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Luminescence dosimetry

Lily Bossin
EPFL, 6 Dec 2024

Who am I?

2015

Master University of Lyon / University of Wuhan

- Master in fundamental physics and solid state physics

February – July 2015 Research assistant, University Aristotle of Thessaloniki

- Optically stimulated luminescence and thermoluminescence properties of meteorites

2015

2015

August 2015 Research assistant, Riso national laboratory

- Investigation on the use of new kinetic models of feldspars for rock surface dating applications

October 2015 – March 2019 PhD candidate, Durham University

- New fortuitous material for luminescence dosimetry following radiological catastrophes

2015

2019

May 2019 – November 2020 Postdoctoral researcher, University of Lausanne

- Development of electron paramagnetic resonance thermochronometry

December 2020, Deputy Head Dosimetry group, Paul Scherrer Institute

2020

The Paul Scherrer Institute (PSI)

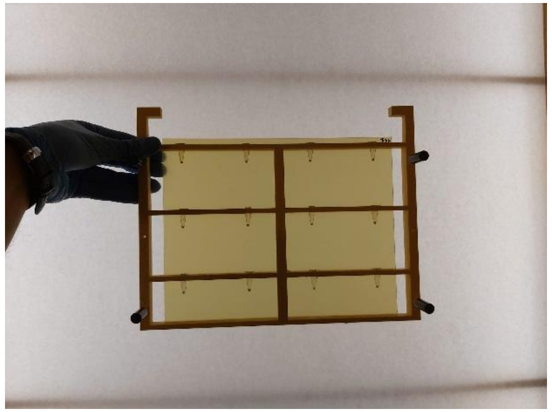
Section Messwesen: Our responsibilities (highlights)

Personal and environmental dosimetry

Calibrations and verifications

Development of calibration methods

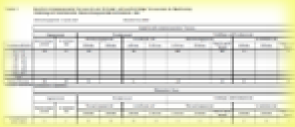
Support for experiments



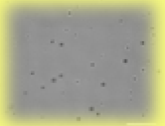
PSI Dosimetry group

External personnal dosimetry for PSI employees and external customers (ETH, research institutes,...)

Reporting to the authorities (central dose registry)



Neutron dosimetry (only service in CH)



Poly-Allyl Diglycol Carbonate (PADC)

Internal dosimetry

Whole body counter

Thyroid counter

Urine and excretion analysis

Research on new instrumentation and methods

BeO OSL system

Fluorescence Nuclear Track Detectors

Supporting research at PSI...

Supporting research at PSI

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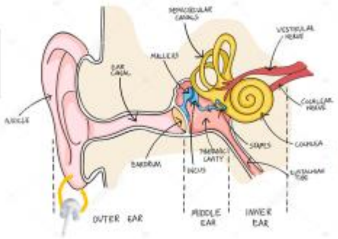
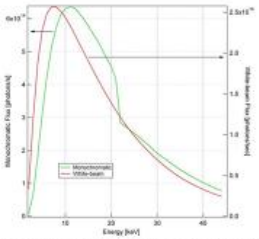
Dosimetry for Dynamic X-Ray Tomography (DYNAMYTE project)



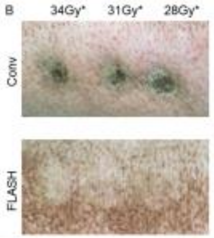
Challenges
- Slow shutter (>1 s opening; long irradiations)
- High doses/dose rates
- Low energy
- Narrow field

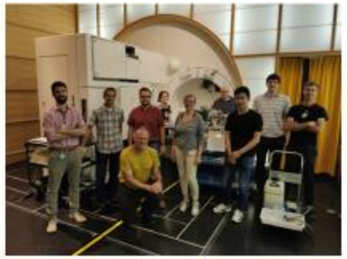


https://www.psi.ch/en/sls/tomcat

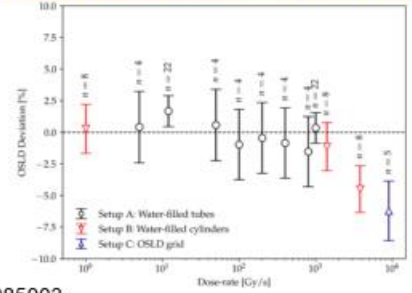
Support for FLASH radiobiology experiments

The advantage of FLASH radiotherapy confirmed in mini-pig and cat-cancer patients





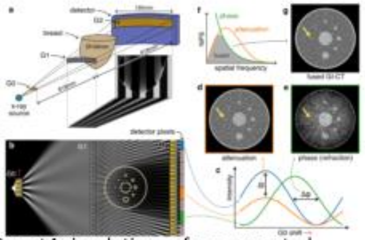
Any dose rate effect in luminescence detectors?



Christensen et al. (2021) Phys. Med. Biol. 66, 085003.

Page 29

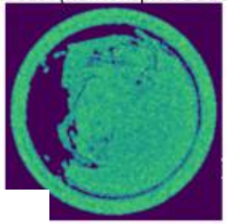
Dosimetry for new imaging modalities



Geant4 simulations of a segmented breast (Kantonspital Baden)

GEANT4

Evaluation of radiation levels in biological samples?



16 cm

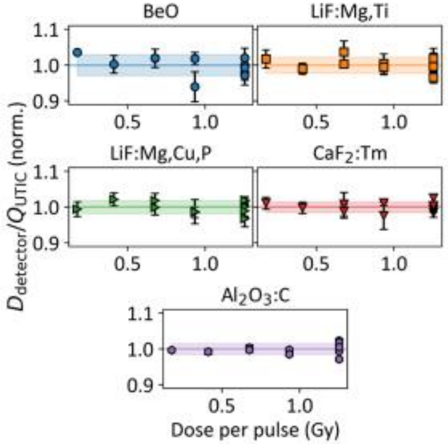
-> CT scan
-> simulations of dose to
- adipose tissue
- glandular tissue
- microcalcifications

Simulations of a segmented ear

16 cm

Dose rate dependence of luminescence detectors (electrons) - USZ

Medical LINAC converted for FLASH studies



Page 32

Outline

- Dosimetry concepts
- Luminescence dosimetry - why?
- TL/OSL/RPL
- Operational properties of luminescence dosimeters
- Case studies and applications

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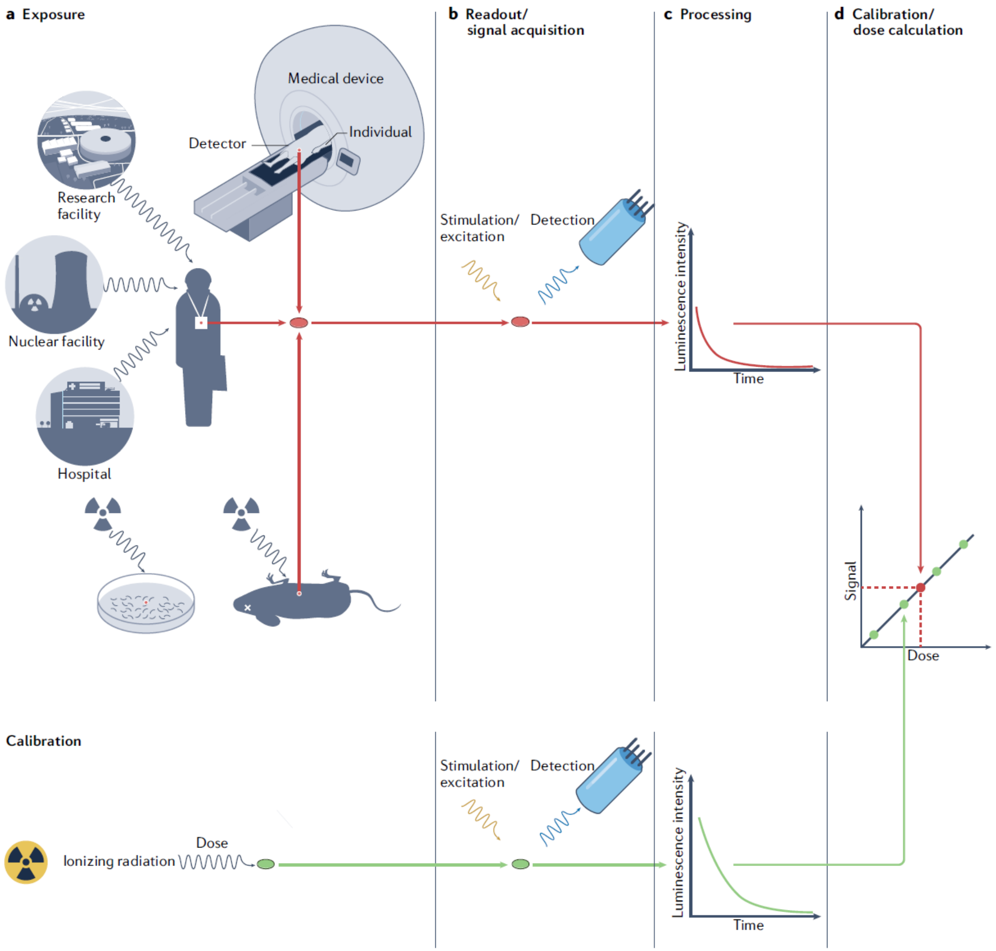
I. Dosimetry concepts

What is dosimetry?

“quantification of the energy deposited in a living or inanimate object from a radiation field in order to estimate, predict or limit the effect of radiation”



Yukihara et al. (2022)

Dosimetry can be…

(find adjectives)

<u>Adjectives</u>

- Passive
- Active
- Computational
- Retrospective
- Accident
- Luminescent
- Personal
- Environmental
- In-vivo
- Clinical
- Off-line
- Real-time
- Three-dimensional
- Neutron
- Gamma
- Etc.

Quantities in dosimetry

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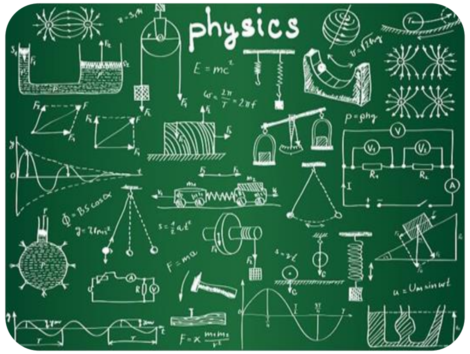


Physical quantities

- Fluence Φ
- Kerma K
- Absorbed dose D
- Linear energy transfer LET
- Etc.

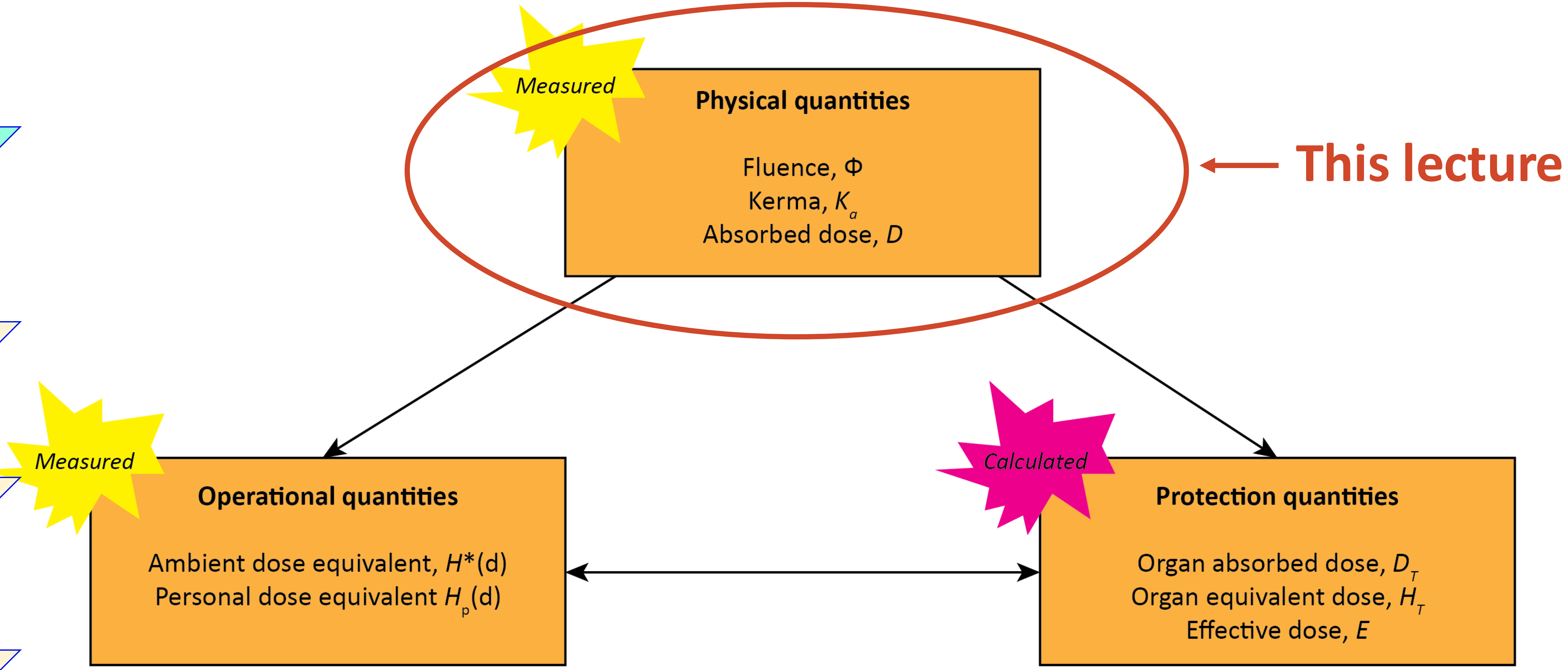
Radiation Protection Quantities

- Organ equivalent dose H_T
- Effective dosis E
- Committed dose E_{50}

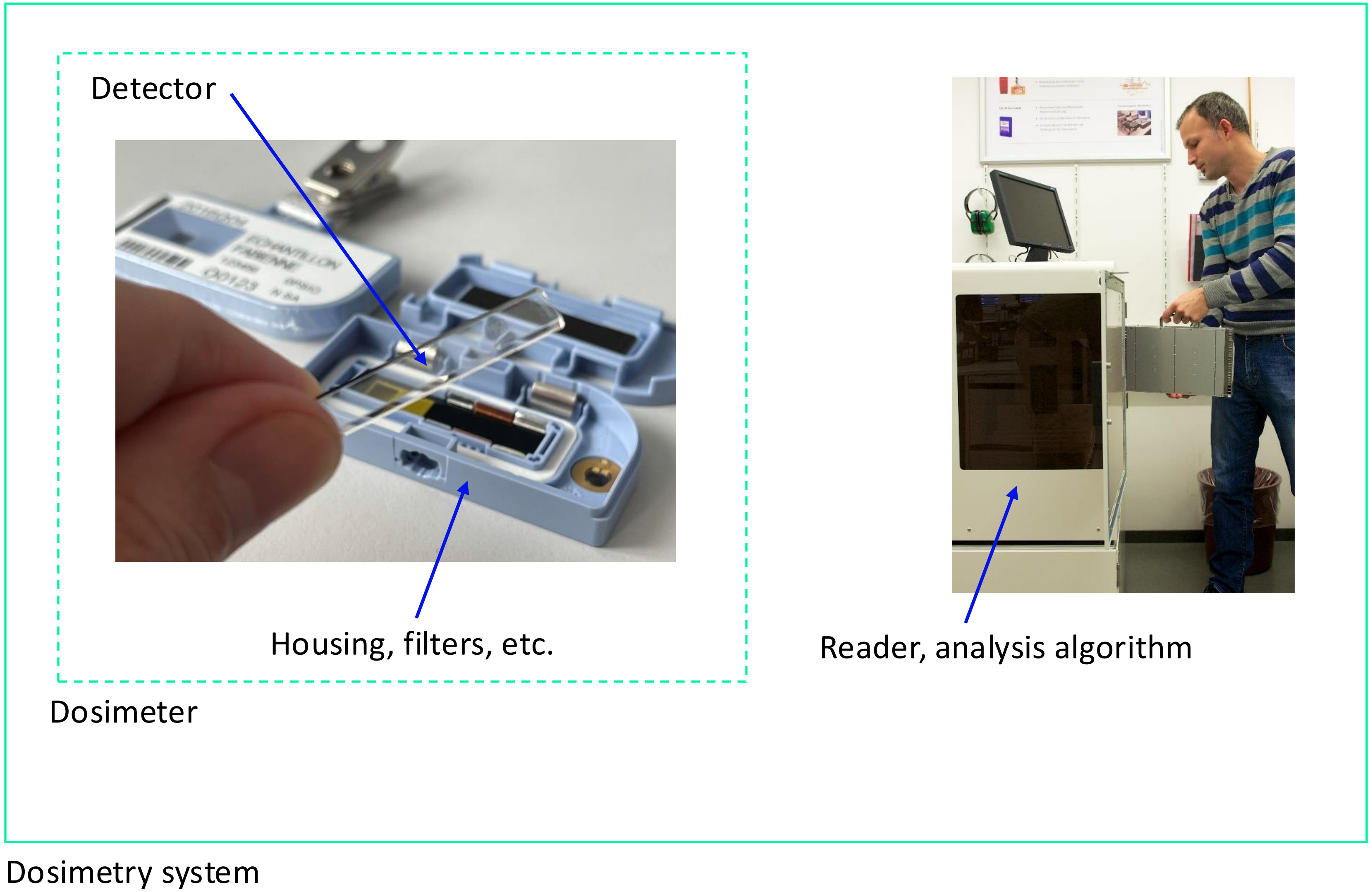
Operational quantities

- Personal dose equivalent $H_p(10)$, $H_p(0.07)$, $H_p(3)$
- Ambient equivalente dose $H^*(10)$

Quantities for radiation protection

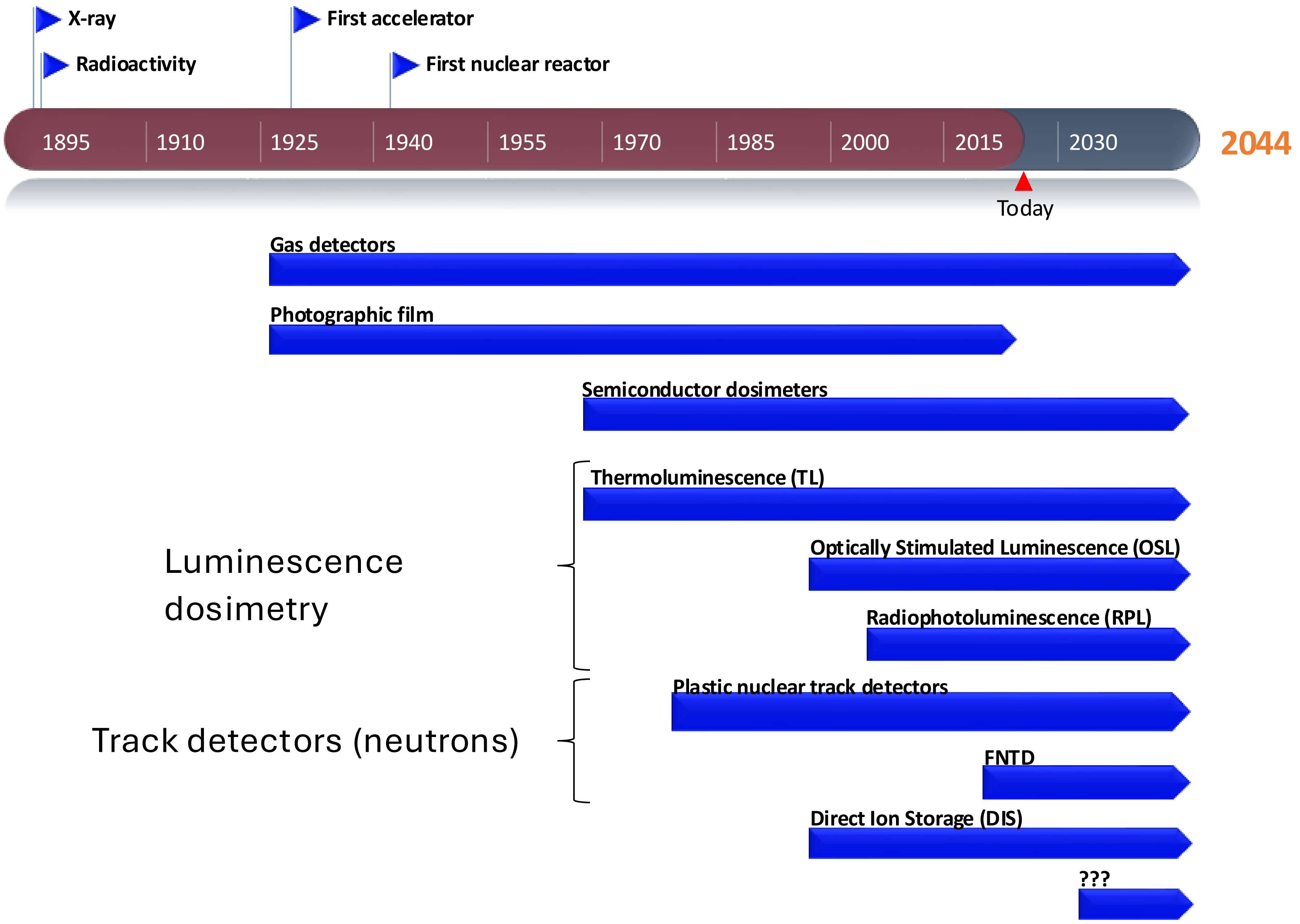


Detector/dosimeter/dosimetry system



What happens inside the detector?
How do we assess the fluence?
How do we assess the LET?
How do we get the absorbed dose?

Technologies in individual monitoring



See: Wernli and Kahilainen (2001), Wernli (2016), Barthe (2001)

II. Luminescence dosimetry - why?

Means to measure ionising radiation

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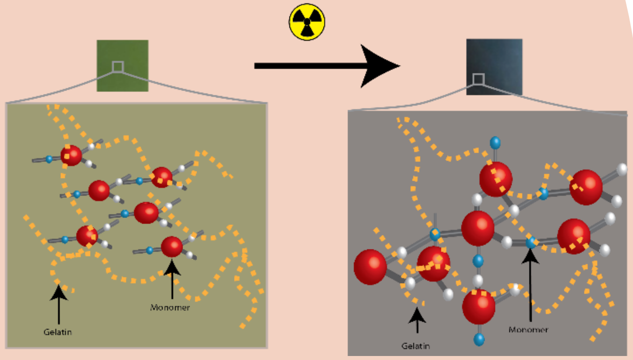
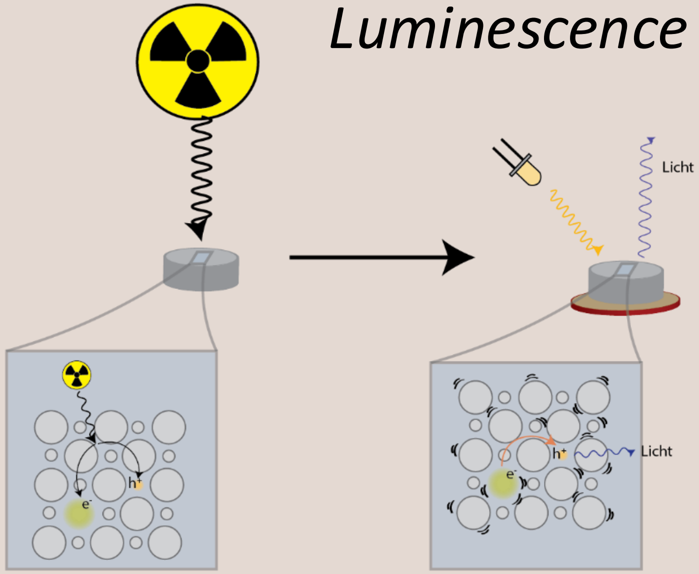
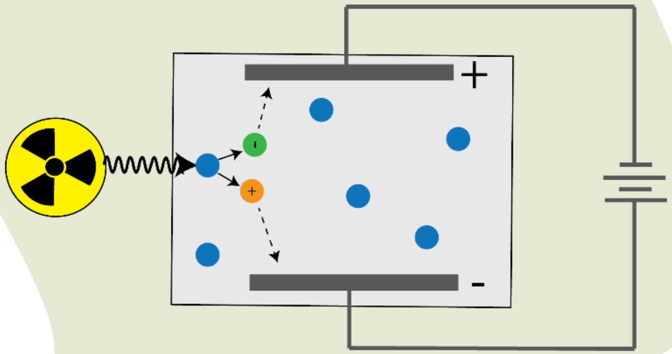
Radiochromic film



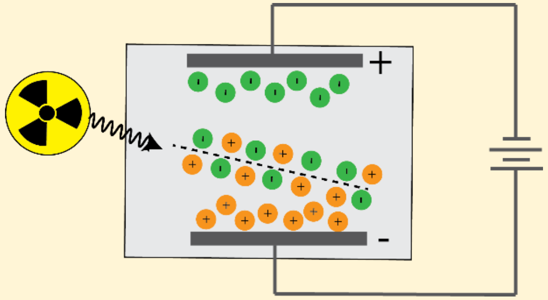
Luminescence



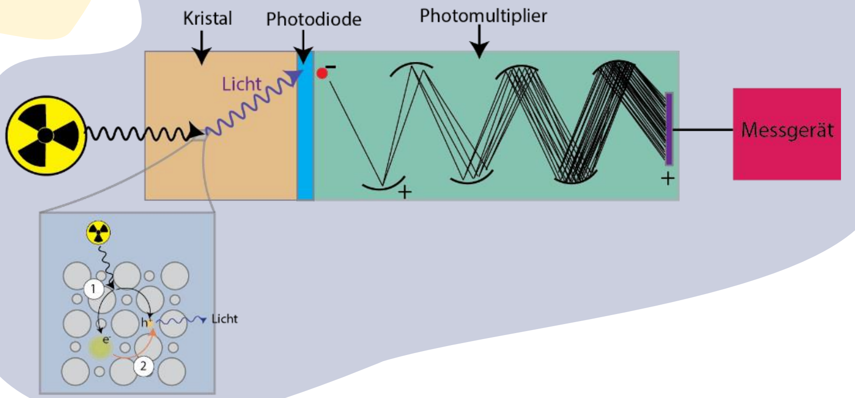
Ionisation chamber



Semi conductor detector



Scintillation counter



Advantages of luminescence dosimetry

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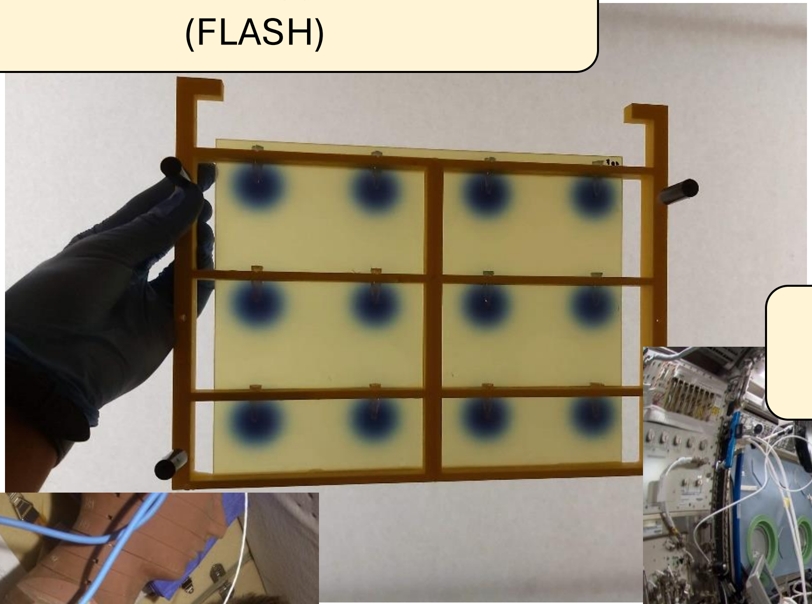
- Small detectors
- No cables
- Precision and accuracy
- Convenience (easy to read)
- Similar to tissue or water
- Minimal influence of magnetic field
- Supposedly minimal influence of dose rate

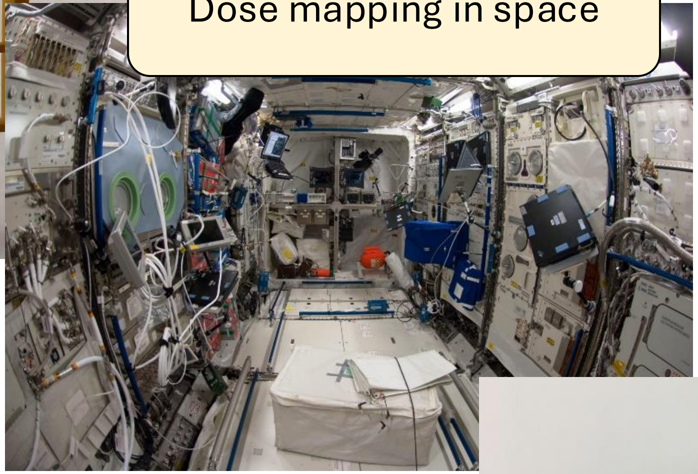
No choice but luminescence dosimetry

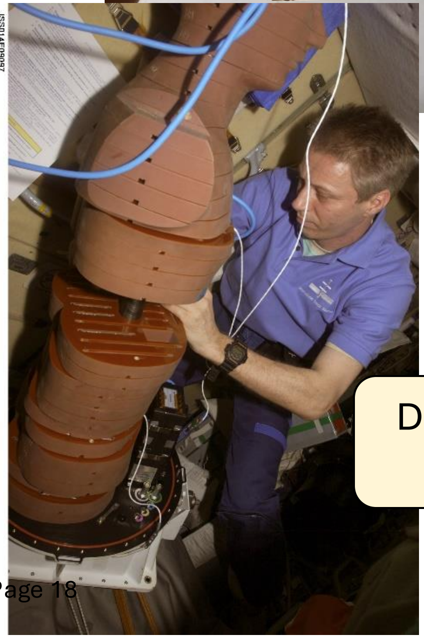
Irradiation of fish eggs with protons (FLASH)



Irradiation of mice with pions

Dose mapping in space





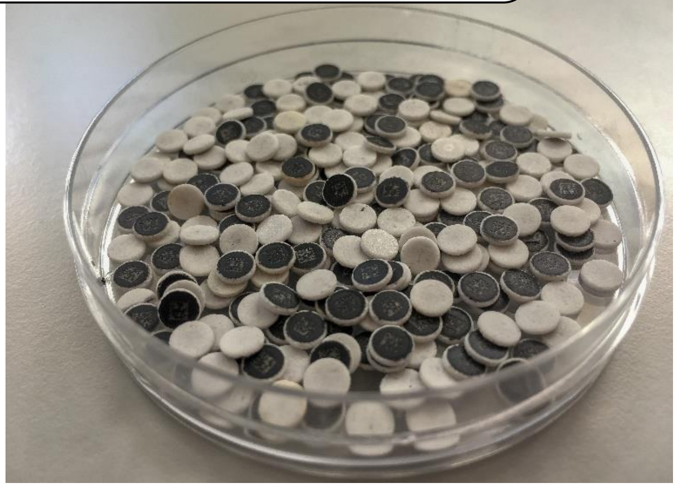
Dose mapping in phantoms

Extremity dosimetry

III. TL/OSL/RPL

Examples of luminescent detectors

Thermoluminescence (TL) detectors
TLDs



Optically stimulated luminescence (OSL) detectors
OSLDs

Radiophotoluminescence (RPL) detectors - RPLGD

How do they work?

A perfect crystal

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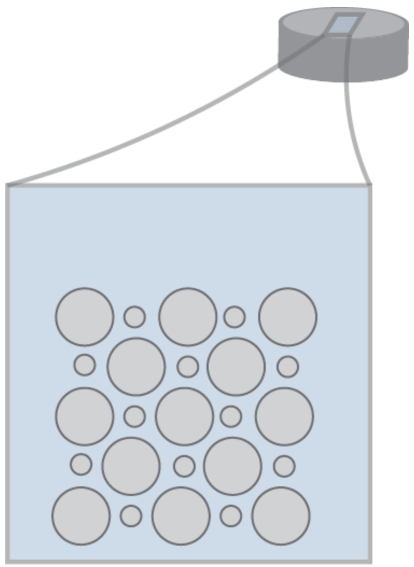
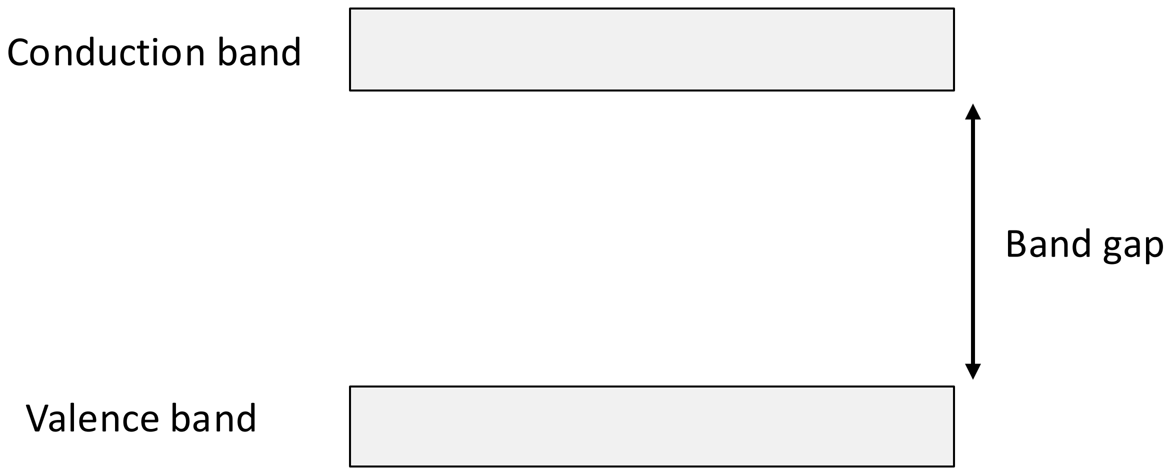
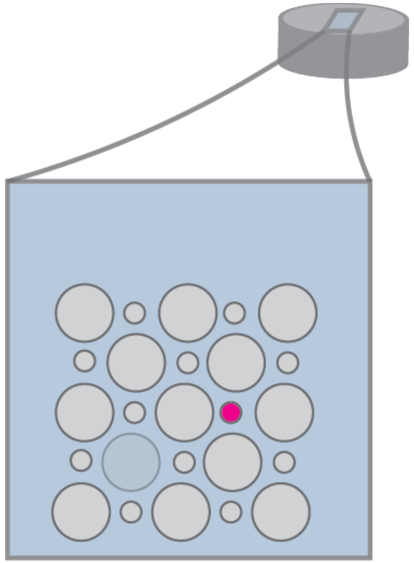


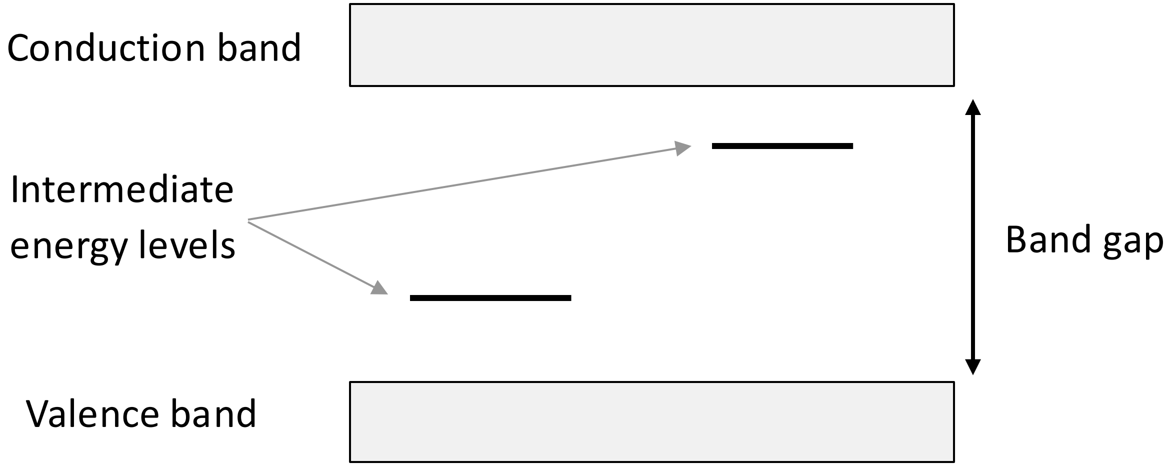
How do you represent the energy levels in a crystal?



An imperfect crystal



How do you represent the energy levels in a crystal?



Imperfections in crystals (dislocations, substitutions, etc) create intermediate energy levels.

An imperfect crystal – exposure to ionising radiation

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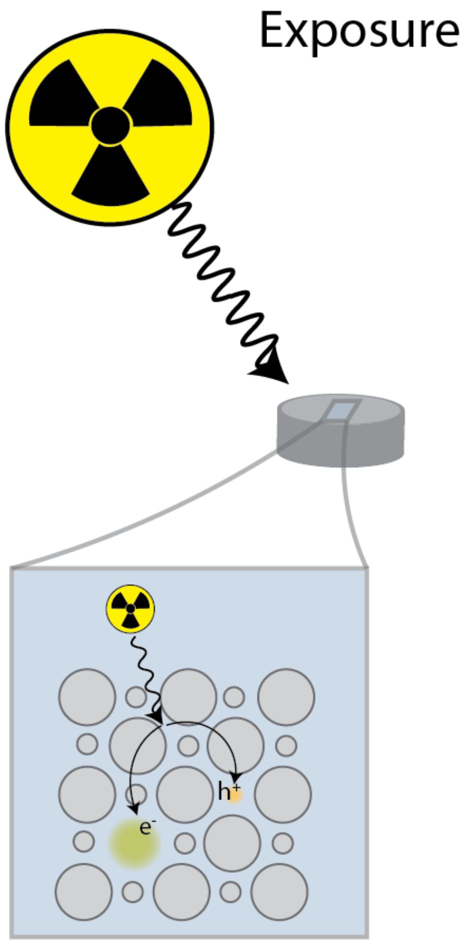
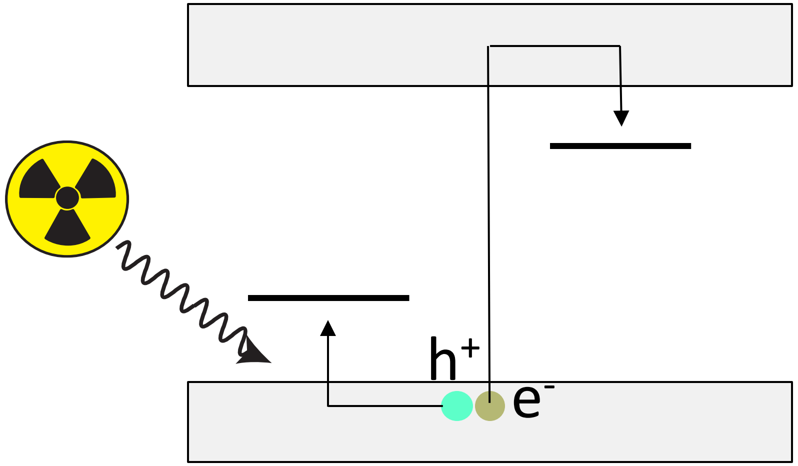
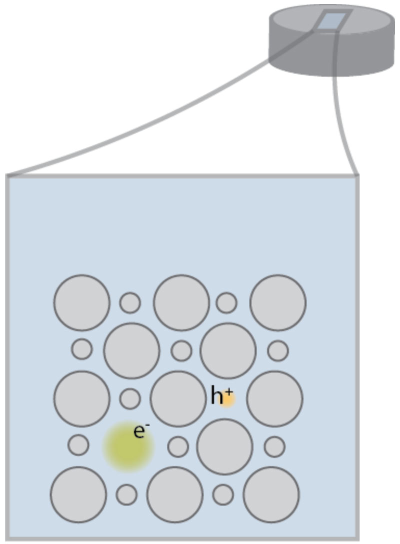
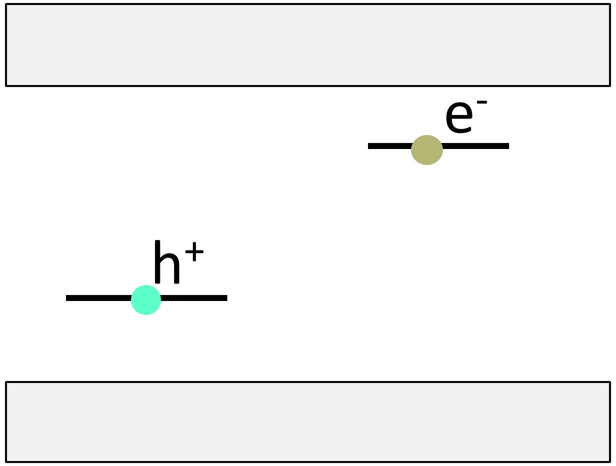




Ionising radiation creates electron-hole pairs. Electrons and holes are released into conduction/valence bands and trapped in defects.

An imperfect crystal – storage





Charges can remain in traps – from fractions of seconds to million of years.

An imperfect crystal – stimulation/readout

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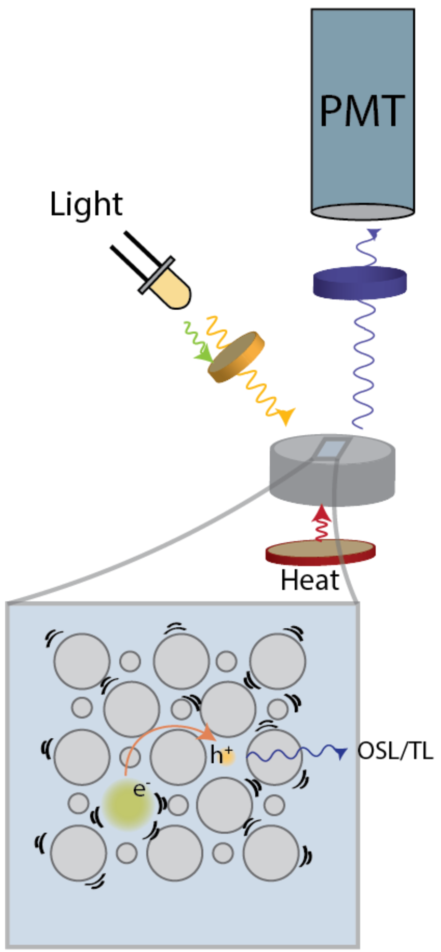
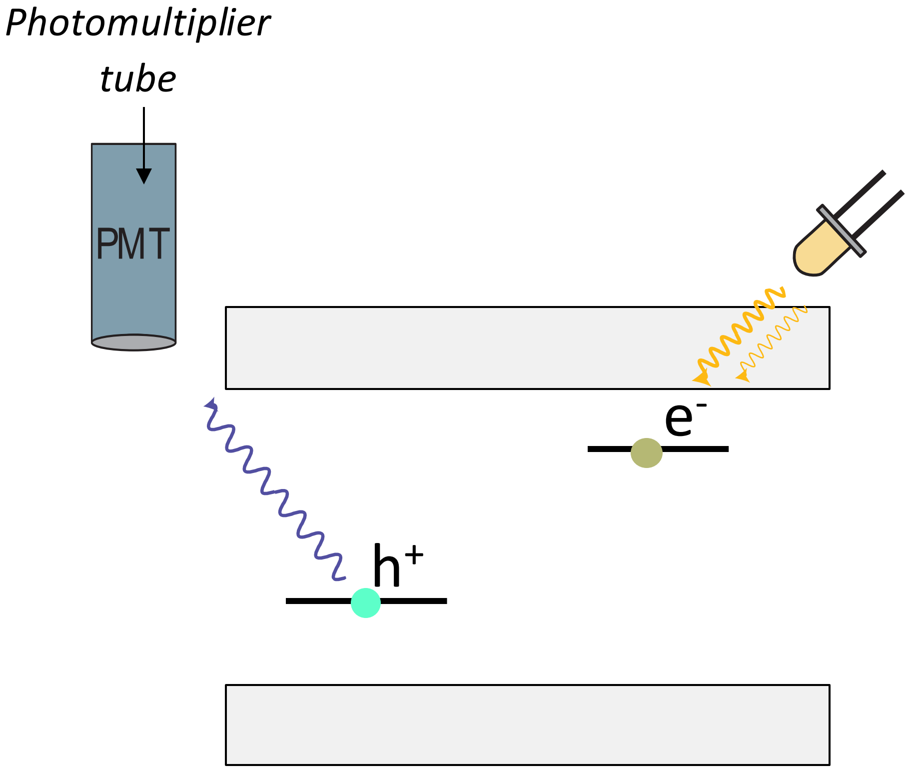
Readout





Upon stimulation, charges are released from their traps. Their travel to a recombination centre. The de-excitation process produces photon, easily detectable in the lab.

Fluorescence vs Phosphorescence vs Stimulated Luminescen

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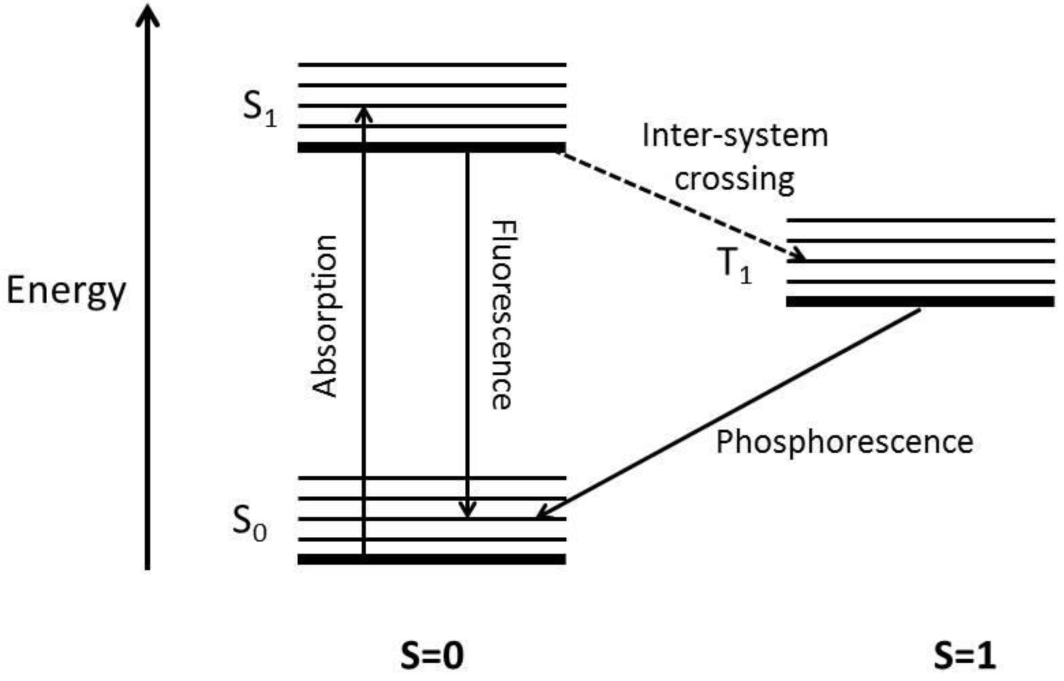
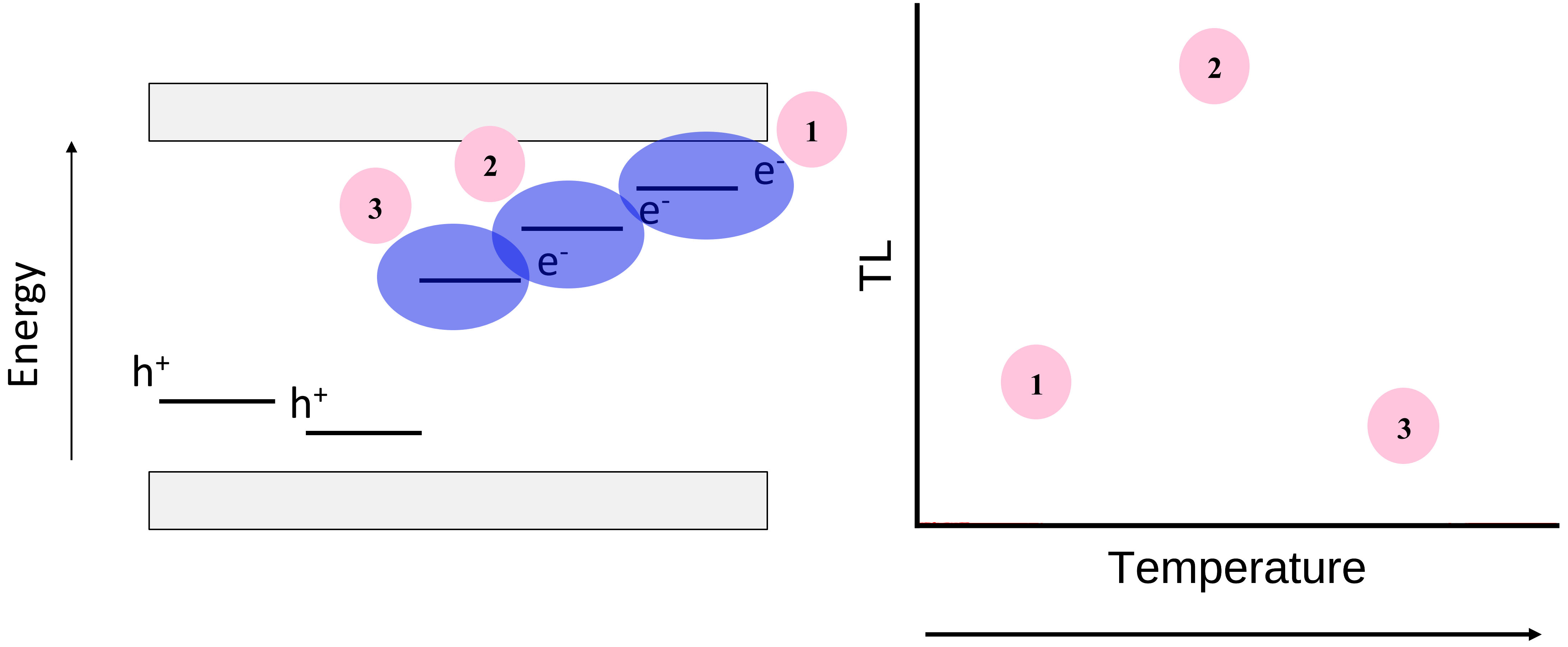


Fluorescence
$\tau < 10^{-8}$ s

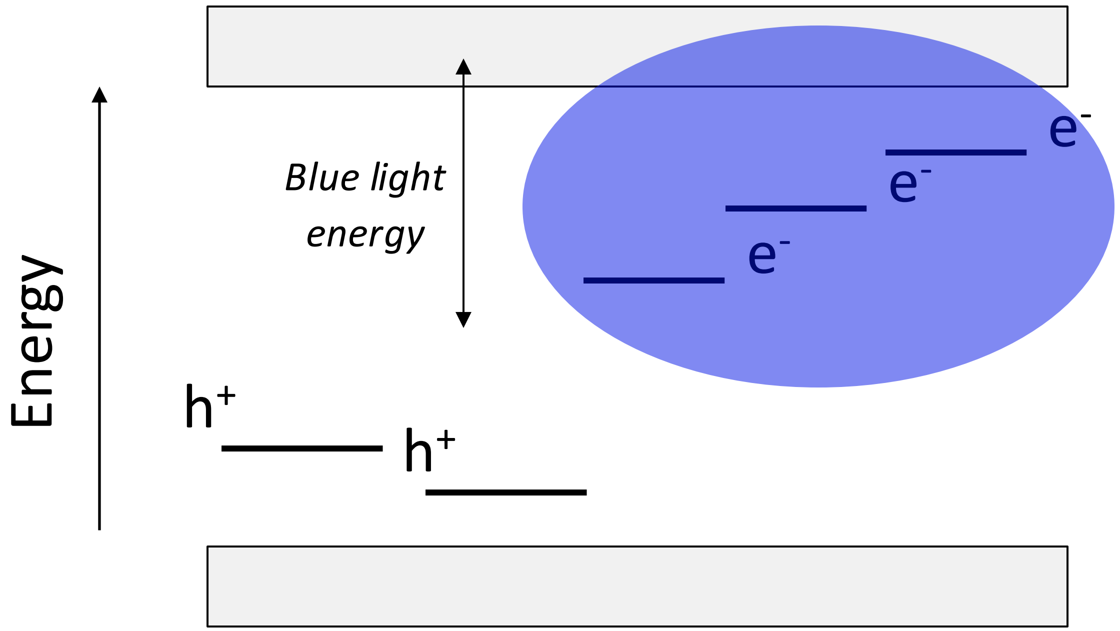
Phosphorescence/stimulated luminescence
10^{-8} s $< \tau < 10^{9}$ years

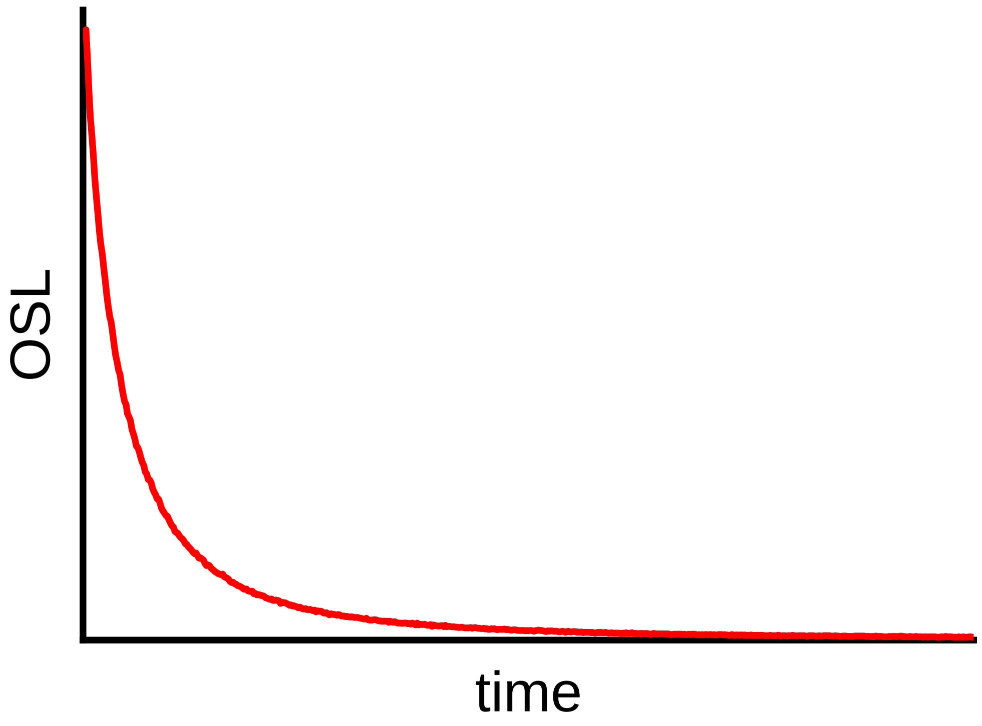
Two main types of stimulation:
1) Thermal stimulation



Thermal stimulation = thermoluminescence (TL)

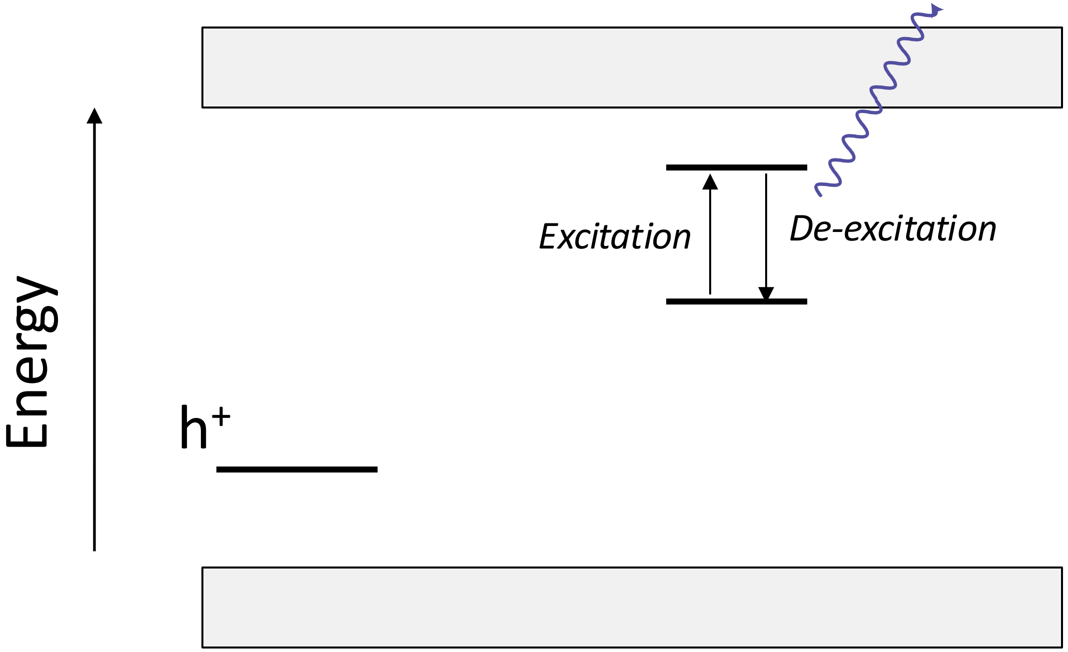
Two main types of stimulation:
2) Optical stimulation

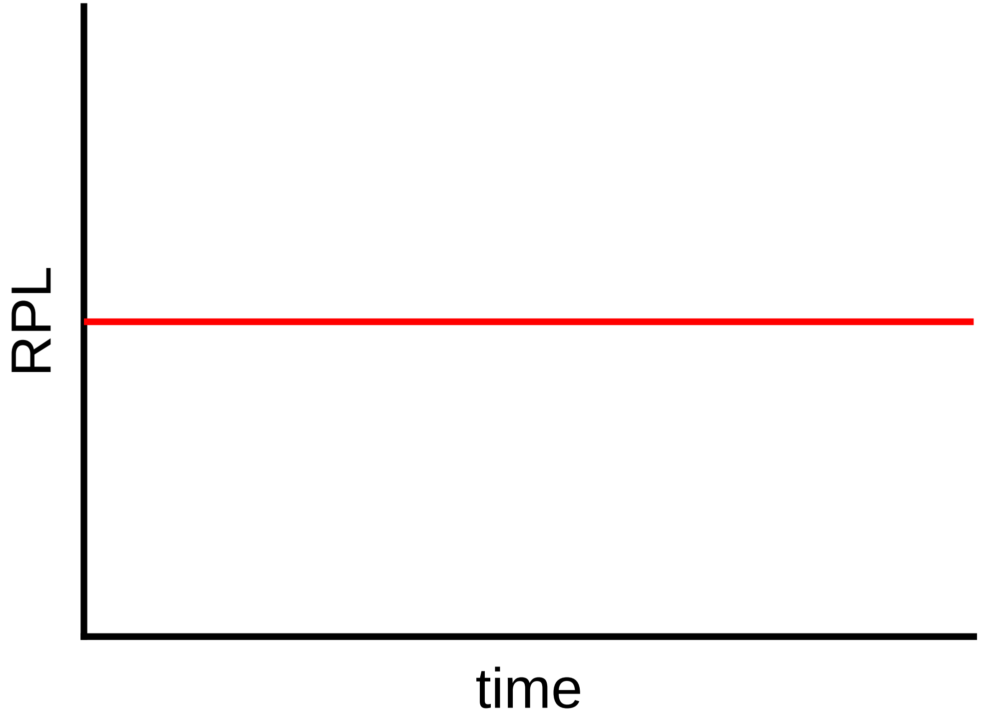




Optical stimulation = optically stimulated luminescence (OSL)

Radiophotoluminescence





Comparison between the techniques

Readout technique	TL	OSL	RPL
• Stimulation • Readout destructive? • Detection window restrained? • Background to take into account? • Signal acquisition? • Affected by thermal quenching?			

Comparison between the techniques

Readout technique	TL	OSL	RPL
Stimulation	Thermal contact		
Readout destructive?	Destructive readout		
Detection window restrained?	Broad detection possible		
Background to take into account?	Blackbody background		
Signal acquisition?	Only integrated luminescence		
Affected by thermal quenching?	Affected by thermal quenching		

Comparison between the techniques

Readout technique	TL	OSL	RPL
Stimulation	Thermal contact	Optical readout	Optical readout
Readout destructive?	Destructive readout	(Semi)destructive readout	Non-destructive readout
Detection window restrained?	Broad detection possible	Need to block stimulation light	Need to block stimulation light
Background to take into account?	Blackbody background	Low background (light leakage)	High background (phosphorescence)
Signal acquisition?	Only integrated luminescence	Time-resolved measurement possible	Time-resolved measurement required
Affected by thermal quenching?	Affected by thermal quenching	Not affected by thermal quenching	Not affected by thermal quenching

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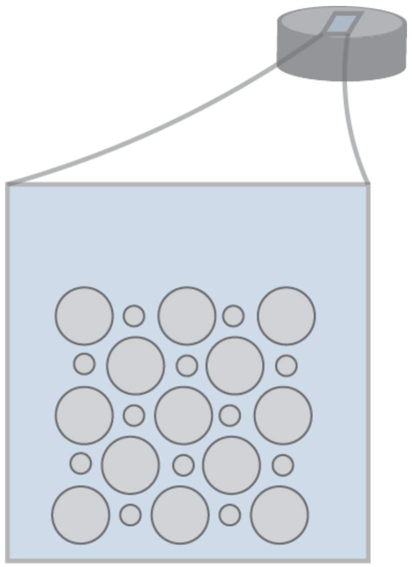
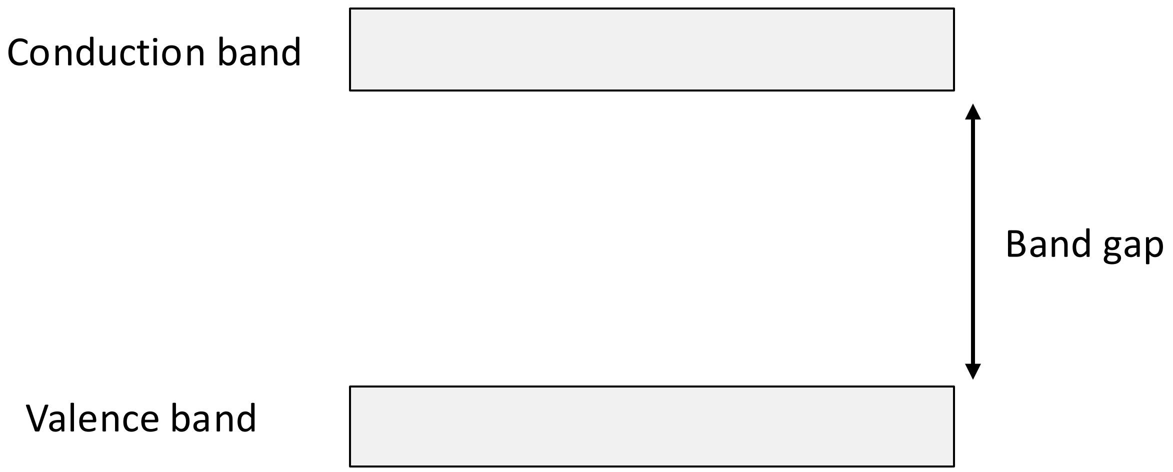
Questions

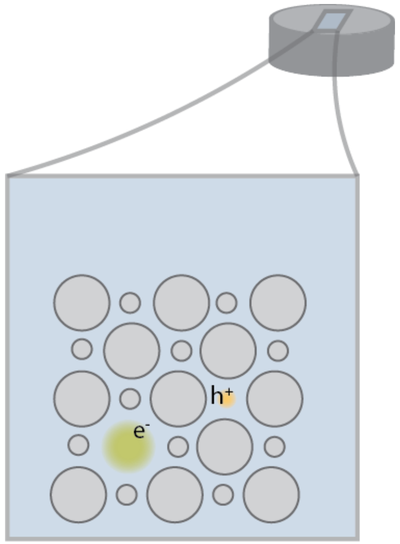
Could we have TL/OSL from a perfect crystal?

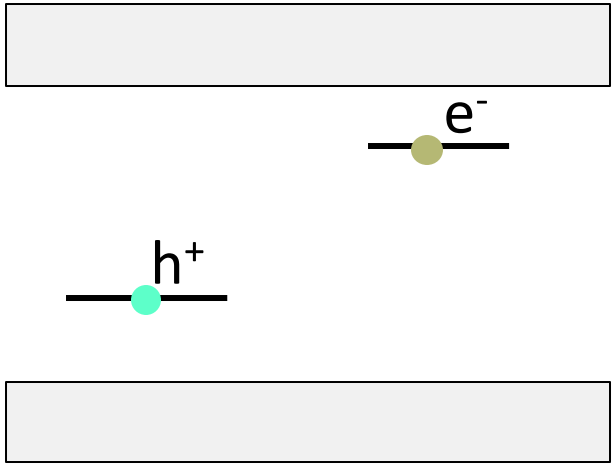




No you wouldn’t. No presence of intermediate energy levels acting as trapping levels.

What governs how long the charges can stay in their traps before the readout?

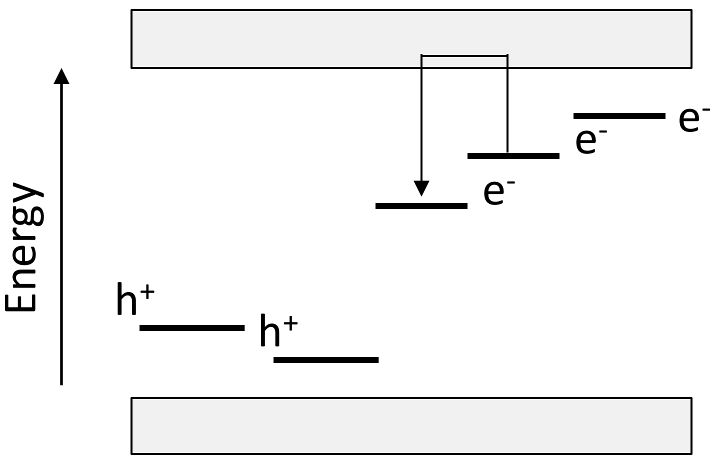




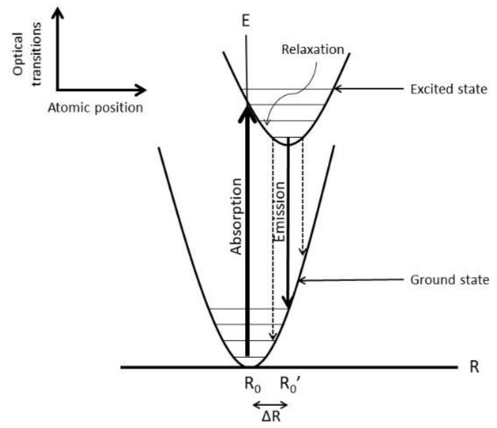
Probability of escape – frequency factor.

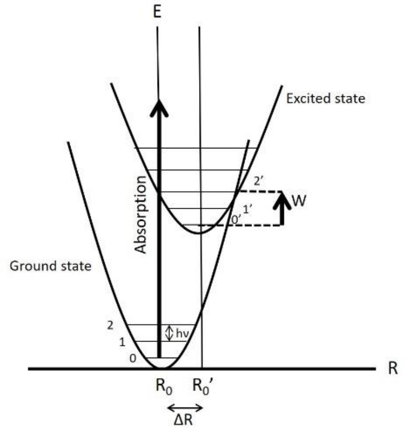
Factors limiting the luminescence efficiency

Competition/Retrapping



Non-radiative recombination





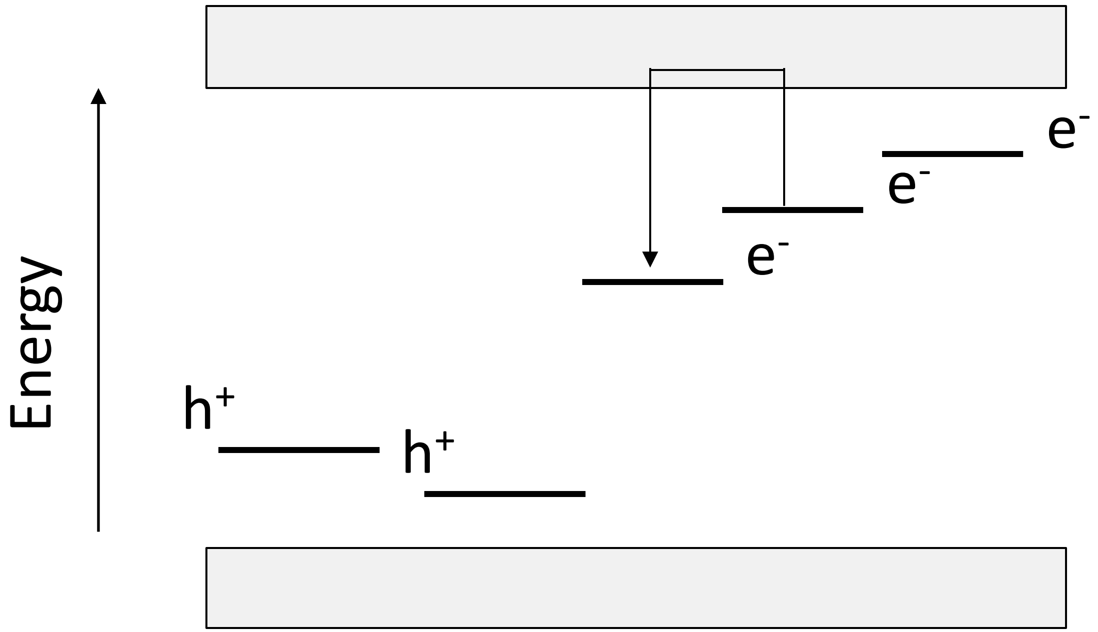
Quenching effects

- **Thermal quenching**
  - → High temperature increases the probablity of non-radiative recombination
- **Concentration quenching**
  - → High number of luminescent centres results in energy transfer
- **Impurity quenching**
  - → «killer» centres introducing non-radiative recombination pathways

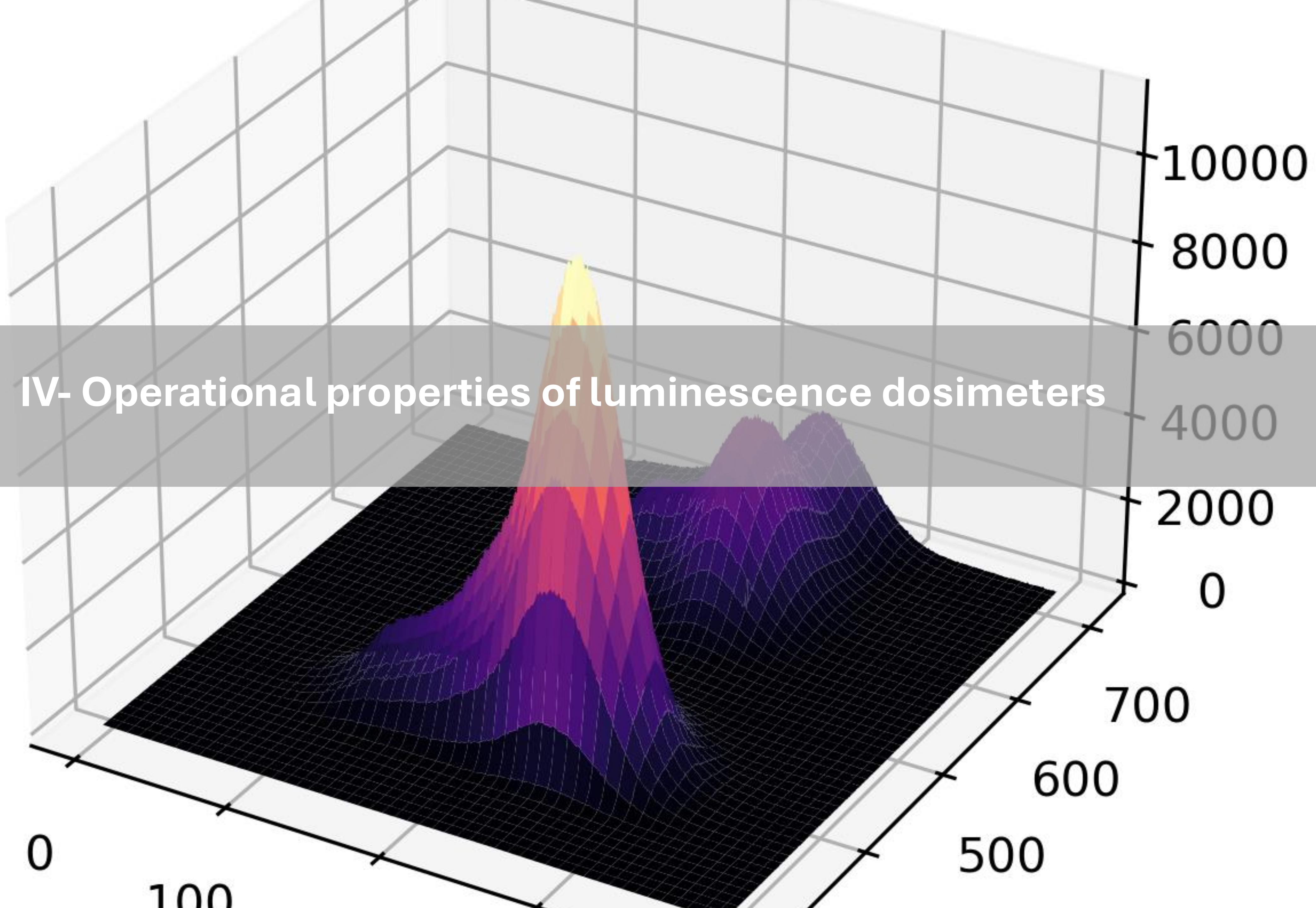
Factors limiting the luminescence efficiency

- I. Dosimetry concept
- II. Luminescence Dosimetry – Why?
- III. TL/OSL/RPL
- IV. Operational properties
- V. Case studies

➢ Average luminescence efficiency: 3-4 % only…



IV- Operational properties of luminescence dosimeters

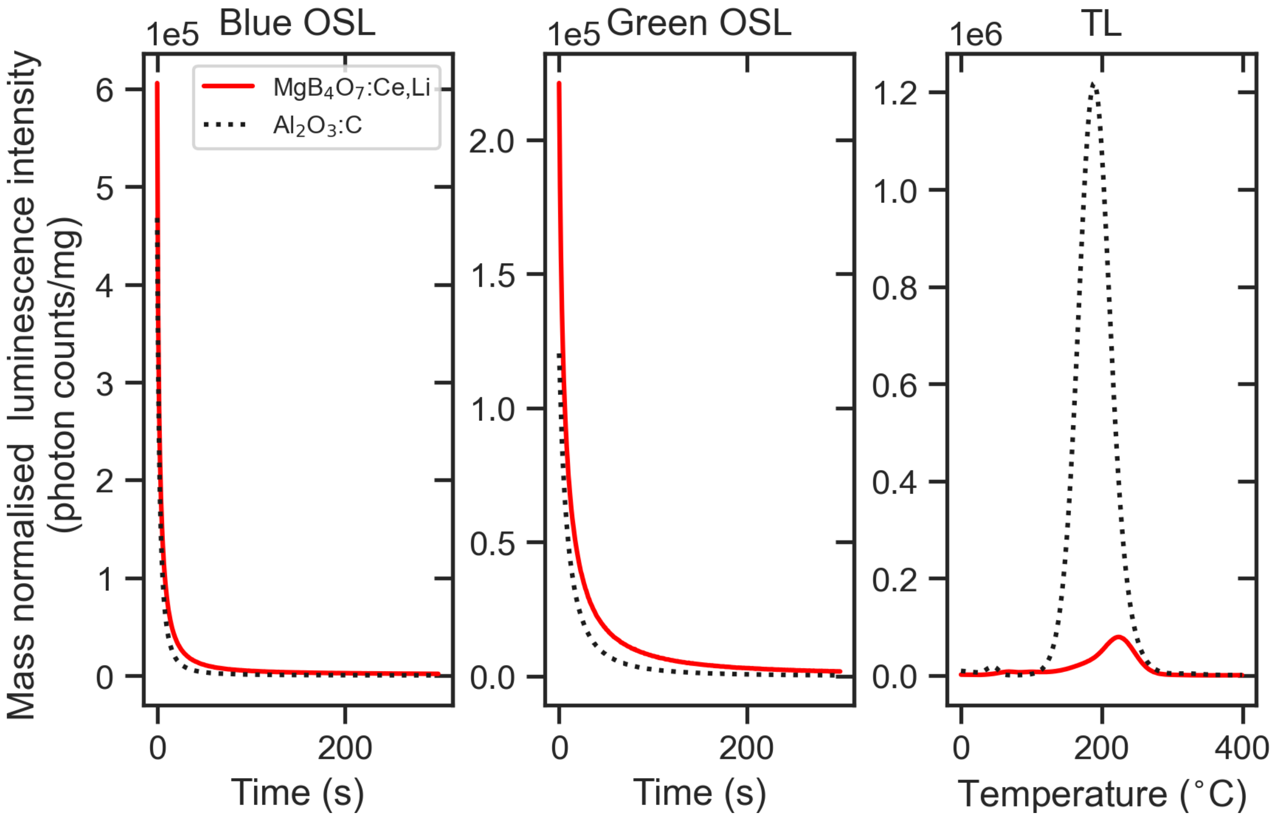


Operational properties of luminescence dosimeters – what you should consider

Is the system I am choosing good enough for my application?

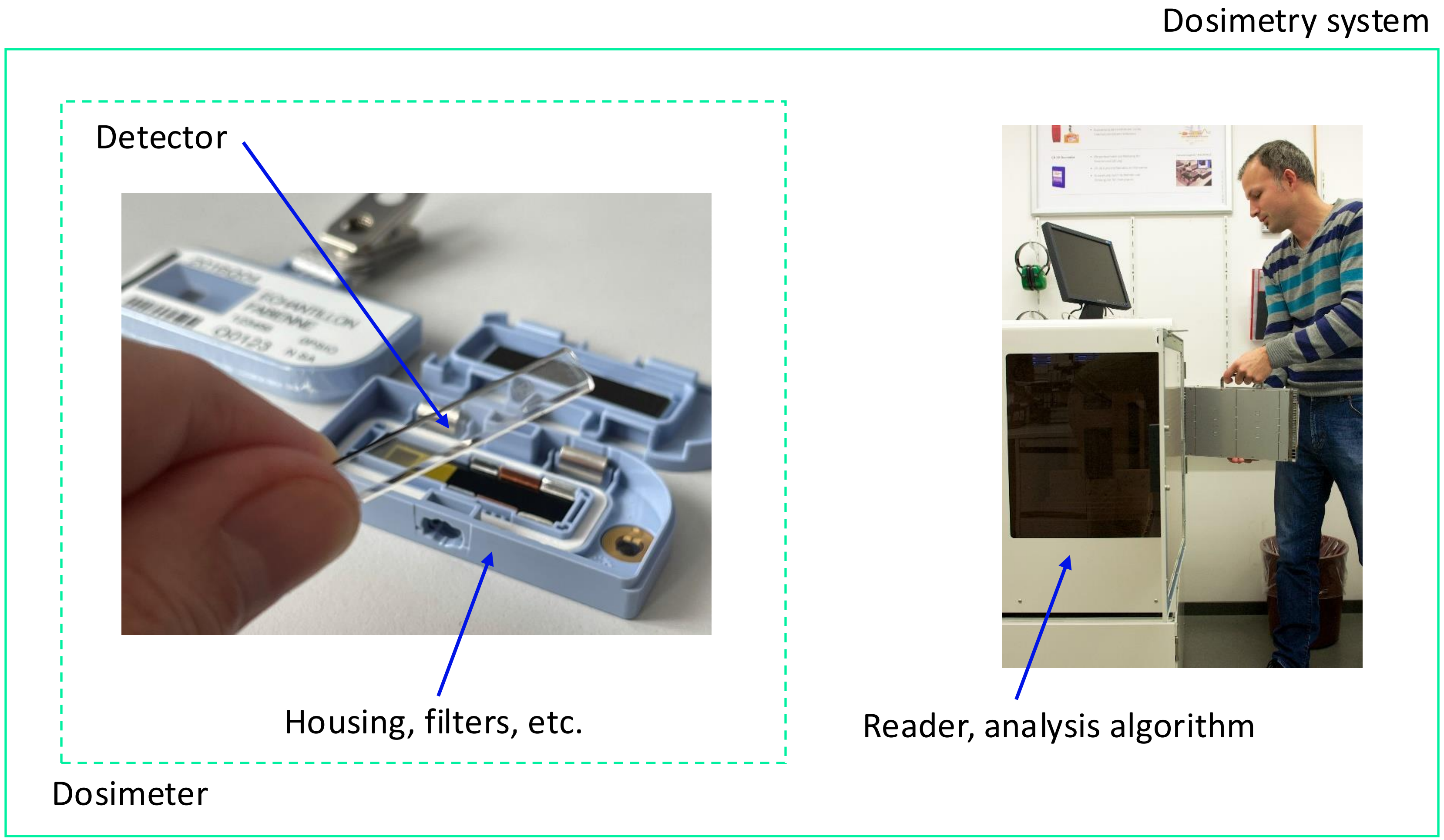
- Do I need to detect low doses (µGy)? High doses (kGy)?
- Do I need to correct for energy response in the range I am interested in?
- Is my signal stable over time?
- …

Sensitivity



Sensitivity of MgB_4O_7:Ce,Li under blue light stimulation, green light stimulation and thermal stimulation compared to that of Al_2O_3:C.

Detection limit



... is a characteristic of the entire system:

- **How much signal does the detector emit**
  - **How well the system can detect it**

Emission spectrum

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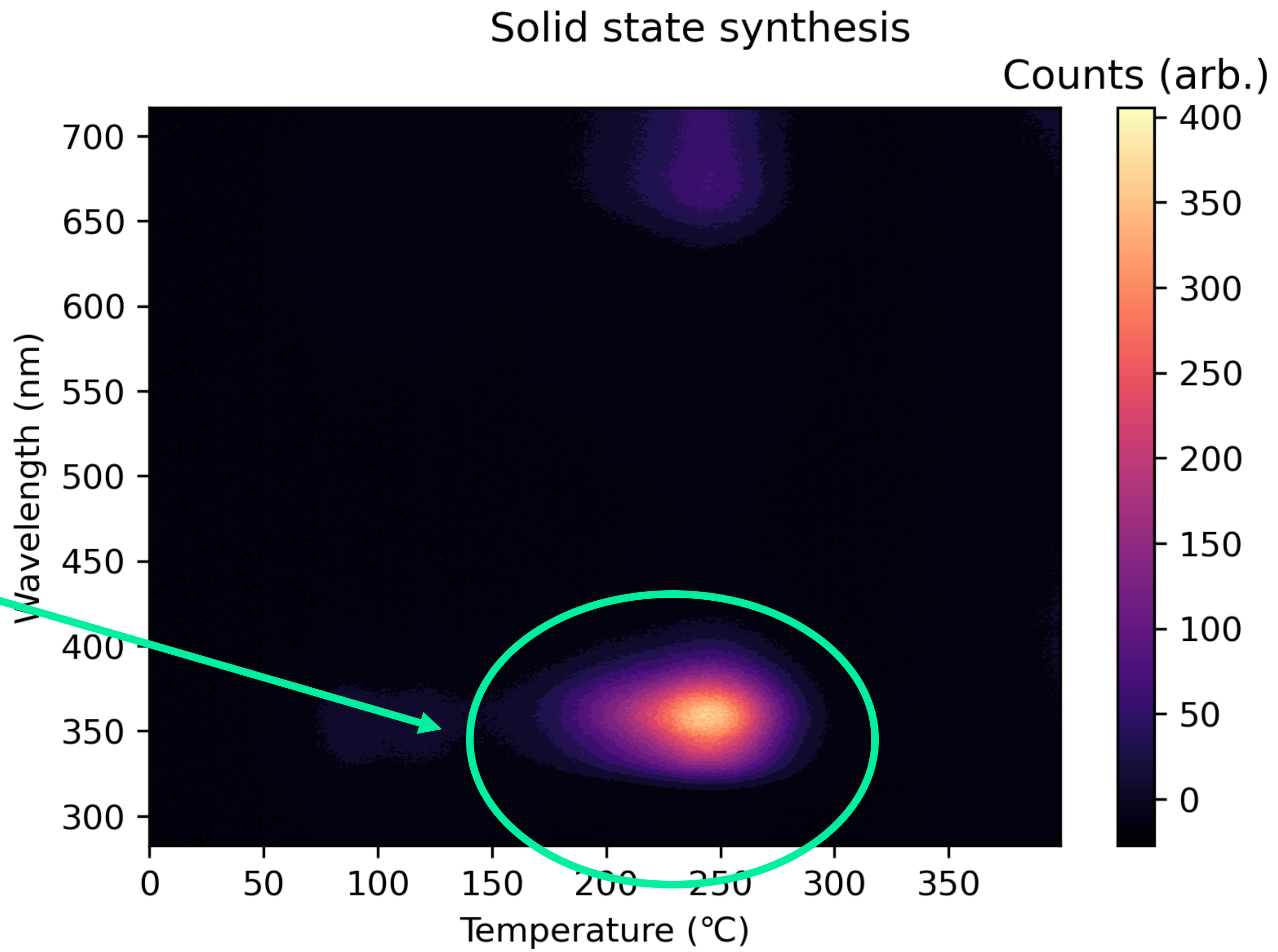
Recombination centre:
Ce^{3+} (≈350 nm)



Adapt detection unit (photomultiplier response, filter) to emission spectrum. For OSL, should differ from stimulation wavelength.

Saturation limit

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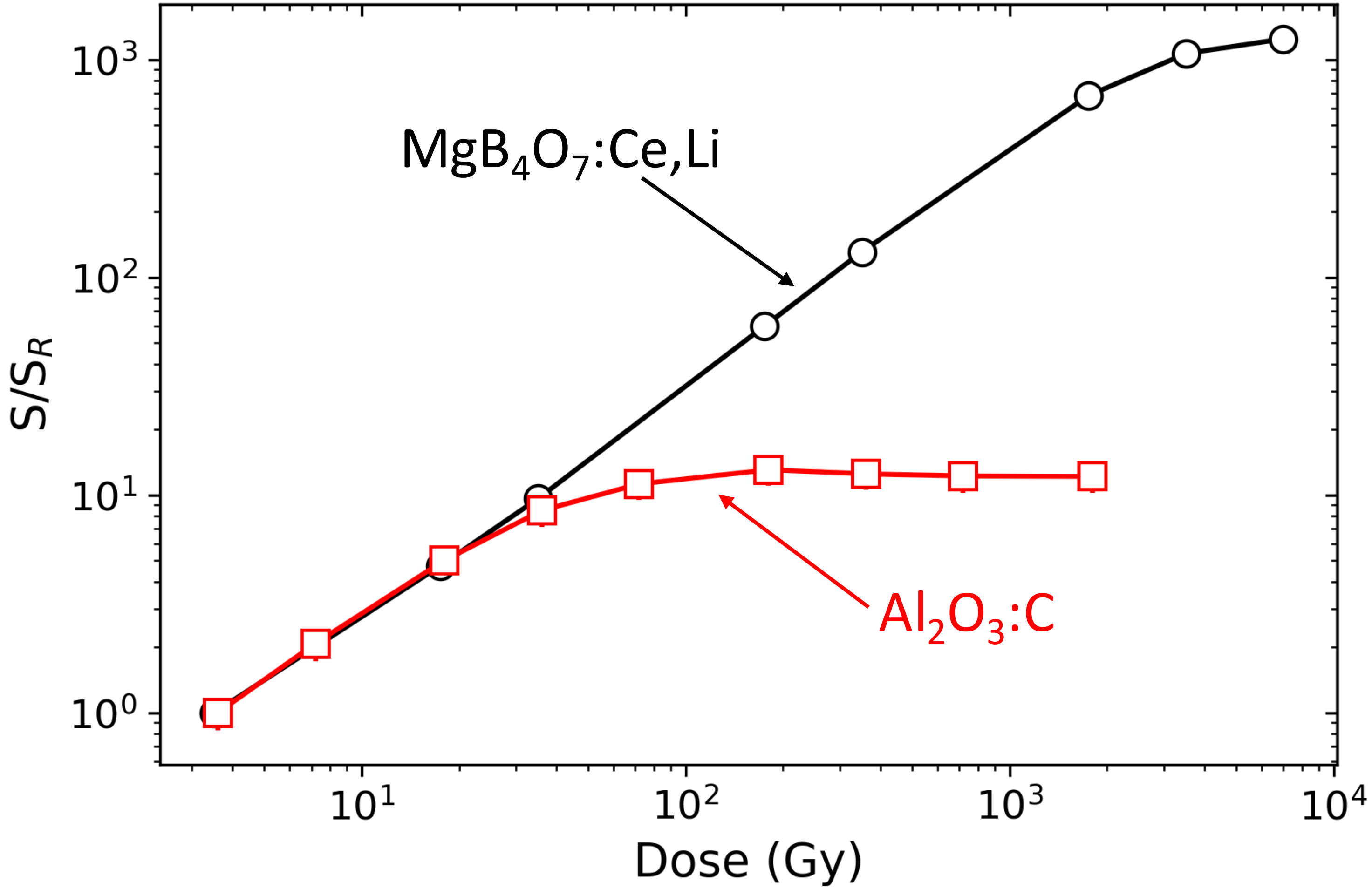
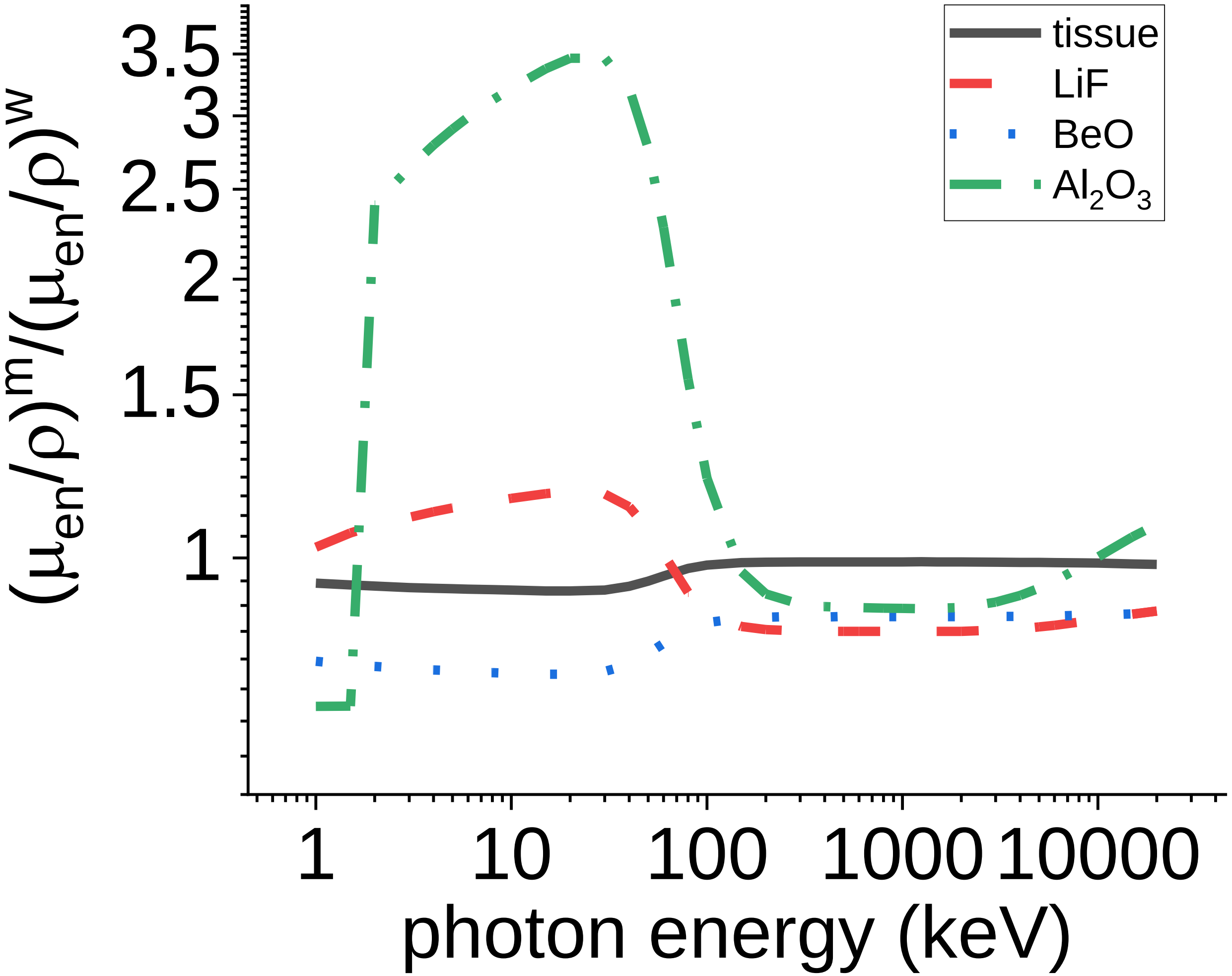


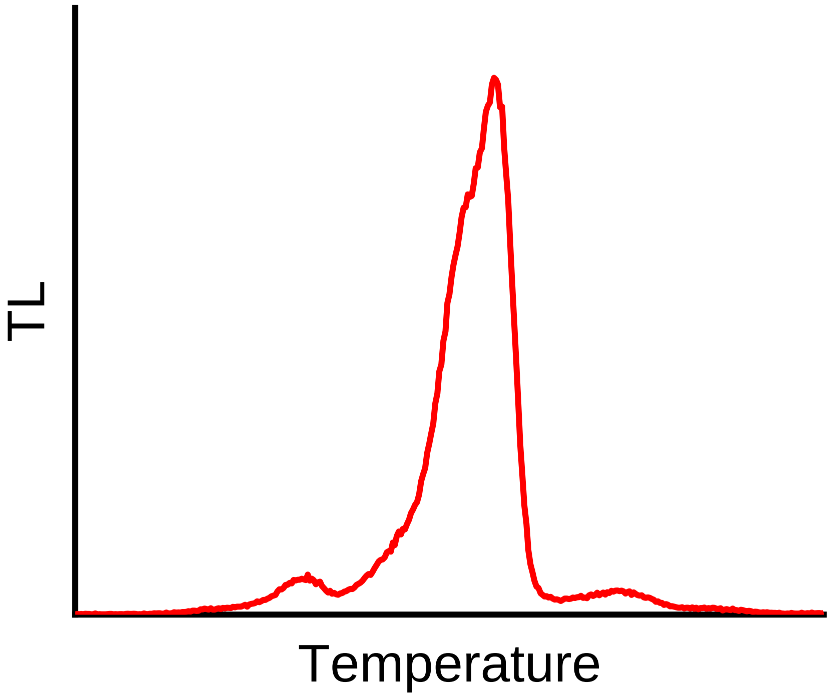
All materials will saturate eventually, some earlier than others.

Energy response



Signal depends on the energy of incoming photon

Stability of the signal



Which peak will fade more rapidly?

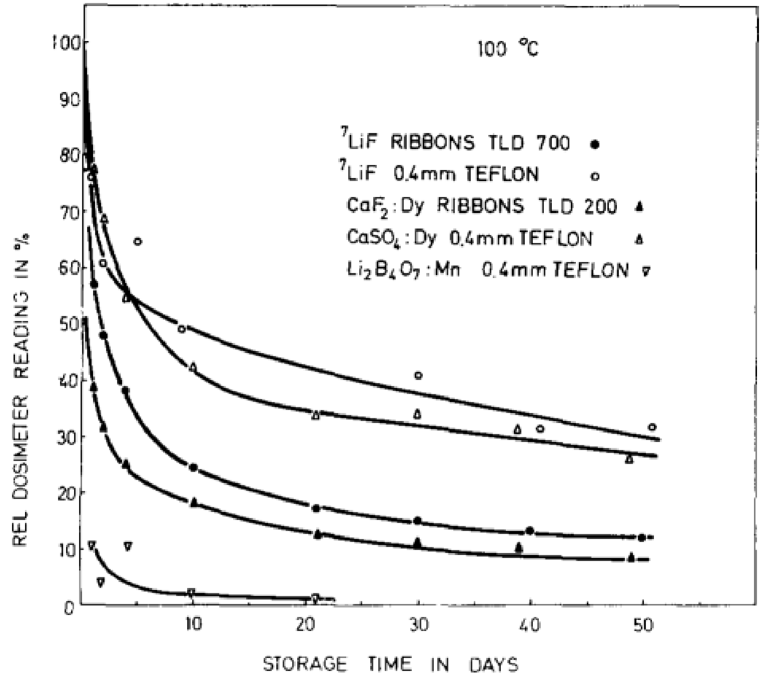


Fig. 16. Fading of TL at 100 °C in TLD 200 and TLD 700 ribbons as well as in LiF: Mg, Ti, in $CaSO_4$: Dy and in $Li_2B_4O_7$: Mn (teflon 0.4 mm thickness).

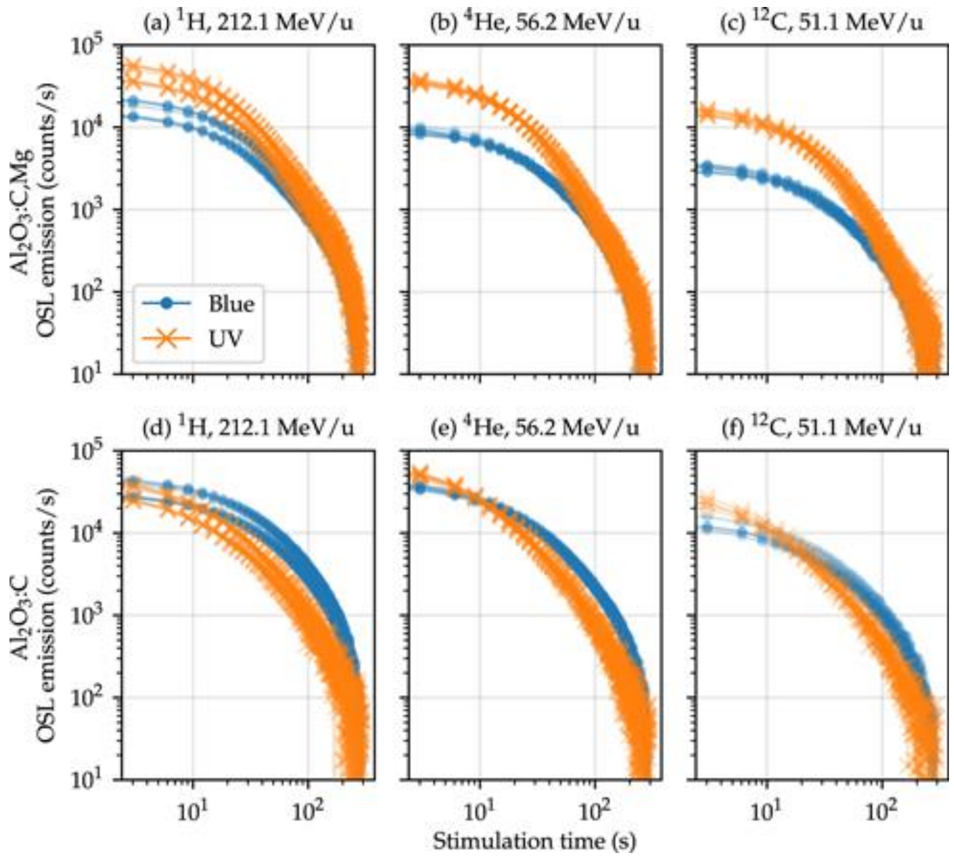
Burgkhardt et al., 1976

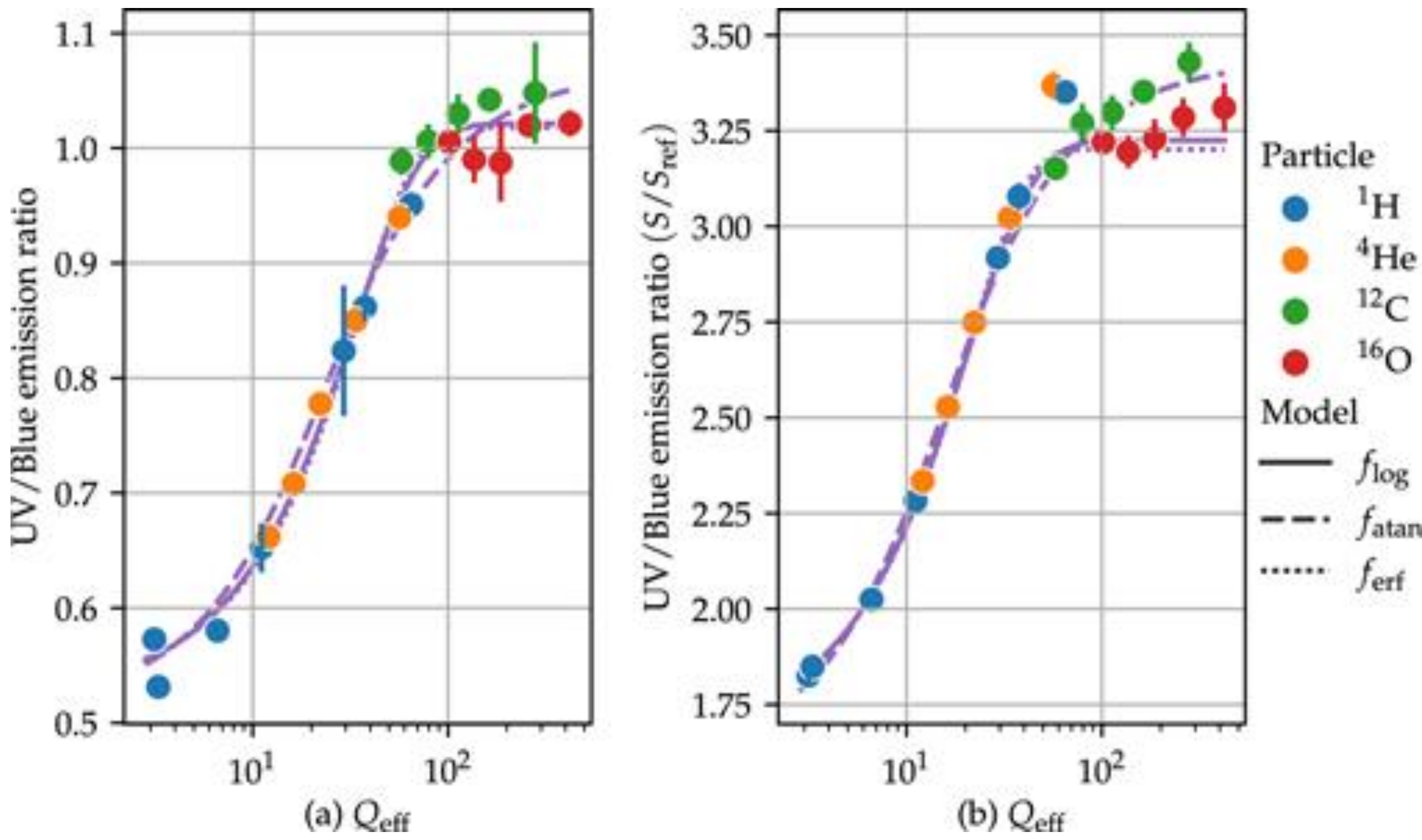
Fading rate depends on trap energy. Fading rate will depend:

- **On material**
- **On trap considered**
- **On storage condition**

Ability to measure linear energy transfer (LET)

Christensen et al., 2023





Al_2O_3:C has two emission bands (blue and UV)…

… and their ratio depends on the LET of the incoming particle.

➢ **Relevance for particle therapy treatment planning.**

Where does this LET dependence come from??

Low LET particle

Deposits energy more uniformly
- ➔ Isolated excitation regions
- ➔ Less localised damage
- ➔ Less local saturation of traps and recombination centre
- ➔ Higher luminescence efficiency

High LET particle

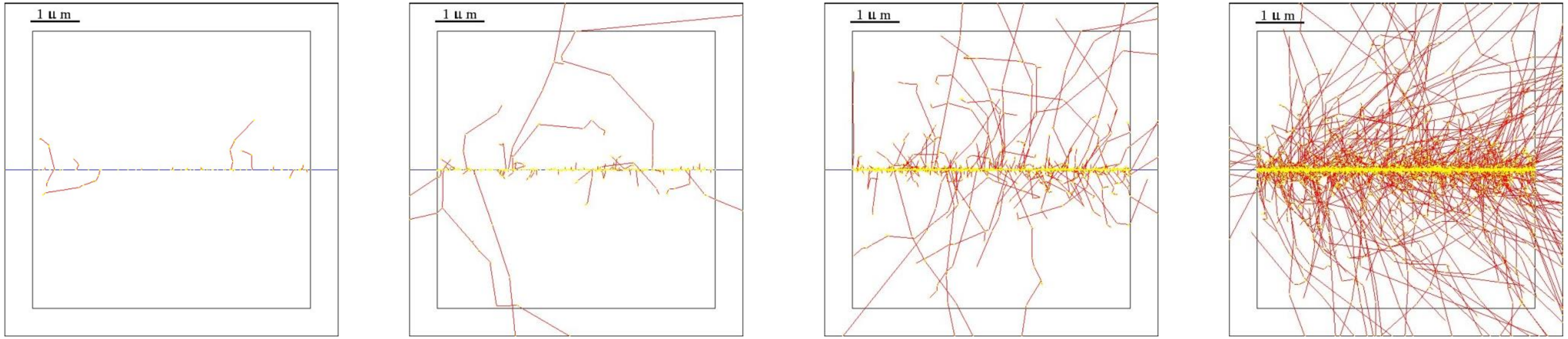
Deposits energy densely amongst its tracks
- ➔ Dense ionisation tracks
- ➔ More localised damage
- ➔ Traps and recomabination along the tracks are easily saturated
- ➔ More competition effects
- ➔ Lower luminescence efficiency

He 144.2 MeV/u — 2.24 keV/μm

O 385.5 MeV/u — 19.8 keV/μm

Ar 450.5 MeV/u — 93.3 keV/μm

Fe 122.9 MeV/u — 421 keV/μm



Courtesy: Dr. Gabriel O. Sawakuchi.

Choose your material wisely

There is no “perfect” luminescence dosimeter that would be able to tick all of those boxes…
→ be wise and select your material.

There are options available, know your material!

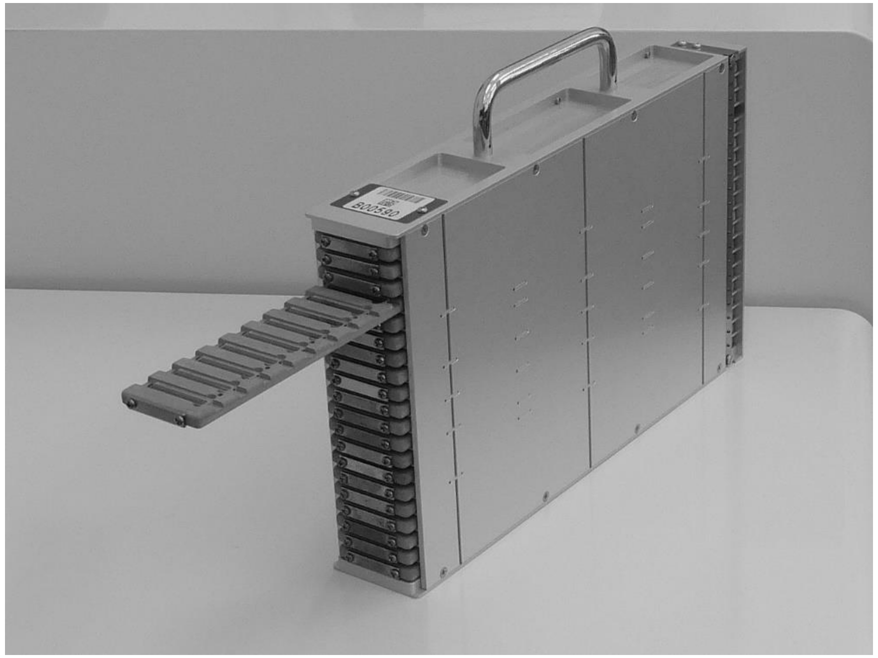
Table 1
Summary of TL and OSL materials most used in dosimetry. Most of the data are from McKeever et al. (1995) with a few updates, as indicated with the additional references; for OSL properties, see (Bøtter-Jensen et al., 2003; Yukihara and McKeever, 2011). The linearity ranges are those summarized in ISO/ASTM 51956 (ISO/ASTM, 2013b), also based on data from McKeever et al. (1995).

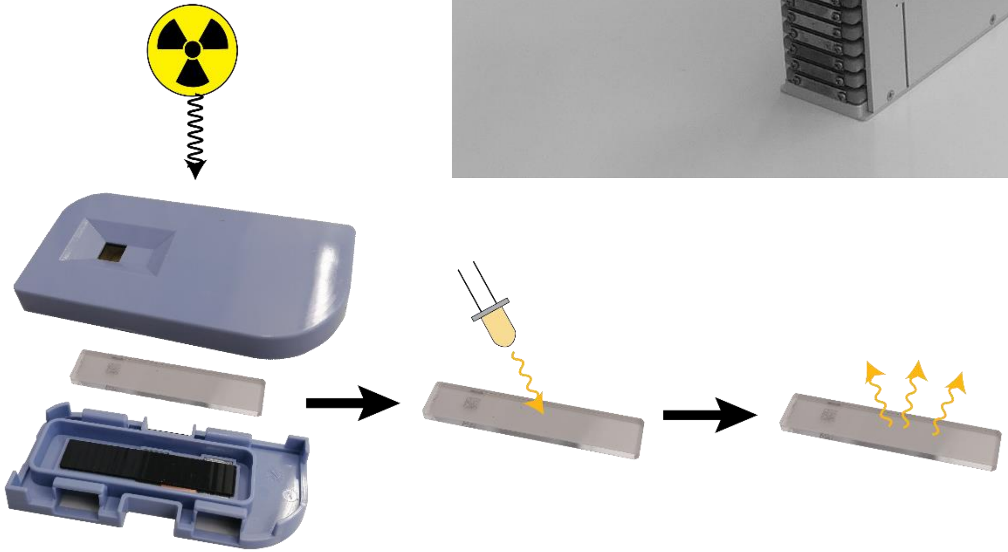
Material	Technique	Z_{eff} (host)	Comments
LiF:Mg,Ti	TL	8.3	Widely used in individual and area monitoring, and in medical dosimetry. TL sensitivity and curve shape influenced by aggregated defects that change with annealing and time. Linear up to 1 Gy, supralinear 1 Gy–10^3 Gy.
LiF:Mg,Cu,P	TL	8.3	High sensitivity, but cannot be heated above 240 °C without loss of sensitivity. Linear up to 10 Gy, then sublinear. High-temperature TL can be used >10^3 Gy.
LiF:Mg,Cu,Si	TL	8.3	Kim et al. (2022)
CaF_2:Mn	TL	16.9	Linear up to 10 Gy, supralinear up to 10^3 Gy.
CaF_2:Dy	TL	16.9	Linear up to 6 Gy, supralinear up to 500 Gy.
CaF_2:Tm	TL	16.9	Linear up to 1 Gy, supralinear up to 10^4 Gy.
Al_2O_3:C	TL/OSL	11.3	High TL and OSL sensitivity, broad, complex single TL peak. Linear up to 1 Gy, supralinear up to 30 Gy.
Al_2O_3:C,Mg	TL/OSL	11.3	Higher concentration of shallow traps in comparison with Al_2O_3:C and more aggregated defects.
Al_2O_3:Mg,Y		11.3	Linear up to 10^4 Gy.
BeO	TL/OSL	7.2	Low TL sensitivity; high OSL sensitivity. Linear up to 1 Gy, supralinear up to 100 Gy.
MgO	TL	10.8	Linear up to 10^4 Gy.
$CaSO_4$:Dy	TL	15.6	Linear up to 10 Gy, supralinear up to 10^3 Gy.
$CaSO_4$:Tm	TL	15.6	Linear up to 10 Gy, supralinear up to 10^3 Gy.
$Li_2B_4O_7$:Mn	TL	7.3	Linear up to 100 Gy, supralinear up to 10^4 Gy.
$Li_2B_4O_7$:Mn,Si	TL	7.3	Danilkin et al. (2006)
$Li_2B_4O_7$:Cu	TL	7.3	Linear up to 10^3 Gy.
MgB_4O_7:Dy	TL	8.5	Linear up to 50 Gy, supralinear up to 5 × 10^3 Gy.
MgB_4O_7:Tm	TL	8.5	Linear up to 50 Gy, supralinear up to 5 × 10^3 Gy.

Yukihara et al., 2022

Instrumentation

Routine readers

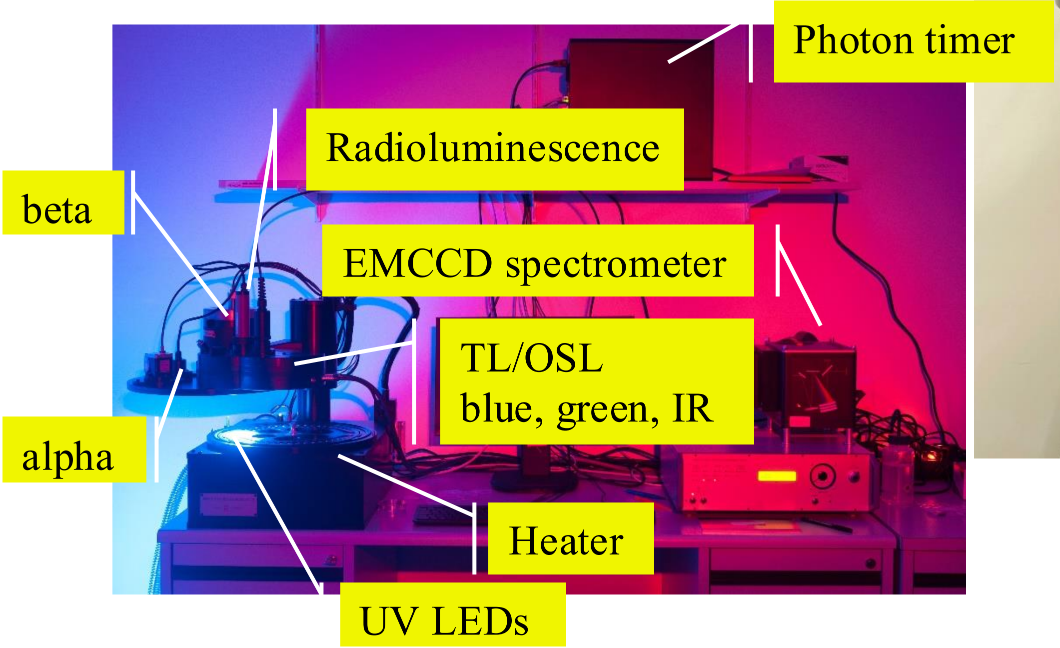


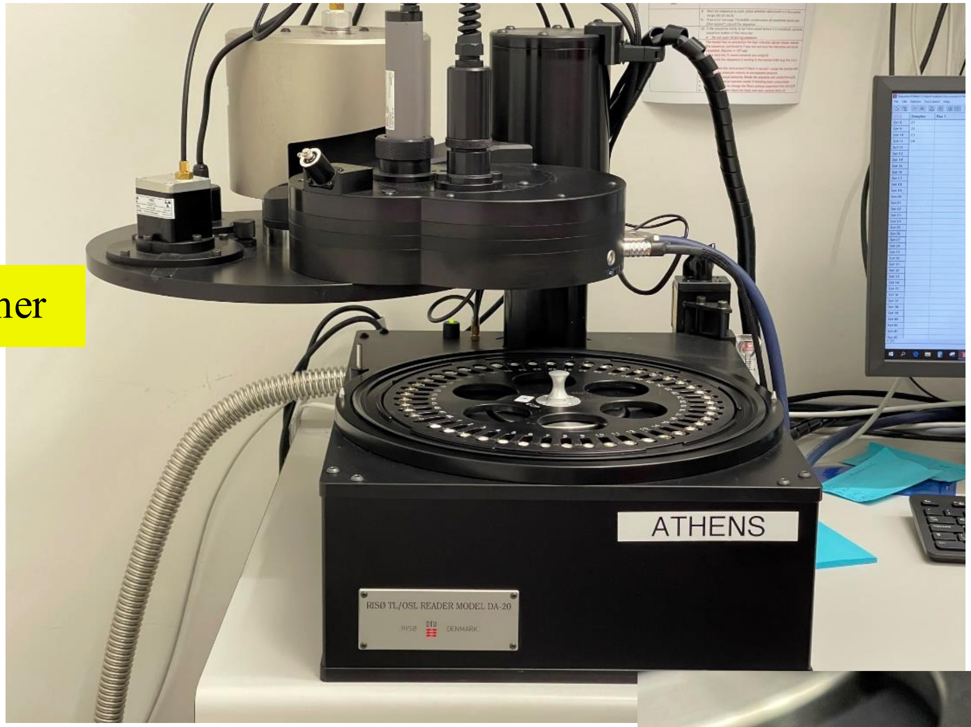


Instrumentation

PSI

Research readers



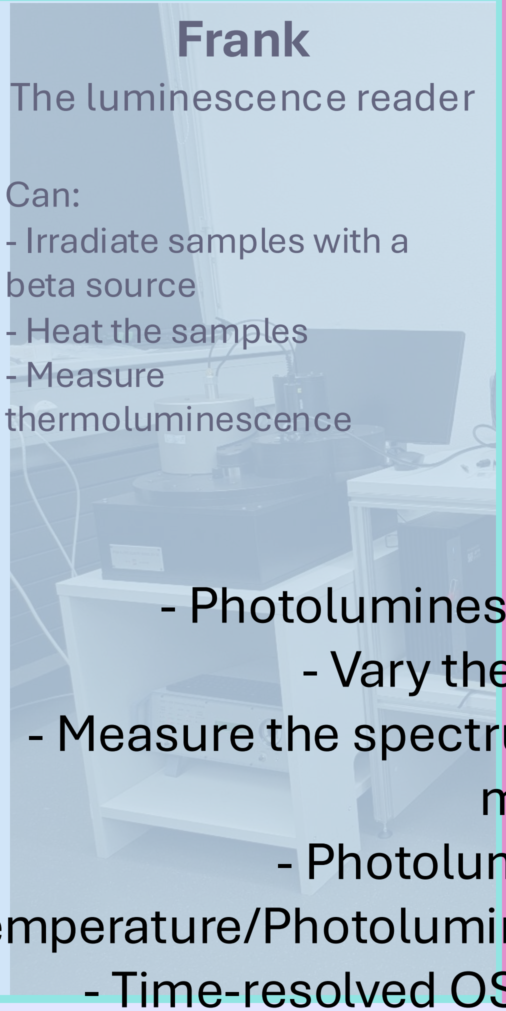


Instrumentation

Frank-and-Stein

Frank

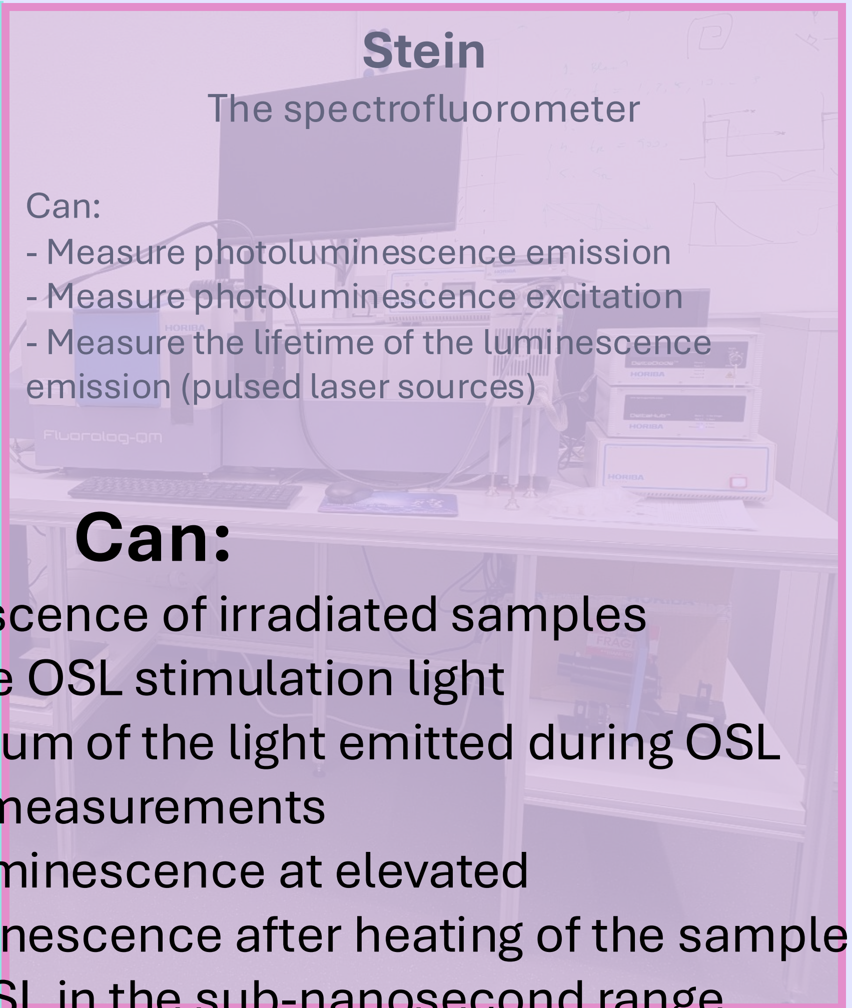
The luminescence reader



Can:
- Irradiate samples with a beta source
- Heat the samples
- Measure thermoluminescence

Stein

The spectrofluorometer

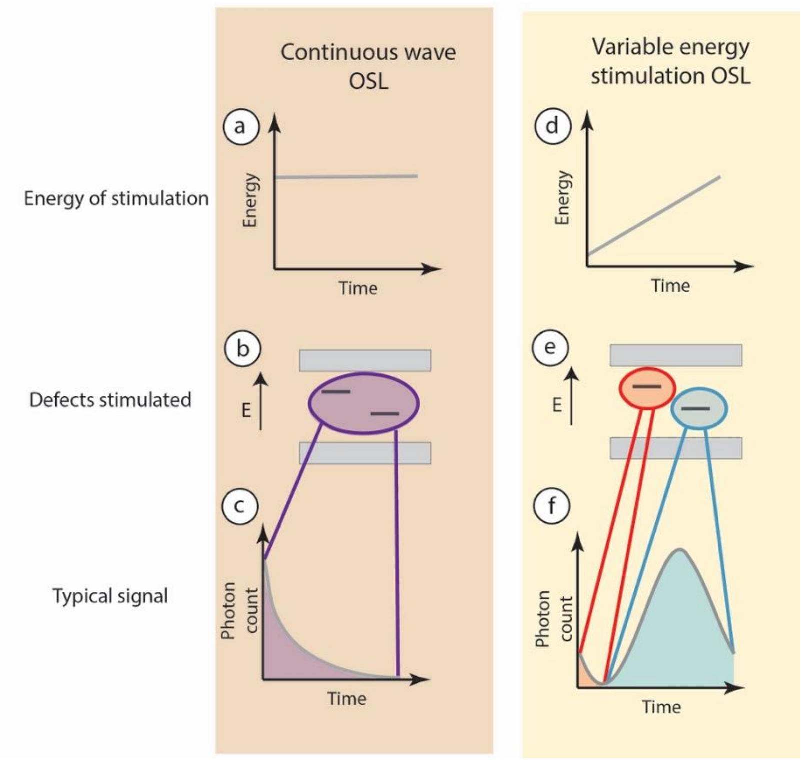


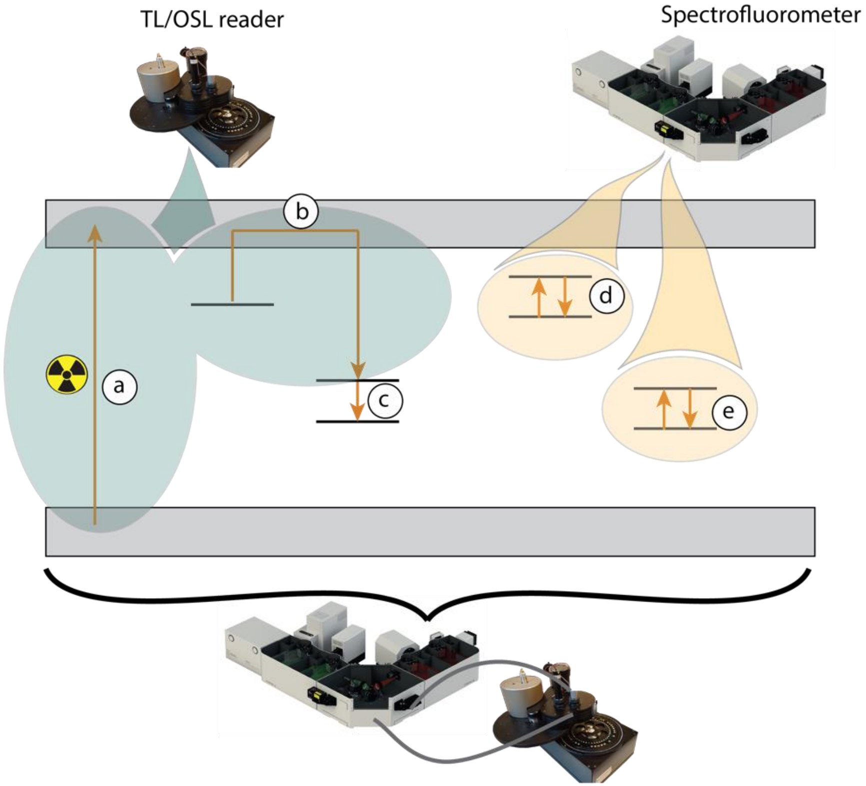
Can:
- Measure photoluminescence emission
- Measure photoluminescence excitation
- Measure the lifetime of the luminescence emission (pulsed laser sources)

Can:

- Photoluminescence of irradiated samples
- Vary the OSL stimulation light
- Measure the spectrum of the light emitted during OSL measurements
- Photoluminescence at elevated temperature/Photoluminescence after heating of the sample
- Time-resolved OSL in the sub-nanosecond range

Frank-and-Stein capabilities





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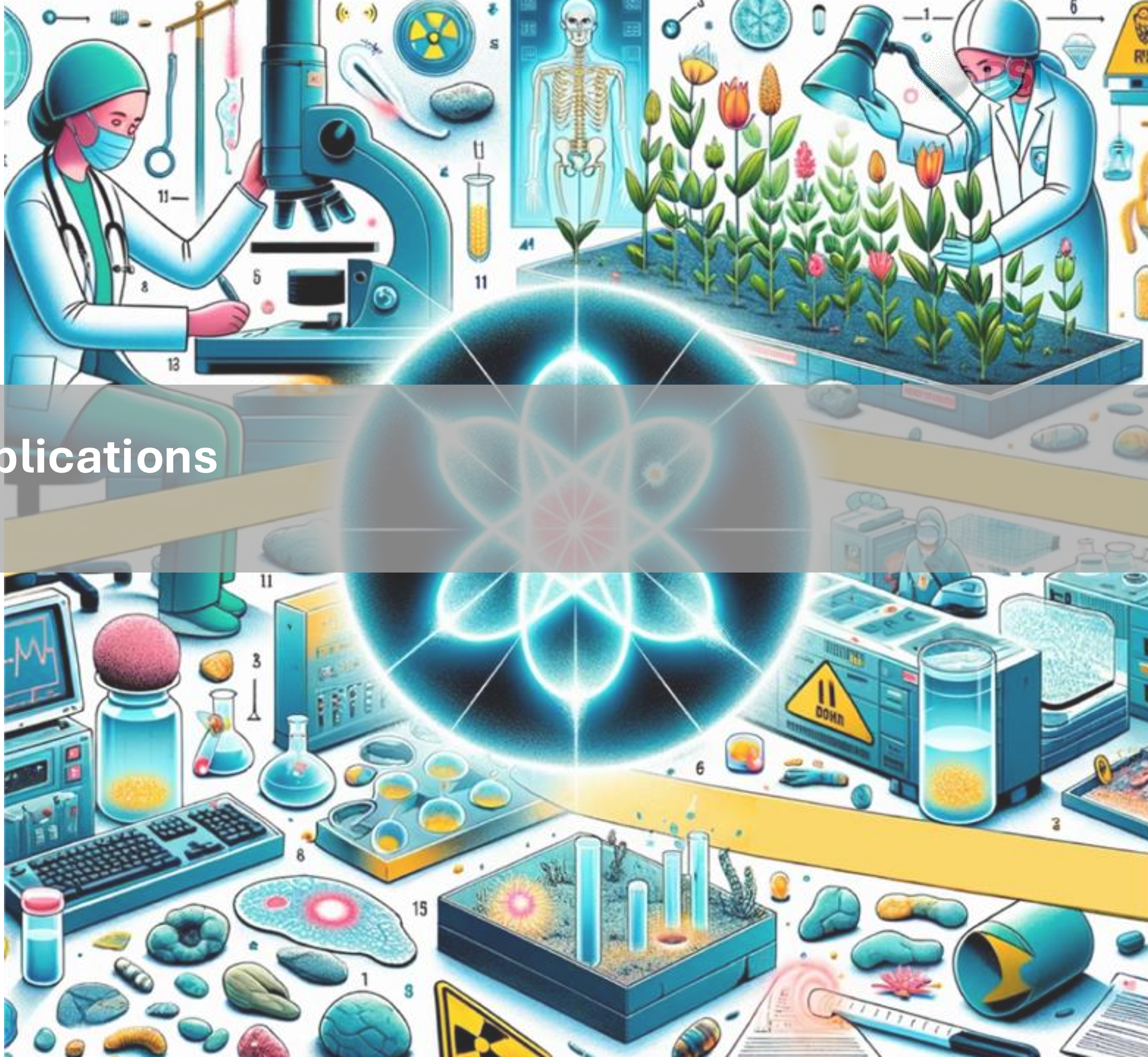
Luminescence dosimetry

Eduardo G. Yukihara[1], *Stephen W. S. McKeever*[2], *Claus E. Andersen*[3], *Adrie J. J. Bos*[4], *Ian K. Bailiff*[5], *Elisabeth M. Yoshimura*[6], *Gabriel O. Sawakuchi*[7], *Lily Bossin*[1] *and Jeppe B. Christensen*[1]

Abstract | Luminescence dosimetry is the process of quantifying the absorbed dose of ionizing radiation using detectors that exhibit luminescence. The luminescence intensity scales with energy absorbed from the radiation field. Calibration enables conversion of the luminescence intensity to the quantity of interest, for example the absorbed dose, kerma and personal dose equivalent. The different techniques available — thermoluminescence (TL), optically stimulated luminescence (OSL) and radiophotoluminescence (RPL) — share a common theoretical framework. Alongside applications in radiation protection, including personal dosimetry and area monitoring, luminescence dosimetry is also used in industry, research and medicine. Examples include quality assurance in radiation therapy, mapping of radiation levels in new accelerators, the estimation of ionizing radiation dose to organs in medicine and accidents, and the characterization of the radiation environment in space. The objective of this Primer is to summarize the fundamental concepts of luminescence dosimetry, the main experimental considerations, analysis procedures, typical results, applications and limitations, with an outlook into potential future advances.

IV. Case studies/Applications



Applications

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Table 3 | **Quantities of interest, quantities estimated and goals of the dosimetry for examples in different fields of application, with useful references when available**

Field	Sub-field	Typical quantities of interest	Quantity estimated	Goal
Radiation protection	Personal dosimetry	$H_p(d)$	Absorbed dose at a depth in the body; such as $d = 0.07$ mm, 3 mm or 10 mm	Conservative estimate of the radiation protection quantity effective dose E (REF.[30])
	Area monitoring	$H^*(d)$	Absorbed dose at a depth in the ICRU sphere; for example $d = 10$ mm	Conservative estimate of the radiation protection quantity effective dose E (REF.[30])
	Space dosimetry	D	Absorbed dose to water D at the dosimeter's position	Quantification of the low-LET part (photons, electrons, high-energy protons) of the complex radiation field[88]
Biology	Radiobiology experiments	D	Absorbed dose D to biological samples	Quantification of the absorbed dose in biological samples, to be correlated with biological endpoints[161]
Accident and epidemiological studies	Retrospective dosimetry	D	Absorbed dose D to minerals in ceramic building material such as brick and tile, or in teeth of human remains	Reconstruction of the dose to groups of individuals for epidemiological studies[89]
	Accident dosimetry	D	Absorbed dose D to fortuitous materials	Reconstruction of the dose to exposed individuals for triage after an accident or for future epidemiological studies[89]
Chronometry, with application to environmental and archaeological sciences	Dating of archaeological artefacts	D	Cumulative absorbed dose D to luminescent mineral grains from natural environmental radiation since a resetting event, for example an anthropogenic heating event	Estimate the age of the resetting event, such as the firing of pottery[38,39]
	Luminescence dating of sediments	D	Cumulative absorbed dose D to luminescent mineral grains from natural environmental radiation since a resetting event, for example exposure to sunlight followed by burial of sediment	Estimate the age of a natural or anthropogenic depositional process[38–40]
	Environmental dosimetry	D	Absorbed dose D to a luminescent detector placed in the environment for a defined measurement period	Estimation of the natural dose rate to the luminescent mineral grains in their burial context from the gamma component and cosmic rays[38,39,162]
Medical	Postal audits	D	Absorbed dose to water D in a specific point in a phantom	Quality assurance programme involving the comparison of the quality of dose delivery from different medical facilities[55]
	In vivo and in/on-phantom dosimetry in radiation therapy and radiodiagnostics	D, K_i, K_e	Absorbed dose to water D at a specific point in the patient or phantom (entrance or exit of the beam, in a cavity close to an organ at risk) incident air kerma K_i, entrance surface air kerma K_e	Quality assurance, demonstration that a certain dose to an organ at risk is not exceeded, verification of doses to patient devices such as a pacemaker[163,164], establishment of guidance levels, optimization[64], provide data for estimation of organ doses and correlation with, or estimation of, secondary effects[64,105]
Industrial applications	Control of instrument output	D, K	Absorbed dose D or air kerma K	Verification of equipment performance
	Radiation hardness	D	Absorbed dose to material D of electronic component, such as silica	Establish the correlation between electronic failure and the exposure to the radiation field
	Food processing and sterilization	D	Absorbed dose D to a material or to water	Quality assurance of the dose delivered in industrial processes[7,165]

LET, linear energy transfer.

Yukihara et al. Nature Rev. Meth. Primer 2:26 (2022).

Applications

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1. Measuring doses following radiological catastrophes
2. Measuring doses and LET for new radiotherapy treatments
3. Constraining the exhumation rate of mountains
4. Characterisation of phosphors

- *Issue addressed*
- *Methodology*
- *Experimental steps*
- *Knowledge gained*

Measuring doses following radiological catastrophes

The application of retrospective luminescence dosimetry in areas affected by fallout from the Semipalatinsk Nuclear Test Site: an evaluation of potential

I.K. Bailiff[*], V. F. Stepanenko[†], H.Y. Göksu[‡], H. Jungner[§], S.B. Balmukhanov[**], T.S. Balmukhanov[**], L.G. Khamidova[†], V.I. Kisilev[††], I.B. Kolyado[††], T.V. Kolizshenkov[†], Y.N. Shoikhet[††], A.F. Tsyb[†].

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I. Dosimetry concept

II. Luminescence Dosimetry – Why?

III. TL/OSL/RPL

IV. Operational properties

V. Case studies

Measuring doses and LET for new radiotherapy treatments

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Improved simultaneous LET and dose measurements in proton therapy

Jeppe Brage Christensen[1]✉, Michele Togno[2], Lily Bossin[1], Oskari Ville Pakari[1], Sairos Safai[2] & Eduardo Gardenali Yukihara[1]

The objective of this study was to improve the precision of linear energy transfer (LET) measurements using Al_2O_3:C optically stimulated luminescence detectors (OSLDs) in proton beams, and, with that, improve OSL dosimetry by correcting the readout for the LET-dependent ionization quenching. The OSLDs were irradiated in spot-scanning proton beams at different doses for fluence-averaged LET values in the (0.4–6.5) $keV\,\mu m^{-1}$ range (in water). A commercial automated OSL reader with a built-in beta source was used for the readouts, which enabled a reference irradiation and readout of each OSLD to establish individual corrections. Pulsed OSL was used to separately measure the blue (F-center) and UV (F^{+}-center) emission bands of Al_2O_3:C and the ratio between them (UV/blue signal) was used for the LET measurements. The average deviation between the simulated and measured LET values along the central beam axis amounts to 5.5% if both the dose and LET are varied, but the average deviation is reduced to 3.5% if the OSLDs are irradiated with the same doses. With the measurement procedure and automated equipment used here, the variation in the signals used for LET estimates and quenching-corrections is reduced from 0.9 to 0.6%. The quenching-corrected OSLD doses are in agreement with ionization chamber measurements within the uncertainties. The automated OSLD corrections are demonstrated to improve the LET estimates and the ionization quenching-corrections in proton dosimetry for a clinically relevant energy range up to 230 MeV. It is also for the first time demonstrated how the LET can be estimated for different doses.

The proton therapy community increasingly focuses on the relative biological effectiveness (RBE) and the effect of the linear energy transfer (LET) distributions in treatment plans[1]. Several detectors, such as semiconductors[2], gas counters[3], and radiochromic films[4], have been proposed to measure the LET of hadrons at conventional dose-rates. Nevertheless, dosimetry in hadron beams remains challenging because of the typical under-response due to the ionization quenching of otherwise dose-rate independent detectors[5–7].

Ionization quenching is occasionally exploited to estimate the LET by comparing the response of two detectors having different quenching characteristics, e.g. scintillators relative to absorbed dose calorimeters[8] or scintillators relative to ionization chambers[9]. Similarly, pairs of differently quenching organic scintillators[10], thermoluminescent detectors[11] or phosphor films[12] have been used to simultaneously estimate dose and LET, which, however, is associated with large uncertainties in regions with steep gradients, e.g. at the distal edge.

The optically stimulated luminescence (OSL) of Al_2O_3:C allows the estimation of both dose and LET, but with the advantage of only requiring a single detector, and has been demonstrated[13] to be dose-rate independent in proton beams up to 150 $kGy\,s^{-1}$.

Al_2O_3:C OSL is associated[14] with a fast UV emission (centered at 335 nm, lifetime < 7 ns) and a much slower blue emission (centered at 420 nm, 35 ms lifetime). The emission in the blue band is traditionally used for dosimetry with a negligible time dependence after irradiation[14]. The emission in the UV band is less favorable for dosimetry, but it has been demonstrated that the ratio of the two emission bands, measured using pulsed OSL (POSL), can be used to establish an LET calibration curve and estimate the LET in proton beams[15–18], and heavier ions[7]. However, previous proton LET studies using Al_2O_3:C focused on LET estimations for constant doses around 0.2 Gy and LETs below the elevated LET at the distal edge[19,20].

Previous studies on the possibility of LET estimation using OSL relied on custom-build readers designed to achieve the type of specialized POSL measurements required for this finality[7,15]. More recently, automated OSL readers capable of POSL measurements, which offers more controlled and stable readout than previous readers,

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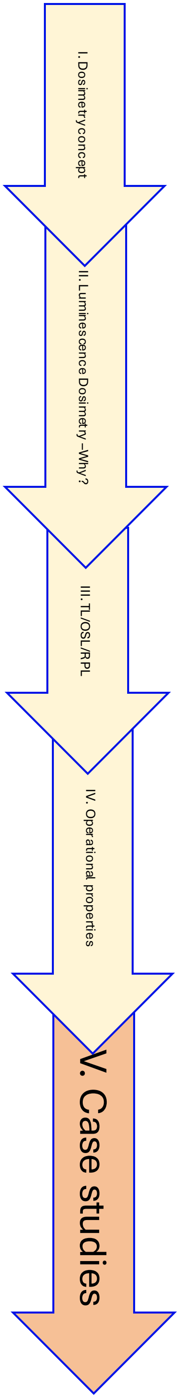

Constraining the exhumation rate of mountains

Chemical Geology 446 (2016) 3–17

Contents lists available at ScienceDirect

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Trapped-charge thermochronometry and thermometry: A status review

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ARTICLE INFO

Article history:
Received 28 September 2015
Received in revised form 16 August 2016
Accepted 18 August 2016
Available online 20 August 2016

Keywords:
Low-temperature thermochronometry
Thermometry
Trapped-charge dating
Electron spin resonance (ESR)
Thermoluminescence (TL)
Optically stimulated luminescence (OSL)
Infra-red stimulated luminescence (IRSL)

ABSTRACT

Trapped-charge dating methods including luminescence and electron spin resonance dating have high potential as low temperature (< 100 °C) thermochronometers. Despite an early proof of concept almost 60 years ago, it is only in the past two decades that thermoluminescence (TL), electron-spin-resonance (ESR), and optically stimulated luminescence (OSL), have begun to gain momentum in geological thermochronometry and thermometry applications. Here we review the physics of trapped-charge dating, the studies that led to its development and its first applications for deriving palaeo-temperatures and/or continuous cooling histories. Analytical protocols, which enable the derivation of sample specific kinetic parameters over laboratory timescales, are also described. The key limitation of trapped-charge thermochronometry is signal saturation, which sets an upper limit of its application to < 1 Ma, thus restricting it to rapidly exhuming terrains (> 200 °C Ma^{-1}), or elevated-temperature underground settings (> 30 °C). Despite this limitation, trapped-charge thermochronometry comprises a diverse suite of versatile methods, and we explore potential future applications and research directions.

1. Introduction

The need to constrain the rate and timing of landscape evolution has led to a continuous growth of thermochronometric techniques, which quantify the thermal histories of rocks (Reiners and Ehlers, 2005). A suite of methods are applicable to different temporal and spatial scales, however constraining recent (<1 Ma) thermal histories at temperatures <100 °C remains challenging. Luminescence and electron-spin-resonance (ESR) dating are trapped-charge dating methods whose thermal sensitivities can span this temporal gap. They are based on the quantification of free electric charge (electrons and holes), which become trapped in the proximity of various defects and impurities in the crystalline lattice of minerals (e.g. quartz, feldspar) as a result of their exposure to environmental radiation (cf. Aitken, 1985). This charge can be evicted by exposure of the crystal to external energy such as heat, light and/or pressure, and hence its concentration can be related to the last exposure of natural materials to high temperature (Aitken et al., 1968; Brown et al., 2009). Therefore, trapped-charge techniques can be used to gain insights into the thermal histories of rocks.

Although the possibility of interpreting trapped charge within natural crystals as records of their thermal histories was initially demonstrated more than half a century ago (Houtermans et al., 1957), this technique received only marginal attention from the geological community, initially for surface palaeothermometry (e.g. Ronca and Zeller, 1965), and later for characterising lunar surface temperatures (e.g. Durrani et al., 1977). With an increasing interest for quantifying recent stages of rock thermal histories from the thermochronological community (Reiners and Ehlers, 2005) and the need to constrain the rate and timing of landscape evolution during the Quaternary, trapped-charge dating methods utilising ESR, thermoluminescence (TL), and optically stimulated luminescence (OSL) were (re)investigated in the context of low-temperature thermochronometry (Grün et al., 1999; Tsuchiya and Fujino, 2000; Herman et al., 2010; Guralnik et al., 2015a; King et al., 2016a). In particular, OSL-thermochronometry has been the focus of rapid development since its introduction in 2010, and has come to be recognized as a new developing field of luminescence dating (Duller, 2015a, 2015b; Roberts and Lian, 2015). In its simplest form, trapped-charge thermochronometry comprises constraining the interplay between (i) the rate of charge trapping, due to exposure to ionising radiation, and (ii) the rate of charge detrapping, due to temperature (Christodoulides et al., 1971). By constraining charge trapping and detrapping rates on a sample-specific basis, the natural concentrations of trapped charge can be translated into ages and their corresponding palaeotemperatures.

Here we aim to provide a brief overview of the underlying physics of trapped-charge dating, describe the common equipment and key measurements of each sub-technique, and trace the development of trapped-charge thermochronometry from early pioneering studies to the current state-of-the-art. At a time when a range of new low-temperature thermochronometric techniques are under development (e.g. Tremblay et al., 2014; Shuster and Cassata, 2015; Amidon et al., 2015),

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http://dx.doi.org/10.1016/j.chemgeo.2016.08.023

Characterisation of phosphors

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Radiation Measurements 158 (2022) 106846

Contents lists available at ScienceDirect

Radiation Measurements

journal homepage: www.elsevier.com/locate/radmeas

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The quest for new thermoluminescence and optically stimulated luminescence materials: Needs, strategies and pitfalls

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ARTICLE INFO

Keywords:
Thermoluminescence
Optically stimulated luminescence
Dosimetry
Synthesis

ABSTRACT

The quest for new materials for thermoluminescence (TL) and optically stimulated luminescence (OSL) dosimetry continues to be a central line of research in luminescence dosimetry, occupying many groups and investigators, and is the topic of many publications. Nevertheless, it has also been a research area with many pitfalls, slow advances in our understanding of the luminescence processes, and rare improvements over existing materials. Therefore, this paper reviews the status of the field with the goal of addressing some fundamental questions: Is there a need for new luminescence materials for TL/OSL dosimetry? Can these materials be designed and, if so, are there strategies or rules that can be followed? What are the common pitfalls and how can they be avoided? By discussing these questions, we hope to contribute to a more guided approach to the development of new luminescent materials for dosimetry applications.

1. Introduction

Thermoluminescence (TL) and Optically Stimulated Luminescence (OSL) are phenomena widely used in radiation dosimetry and applied in different fields, such as personal and environmental dosimetry, medical dosimetry, imaging of ionizing radiation dose, archeological and geological dating and assessment of the severity of radiation accidents (McKeever, 1985; McKeever et al., 1995; Chen and McKeever, 1997; Bøtter-Jensen et al., 2003; Chen and Pagonis, 2011; Yukihara and McKeever, 2011; Yukihara et al., 2022b). Besides dosimetry applications, TL materials have also been explored as particle temperature sensors (Talghader et al., 2016; Yukihara et al., 2018), and OSL materials have been examined as rechargeable persistent phosphors for bio-imaging applications (Xu et al., 2018). OSL materials are also used as photostimulable phosphors in computed radiography (Leblans et al., 2011).

In such TL/OSL applications a key role is played by the luminescent material. Since the work on TL dosimetry materials by Daniels and colleagues and on OSL dosimetry materials by Antonov-Romanovskii in the 1950s (Daniels et al., 1953; Antonov-Romanovskii et al., 1955) there has been a continuous and extensive search for the "ideal" luminescent material that exhibits a linear dose-response relationship over the widest possible dose range, a high sensitivity, along with good neutron/gamma discrimination, tissue equivalency, reproducibility, and stability of the luminescent signal. With the expansion of TL/OSL to applications beyond personal and environmental dosimetry, the concept of the "ideal" material also has to be revised according to new applications. The historical development, properties and uses of various TL materials have been summarized in McKeever et al. (1995). Since then other reviews can be found for TL (Bhatt and Kulkarni, 2014) and for OSL materials (Pradhan et al., 2008; Nanto, 2018; Yanagida et al., 2019; Yuan et al., 2020).

Although many materials show promising TL/OSL properties, few have been used routinely or commercially in dosimetry (see Table 1). Available TL dosimetric materials are mostly limited to doped compounds of fluorides (LiF, CaF_2), simple oxides (Al_2O_3, BeO, MgO), borates (MgB_4O_7, and $Li_2B_4O_7$) and sulfates ($CaSO_4$). In the case of OSL, only two OSL materials are used in commercial dosimetry systems: Al_2O_3:C and BeO. Both are highly sensitive to ionizing radiation. For computed radiography other OSL materials such as BaFBr:Eu and CsBr:Eu are also available (Leblans et al., 2011; Nanto, 2018), but these were designed not for dosimetry, but for X-ray imaging, and have high effective atomic numbers ($Z_{eff} \geq 30$–50). Several other materials have been investigated for OSL dosimetry (Pradhan et al., 2008; Oliveira and

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https://doi.org/10.1016/j.radmeas.2022.106846
Received 21 March 2022; Received in revised form 26 July 2022; Accepted 16 August 2022
Available online 20 August 2022

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Questions?

Master projects available in our group!!

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