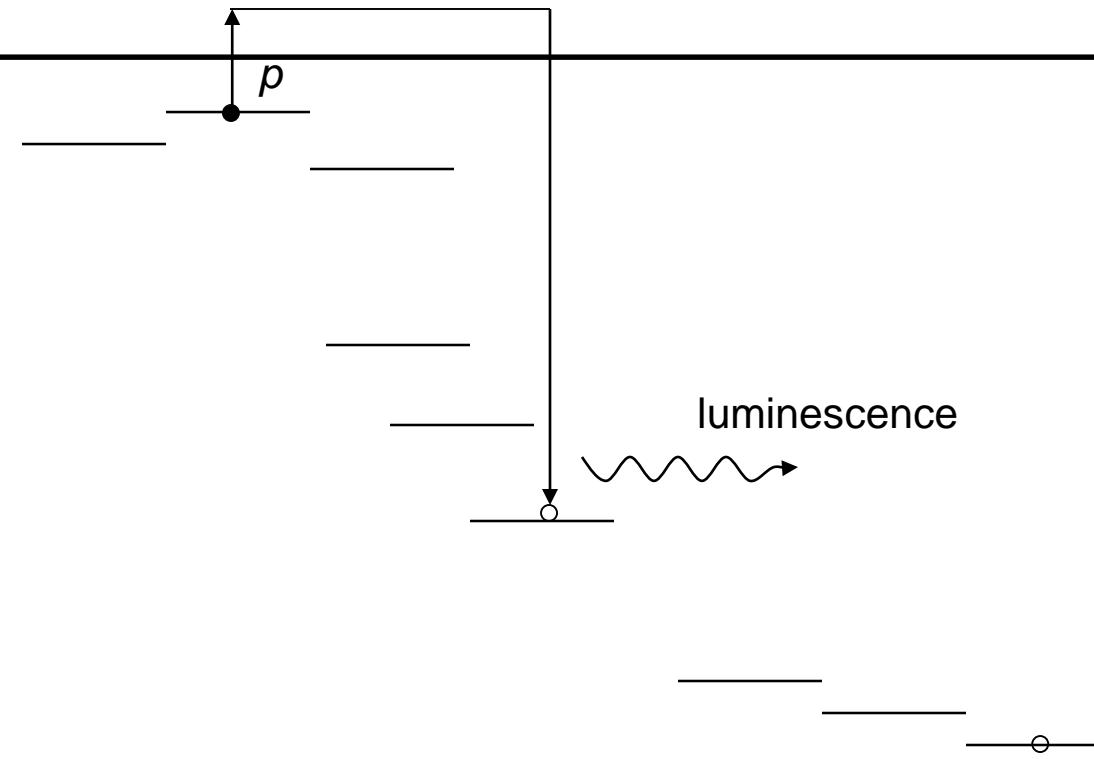


Valence band

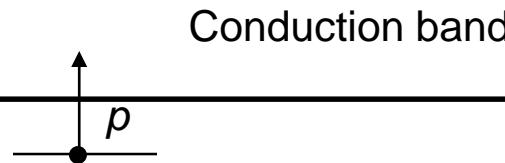
Thermal/optical stimulation



Recombination



Thermal/optical stimulation



Thermal stimulus

$$p = s e^{-\frac{E}{kT}}$$

- E = Activation energy
- s = Frequency factor
- k = Boltzmann constant
- T = Temperature

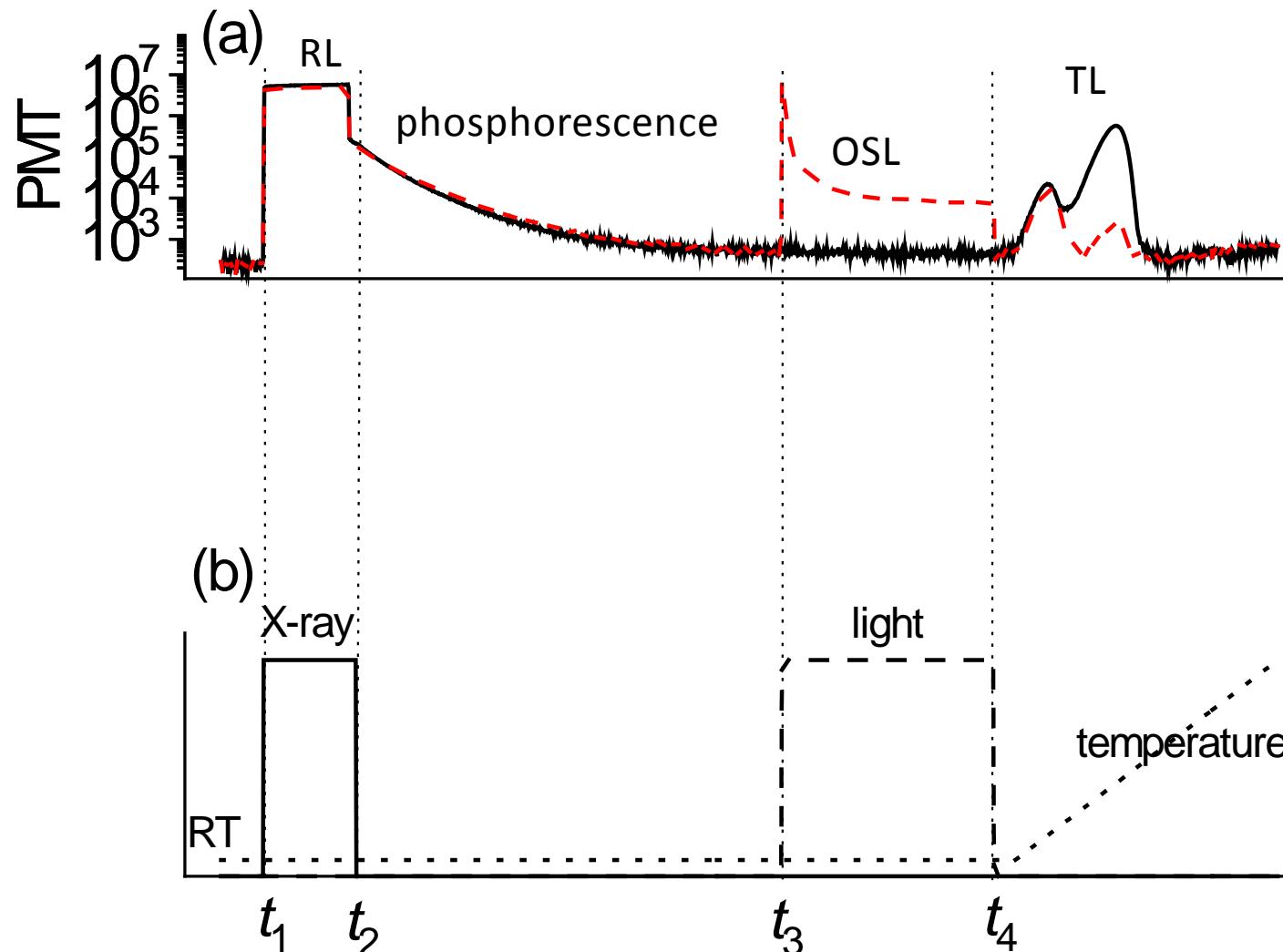
Optical stimulus

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

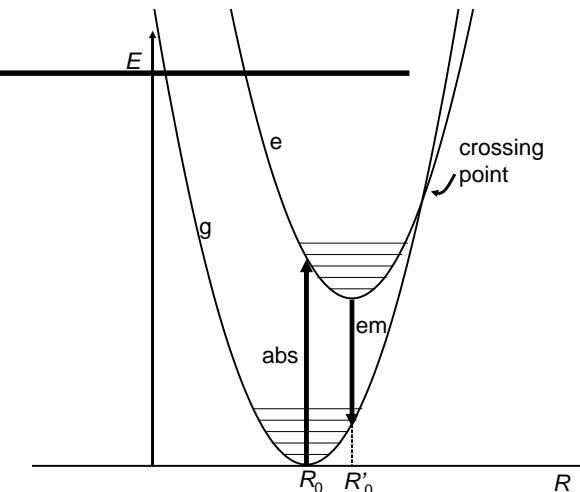
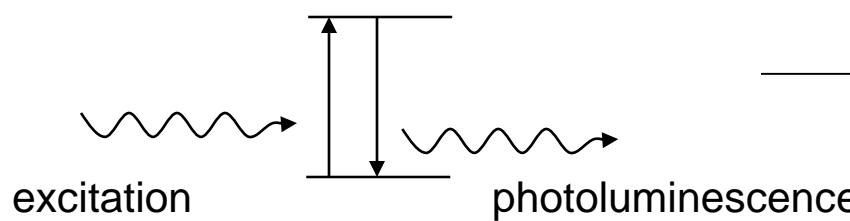
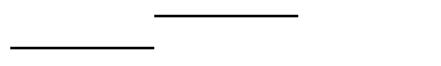
- σ = photoionization cross-section (cm^2)
- ϕ = photon flux ($\text{fótons cm}^{-2} \text{ s}^{-1}$)
- λ = wavelength

$$E(\text{eV}) \approx \frac{1240}{\lambda(\text{nm})}$$

Other processes



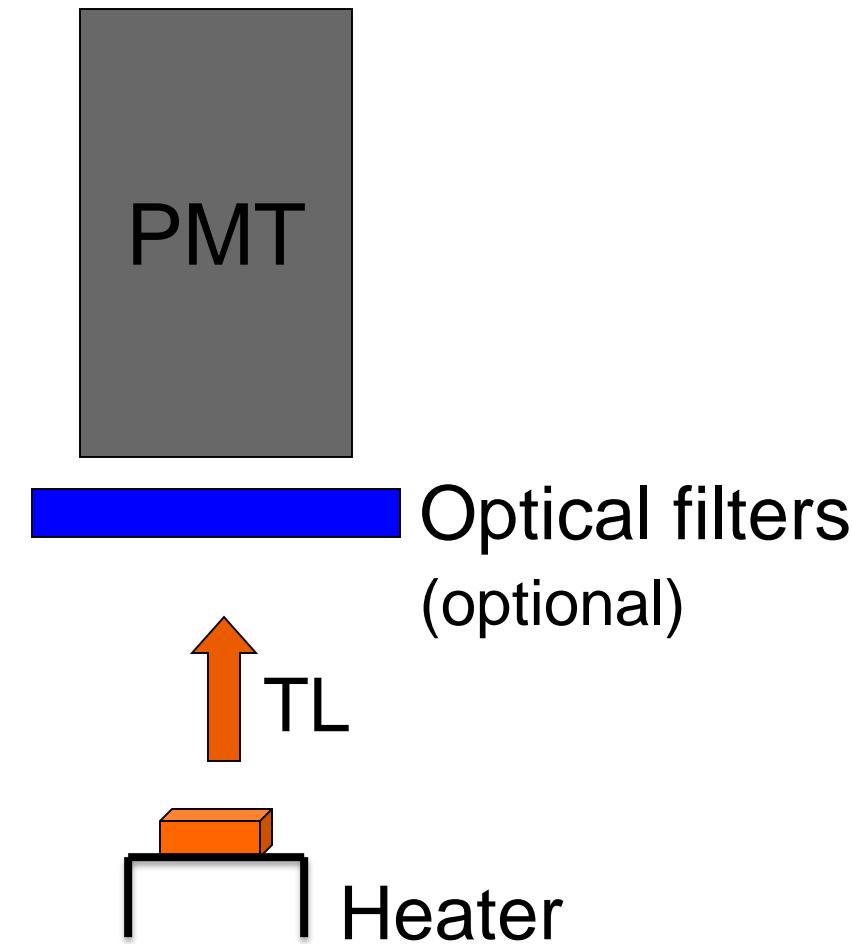
Radiophotoluminescence



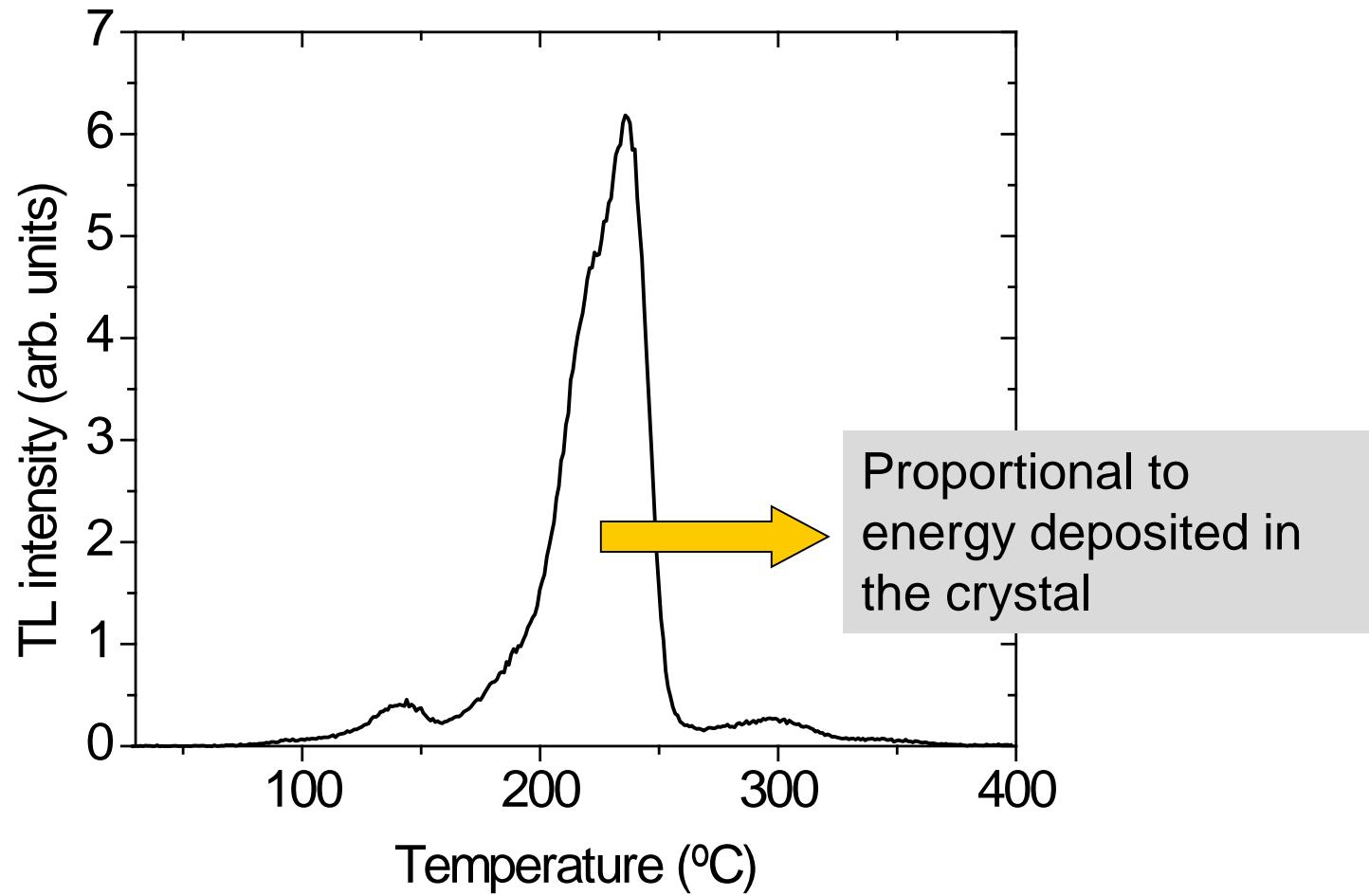
How to measure TL/OSL?



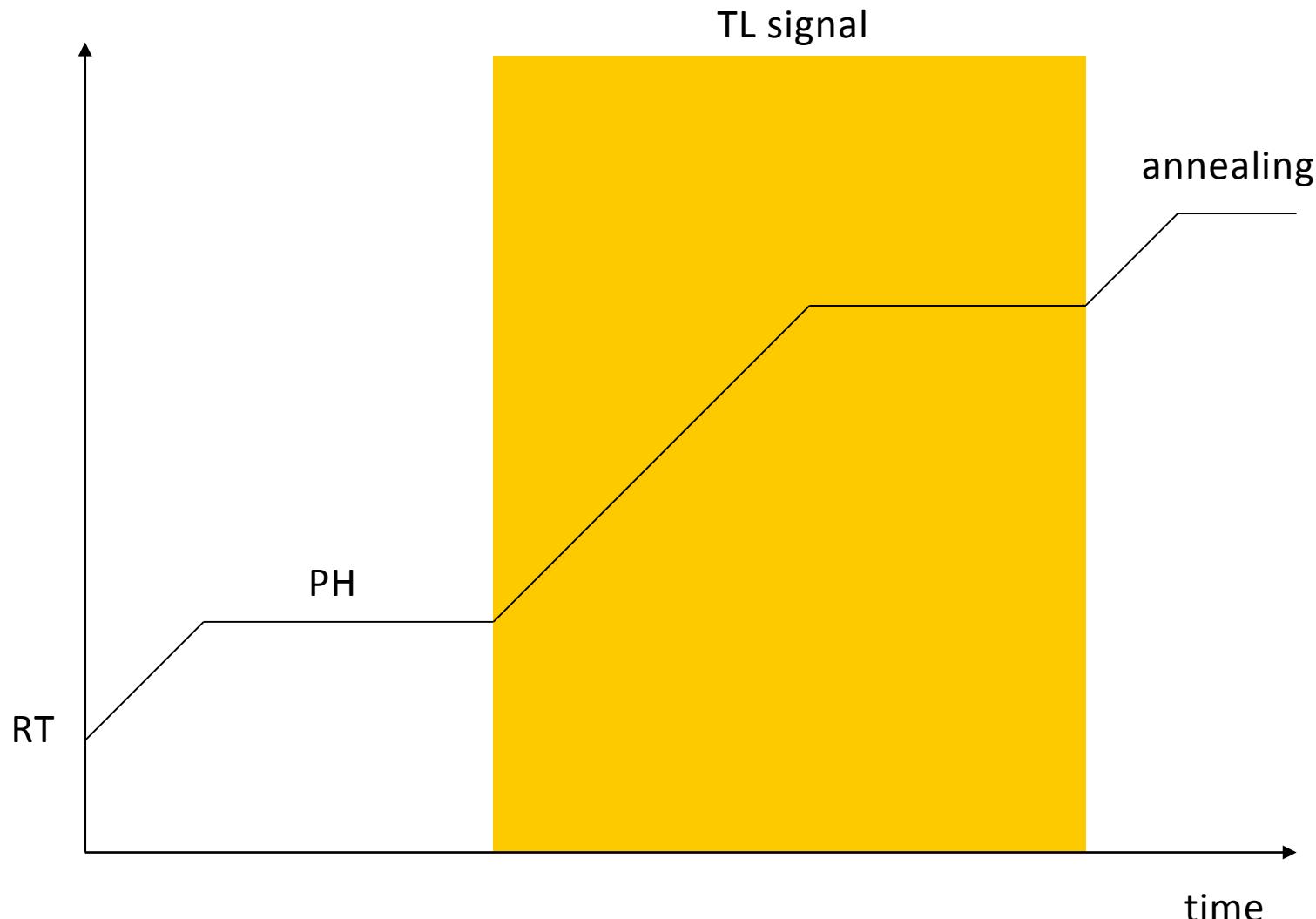
TL readout



TL readout

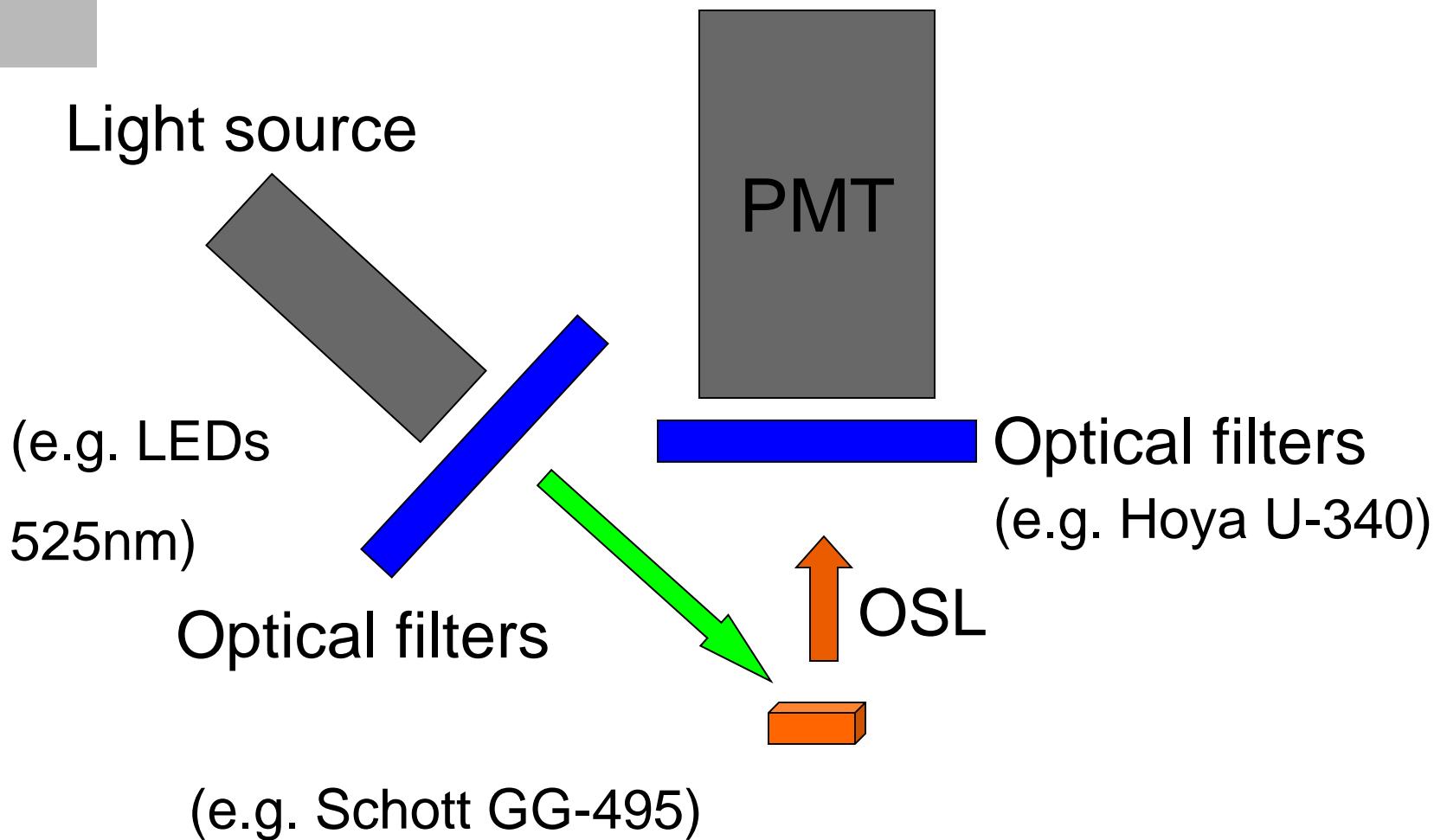


Time-temperature profiles (TTP)

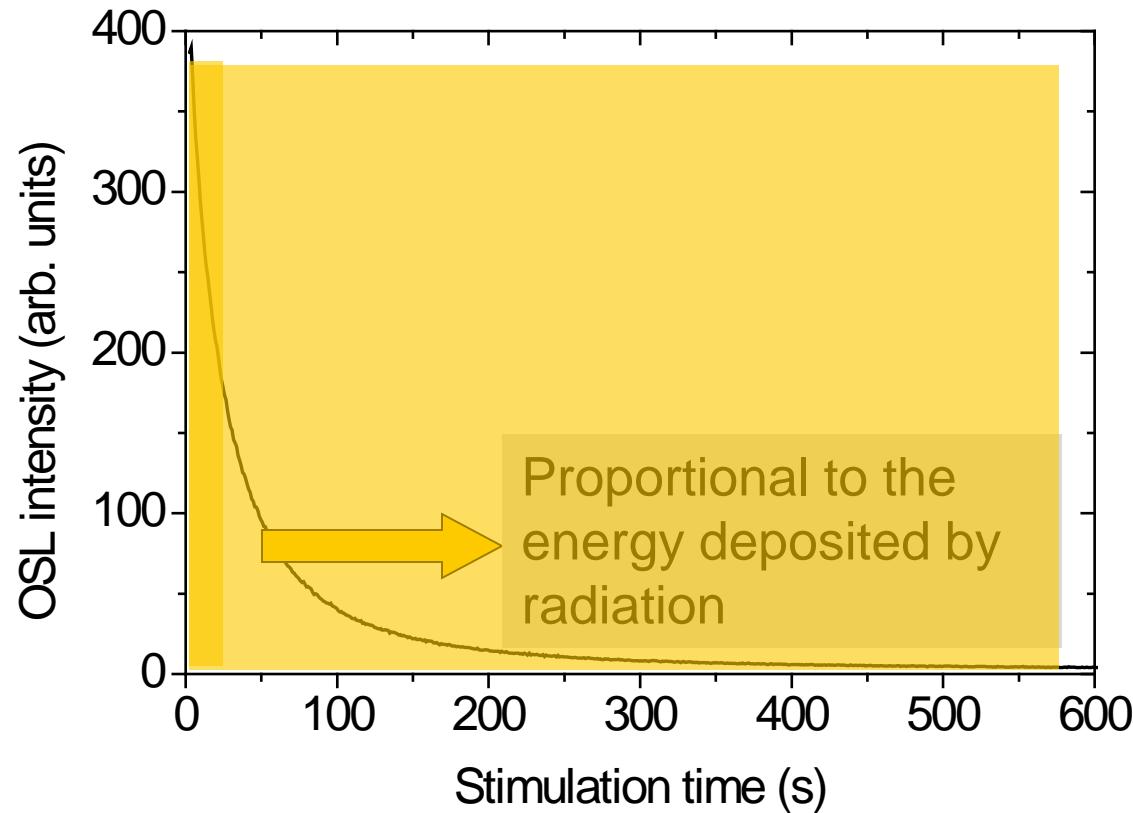


Examples of TL systems

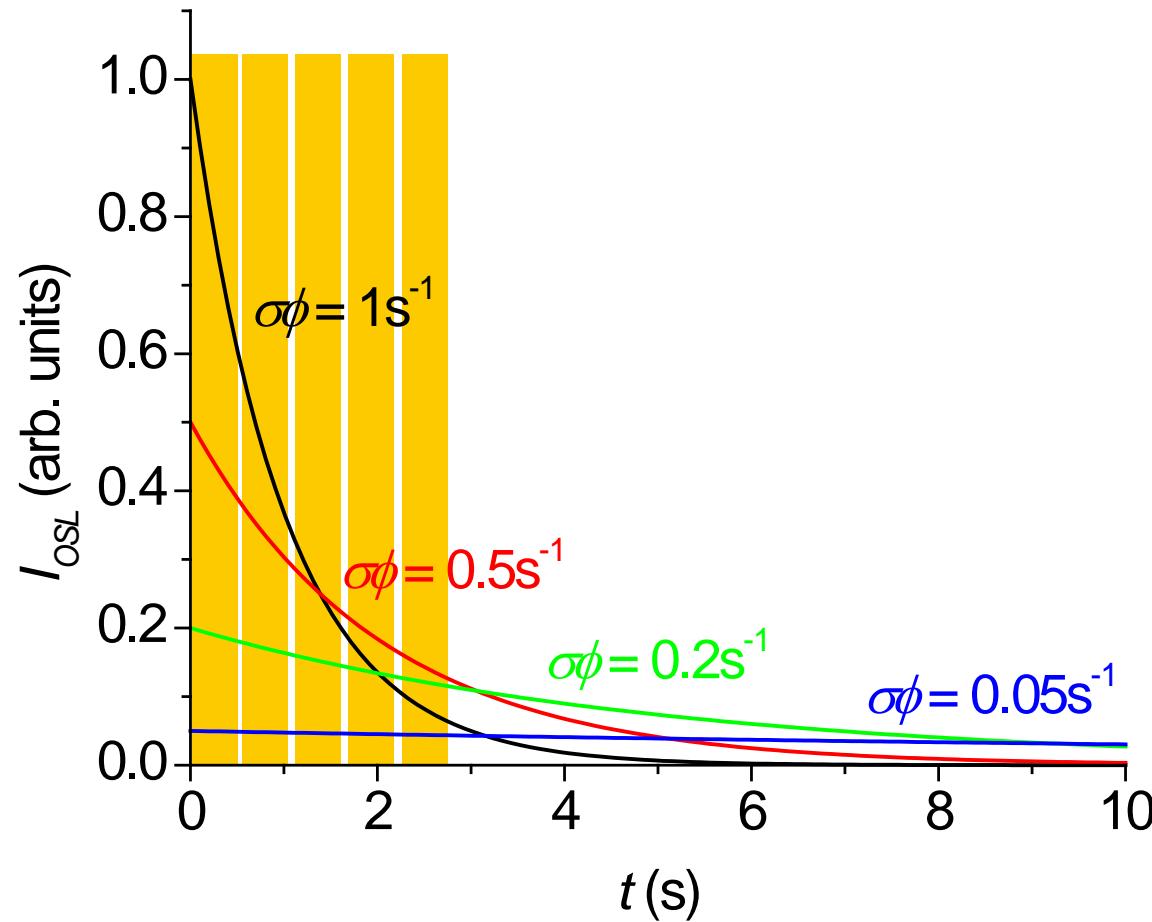




OSL readout



OSL dependence with stimulation intensity



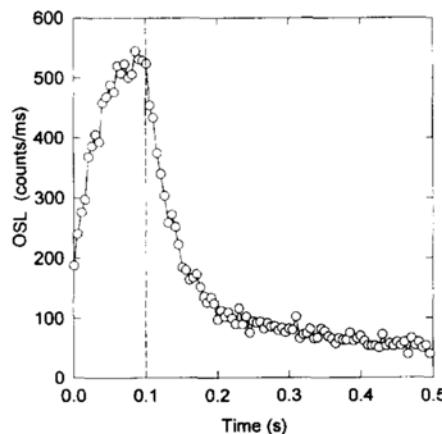
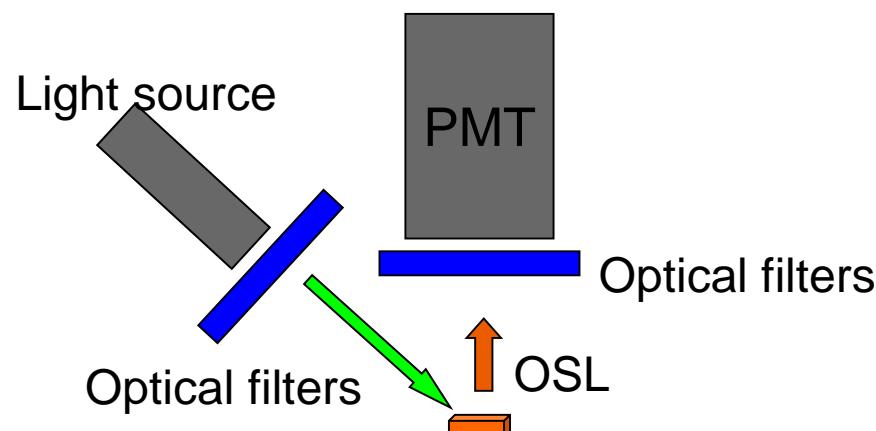


Fig. 2. Time-resolved OSL from $\alpha\text{-Al}_2\text{O}_3:\text{C}$ following irradiation at room temperature with a dose of 0.04 Gy $^{90}\text{Sr}/^{90}\text{Y}$. A single laser pulse (150 mW, 100 ms wide) was used to excite the OSL. The measurement temperature was 25°C.

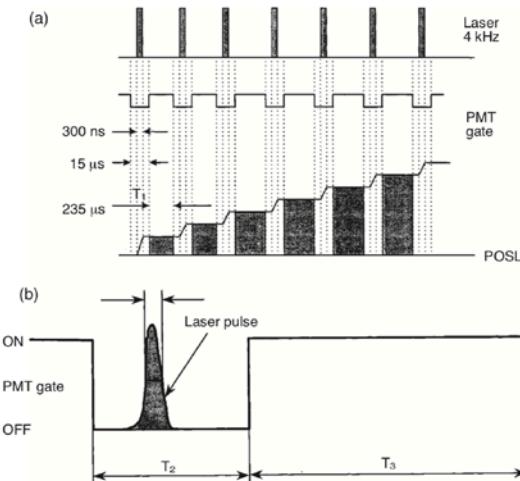
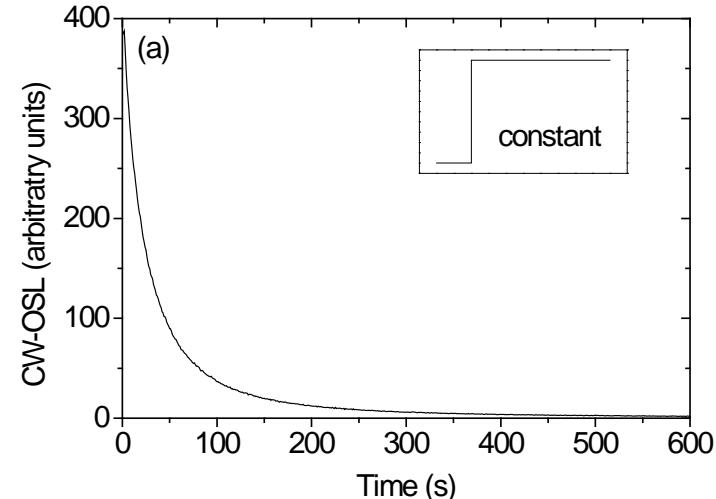
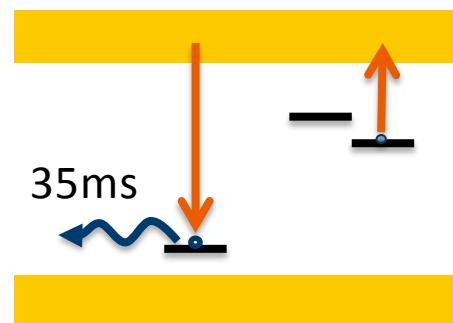


Figure 1. (a) Schematic of the timing diagram to synchronise the POSL measurement with the laser pulses. (b) Detail indicating the actual laser pulse and the PMT gate, with the pulse width T_1 , the 'dead time' T_2 , and the 'acquisition time' T_3 .

Akselrod & McKeever. **A radiation dosimetry method using pulsed optically stimulated luminescence.** Radiat. Prot. Dosim. 81, 167-176 (1999).

Example of Al₂O₃:C OSL system

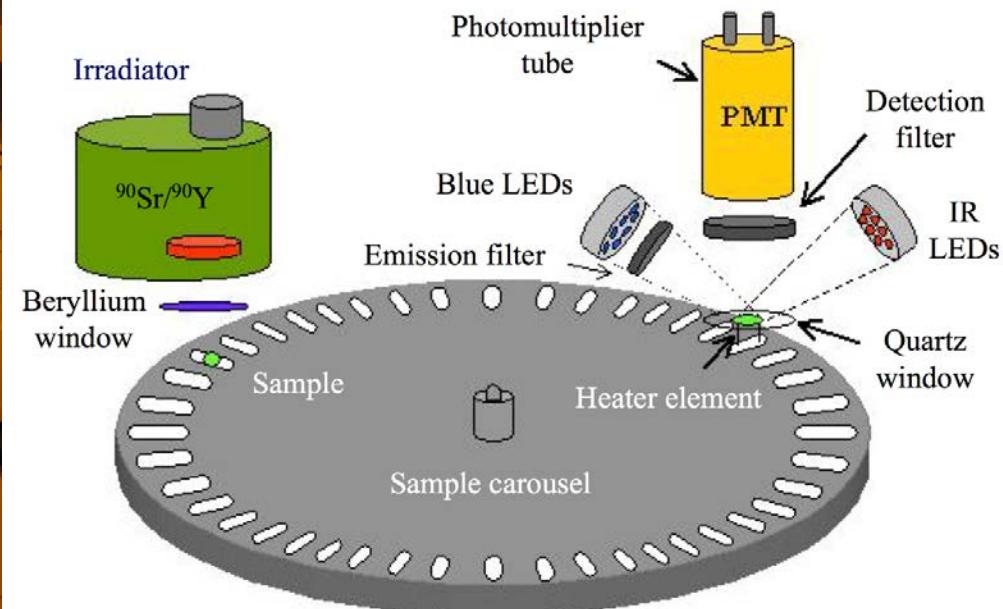
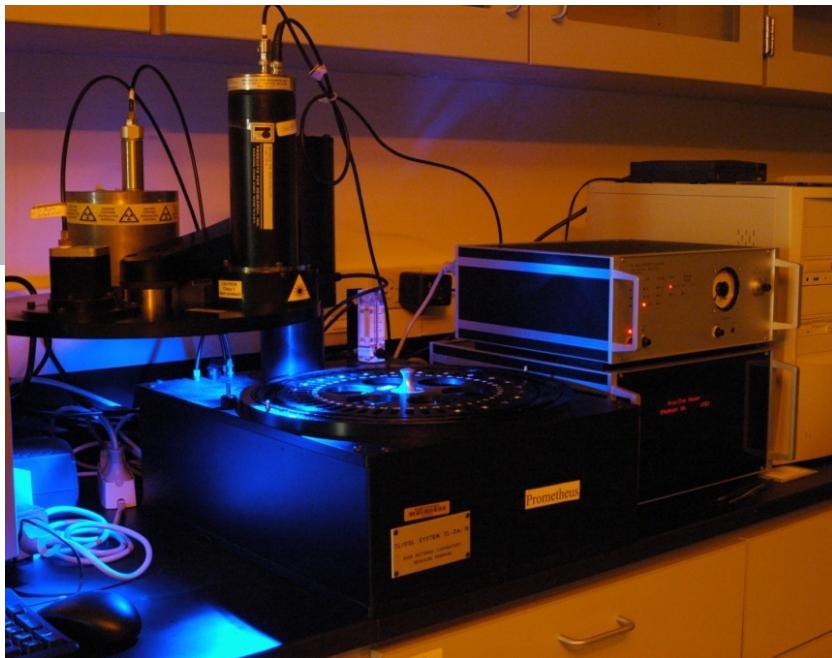


Courtesy: Landauer Inc.

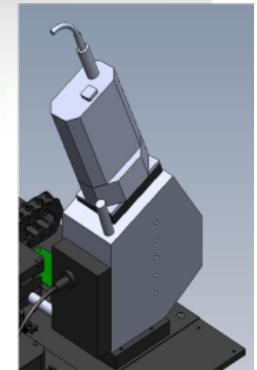
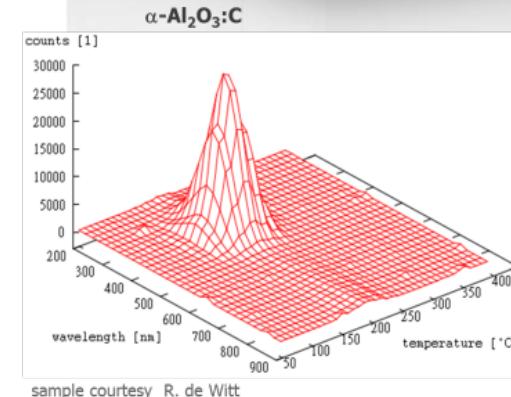
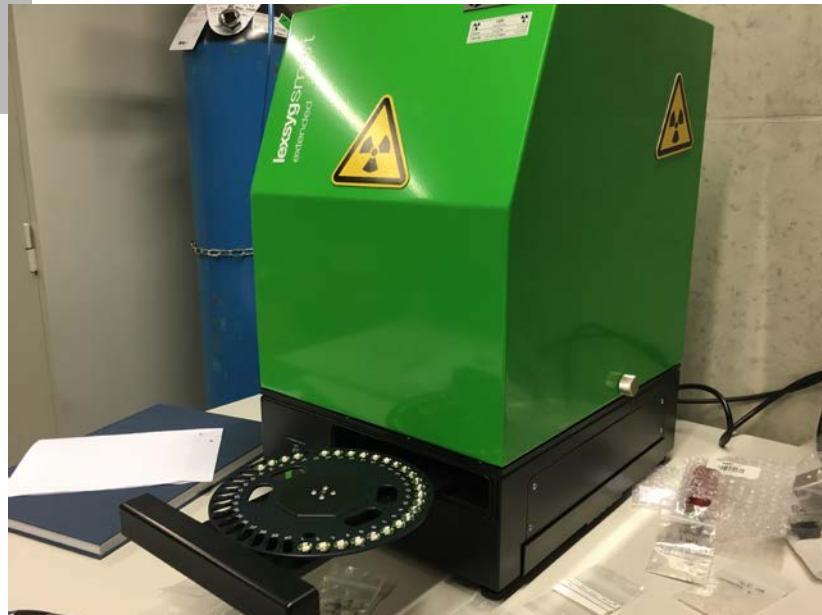
Example of BeO OSL system



Automated TL/OSL readers



Automated TL/OSL readers



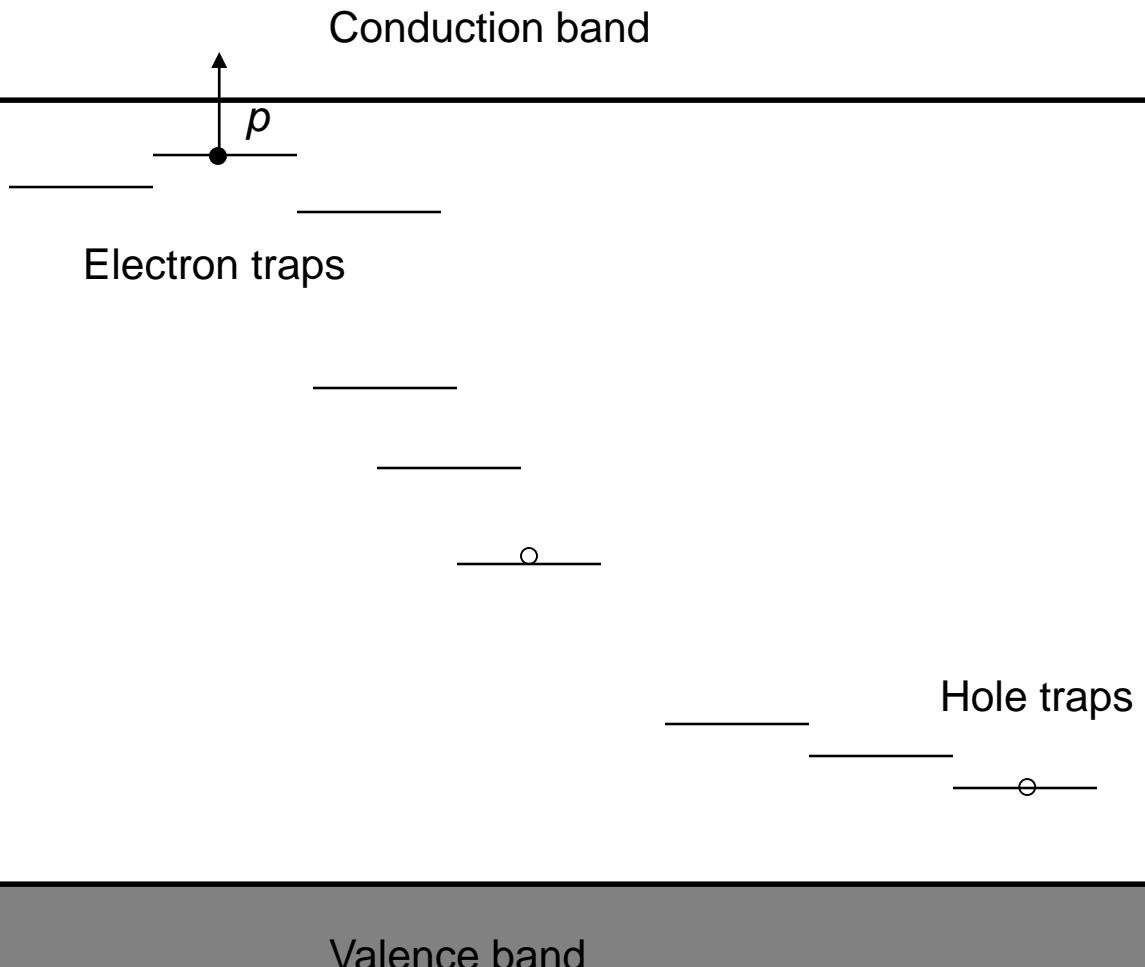
What are the main characteristics?



Important characteristics

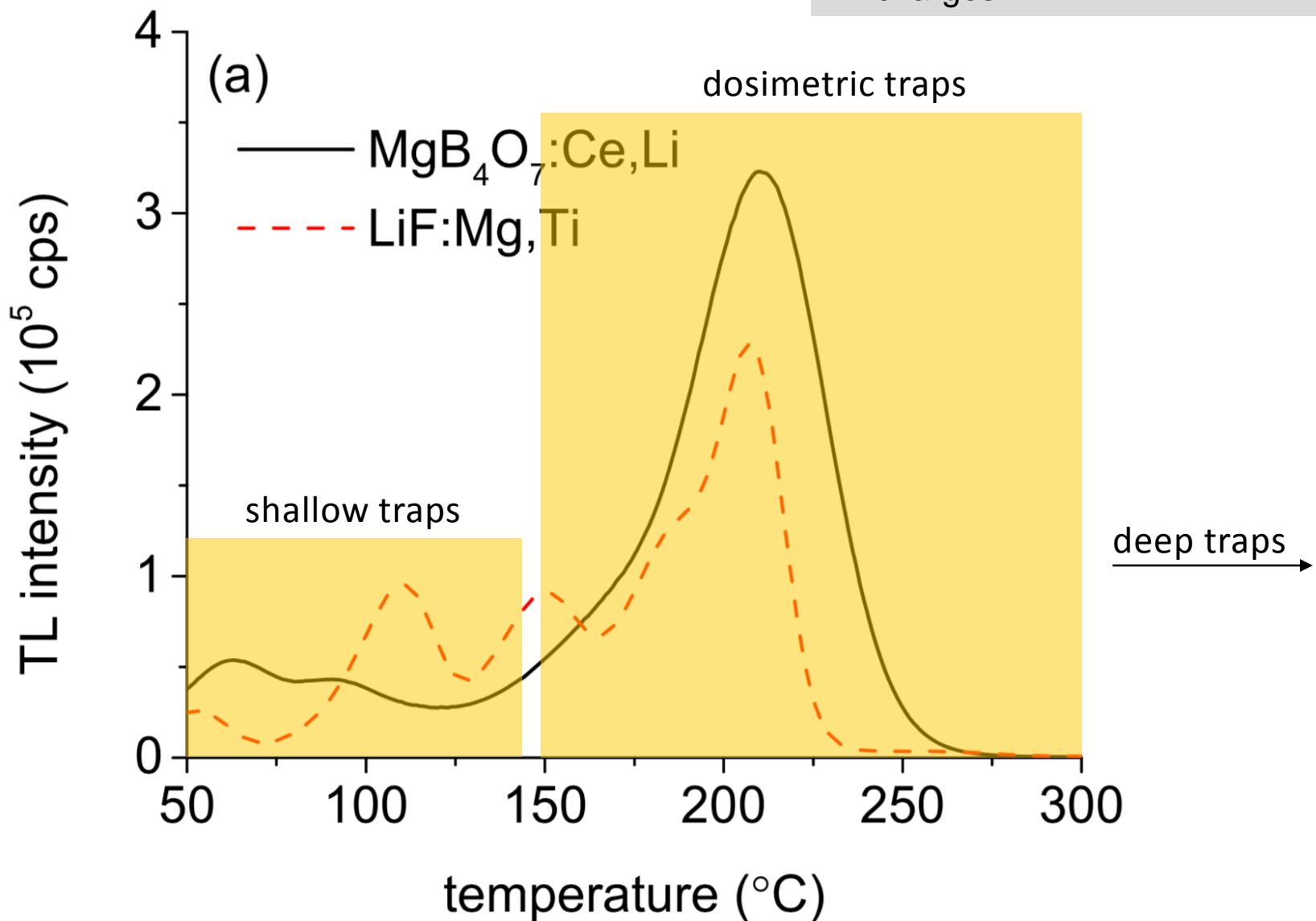
- TL curve (thermal stability)
- OSL stimulation spectrum (optical stability)
- Emission spectrum (recombination/luminescence centers)
- Luminescence lifetime (CW-OSL vs. POSL)
- Mathematical models

Detrapping process

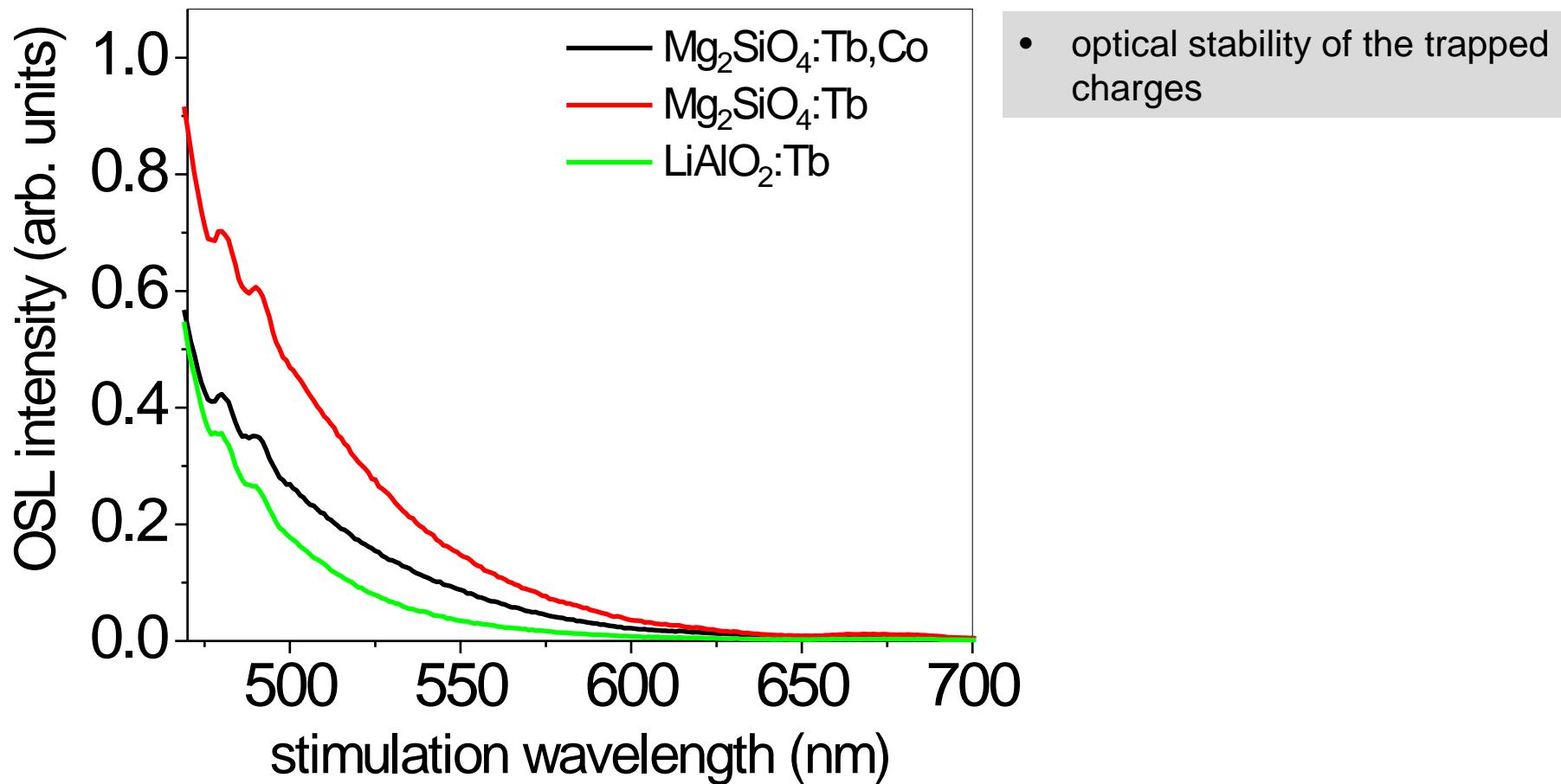


TL curves

- thermal stability of trapped charges

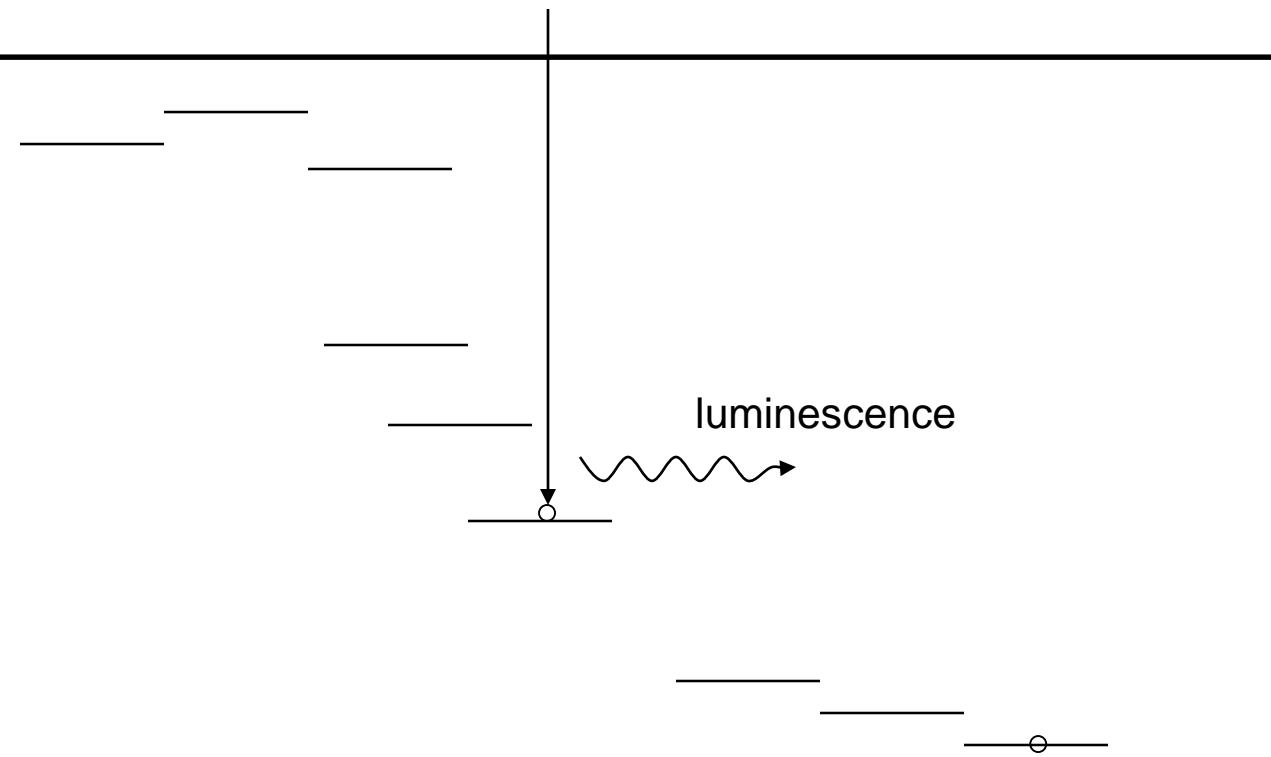


OSL stimulation spectrum

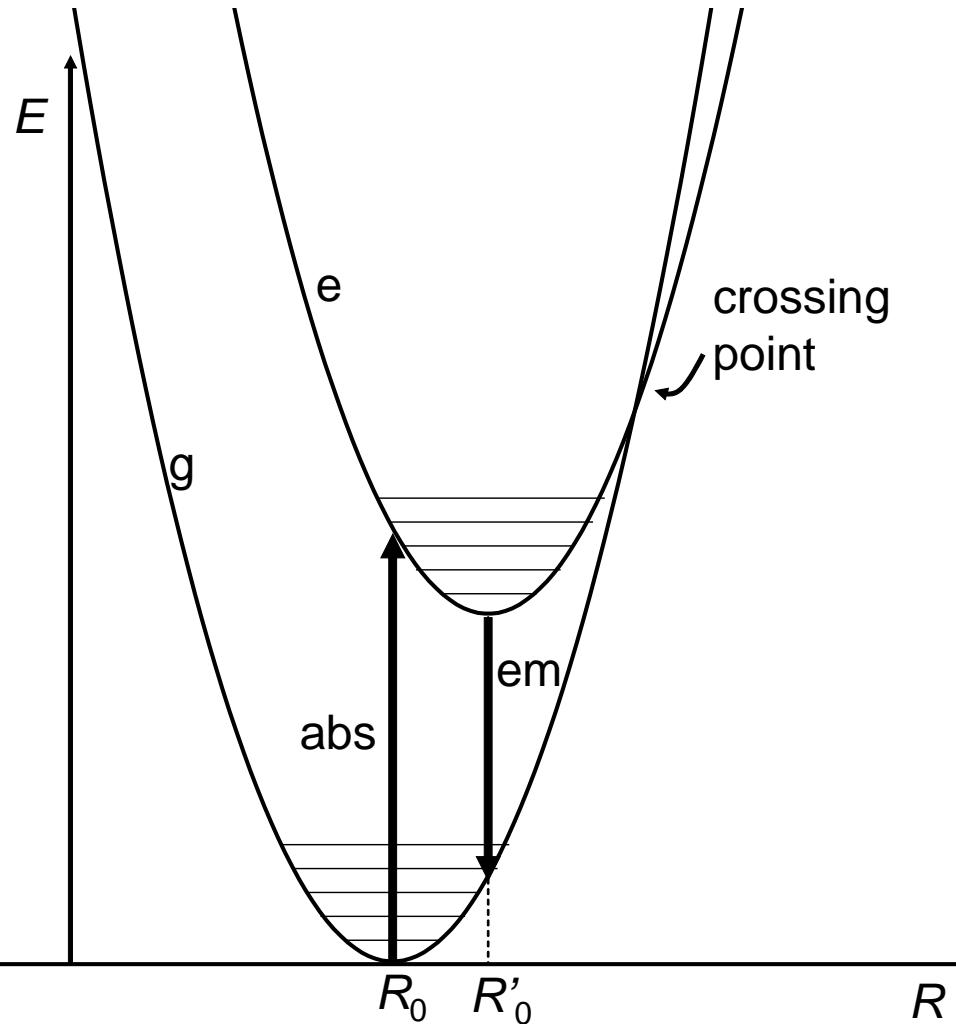


- optical stability of the trapped charges

Recombination process



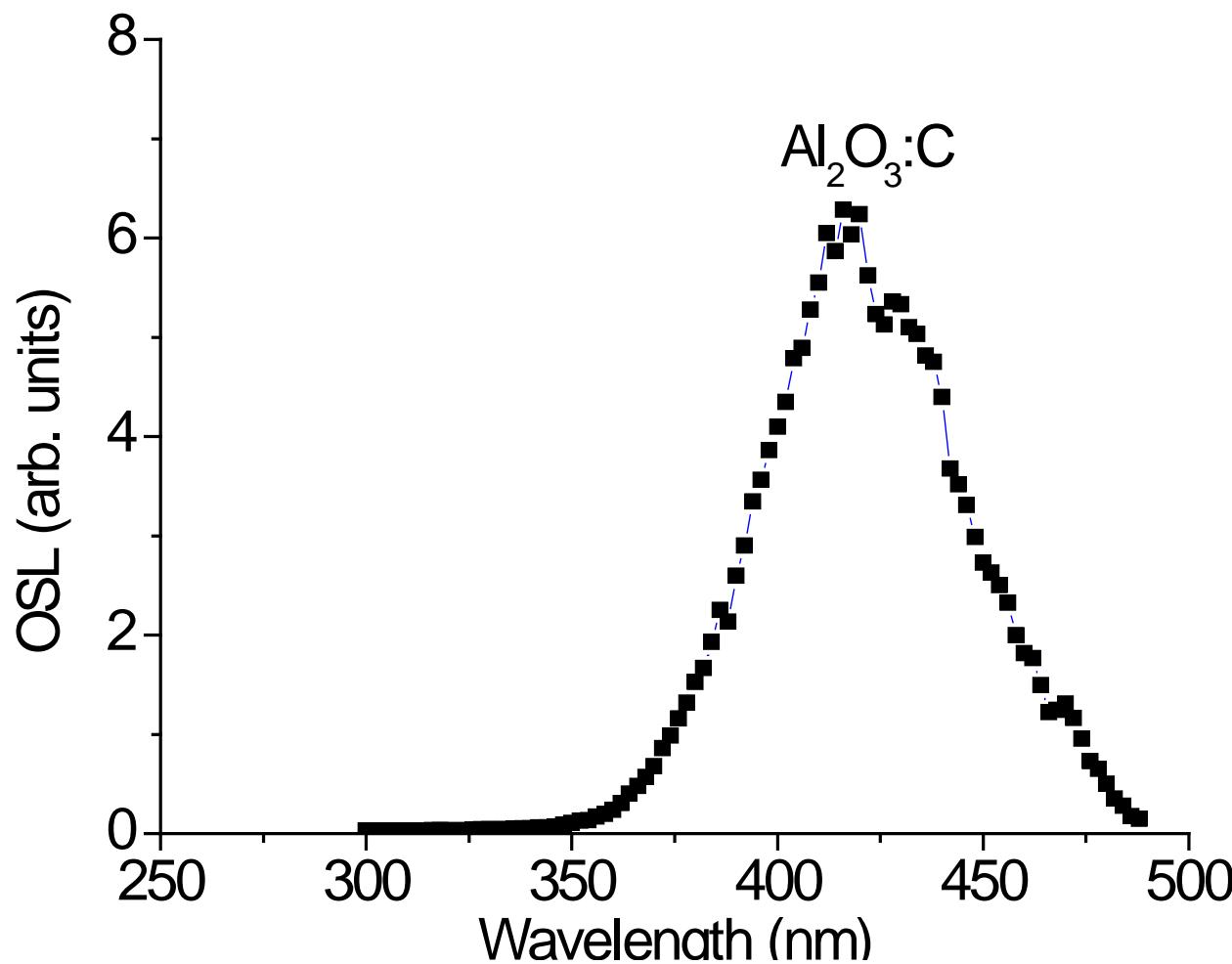
Luminescence



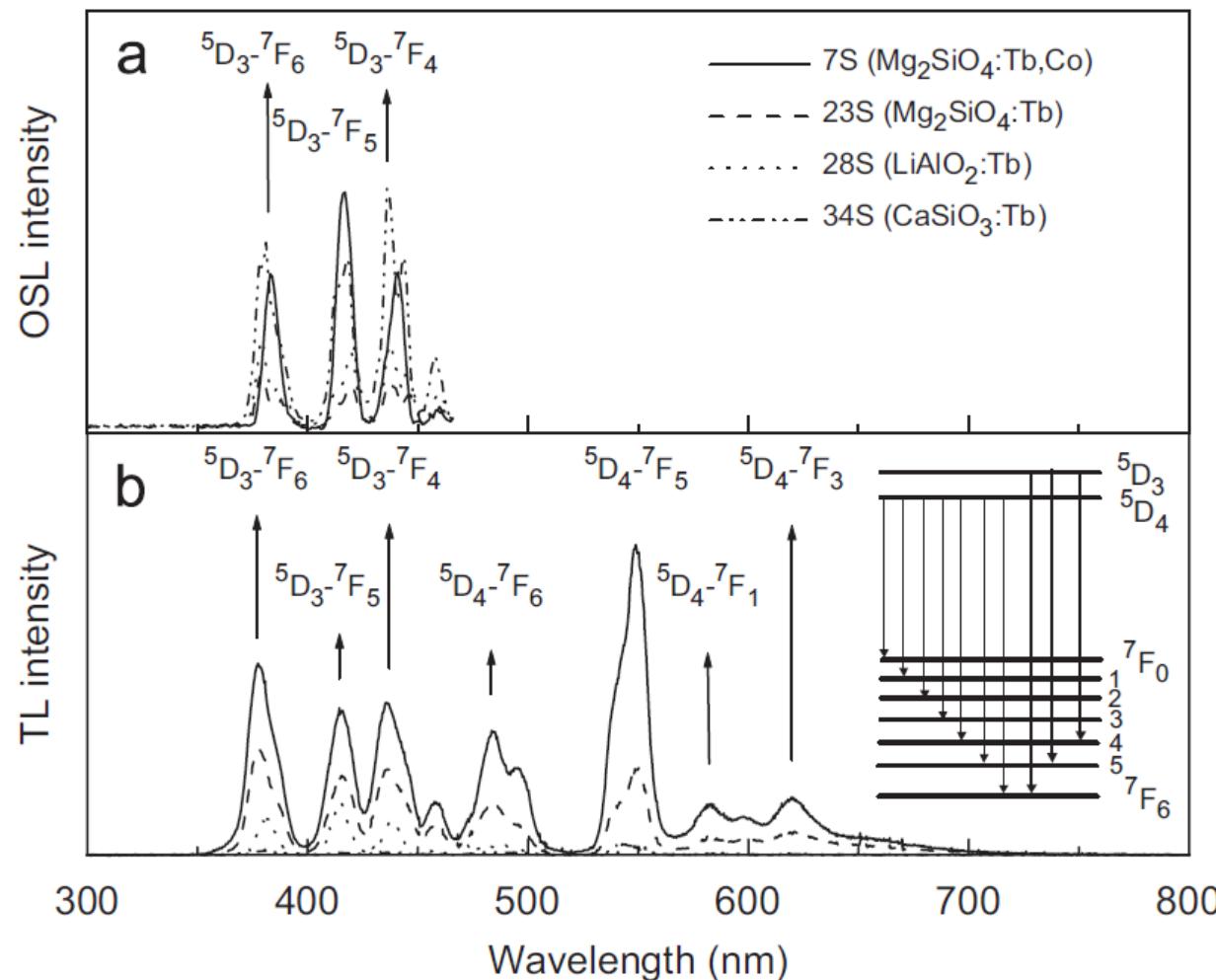
Note that relationship between energy levels determines:

- Width of the emission band
- Stokes-shift
- Relationship between radiative and non-radiative processes

OSL emission spectrum

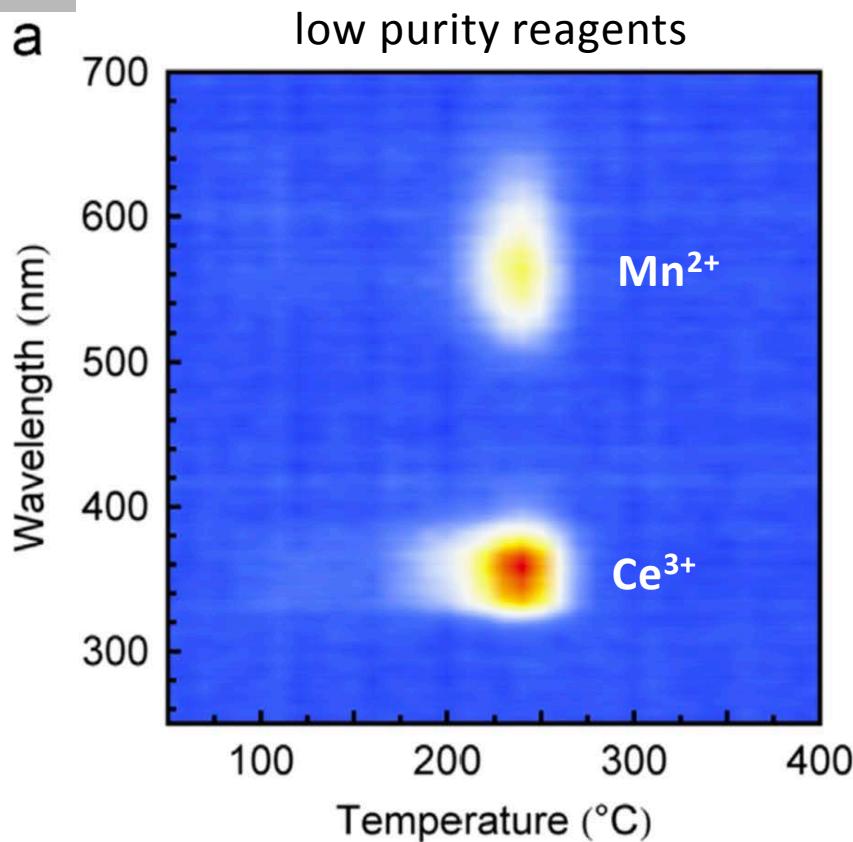
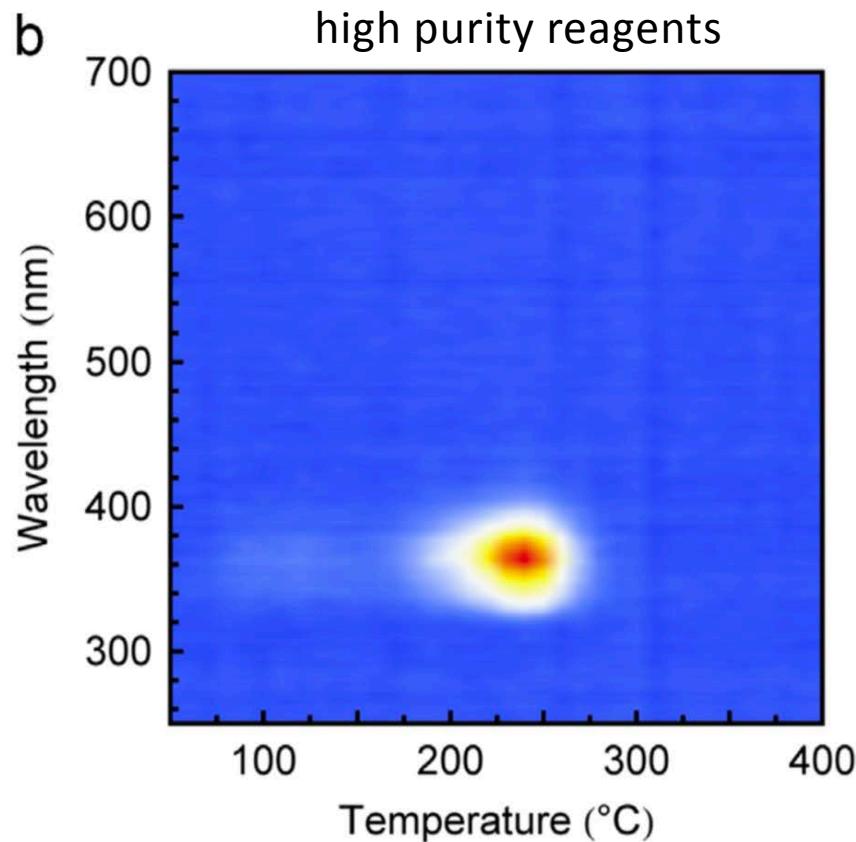


TL and OSL emission spectrum



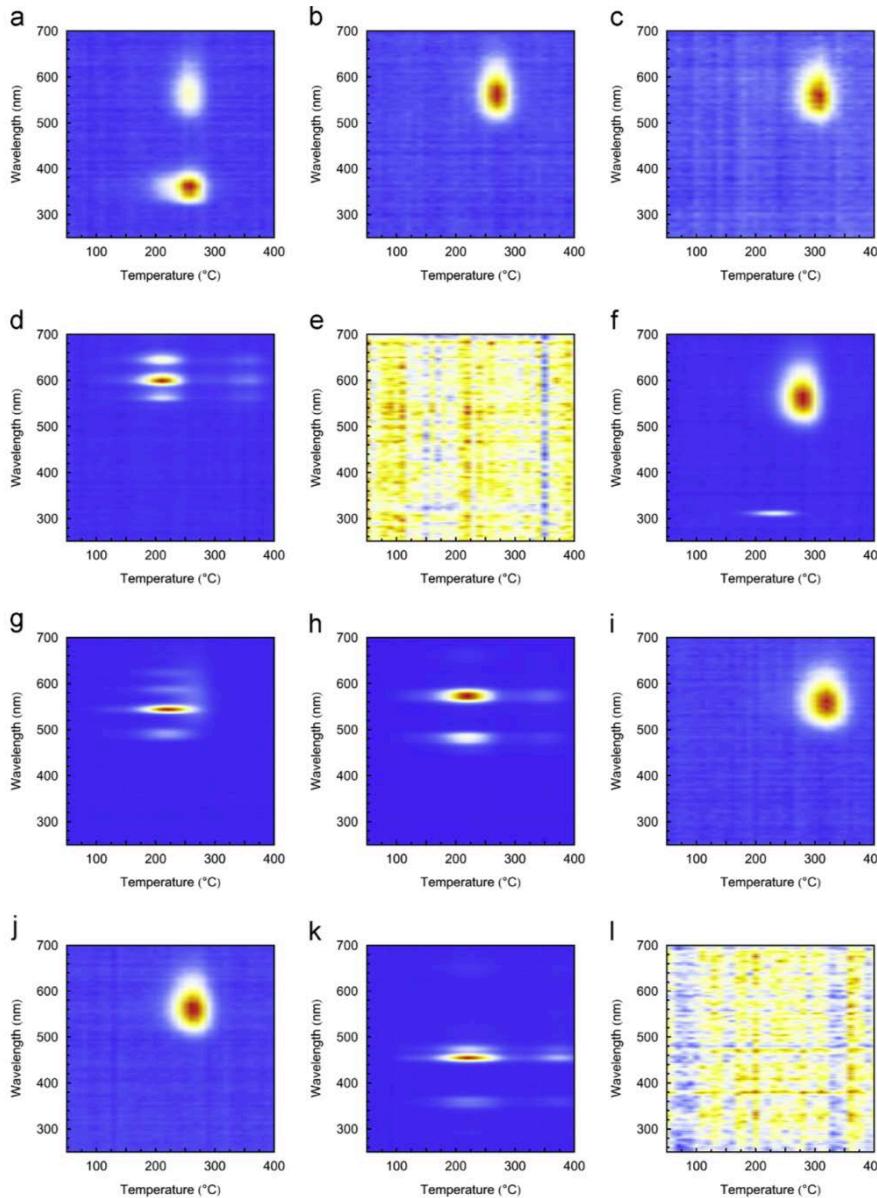
Mittani, J.C., Prokić, M., Yukihara, E.G., 2008. Optically stimulated luminescence and thermoluminescence of terbium-activated silicates and aluminates. Radiat. Meas. 43, 323-326.

MgB₄O₇:Ce,Li

a**b**

Yukihara, E.G., Milliken, E.D., Doull, B.A., 2014. Thermally stimulated and recombination processes in MgB₄O₇ investigated by systematic lanthanide doping. J. Lumin. 154, 251-259.

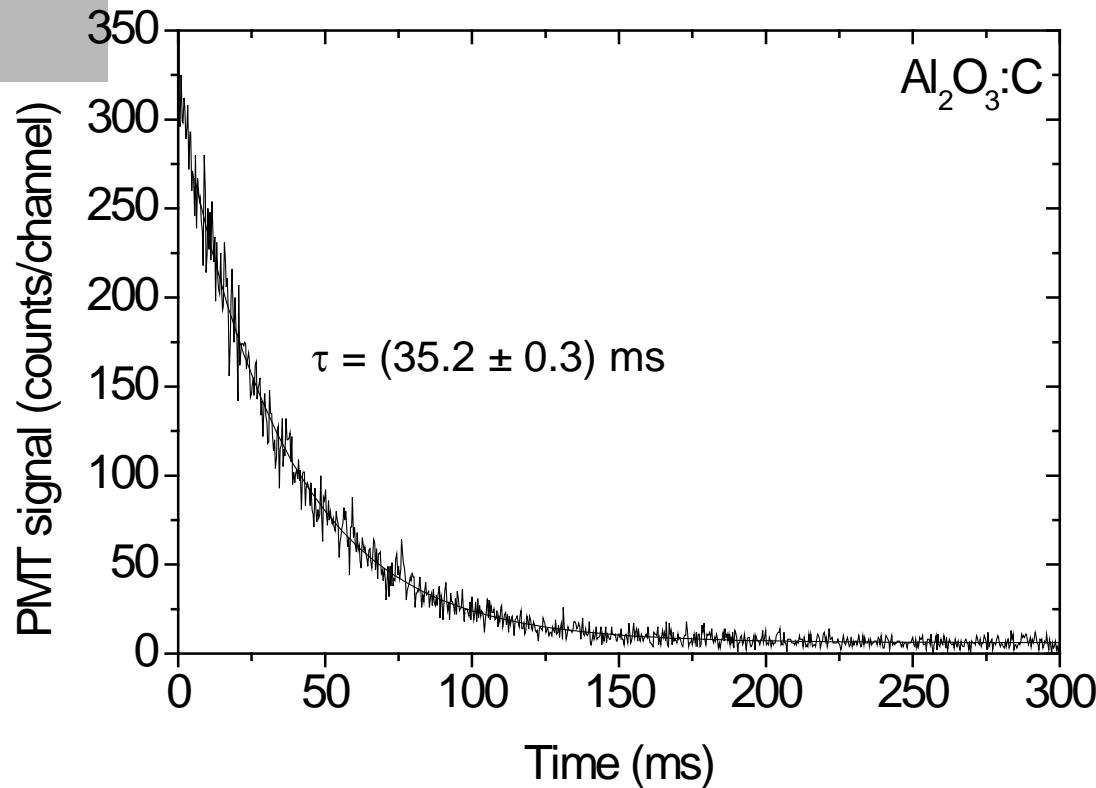
Systematic characterization of TL emission



- Detection window
- Underlying recombination processes

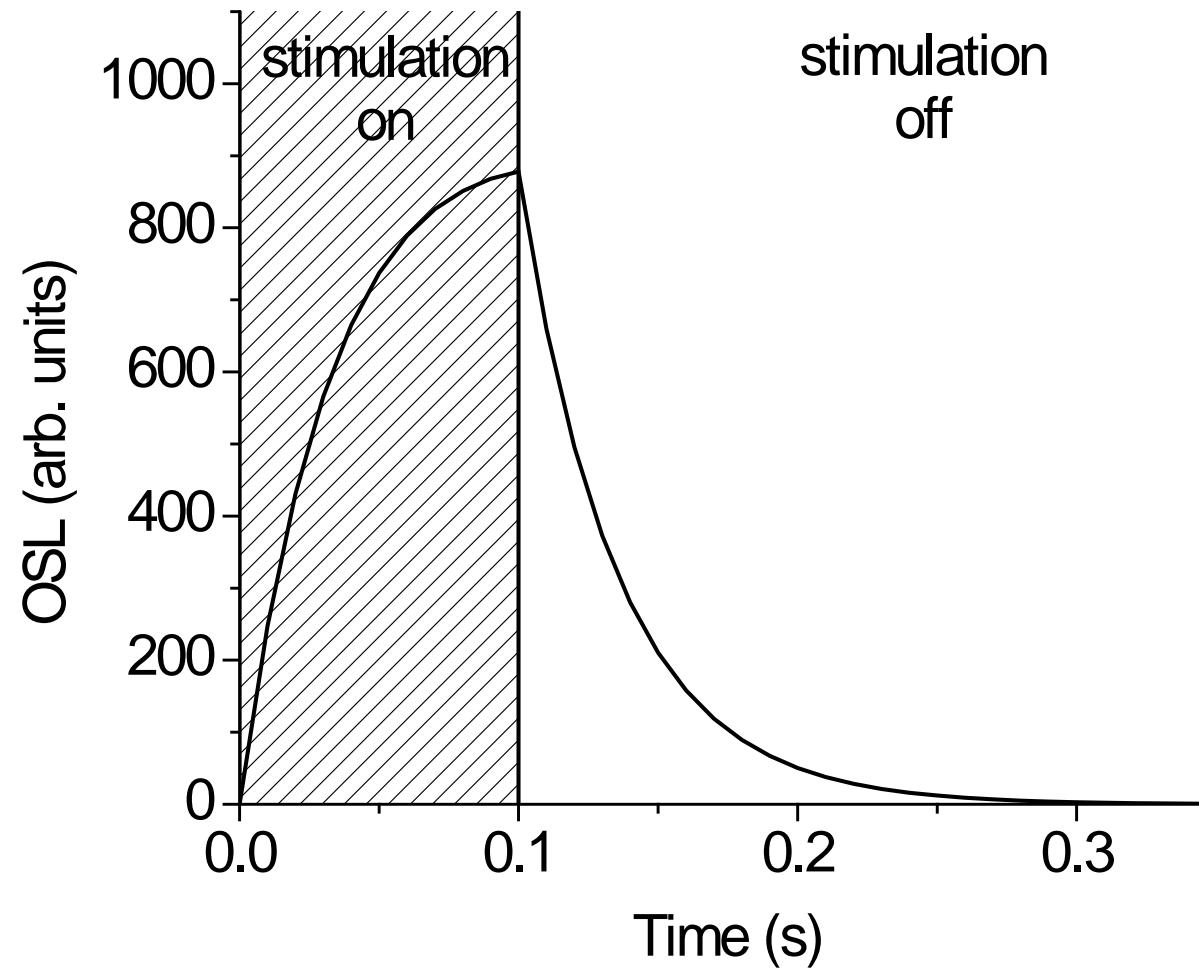
Yukihara, E.G., Milliken, E.D., Doull, B.A., 2014. Thermally stimulated and recombination processes in MgB₄O₇ investigated by systematic lanthanide doping. *J. Lumin.* 154, 251-259.

Luminescence lifetime

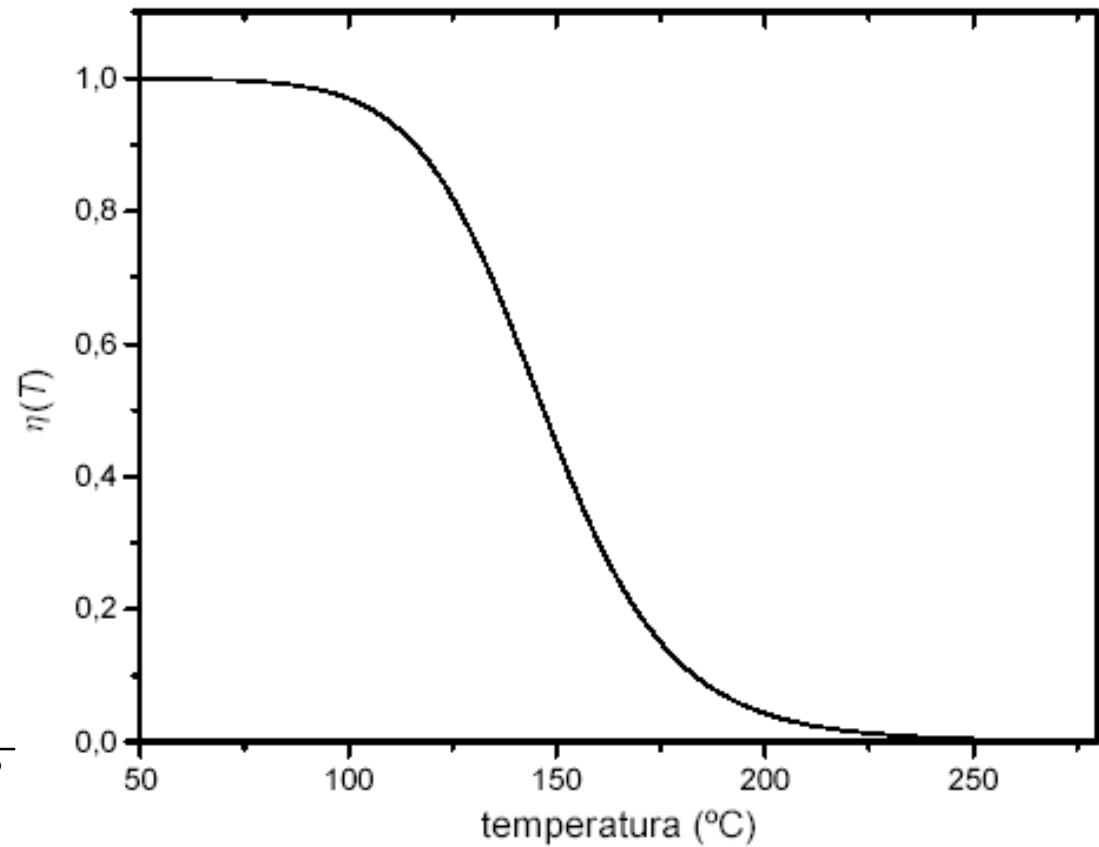
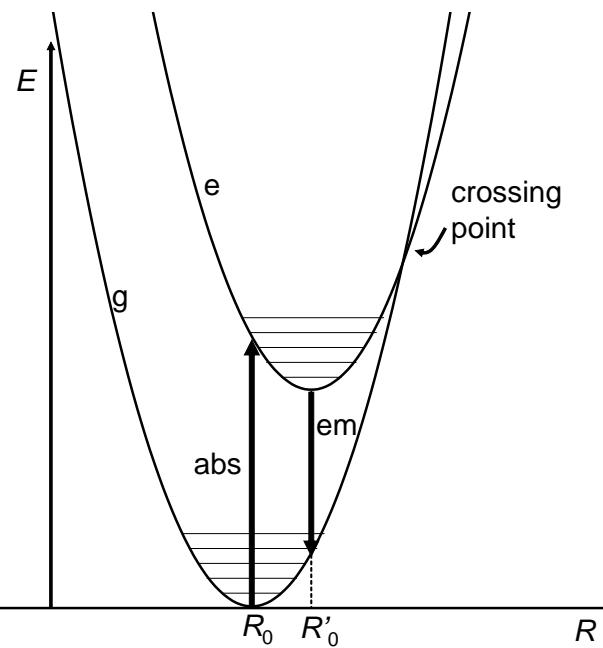


$$N_e(t) = N_e(0) e^{-\frac{t}{\tau_R}}$$

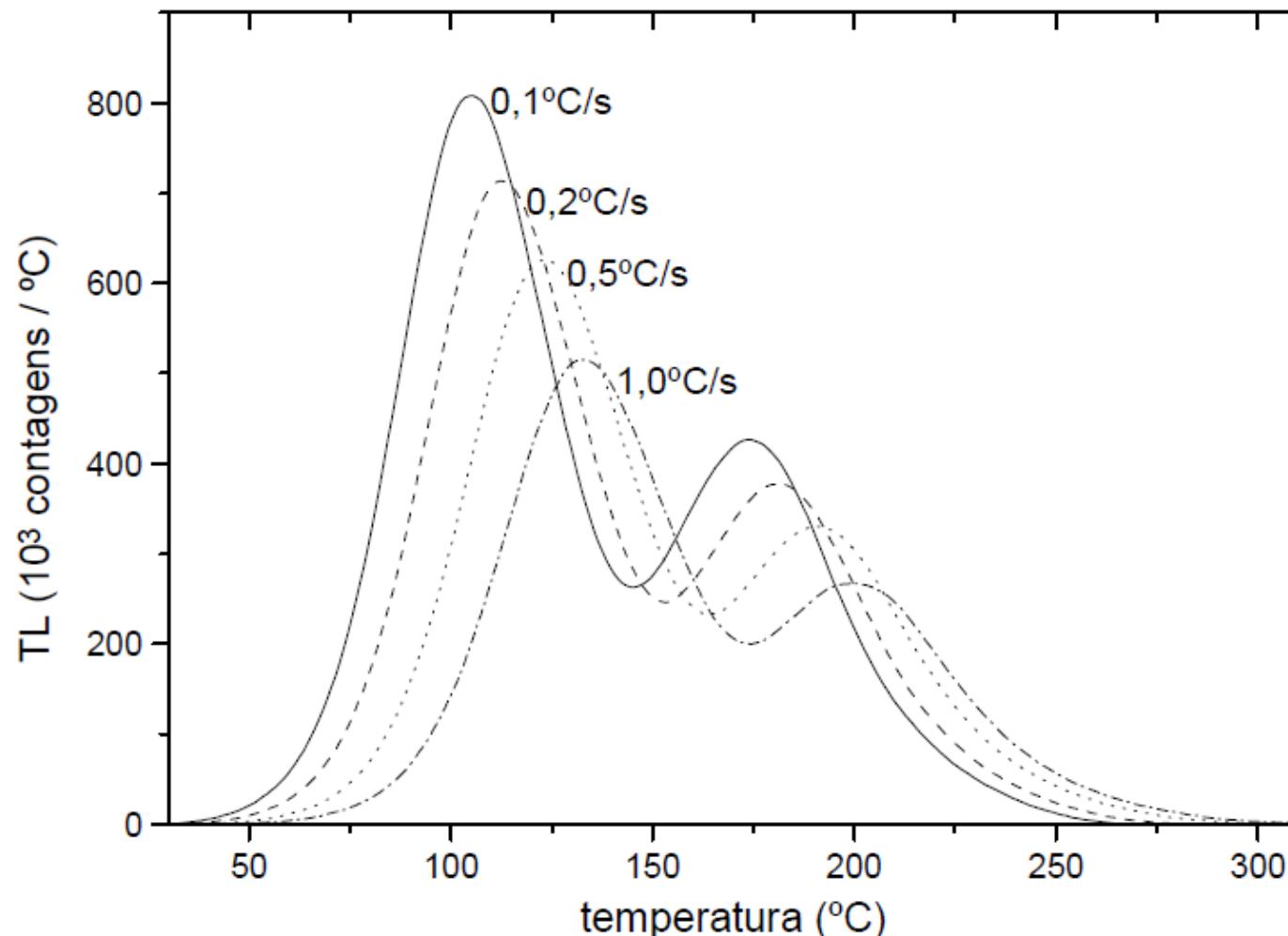
Luminescence lifetime and POSL



Thermal quenching

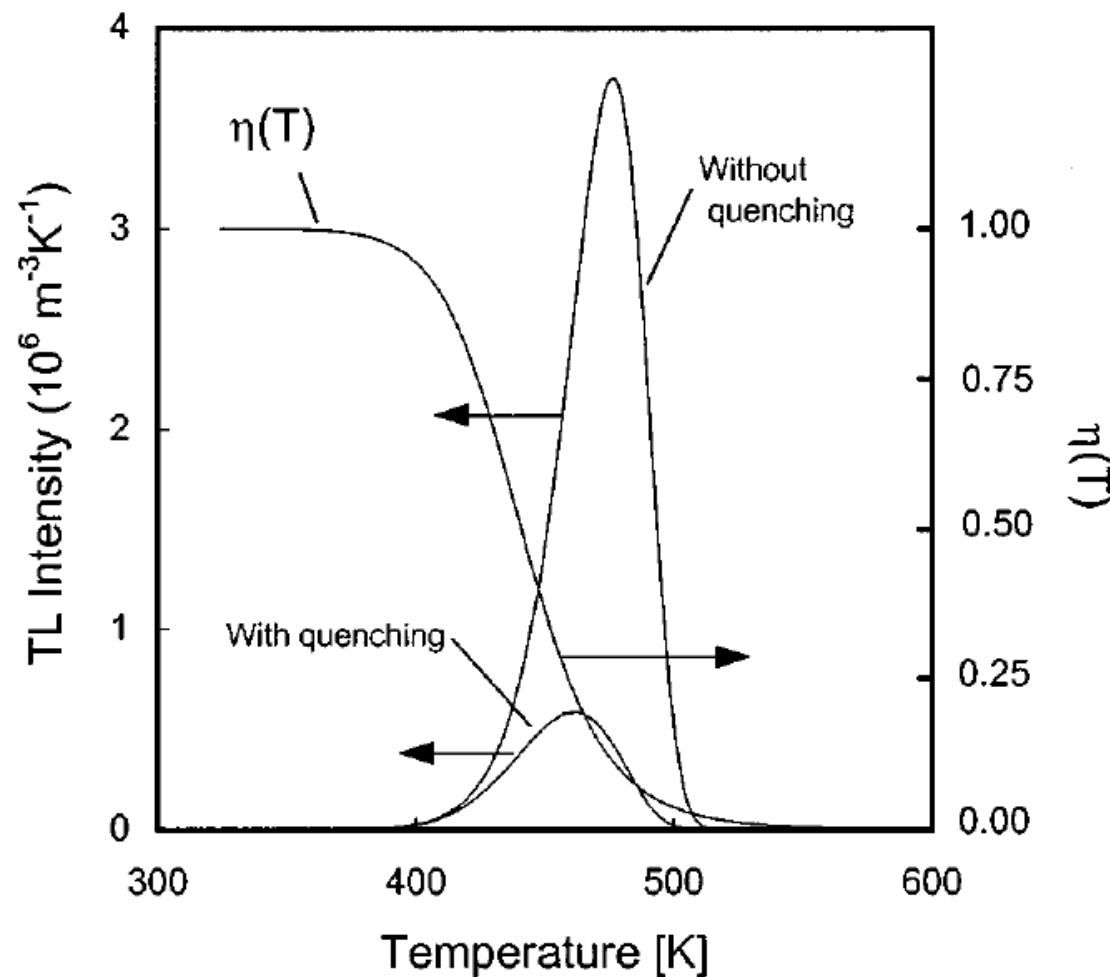


Thermal quenching



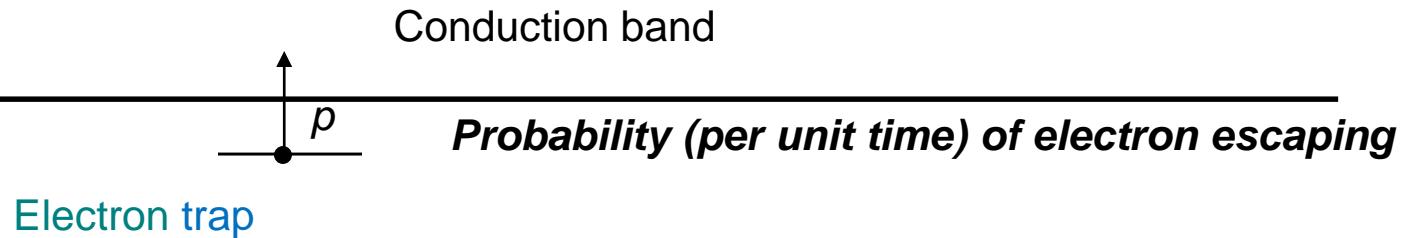
Yukihara, E.G., 2001. Desvendando a cor e a termoluminescência do topázio. Ph. D. Thesis, Universidade de São Paulo.

Thermal quenching



Akselrod, M.S., Agersnap Larsen, N., Whitley, V.H. and McKeever, S.W.S., 1998. Thermal quenching of F-center luminescence in $\text{Al}_2\text{O}_3:\text{C}$. *C. J. Appl. Phys.* 84, 3364-3373.

TL models



$$p = s e^{-\frac{E}{kT}}$$

- E = Activation energy
- s = Frequency factor
- k = Boltzmann constant
- T = Temperature

Recombination center

Hole traps

Valence band

First-order TL model (RW)

$$T(t) = T_0 + \beta t$$

$$I_{TL} \propto \left| \frac{dm}{dt} \right| = \left| \frac{dn}{dt} \right| = np$$

T_0 = temperature at $t = 0$

β = heating rate

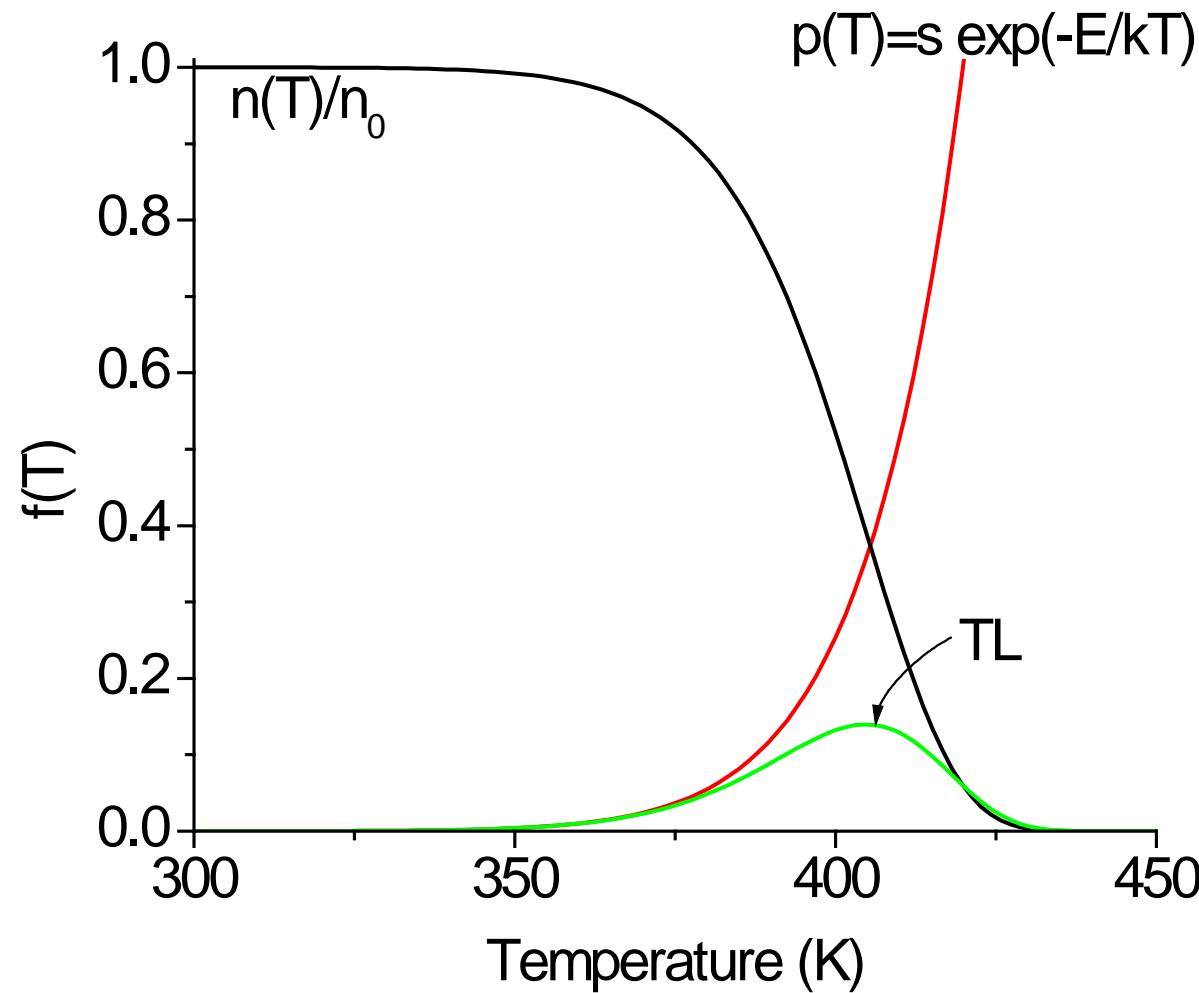
$$\beta = \frac{dT}{dt}$$

$$\frac{dn}{dt} = \frac{dn}{dT} \frac{dT}{dt} = \frac{dn}{dT} \beta = -np$$

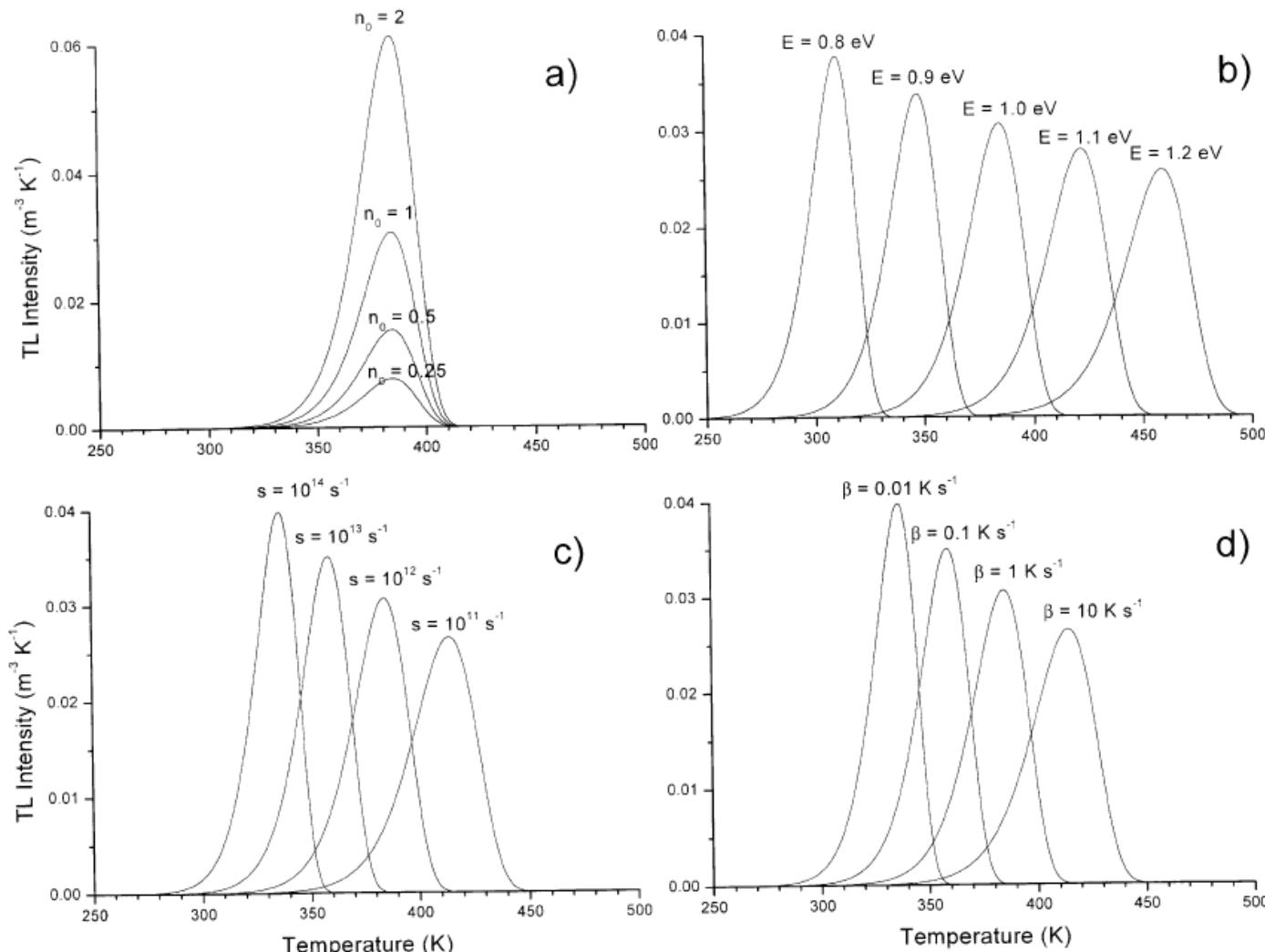
$$n = n_0 \exp \left[-\frac{s}{\beta} \int_{T_0}^T e^{-E/k\theta} d\theta \right]$$

$$I_{TL}^{RW}(T) \propto n_0 \frac{s}{\beta} \exp \left(-\frac{E}{kT} \right) \exp \left[-\frac{s}{\beta} \int_{T_0}^T \exp \left(-\frac{E}{k\theta} \right) d\theta \right]$$

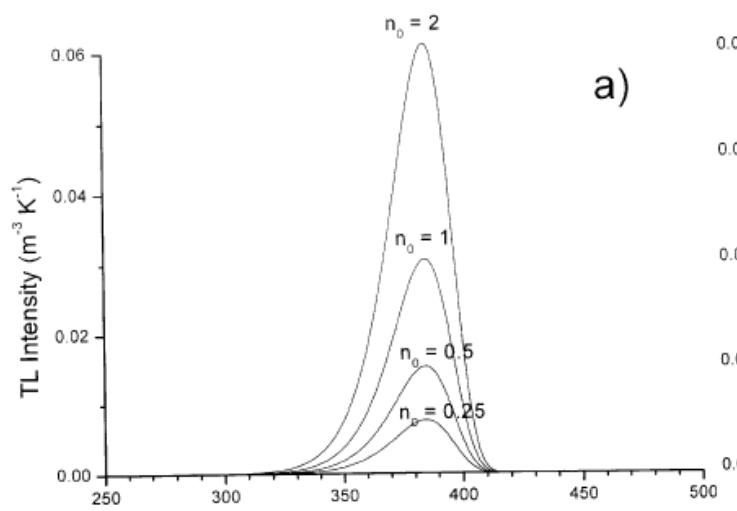
First-order TL model (RW)



First-order TL model (RW)



First-order TL model (RW)



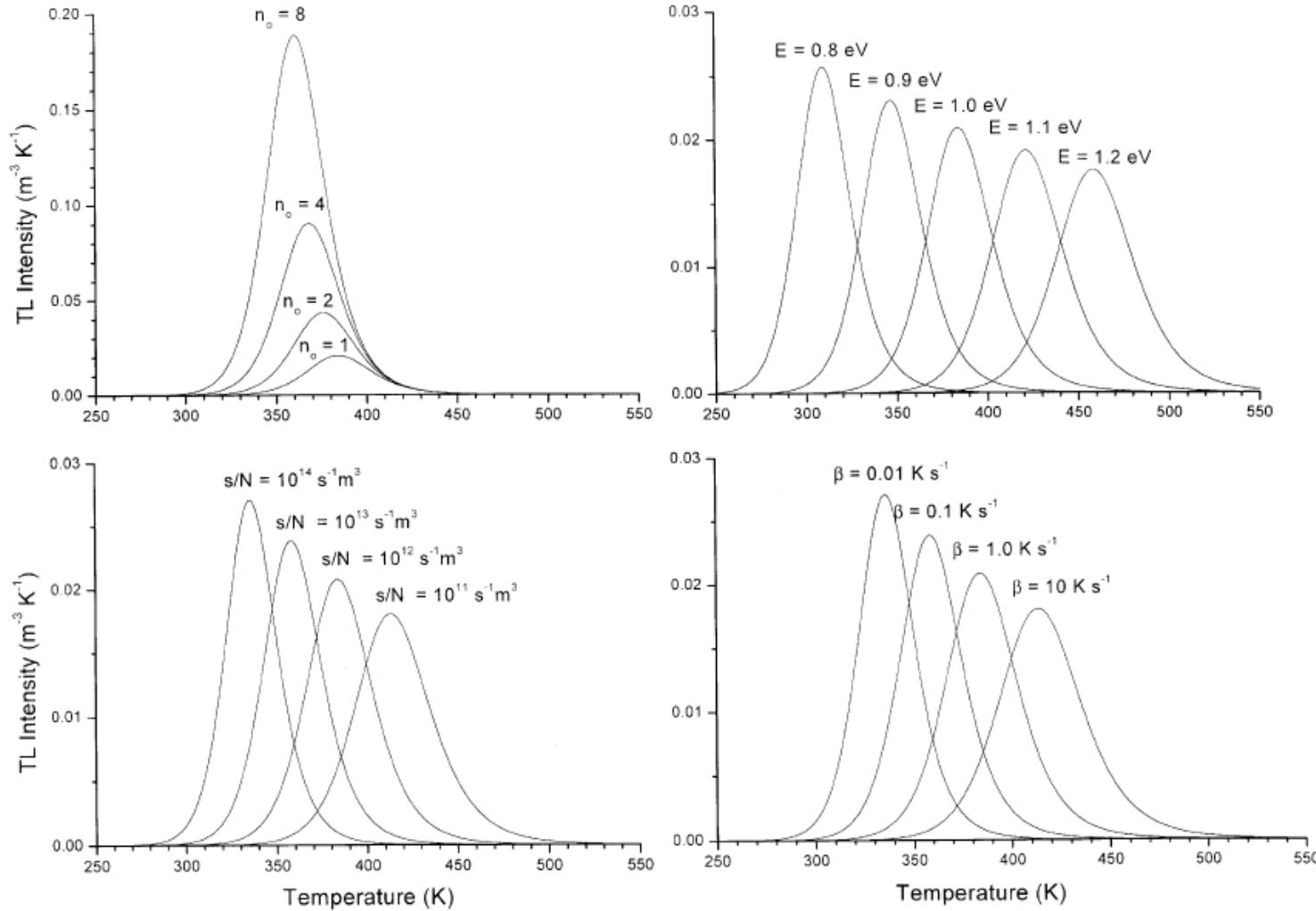
- Recombination is instantaneous
- TL peak is asymmetric
- TL peak position is not dependence on trap occupancy

Second-order TL model (GG)

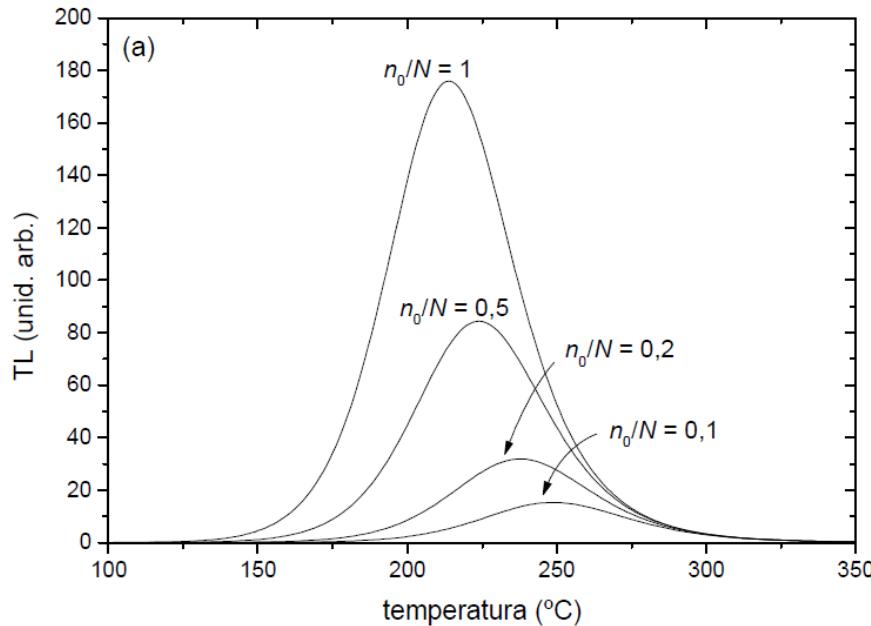
$$I_{TL}^{GG}(t) \propto \left| \frac{dm}{dt} \right| = \left| \frac{dn}{dt} \right| = \frac{n^2 p}{N}$$

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

Second-order TL model (GG)



Second-order TL model (GG)



- Recombination is delayed
- TL peak is more symmetrical than 1st order
- TL peak position depends on trap occupancy

TL models

1st order

$$I_{TL}^{RW}(t) \propto np$$

2nd order

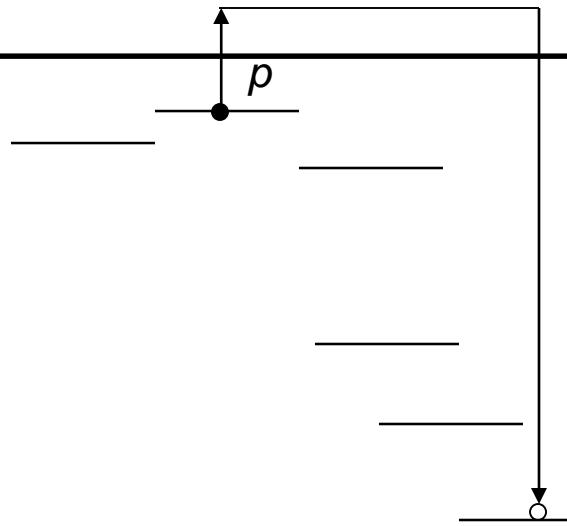
$$I_{TL}^{GG}(t) \propto \frac{n^2 p}{N}$$

GOK

$$I_{TL}^{GOK}(t) \propto \frac{n^b}{N^{b-1}} p$$

$$I_{TL}^{GOK}(t) \propto \frac{n_0^b}{N^{b-1}} s e^{-E/kT} \left[1 + (b-1) \left(\frac{n_0}{N} \right)^{b-1} \frac{1}{\beta} \int_{T_0}^T \exp\left(-\frac{E}{k\theta}\right) d\theta \right]^{-\frac{b}{b-1}}$$

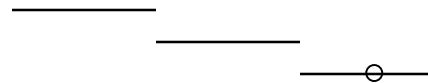
First-order OSL model



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

- σ = photoionization cross-section
- ϕ = photon flux ($\text{cm}^{-2} \text{ s}^{-1}$)
- λ = wavelength

luminescence



First-order OSL model

$n(t)$ = concentration of trapped electrons

n_0 = initial concentration of trapped electrons

$$\frac{dn}{dt} = -np$$

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

$$\frac{1}{n} dn = -p dt$$

$$\ln n - \ln n_0 = -pt$$

$$\ln\left(\frac{n}{n_0}\right) = -pt$$

$$n = n_0 e^{-pt} = n_0 e^{-\sigma\phi t}$$

First-order OSL model

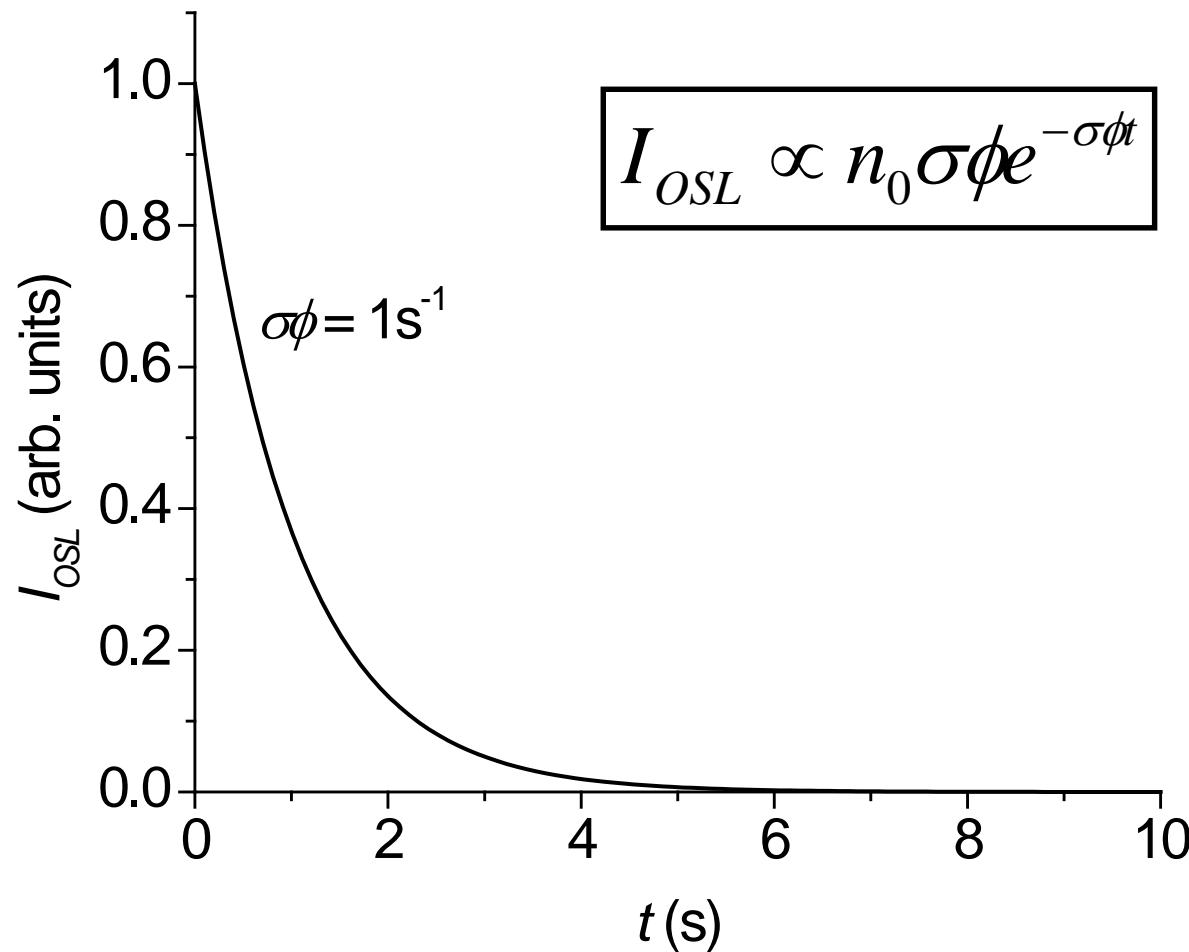
$$\frac{dn}{dt} = -np$$

$$I_{OSL} \propto \left| \frac{dn}{dt} \right| = np = (n_0 e^{-\sigma\phi t}) (\sigma\phi)$$

$$n = n_0 e^{-\sigma\phi t}$$

$$I_{OSL} \propto n_0 \sigma \phi e^{-\sigma\phi t}$$

First-order OSL model



First-order OSL model

type of defect and wavelength

$$I_{OSL} \propto n_0 \sigma \phi e^{-\sigma \phi t}$$

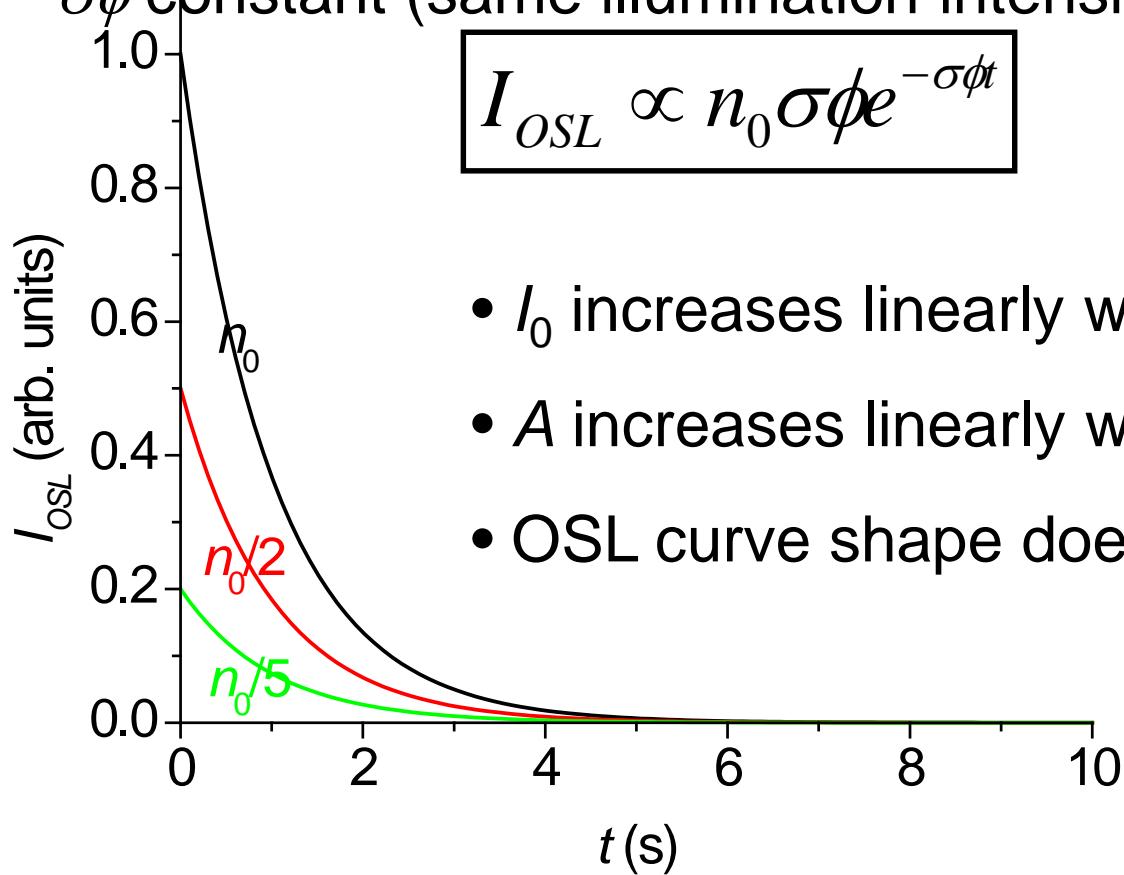
dose Illumination intensity

First-order OSL model

Dose dependence

$\sigma\phi$ constant (same illumination intensity)

$$I_{OSL} \propto n_0 \sigma\phi e^{-\sigma\phi t}$$



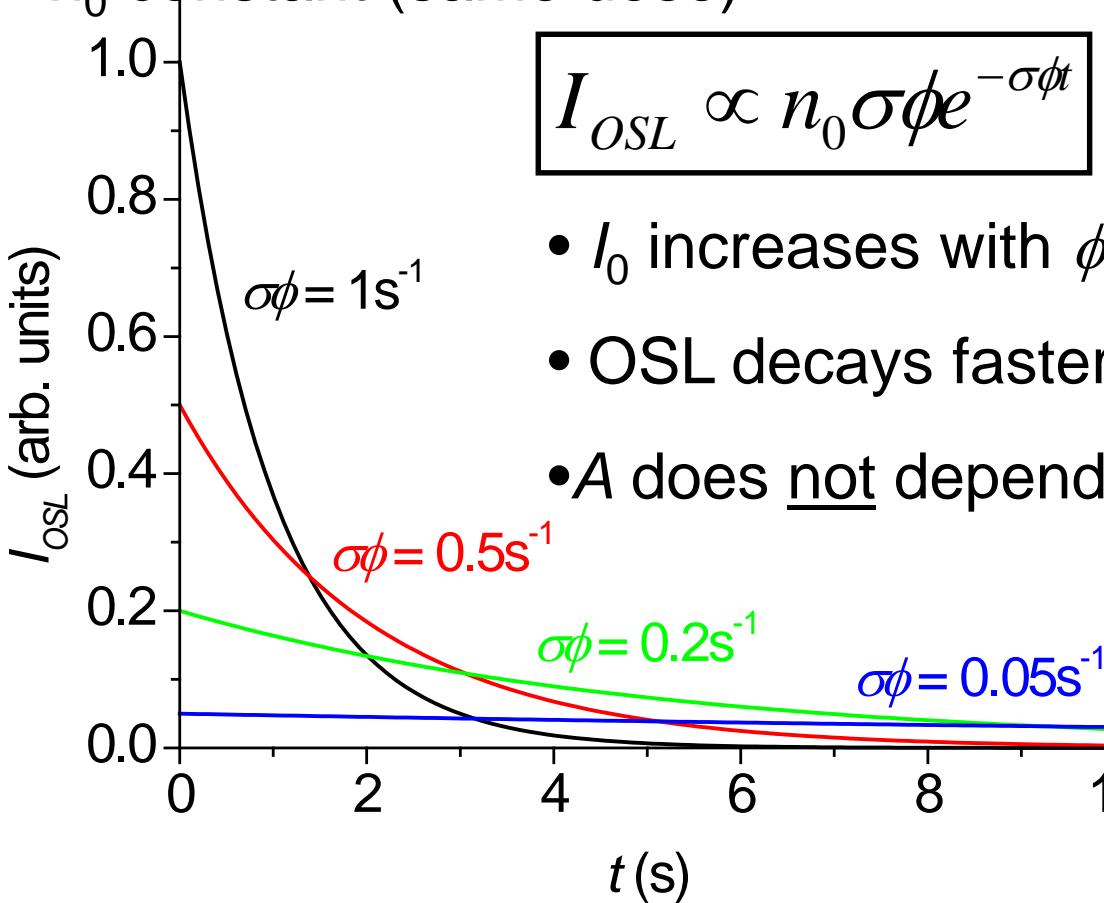
- I_0 increases linearly with dose
- A increases linearly with dose
- OSL curve shape does not depend on dose

First-order OSL model

Dependence with stimulation

n_0 constant (same dose)

$$I_{OSL} \propto n_0 \sigma \phi e^{-\sigma \phi t}$$



- I_0 increases with ϕ
- OSL decays faster with increase in ϕ
- A does not depend on ϕ

First-order OSL model

Dependence with stimulation

n_0 constant (same dose)

$$A = \int_0^{\infty} I_{OSL} dt \propto \int_0^{\infty} n_0 \sigma \phi e^{-\sigma \phi t} dt = n_0 \sigma \phi \int_0^{\infty} e^{-\sigma \phi t} dt$$

$$A \propto n_0 \sigma \phi \left[-\frac{e^{-\sigma \phi t}}{\sigma \phi} \right]_0^{\infty} = n_0 (-0 + 1)$$

$$A \propto n_0$$

Other OSL models

1st order

$$I_{OSL}^{RW}(t) \propto np$$

2nd order

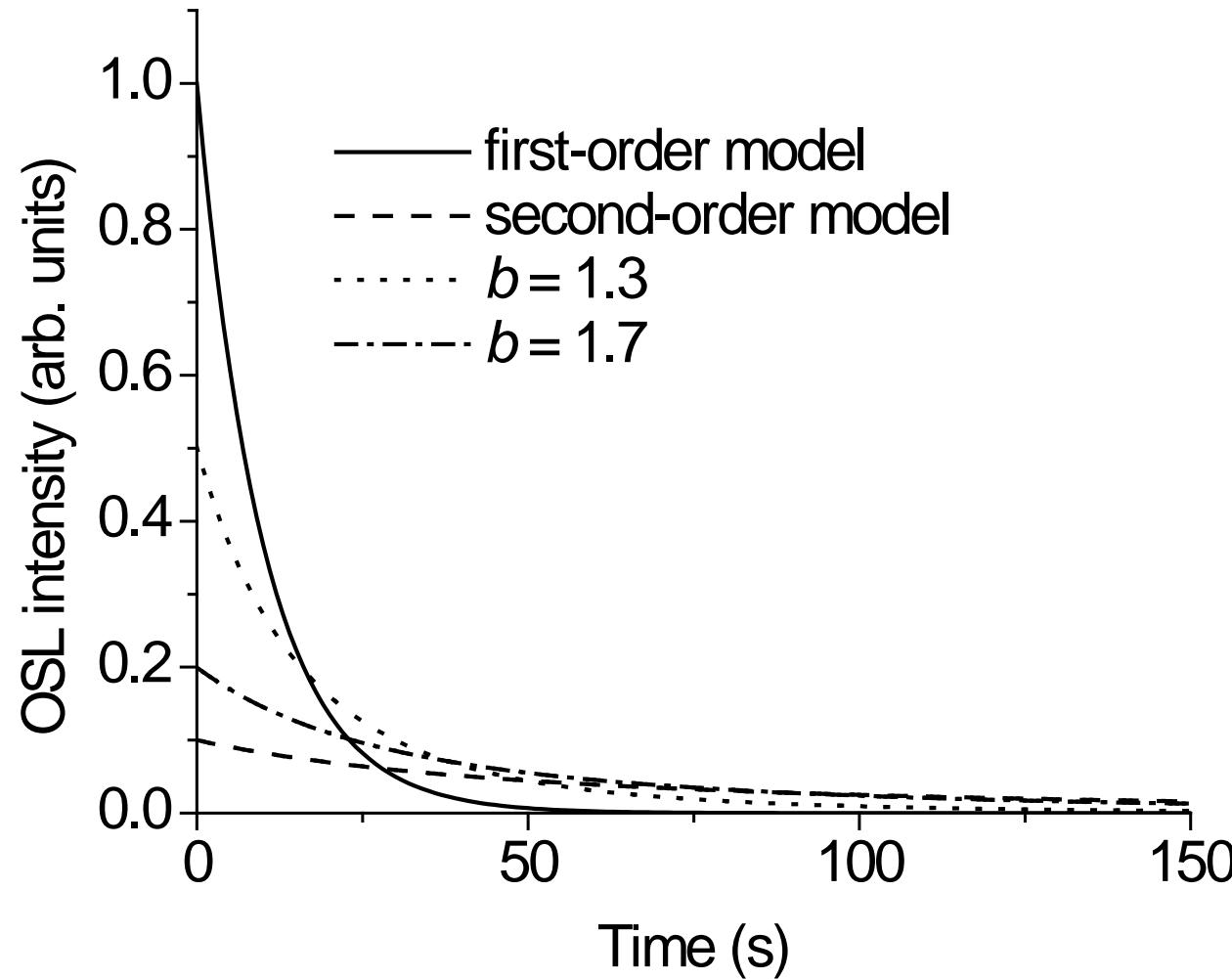
$$I_{OSL}^{GG}(t) \propto \frac{n^2 p}{N}$$

GOK

$$I_{OSL}^{GOK}(t) \propto \frac{n^b}{N^{b-1}} p$$

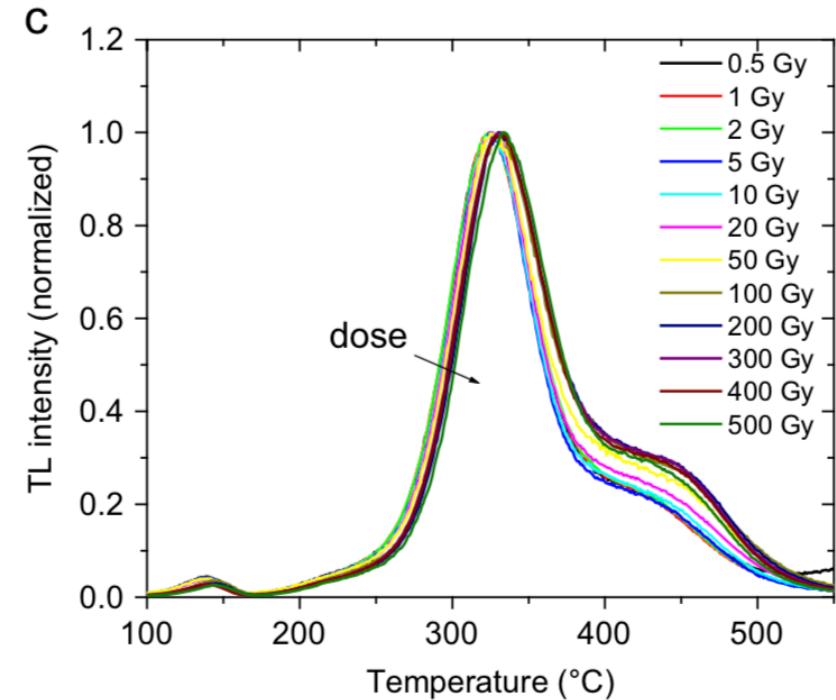
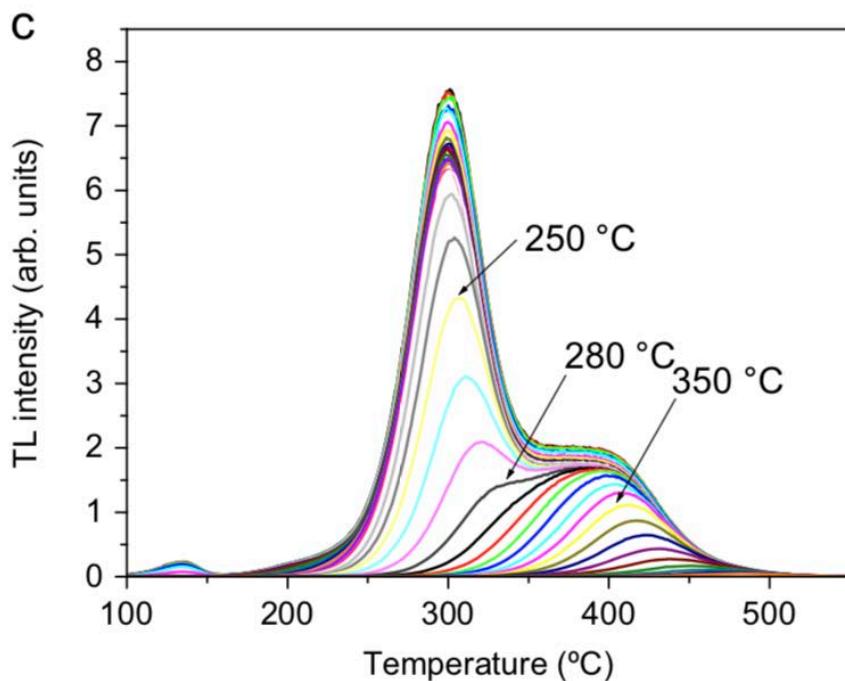
$$I_{OSL}^{GOK}(t) \propto \frac{n_0^b p}{N^{b-1}} \left[1 + (b-1) \left(\frac{n_0}{N} \right)^{b-1} pt \right]^{-\frac{b}{b-1}}$$

Other OSL models



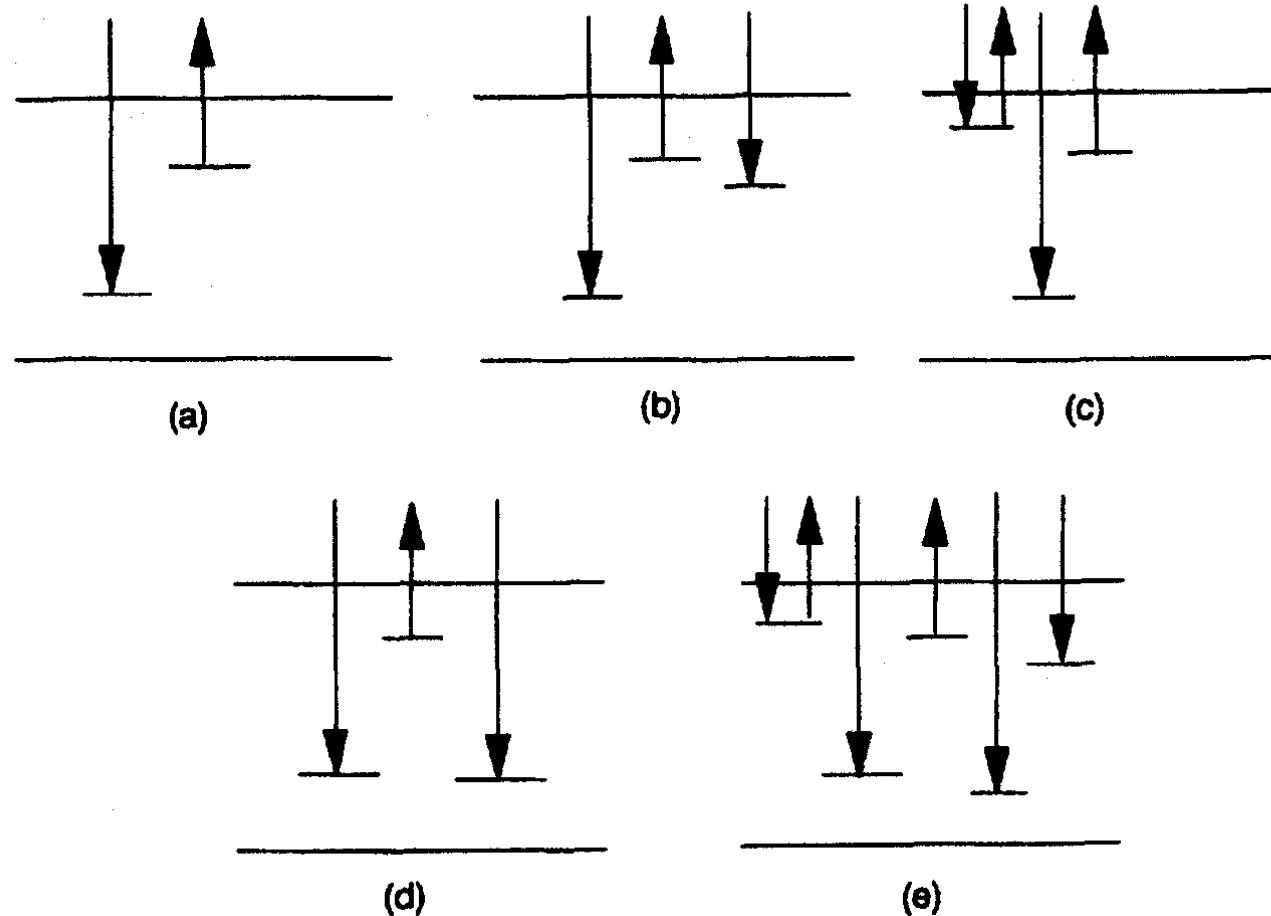
Word of caution about $b \neq 1$!

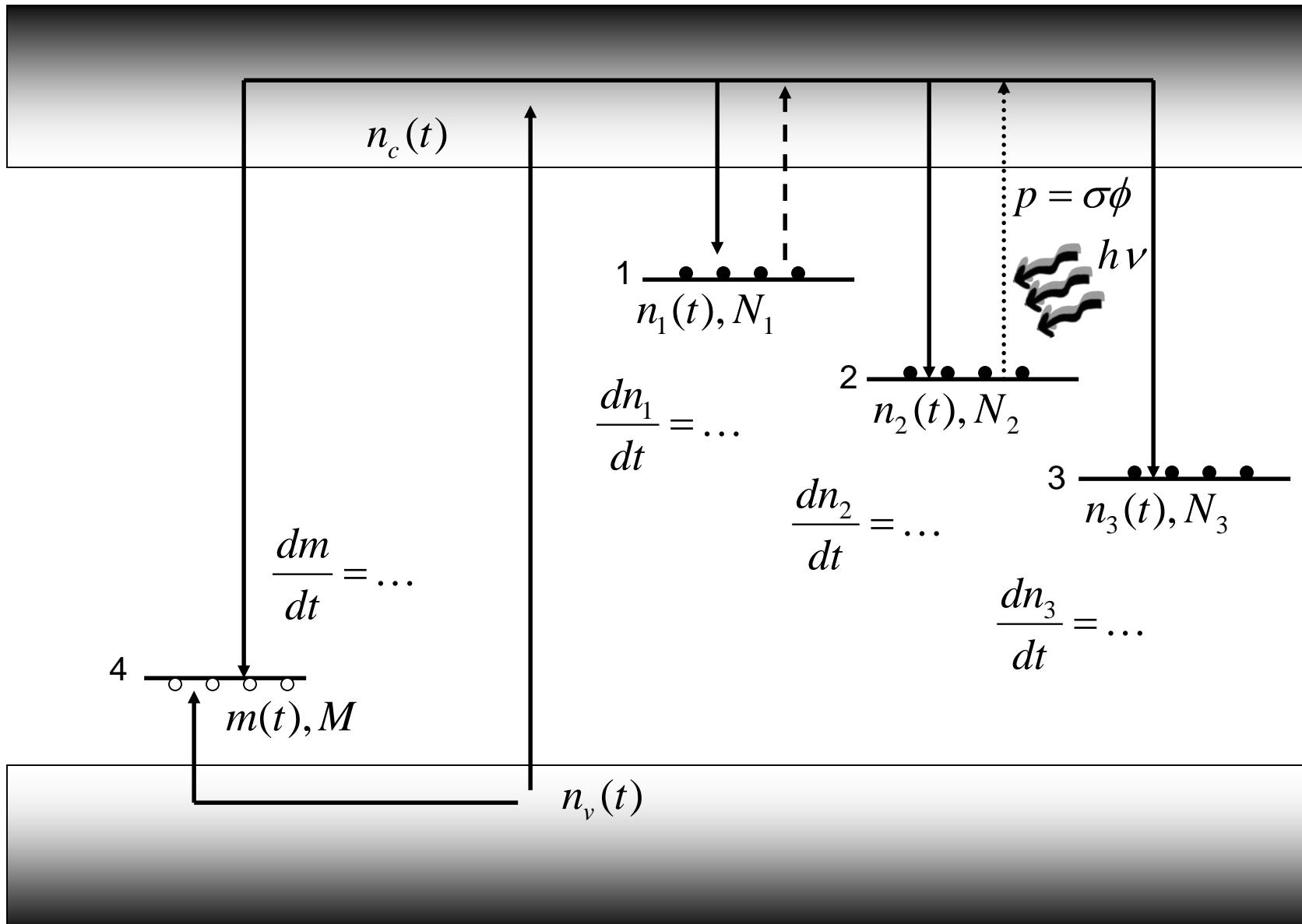
- Rarely seen in nature (why?)
- Implies TL peak shift to lower temperatures with dose
- b may vary even during TL readout (fitting with fixed b is meaningless)
- Superposition principle is not valid (fitting of multiple peaks is meaningless)



Doull, B.A., Oliveira, L.C., Wang, D.Y., Milliken, E.D., Yukihara, E.G., 2014. Thermoluminescent properties of lithium borate, magnesium borate and calcium sulfate developed for temperature sensing. *J. Lumin.* 146, 408-417.

Other models





Approaching the problem

$$\frac{dn_1(t)}{dt} = -n_1(t)s_1 e^{-\frac{E_1}{kT}} + [N_1 - n_1(t)]A_1 n_c(t)$$

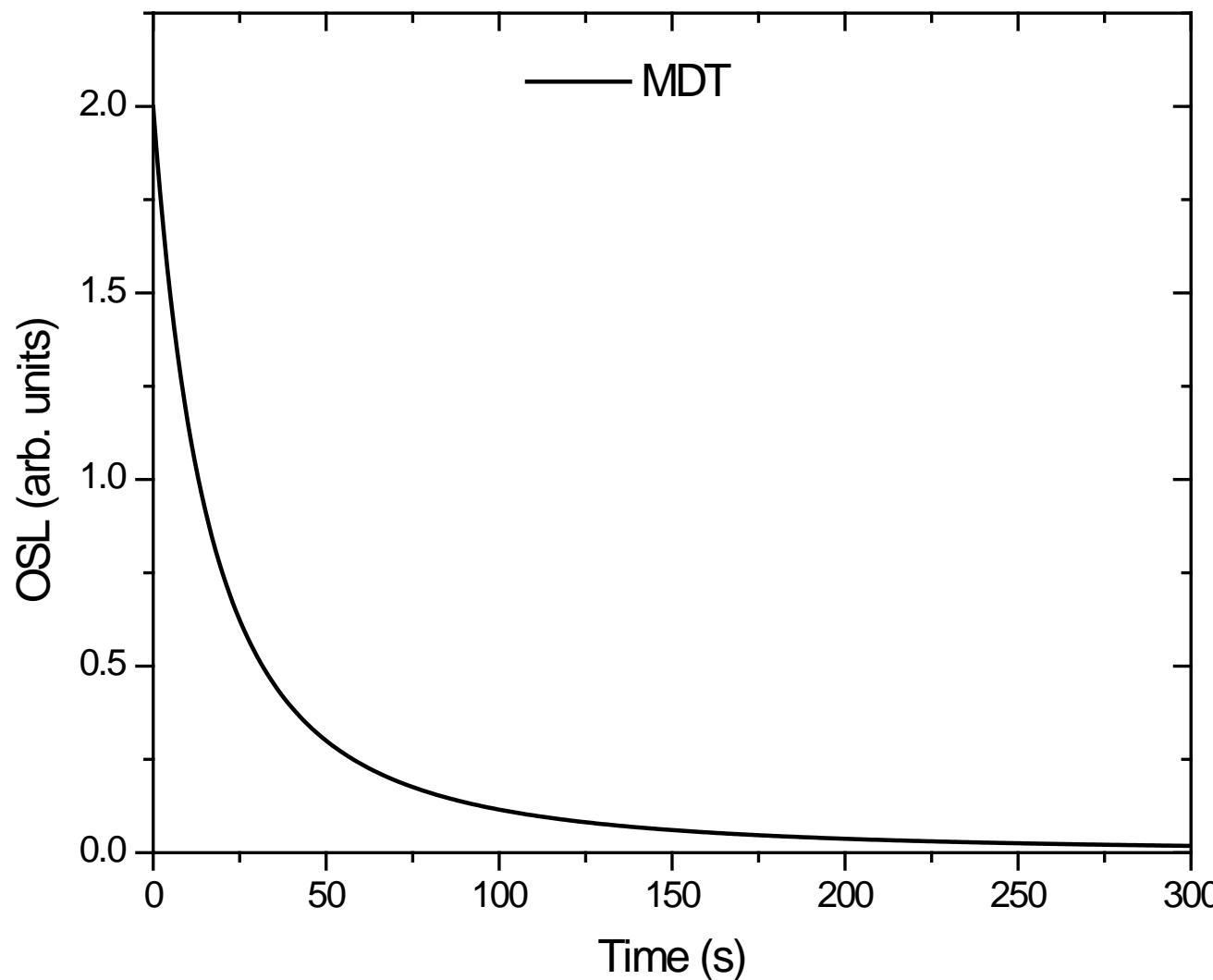
$$\frac{dn_2(t)}{dt} = -n_2(t)\sigma_2 \phi + [N_2 - n_2(t)]A_2 n_c(t)$$

$$\frac{dn_3(t)}{dt} = [N_3 - n_3(t)]A_3 n_c(t)$$

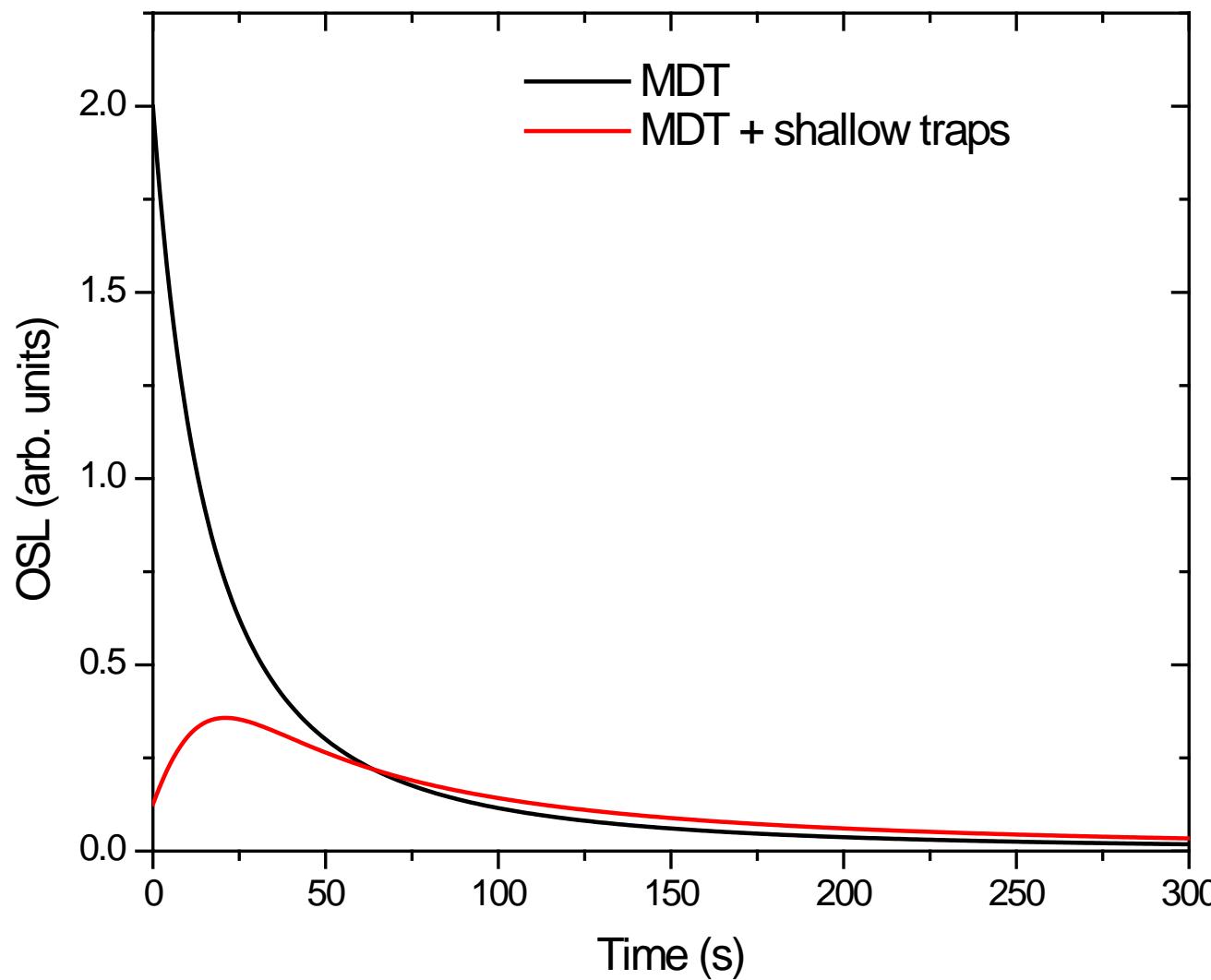
$$\frac{dm(t)}{dt} = -m(t)A_m n_c(t) + f$$

$$n_c(t) = \frac{f + n_1(t)s_1 e^{-\frac{E_1}{kT}} + n_2(t)\sigma_2 \phi}{[N_1 - n_1(t)]A_1 + [N_2 - n_2(t)]A_2 + [N_3 - n_3(t)]A_3 + m(t)A_m}$$

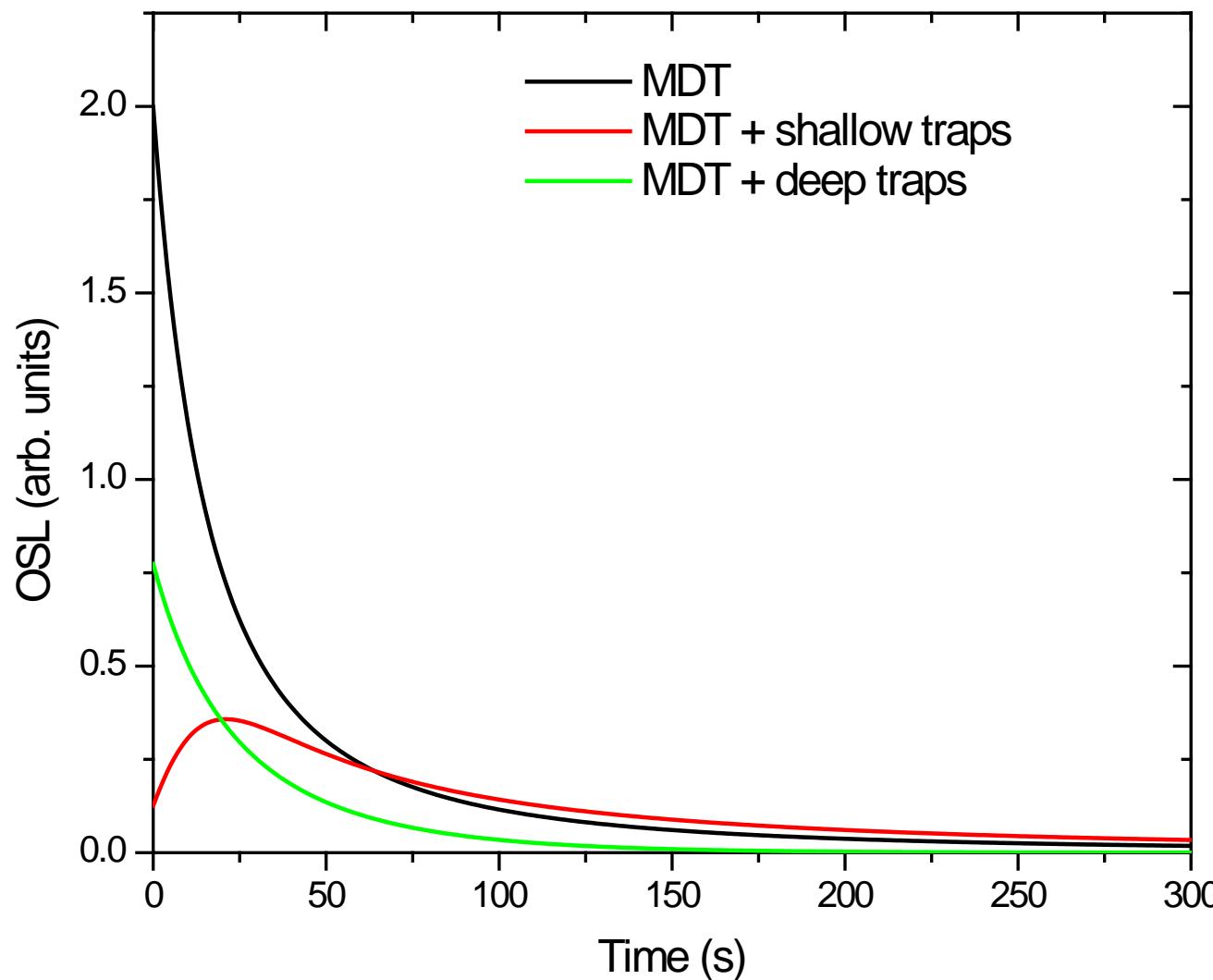
Effect of shallow traps and deep traps on OSL



Effect of shallow traps and deep traps on OSL



Effect of shallow traps and deep traps on OSL



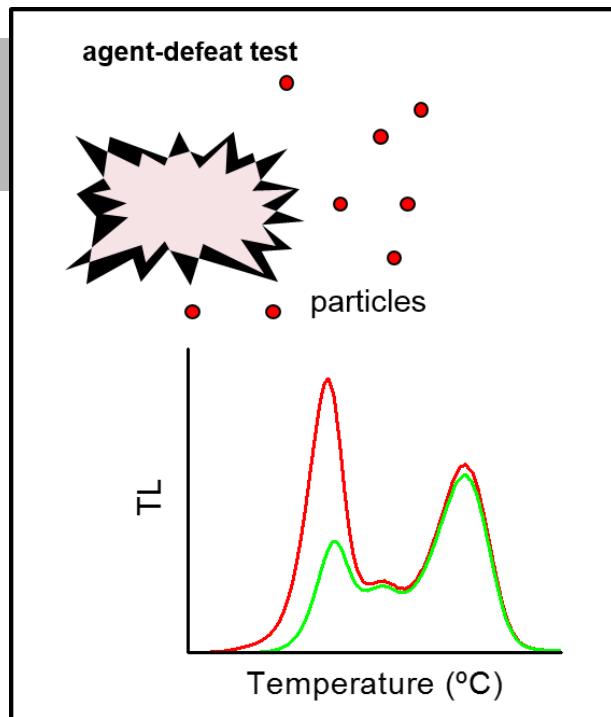
Advantages/disadvantages

TL

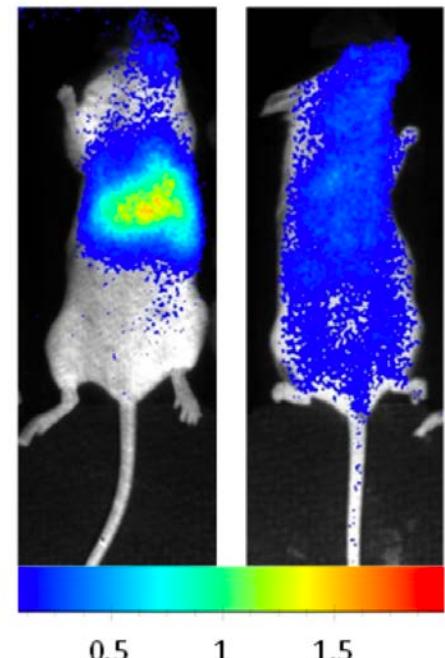
- Relatively slow (~seconds)
- Wide availability of materials
- Possibly light insensitive
- Intrinsic neutron sensitivity (Li, B)
- Easy to select stable signal

OSL

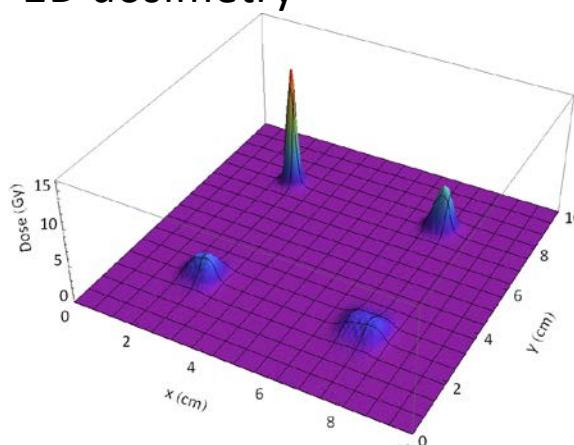
- Fast readout (~1 s)
- Only $\text{Al}_2\text{O}_3:\text{C}$ and BeO
- Sensitive to light
- Neutron insensitive / need converters
- Shallow traps leads to fading

TL temperature sensing

J. J. Talghader, M. L. Mah, E. G. Yukihara, A. C. Coleman, *Microsystems & Nanoengineering* 2 (2016) 16037.

new TL/OSL materials**persistent phosphors**

M. Pellerin, E. Glais, T. Lecuyer, J. Xu, J. Seguin, S. Tanabe, C. Chanéac, B. Viana, C. Richard, J. Lumin. 202 (2018) 83-88.

2D dosimetry

Ahmed, M.F., Shrestha, N., Ahmad, S., Schnell, E., Akselrod, M.S., Yukihara, E.G., 2017. Demonstration of 2D dosimetry using Al₂O₃ optically stimulated luminescence films for therapeutic megavoltage x-ray and ion beams. *Radiat. Meas.* 106, 315-320.

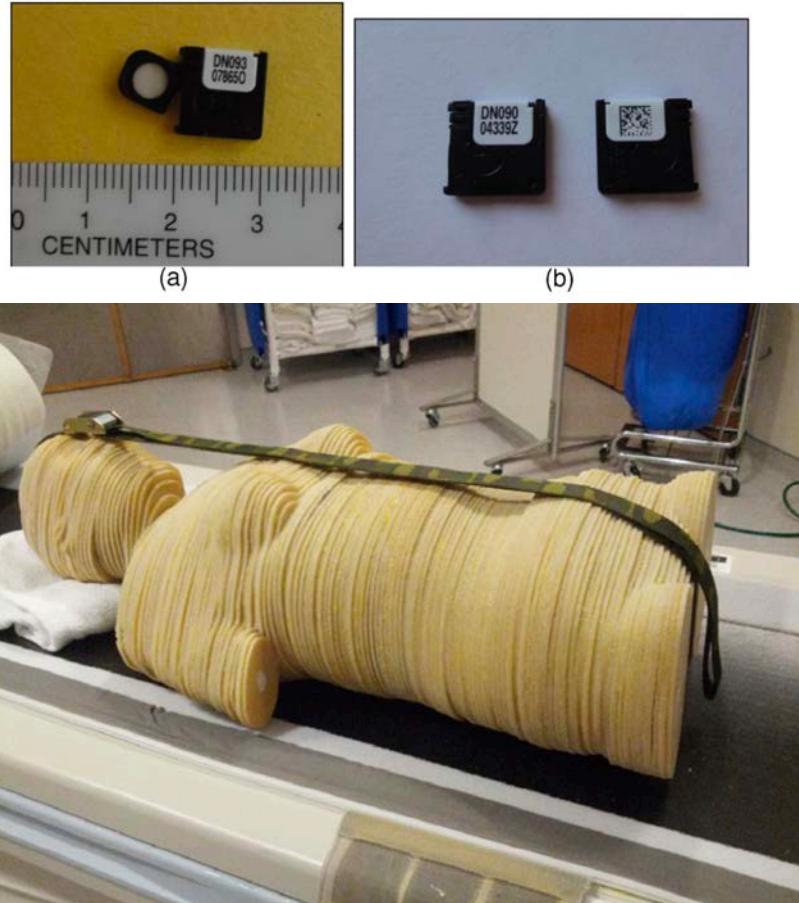
Trends

space dosimetry



Berger et al., 2016. DOSIS & DOSIS 3D: long-term dose monitoring onboard the Columbus Laboratory of the International Space Station (ISS). *J. Space Weather Space Clim.* 6, A39.

medical applications



Scarboro, S.B., Cody, D., Alvarez, P., Followill, D., Court, L., Stingo, F.C., Zhang, D., McNitt-Gray, M., Kry, S.F., 2015. Characterization of the nanoDot OSLD dosimeter in CT. *Med Phys* 42, 1797-1807.

Stepusin, E.J., Long, D.J., Ficarrotta, K.R., Hintenlang, D.E., Bolch, W.E., 2017. Physical validation of a Monte Carlo-based, phantom-derived approach to computed tomography organ dosimetry under tube current modulation. *Med Phys* 44, 5423-5432.

Wir schaffen Wissen – heute für morgen

We discussed...

- ... fundamentals of the TL/OSL process
- ... main TL/OSL characteristics
- ... advantages and disadvantages
- ... trends



Thank you for attention

Many thanks to...

- Martin Nikl
- LUMDETR scientific committee



eduardo.yukihara@psi.ch