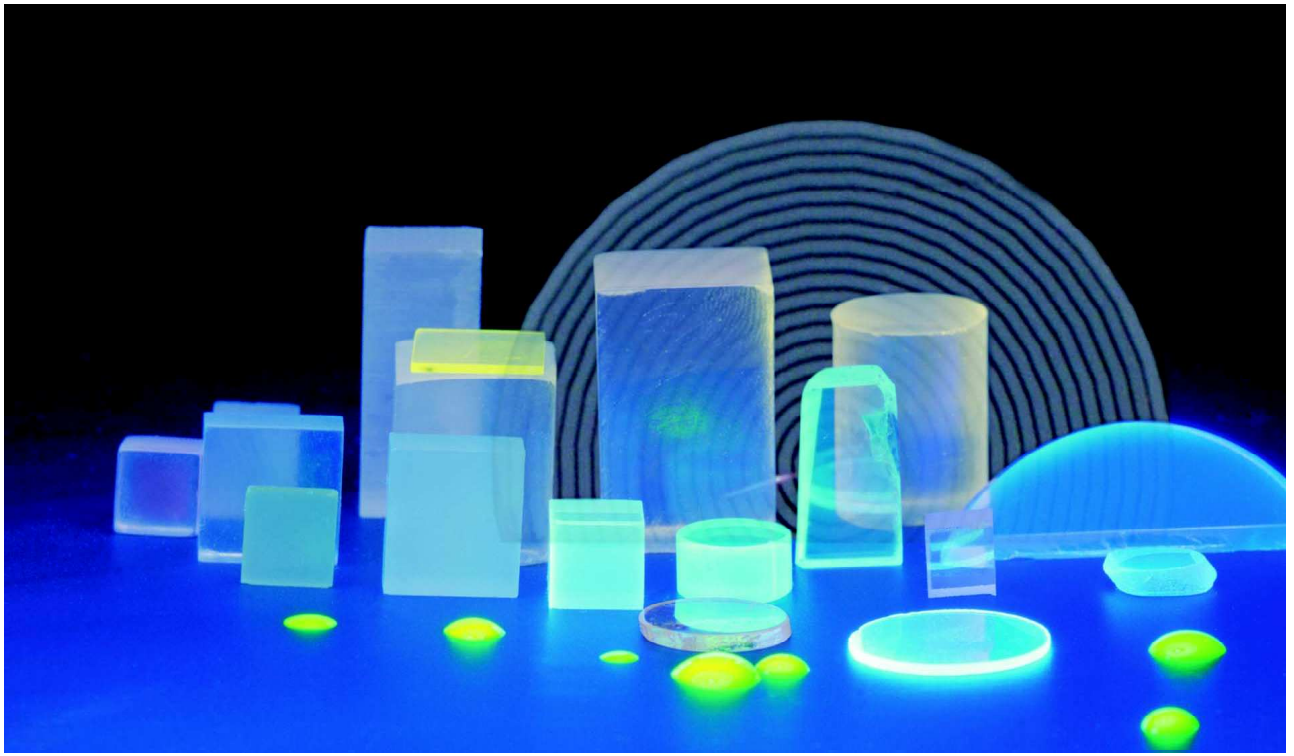
Current status and future prospects of inorganic scintillator research

Pieter Dorenbos



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Interfaculty Reactor Institute

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Outline

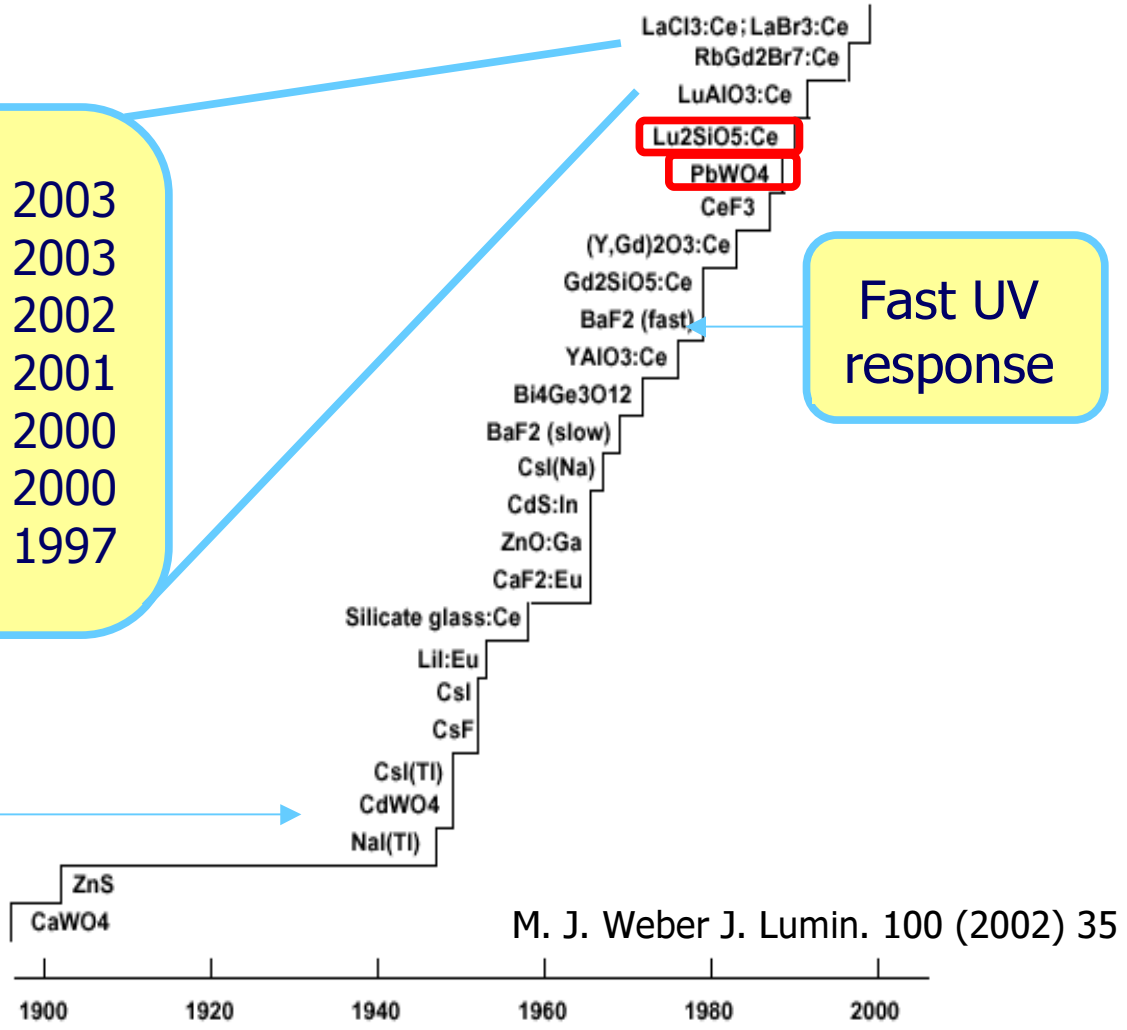
- Scintillators for low energy (<3 MeV) gamma detection
  - Ce^{3+} activated scintillators
  - intrinsic scintillators
- Scintillators for electromagnetic calorimeters (GeV-TeV)
- Requirements
  - speed
  - light output and energy resolution
  - density
- Future prospects and directions of research

History of scintillators

Cs_2LiYCl_6:Ce	2003
LuI_3:Ce	2003
K_2LaI_5:Ce	2002
$LaBr_3$:Ce	2001
$LaCl_3$:Ce	2000
$Lu_2Si_2O_7$:Ce	2000
$RbGd_2Br_7$:Ce	1997

Fast UV response

Invention of the photomultiplier tube



M. J. Weber J. Lumin. 100 (2002) 35

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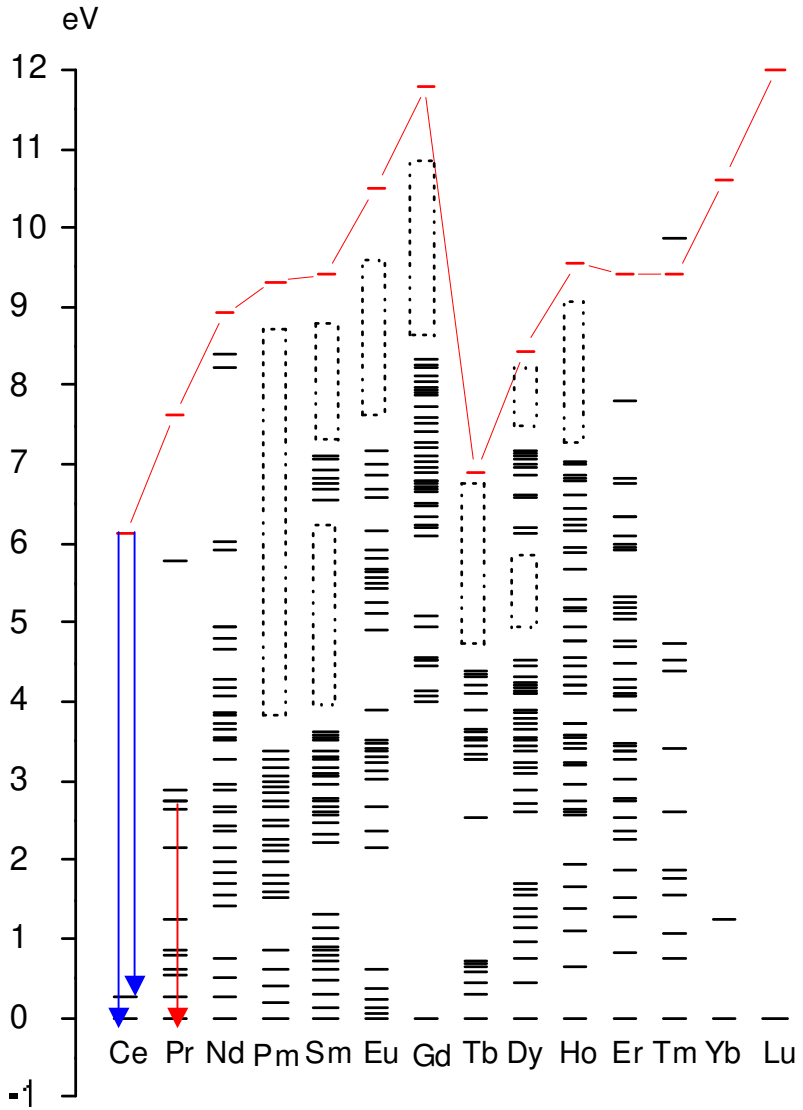

- allowed 5d→4f emission
- fast τ = 15-60 ns
- absence of slow 4f →4f emission
- λ_{em} depends on host
  - fluorides ≈300 nm
  - oxides ≈400 nm
  - sulfides ≈500 nm
- dopant in high density La, Gd, and Lu-compounds

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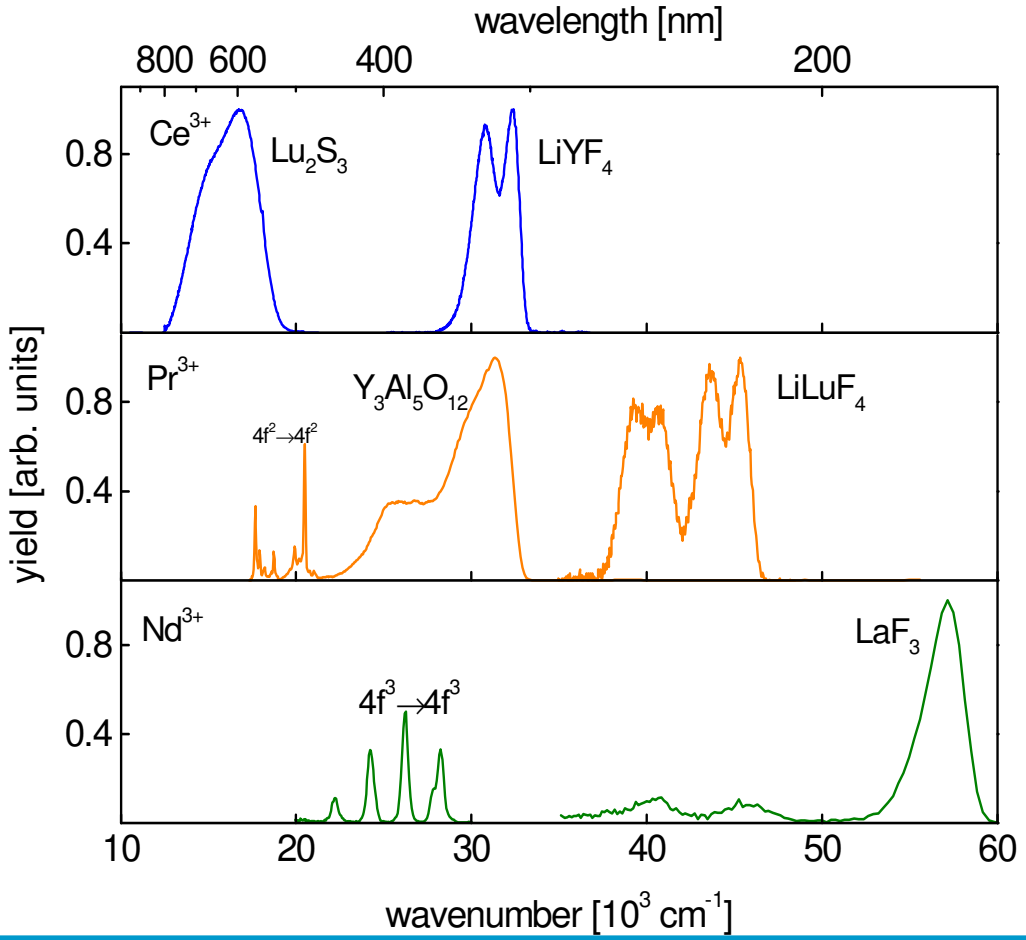
X-ray excited emission spectra of Ce^{3+}, Pr^{3+}, and Nd^{3+}



λ_{em} depends on

- lanthanide
- host

- 250 nm < λ^{Ce} < 700 nm
- 210 nm < λ^{Pr} < 400 nm
- λ^{Nd} < 200
- slow 4f→4f emission Pr and Nd

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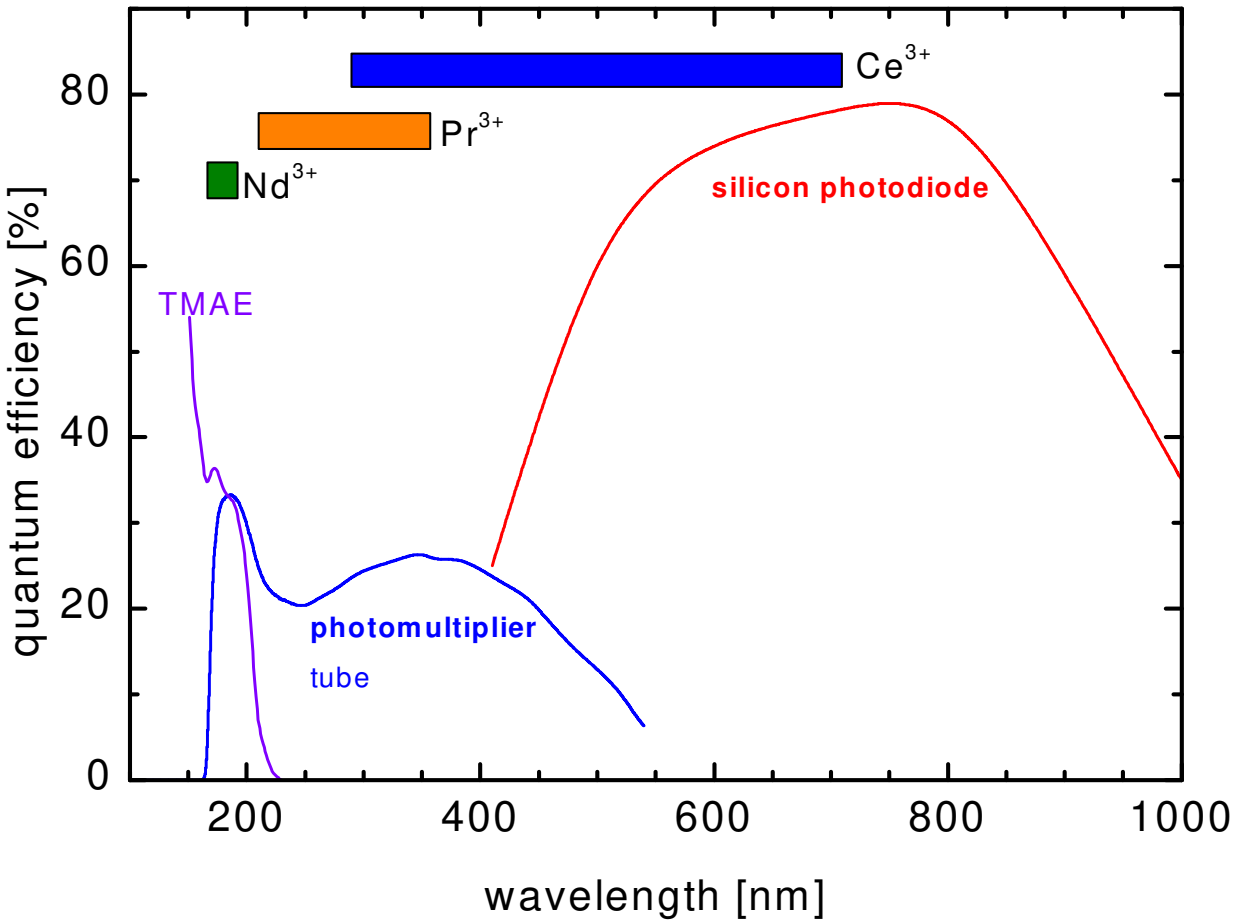
df emission in Ce, Pr, Nd



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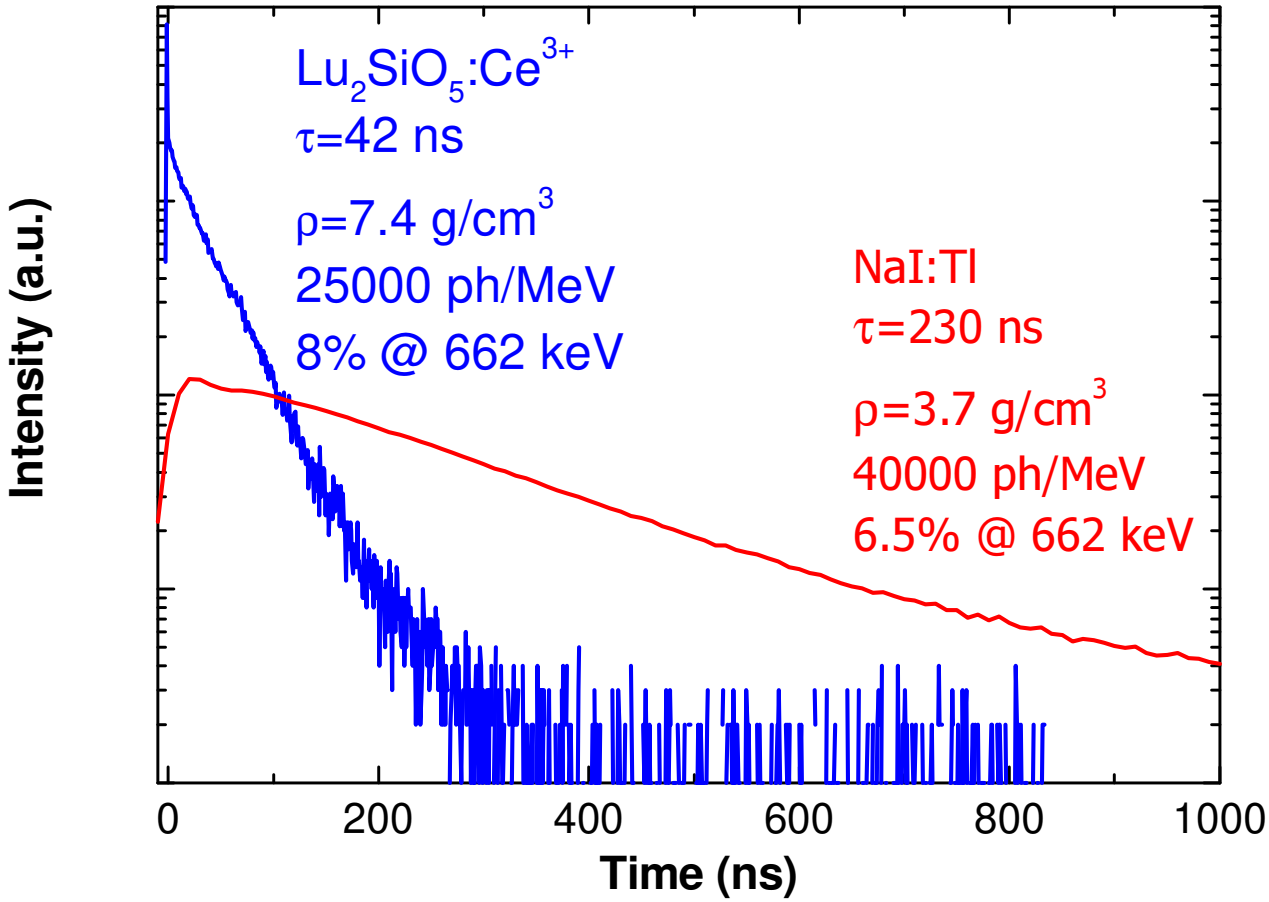
Lu2SiO5:Ce3+
τ=42 ns
ρ=7.4 g/cm3
25000 ph/MeV
8% @ 662 keV
NaI:Tl
τ=230 ns
ρ=3.7 g/cm3
40000 ph/MeV
6.5% @ 662 keV
Intensity (a.u.)
0
200
400
600
800
1000
Time (ns)

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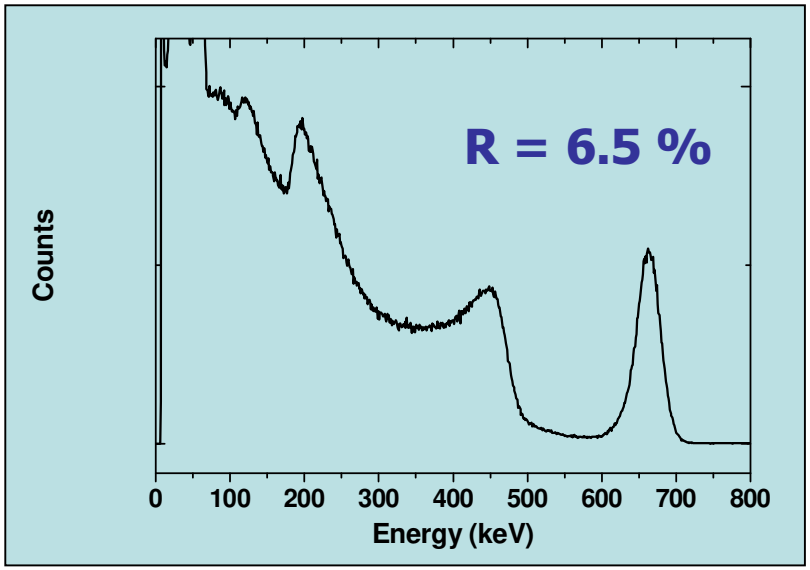
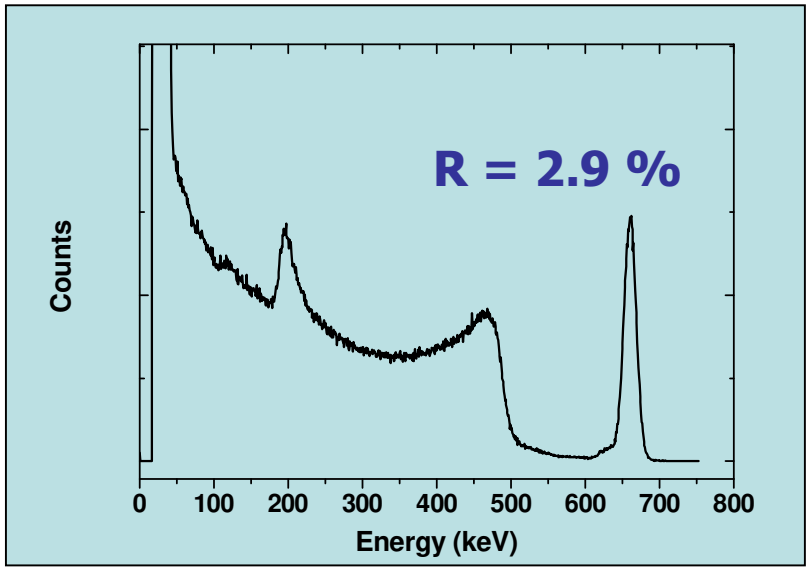
Scintillator performances

Pulse-height spectra (662 keV gamma rays)





NaI:Tl

43. 10^3 ph/MeV

230 ns

3.7 g/cm^3

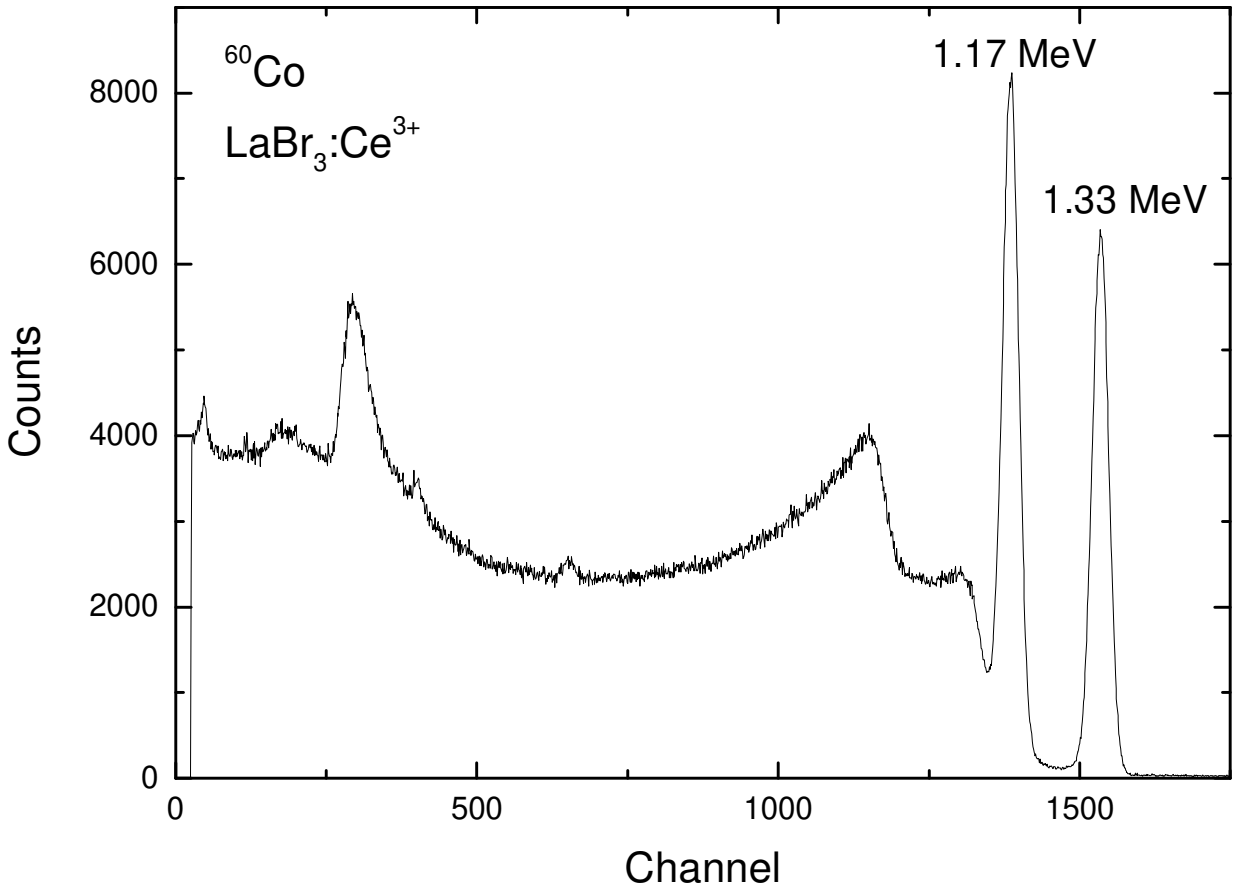
$LaBr_3$:Ce

61. 10^3 ph/MeV

18 ns

5.3 g/cm^3

^{60}Co spectrum measured with prototype $LaBr_3$:Ce scintillator



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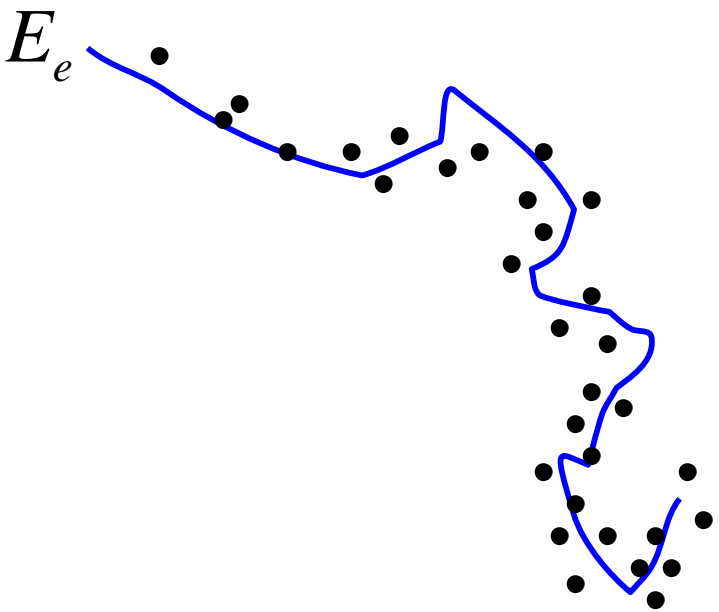
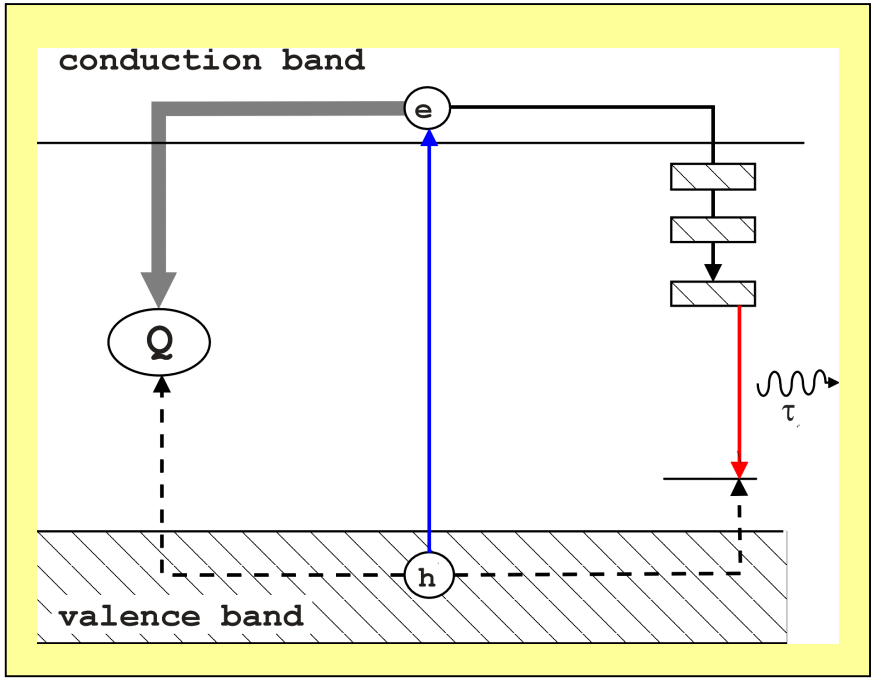
Speed of impurity activated scintillators

$$\tau_r = f\left(\tau_i, \tau_t, \tau_v, \tau_{lc}, \tau_{pd}\right)$$





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Radiative decay rate

$$\Gamma_v = \frac{1}{\tau_v} \propto \frac{n}{\lambda^3}\left(\frac{n^2+2}{3}\right)^2 \sum_f \left|\langle f|\mu|i\rangle\right|^2$$

- dipole and spin allowed transitions
- lanthanide 5d→4f emission Ce^{3+}, Pr^{3+}, Nd^{3+}, Eu^{2+}
- s^2-elements Tl^+, Pb^{2+}, Bi^{3+} (spin forbidden)
- charge transfer luminescence
  - core valence luminescence
  - $Yb^{2+} + h_{VB} \rightarrow Yb^{3+}$

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Limits to the decay time of impurity activated scintillators

activator	Dip. All.	Spin all.	τ (ns)
Ce^{3+}	d-f	Yes	17-60
Pr^{3+}	d-f	Yes	10-30
Nd^{3+}	d-f	Yes	5-15
Eu^{2+}	d-f	Yes	700-1000
Tl^{+}	p-s	No	> 200
Pb^{2+}	p-s	No	> 200
Bi^{3+}	p-s	No	> 200
STE	π-σ	No	>1000
CVL	CT	Yes	≈ 1

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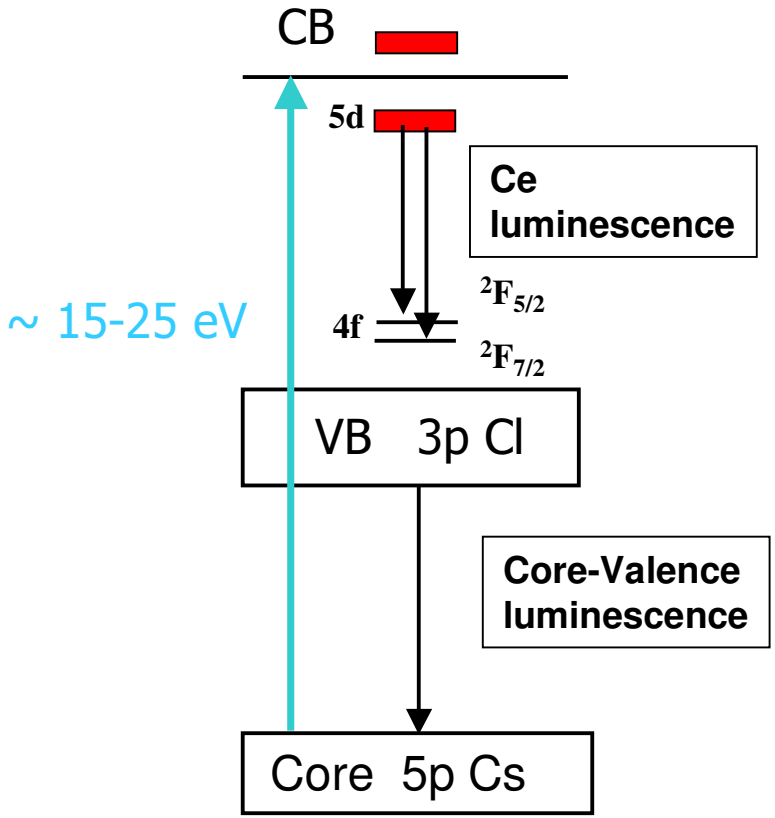
Core-valence Luminescence in $Cs_2LiYCl_6:Ce^{3+}$

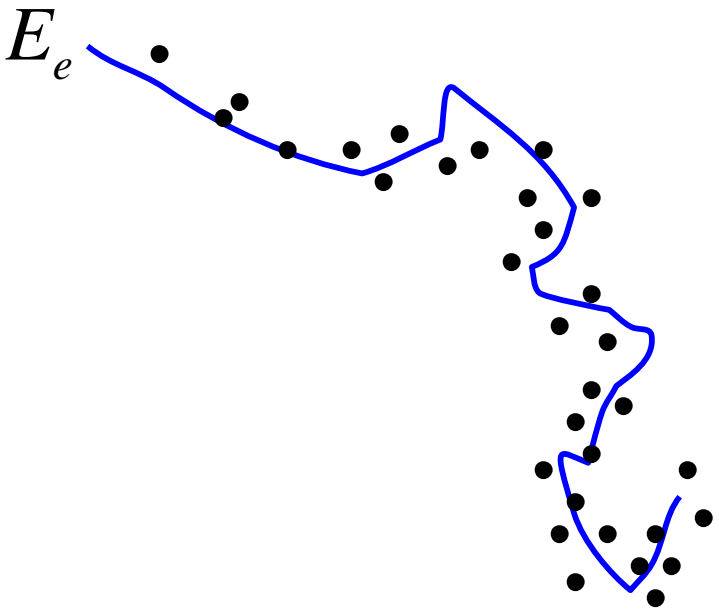


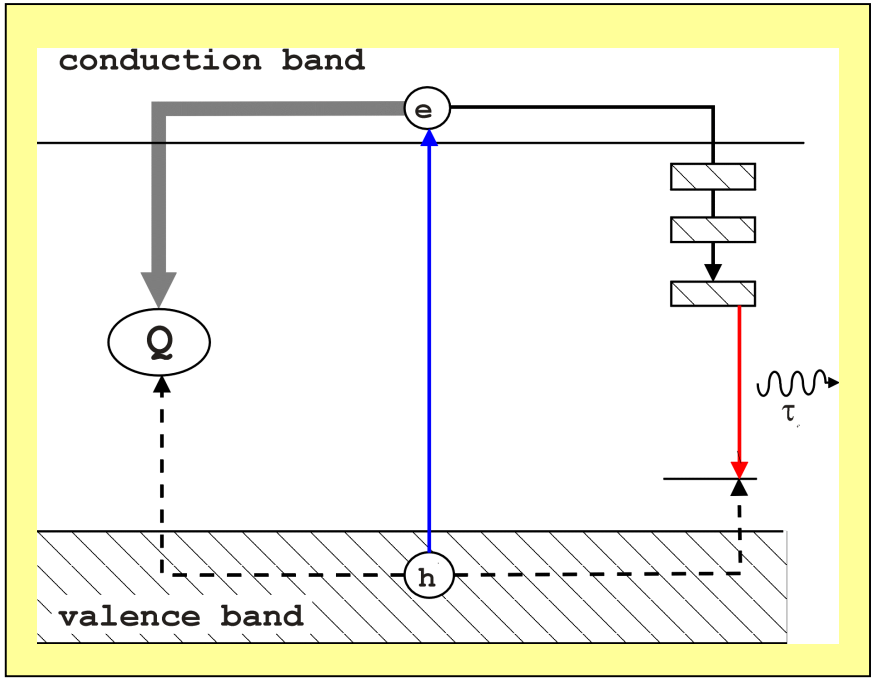
Properties of CVL materials

- very fast decay 0.8-2 ns
- poor light yield 700-2000 photons/MeV
- CVL is only possible in K, Rb, and Cs halides, and BaF_2
- it has never been observed in oxides
- relatively low density

Efficiency of impurity activated scintillators

$$N_{dph} = f\left(N_{eh}, S, Q, \eta_{lc}, \eta_{pd}\right)$$





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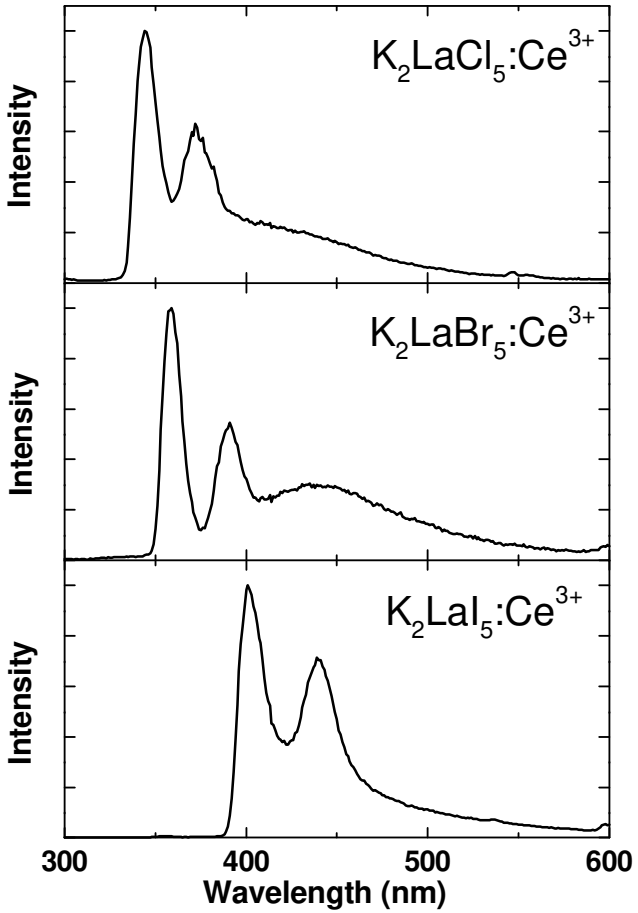
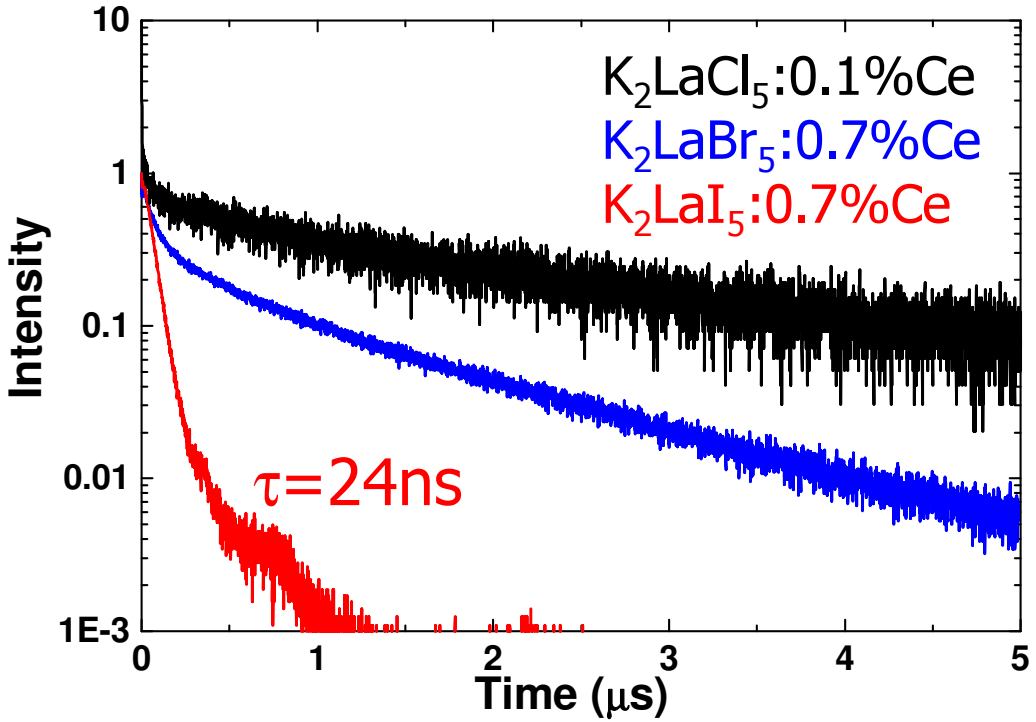
Intrinsic emission and slow transfer





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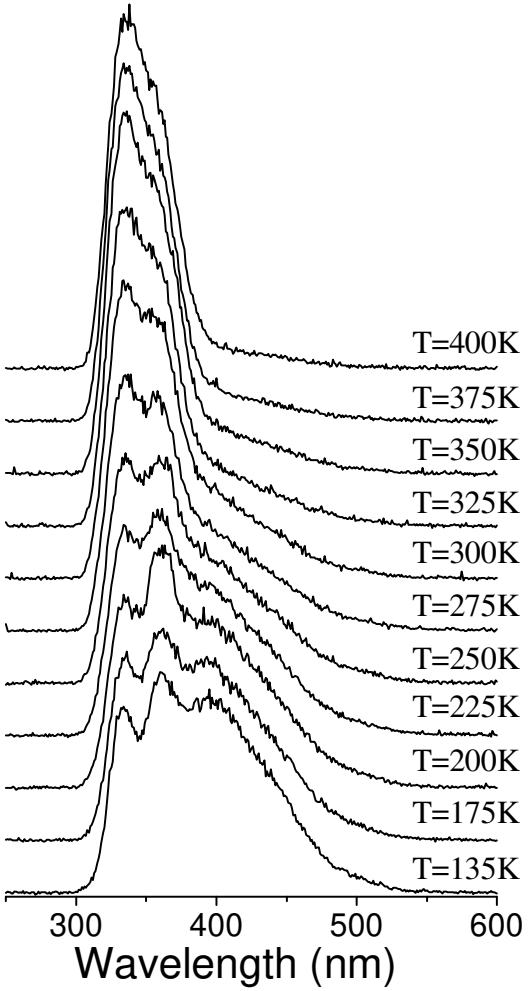
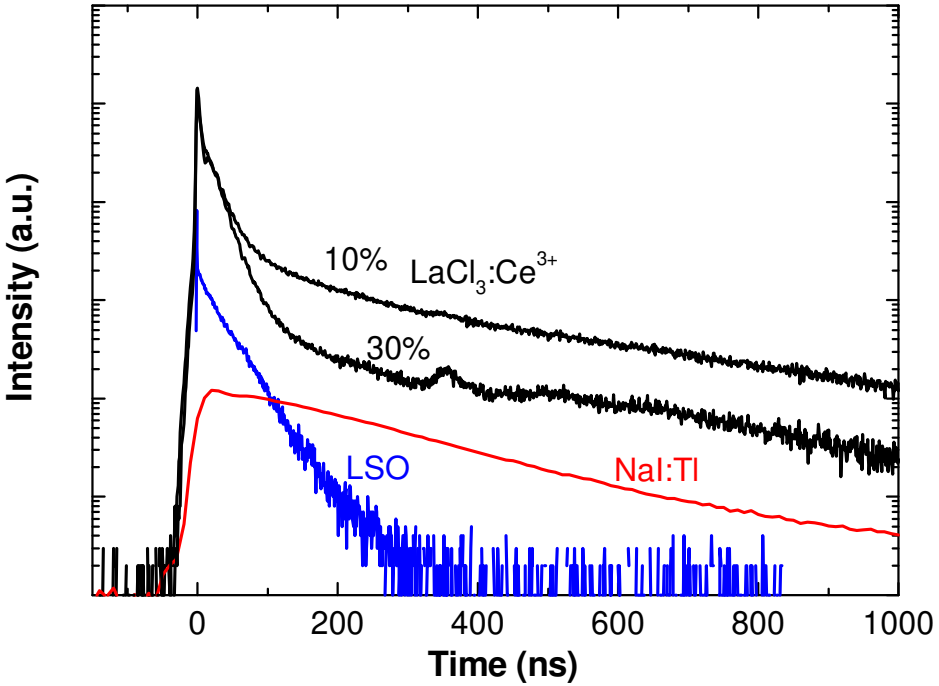
$LaCl_3:Ce^{3+}$ emission and decay





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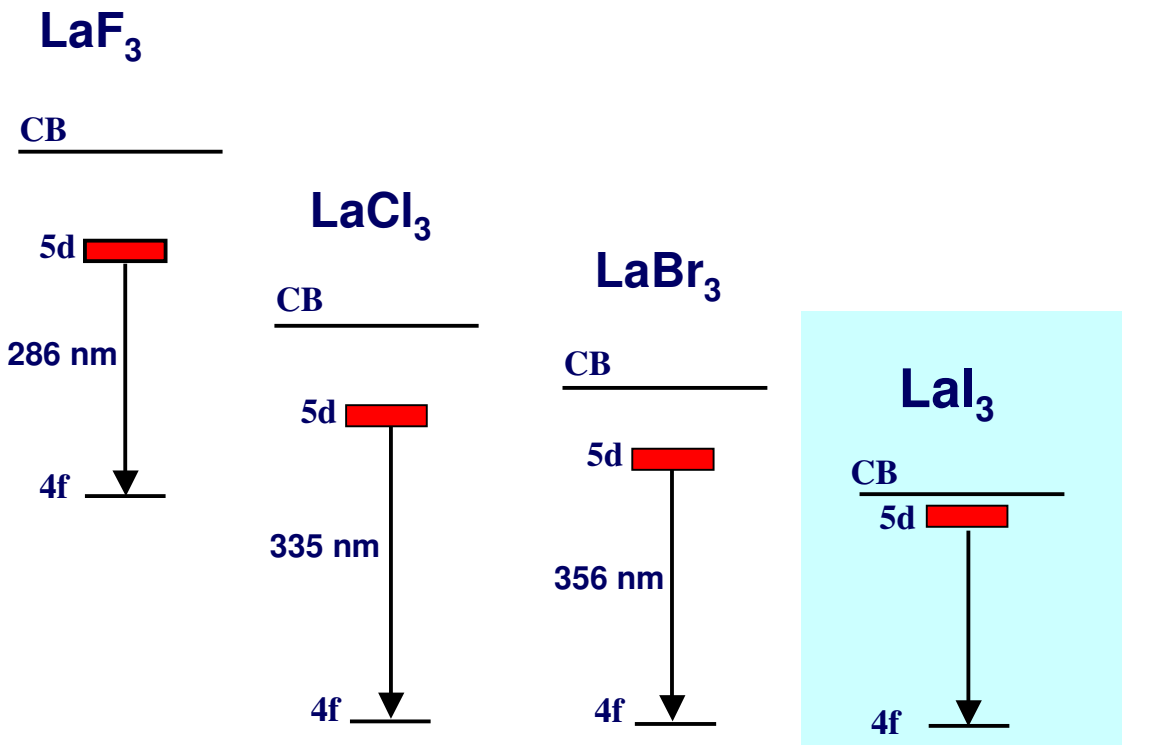
The LaX_3:Ce series

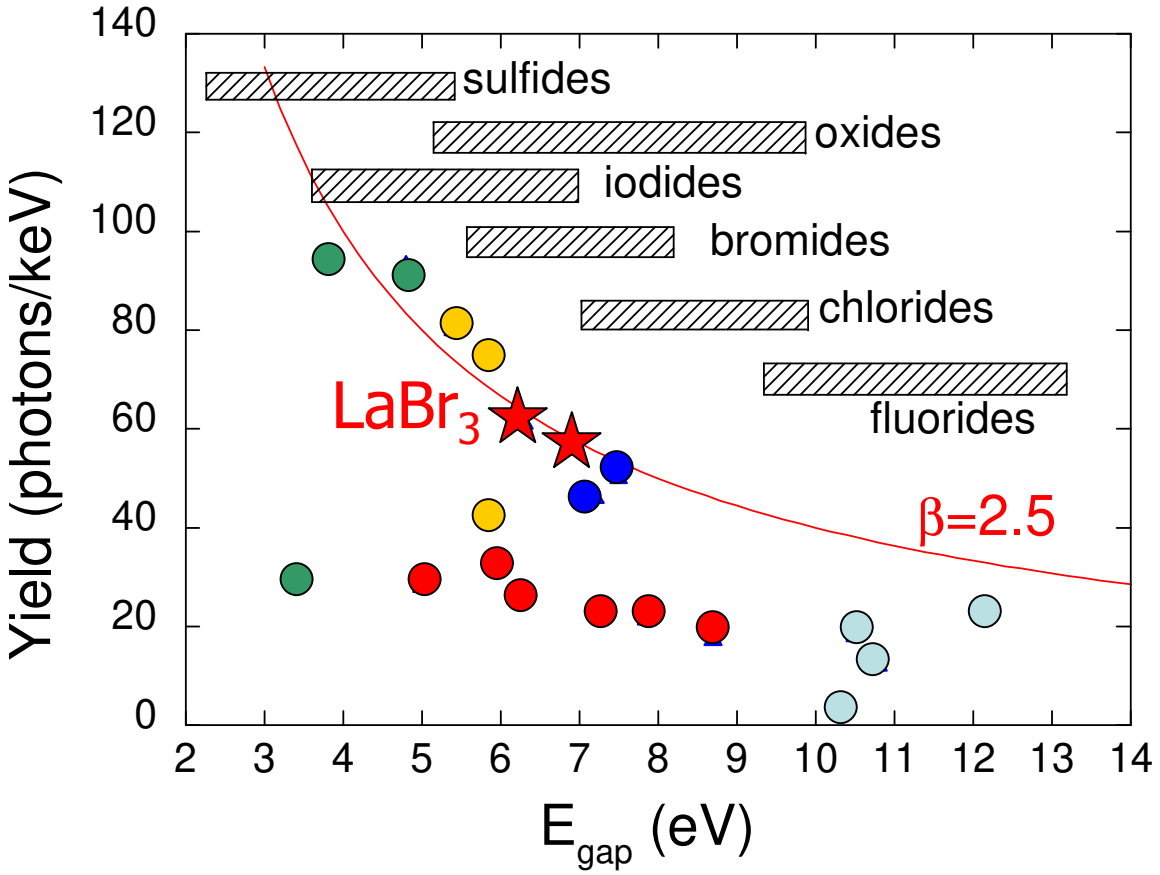
Number of thermalised electron-hole pairs

$\propto$ E_γ / 2.5 E_g



Light yield (10^3 ph/MeV)	**2 (S<<1)**	**49**	**61**	0 (Q<<1)

Limits to the light output



$$N_{eh} = \frac{E_\gamma}{\beta E_{gap}}$$

$$\frac{N_{ph}}{E_\gamma} = \frac{SQ}{\beta E_g} = \frac{1}{\beta E_g}$$

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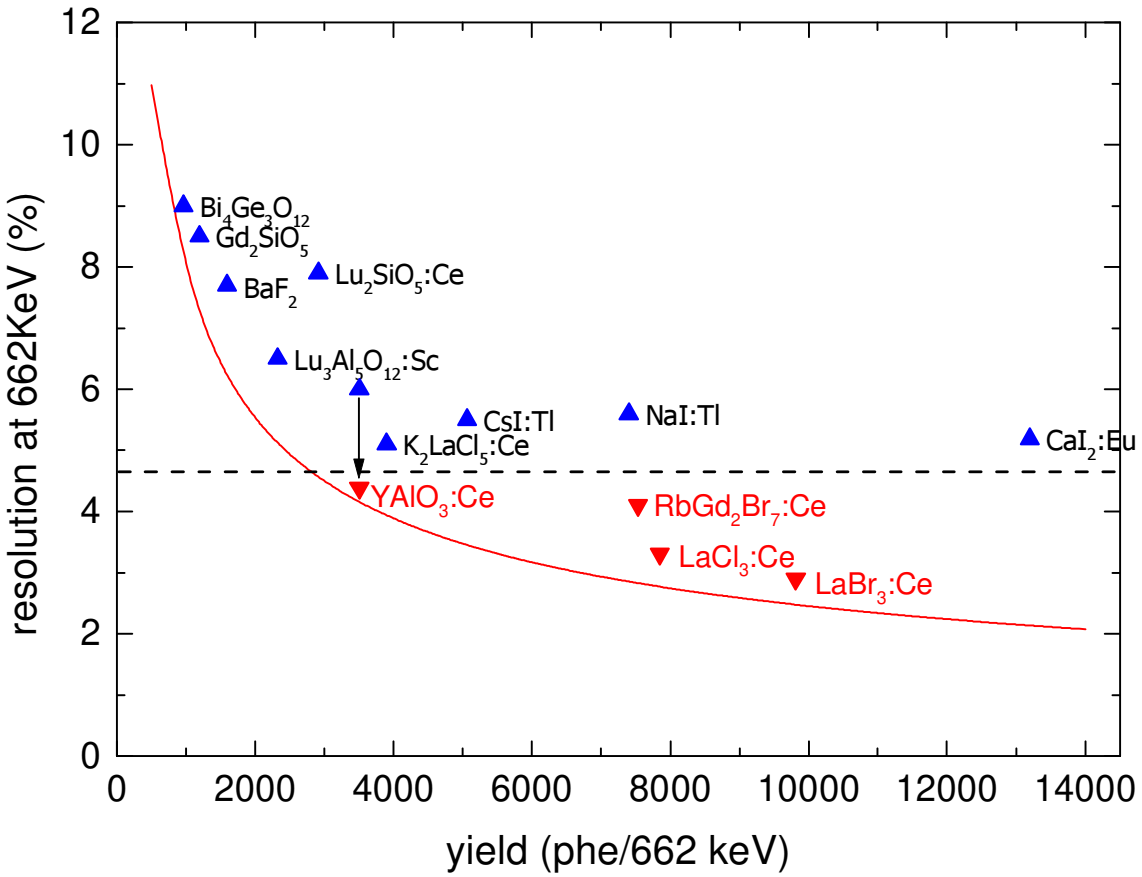
Limits to the energy resolution



$$\frac{\Delta E}{E} = 2.34\sqrt{\frac{1+v(M)}{N_{dph}}}$$

Progress of past five years

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Future prospects impurity activated scintillators

- Study of small band gap (3-4 eV) materials
  - > 100.000 ph/MeV is feasible
  - energy resolution below 2% @ 662 keV
  - PD and APD readout of long wavelength Ce^{3+} emission
- present study
  - $LaI_3:Ce^{3+}$ $E_g \approx 3.5$ eV 450, 500 nm
  - $K_2LaI_5:Ce^{3+}$ $E_g \approx 4.5$ eV 405, 445 nm
  - $LuI_3:Ce^{3+}$ $E_g \approx 4.1$ eV 475, 520 nm

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Scintillator requirements for the ELOISATRON calorimeter

- Bunch spacing: $\tau_b < 20$ ns
- requirement: $\tau_r = f\left(\tau_i, \tau_t, \tau_v, \tau_{lc}, \tau_{pd}\right) < \tau_b / 4 = 5ns$
- crystal length: > 25 cm
- creation time ionization track: $\tau_i \approx$ 1 ns
- light collection time: $\tau_{lc} \approx 3$ ns
- ultimate response time: $\tau_r \approx$ 4 ns
- scintillation decay: $\tau_v < 4$ ns

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ELOISATRON calorimeter speed considerations

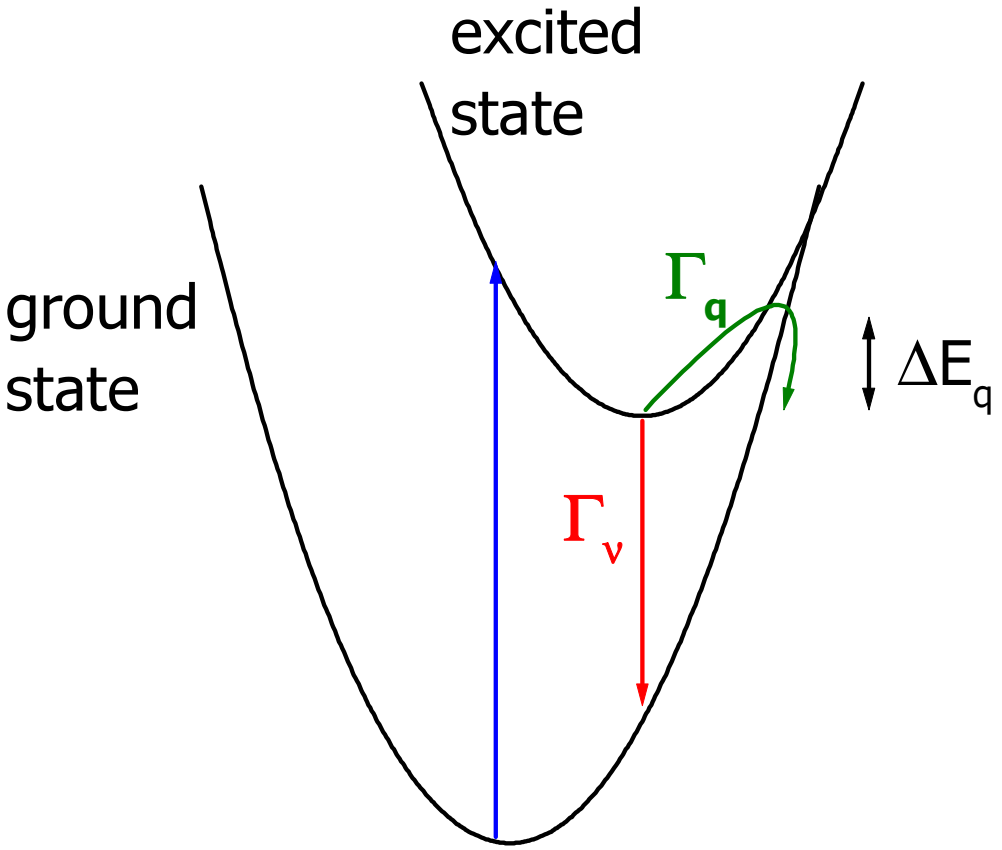
- Ce^{3+} is too slow
- Pr^{3+} and Nd^{3+} are faster, but in addition slow 4f→4f emission. Emission at too high energy for read-out
- CVL is fast (ns) but limited to too low density (halide) materials
- Remaining option
  - quenched luminescence

Internal luminescence quenching

$$\Gamma = \frac{1}{\tau} = \Gamma_v + \Gamma_q$$

$$\Gamma_q(T) = \Gamma_0 \exp\left(\frac{-\Delta E_q}{k_B T}\right)$$

$$I = \frac{\Gamma_v}{\Gamma_v + \Gamma_q}$$



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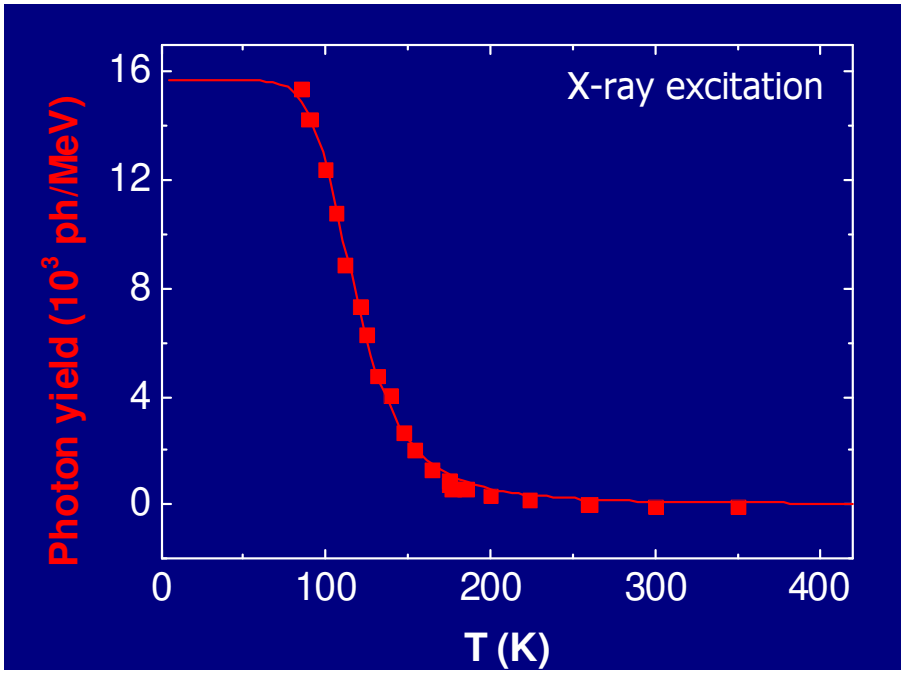
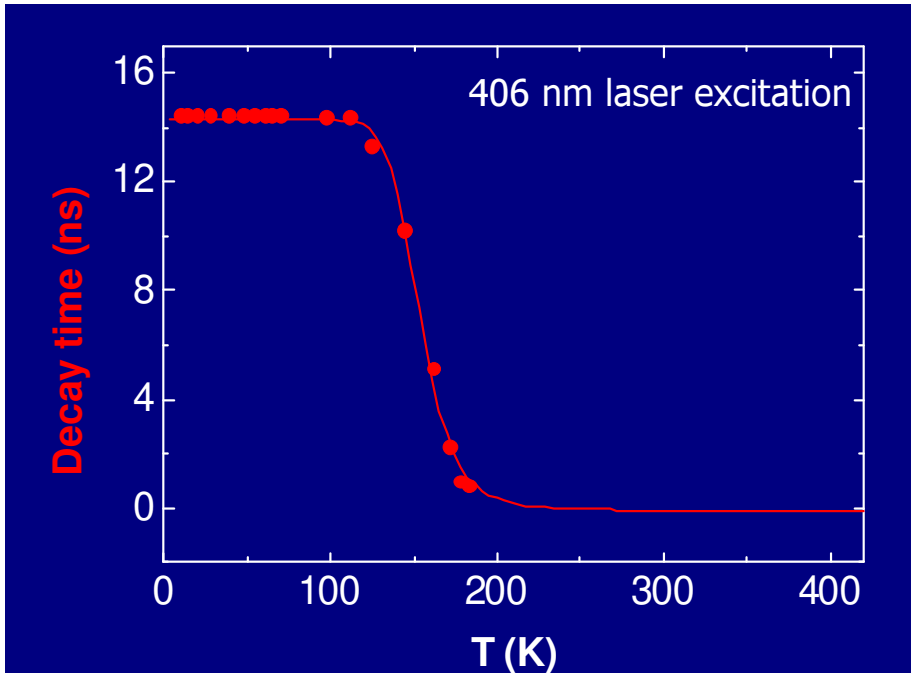
LaI_3:Ce; quenching via the conduction band?

T evolution





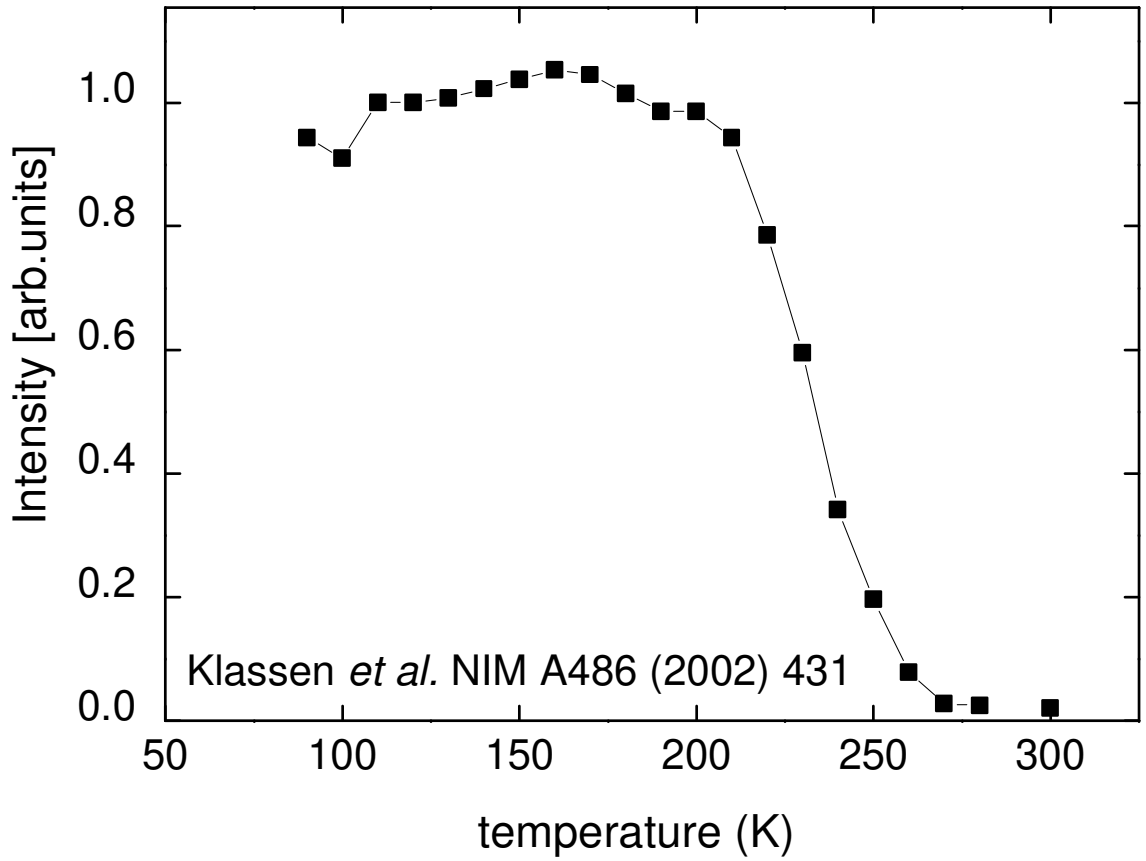
$$I(T)=\frac{I_0}{1+\frac{\Gamma_0}{\Gamma_\nu}\exp\left(-\frac{\Delta E_q}{kT}\right)}$$

➡ ΔE_q=0.08 eV

$$\tau=\frac{1/\Gamma_\nu}{1+\frac{\Gamma_0}{\Gamma_\nu}\exp\left(-\frac{\Delta E_q}{kT}\right)}$$

➡ ΔE_q=0.20 eV

Quenching of $PbWO_4$ emission



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ELOISATRON calorimeter light yield and resolution considerations

$$\frac{\sigma(E)}{E} = \frac{a}{\sqrt{E}} \oplus \frac{b}{E} \oplus c$$

- at TeV energies stochastic term and noise cont
- temperature fluctuations

Conclusion

crystal quality is more important than light yield

N.B. 1 photon/MeV$\rightarrow 10^6$ photons/TeV

light output can be traded off against faster decay

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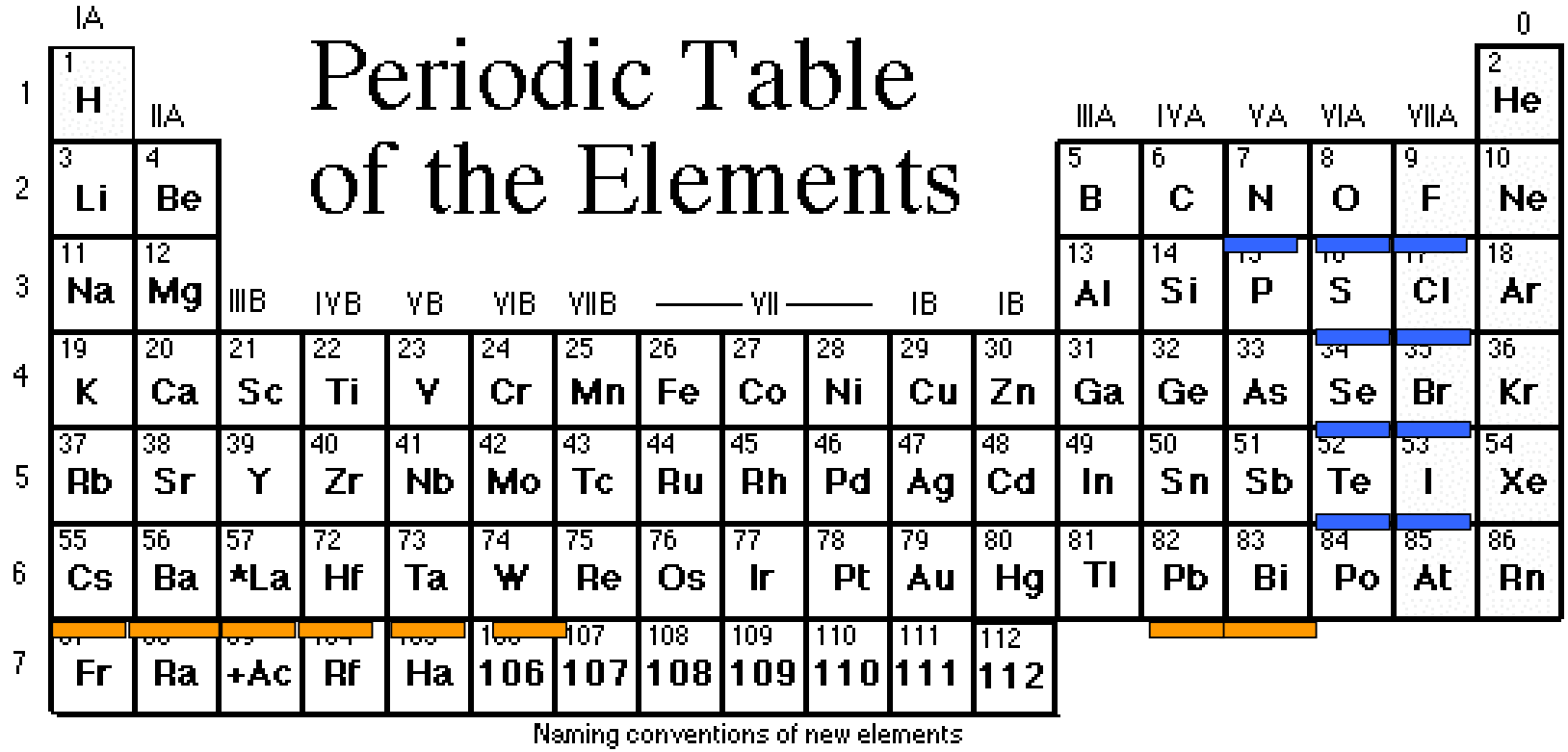
ELOISATRON calorimeter density considerations

- Requirement: optically transparent + high density
- transparency
  - wide band gap inorganic compound
  - absence of optically active electrons
    - ions with closed shell configuration
- Density
  - high atomic number of cations and/or anions
  - small radius of cations and anions
  - high ratio of cations to anions
  - large packing fraction of the lattice

Periodic Table of the Elements



Naming conventions of new elements

* Lanthanide Series	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu
+ Actinide Series	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr

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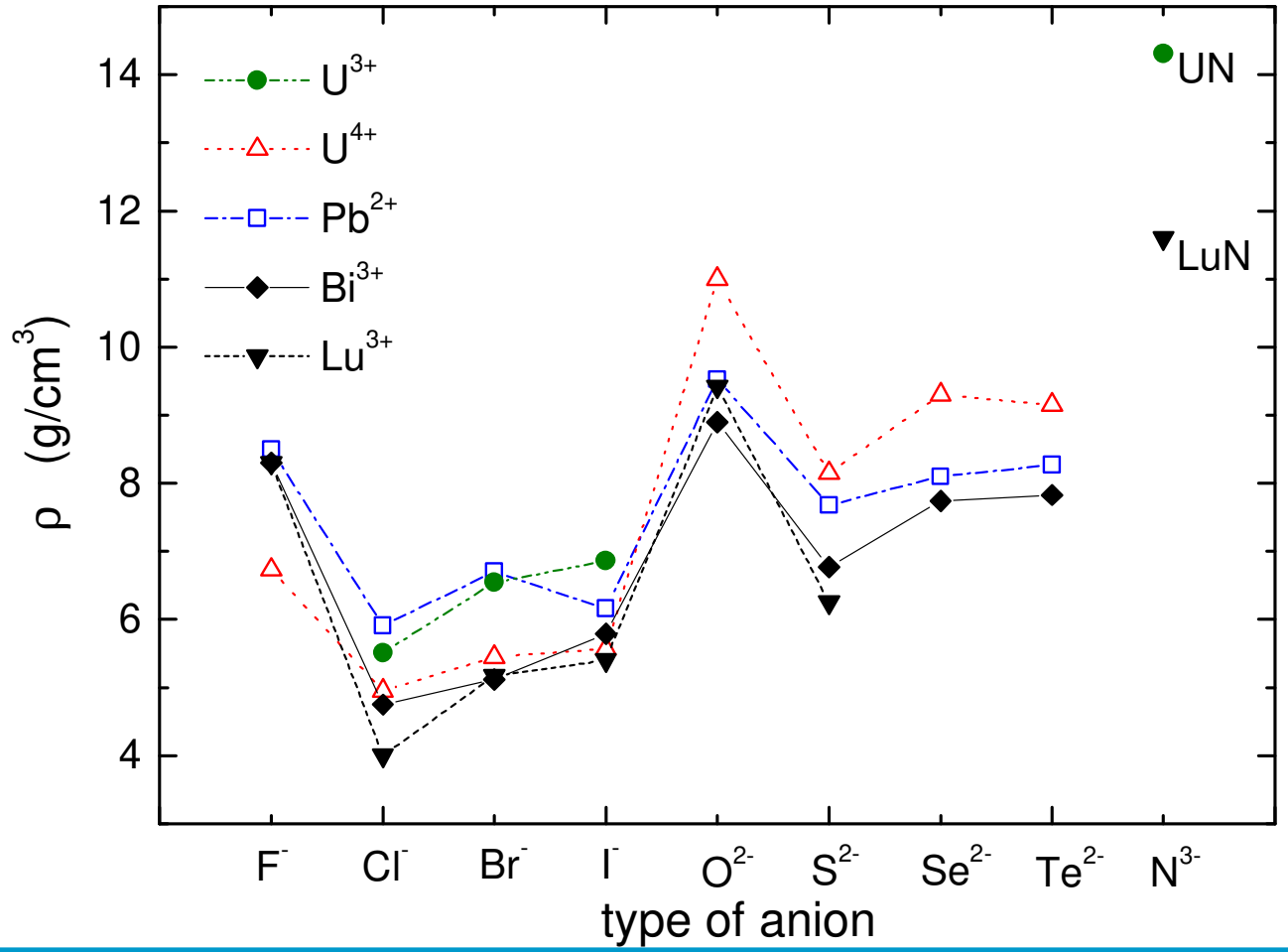
Density of binary compounds



$$F > Cl \leq Br \leq I$$

$$O > S \leq Se \leq Te$$

$$N > O > F$$

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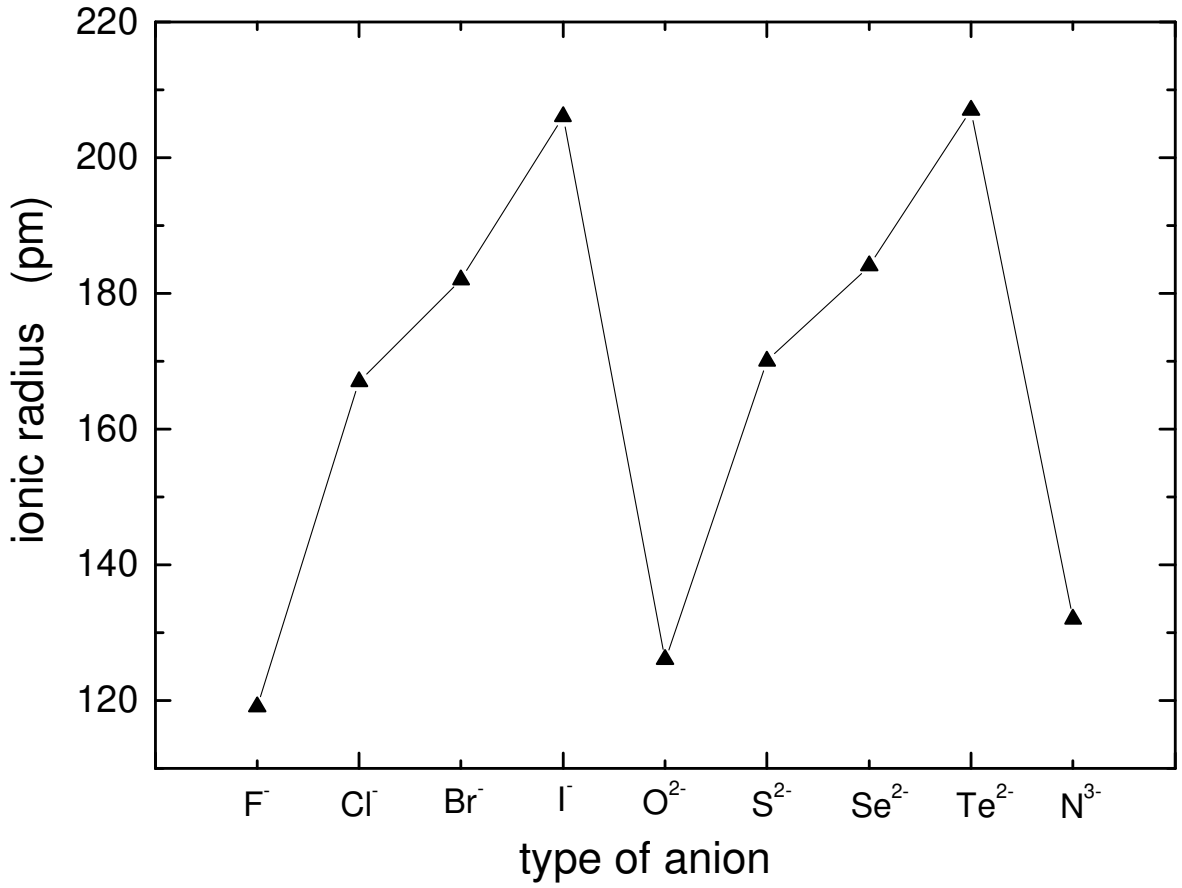
Ionic radii of anions in inorganic compounds



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What compound can do the job?

- High density of fluorides, oxides, and nitrides is related with small ionic radius
- Density LuF_3<Lu_2O_3<LuN because of higher charge of anion→larger cation/anion ratio
- Nitride compounds is not a feasible option for calorimeter

Conclusion
The highest density compounds must be found amongst the oxides

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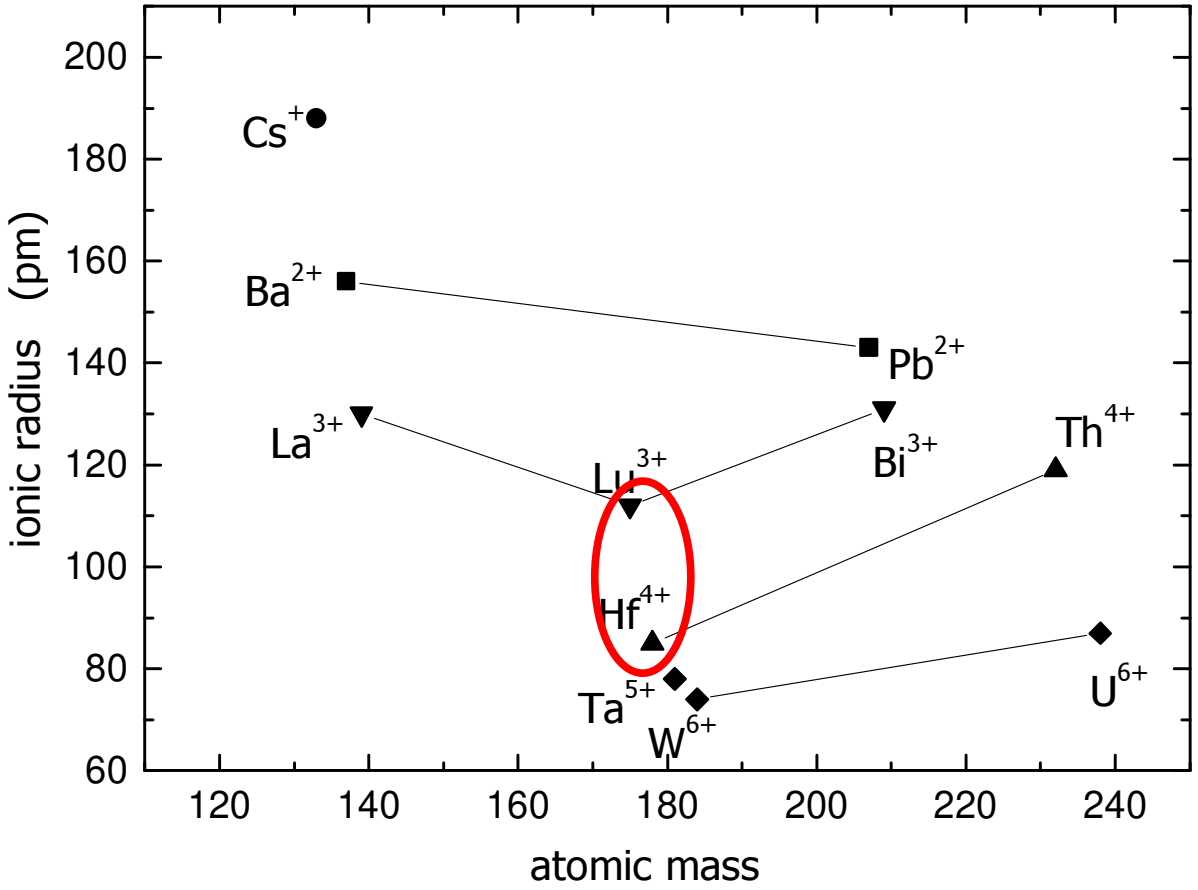
Ionic radii of high atomic number cations



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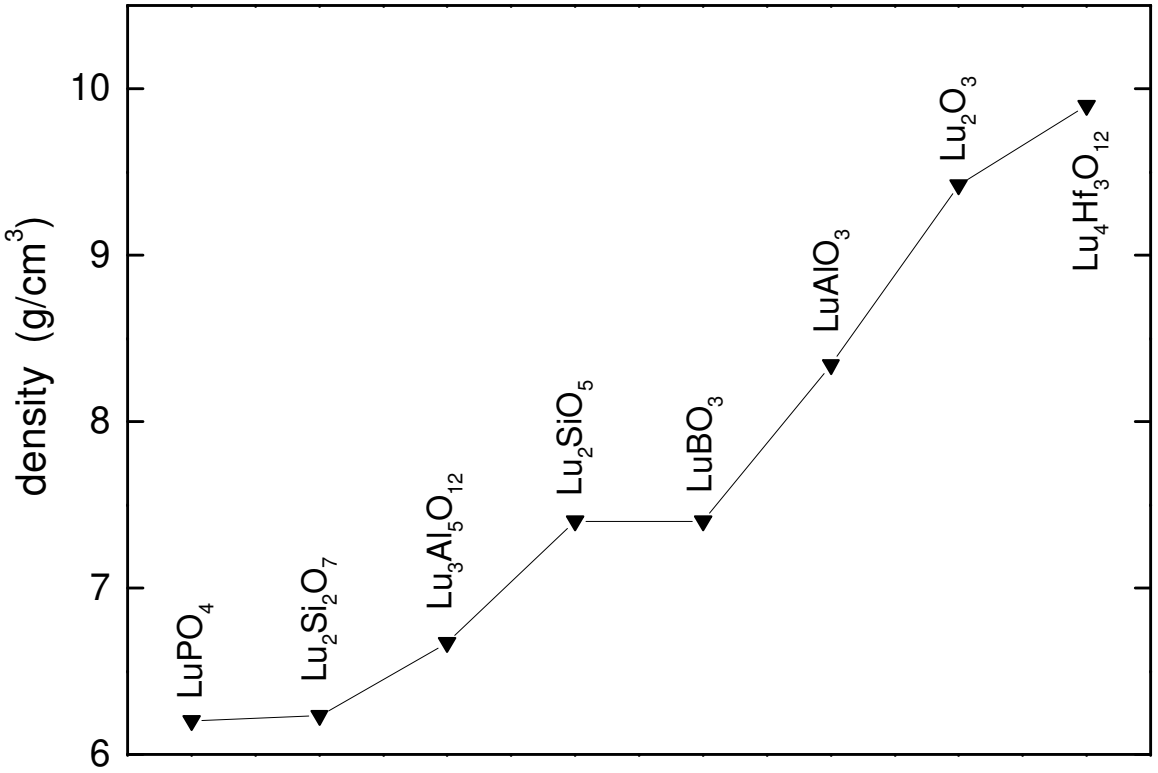
Density of ternary Lu-based compounds



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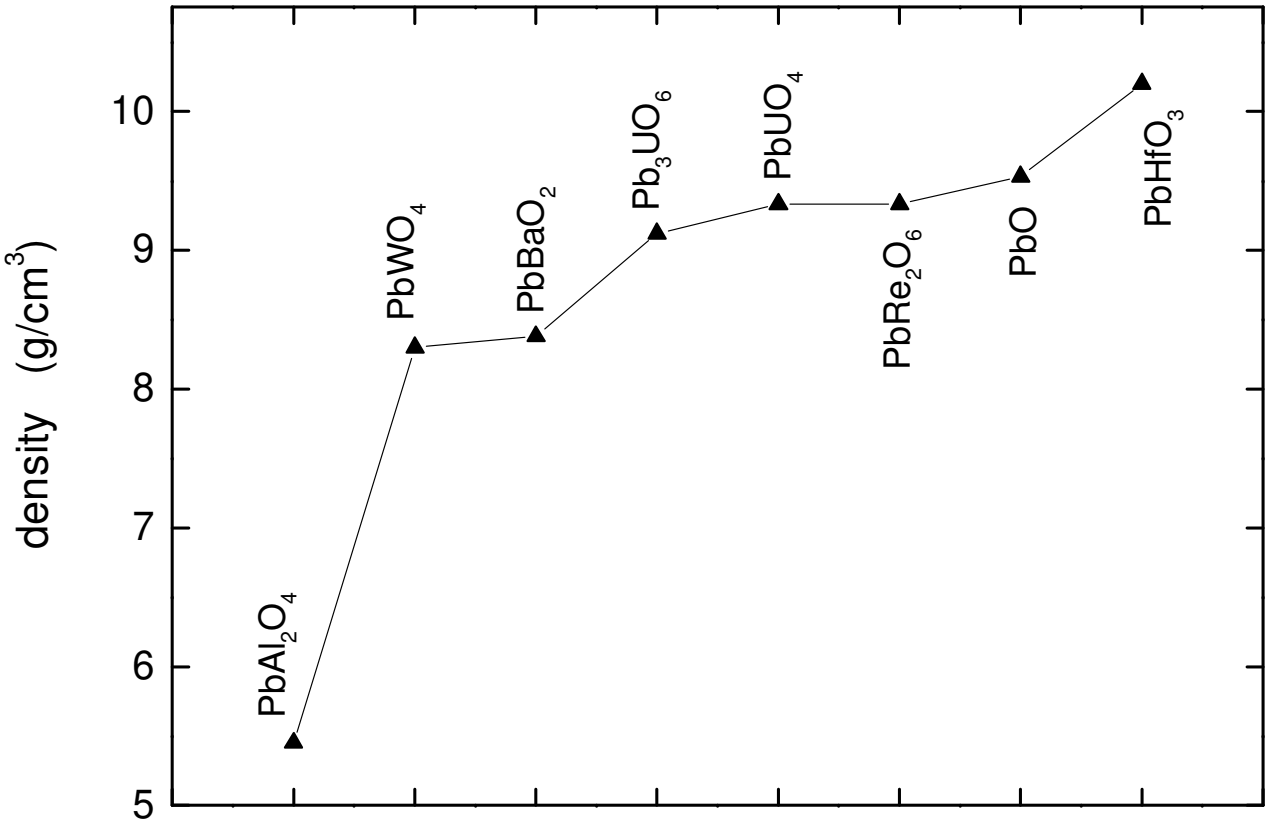
Density of ternary Pb-based compounds



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Future prospects and directions for research

- Ultimate density of 10 g/cm^3 is feasible
- to obtain τ_v< 4ns, a quenched luminescence mechanism is needed
- impurity activation may introduce problems with
  - transfer time
  - concentration gradients $\rightarrow$inhomogeneity
  - radiation hardness
- A quenched intrinsic luminescence mechanism is the best option

Future prospects and directions for research

- $^3P_1 \rightarrow {}^1S_0$ emission in Pb^{2+} or Bi^{3+}
- Pb-compounds are more dense than Bi-compounds
- Pb-compounds with similar properties as $PbWO_4$ but higher density should be searched for.
- Ce^{3+} emission in Pb-compounds has never been observed
- $PbHfO_3$ has a density of 10.2 g/cm^3