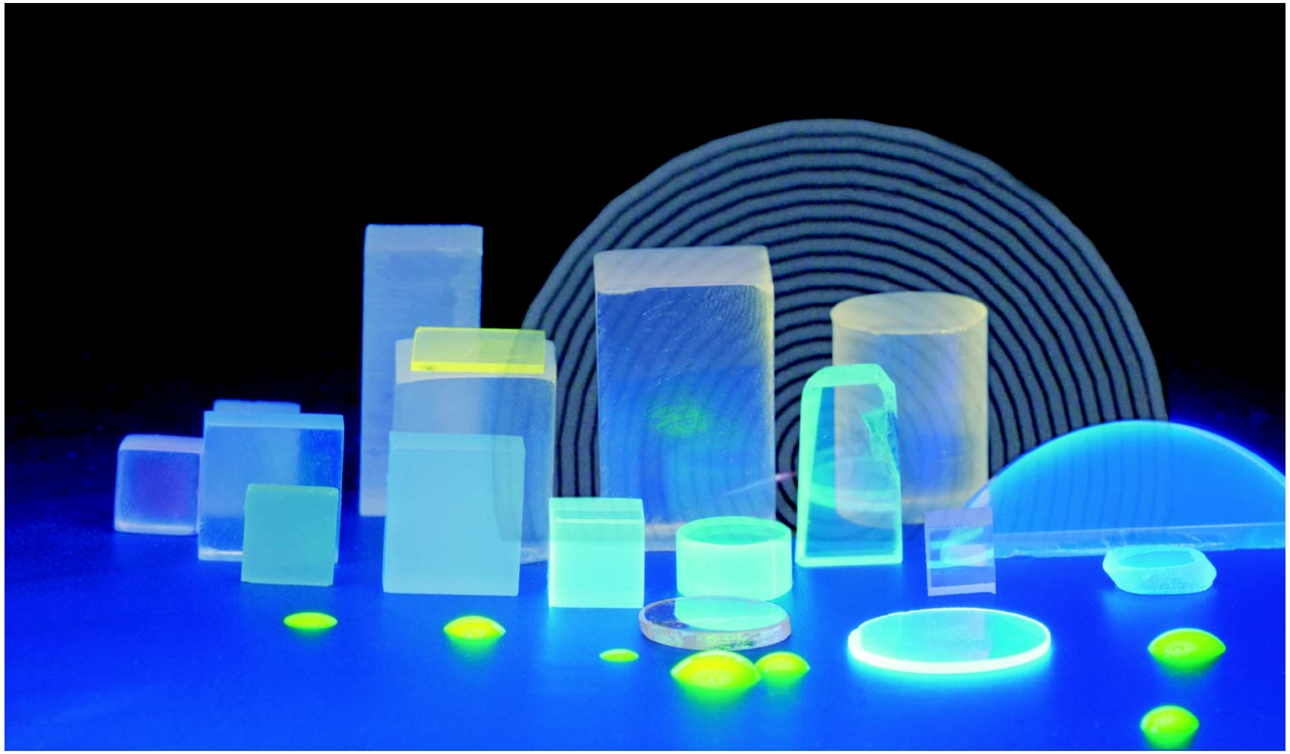
Current status and future prospects of inorganic scintillator research

Pieter Dorenbos



October 7, 2003

Interfaculty Reactor Institute

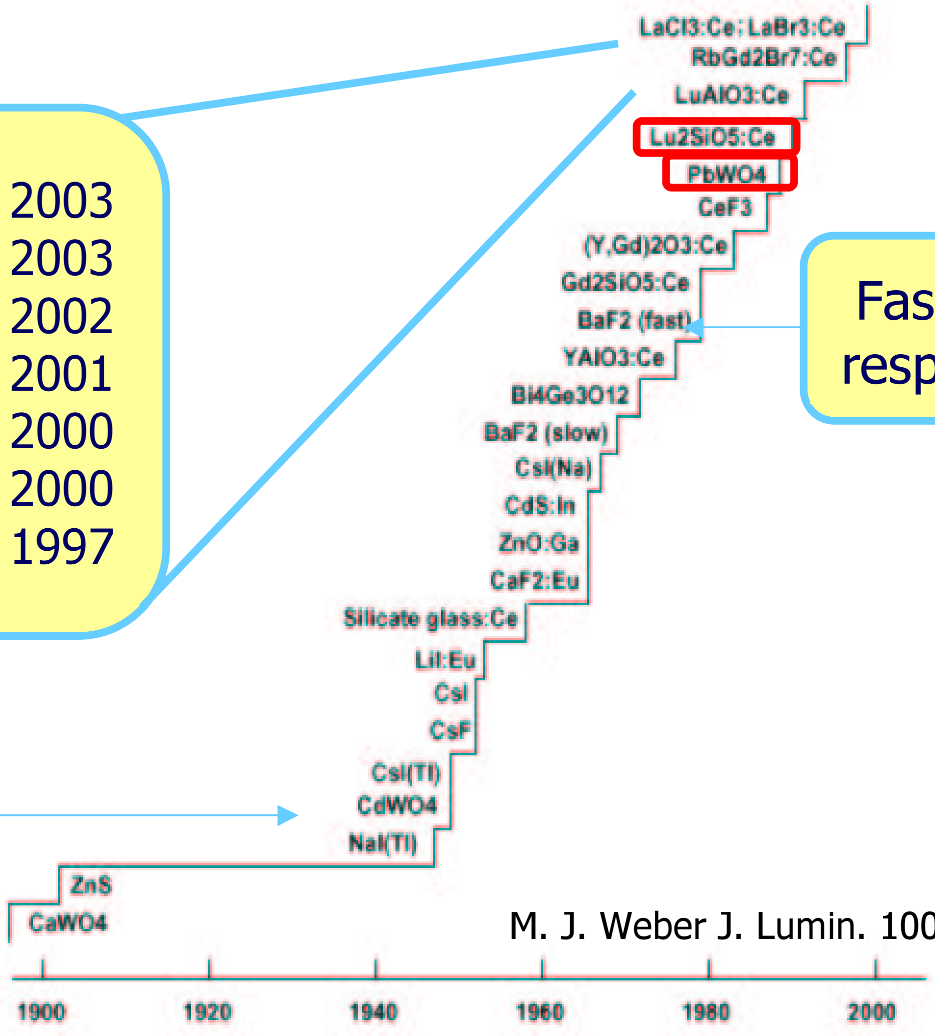
TU Delft
Technische Universiteit Delft

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

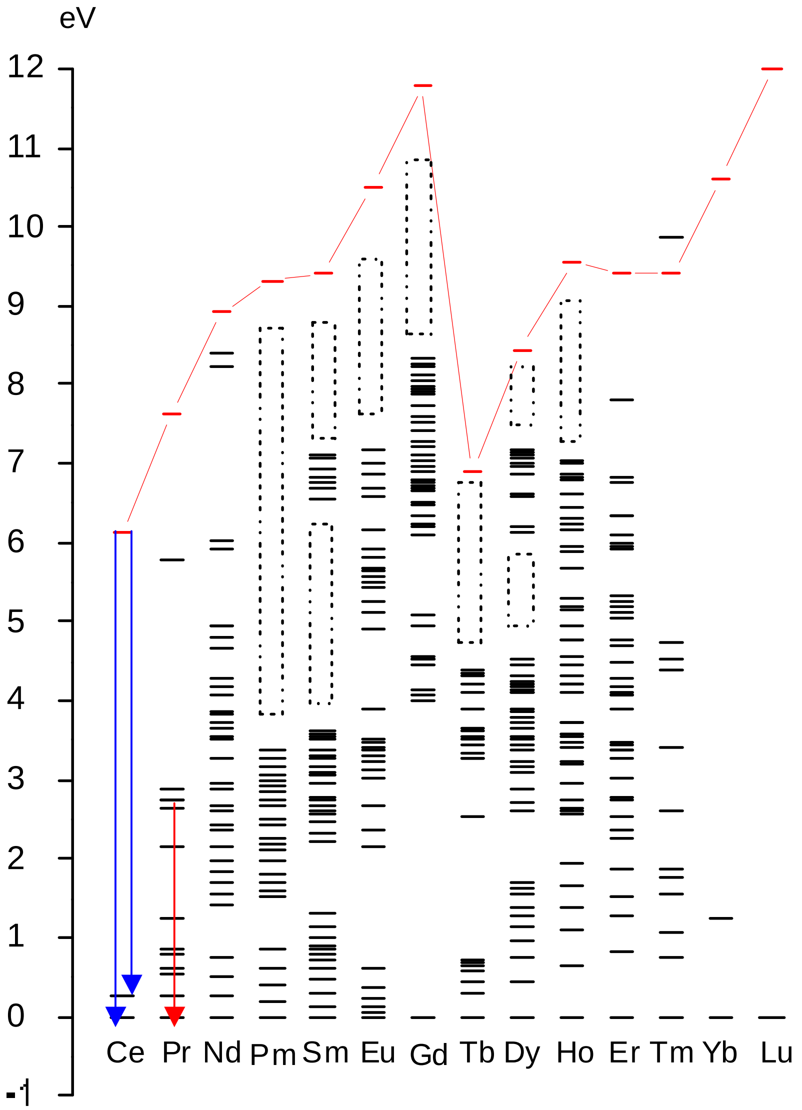
Cs_2LiYCl_6:Ce	2003
LuI_3:Ce	2003
K_2LaI_5:Ce	2002
$LaBr_3$:Ce	2001
$LaCl_3$:Ce	2000
$Lu_2Si_2O_7$:Ce	2000
$RbGd_2Br_7$:Ce	1997



Fast UV response

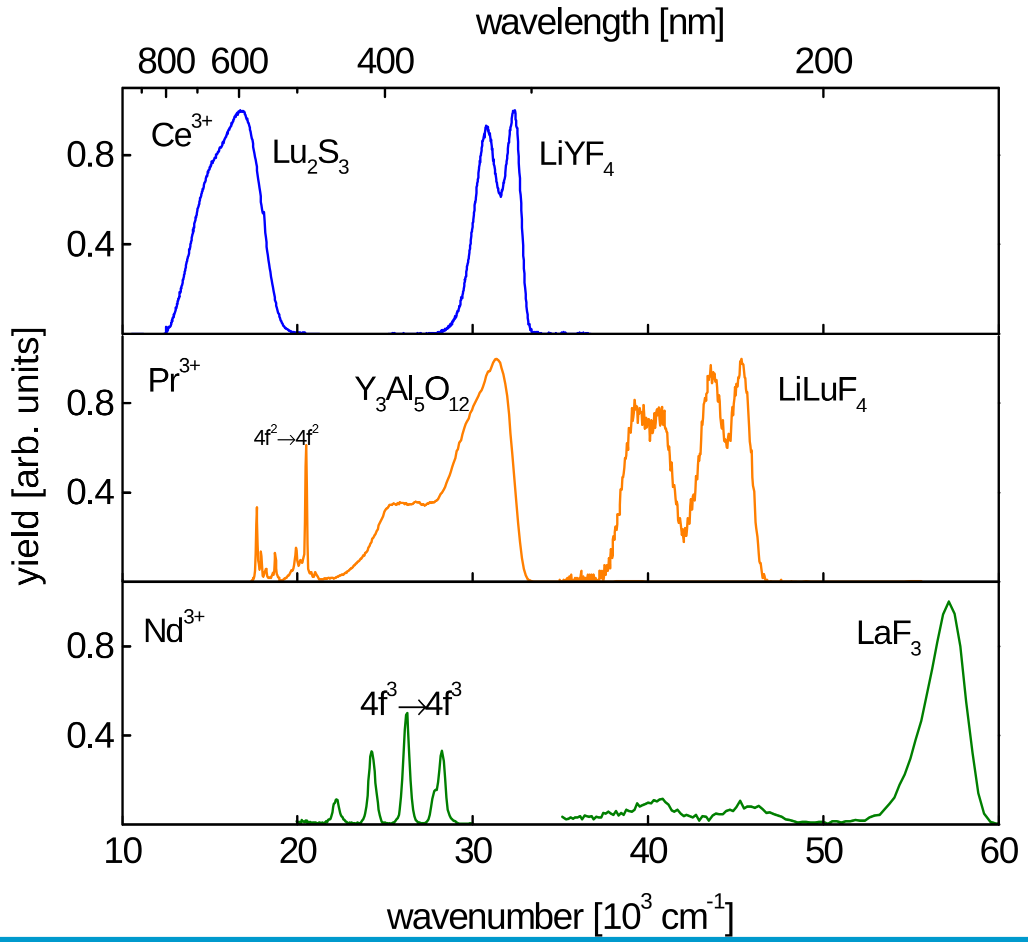
Invention of the photomultiplier tube

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



- 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

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

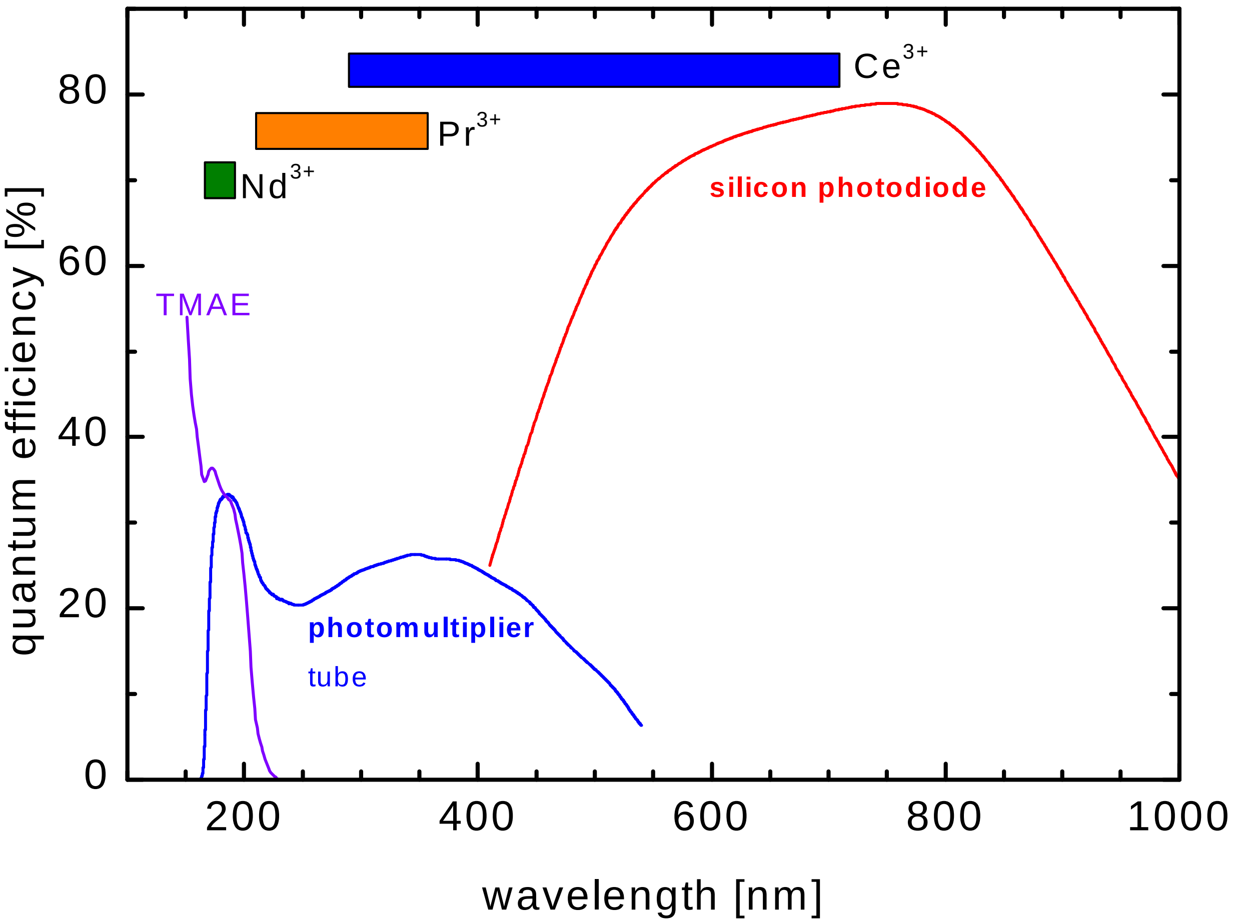


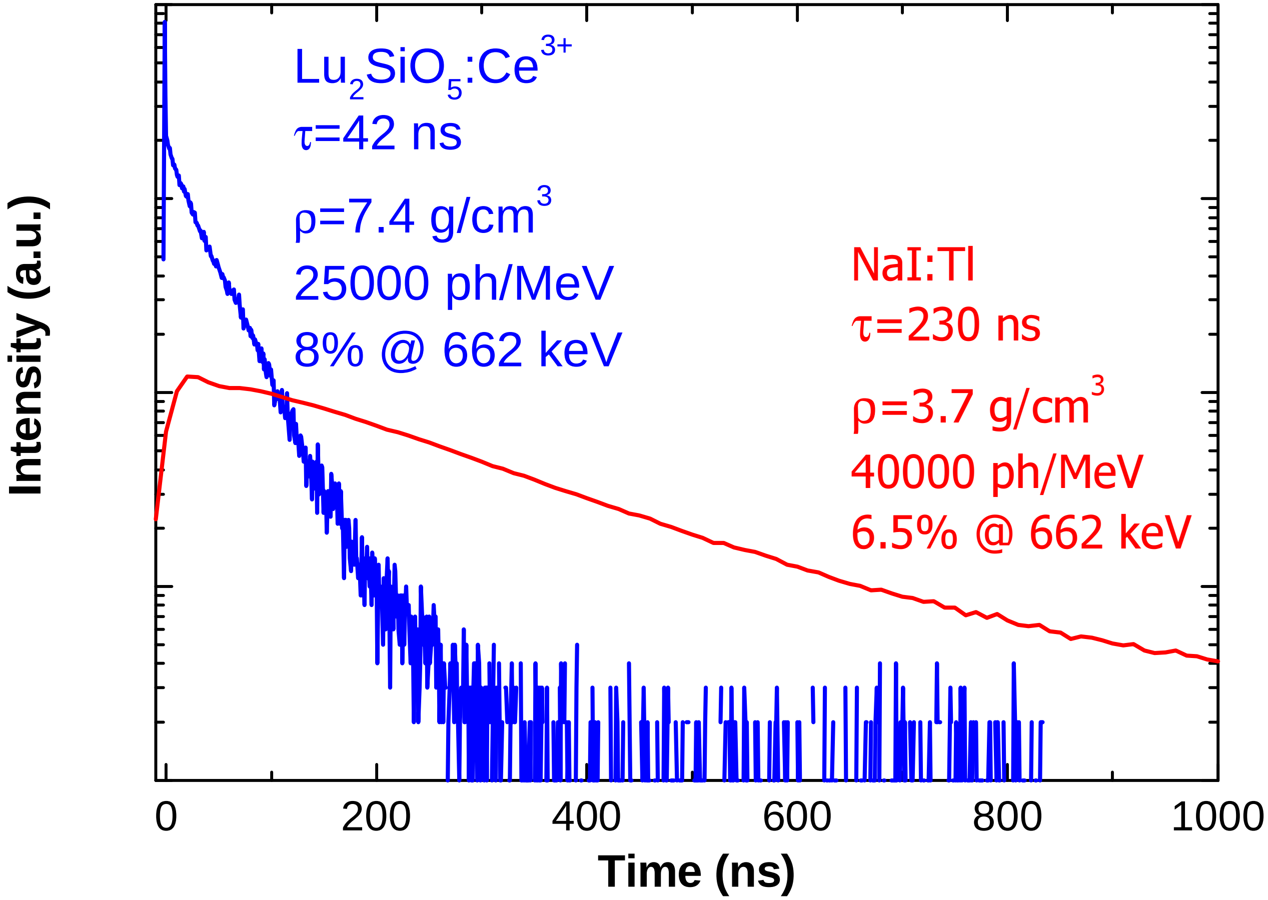
λ_{em} depends on

  - lanthanide
  - host

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

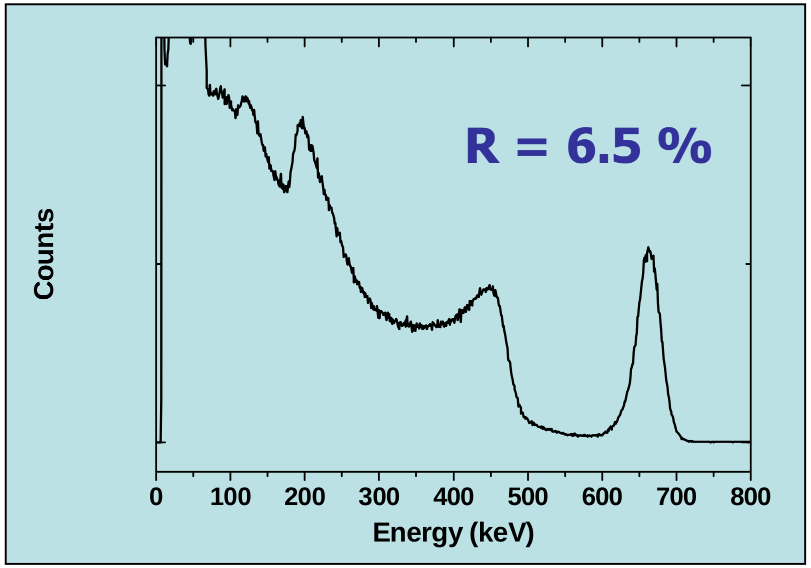
df emission in Ce, Pr, Nd

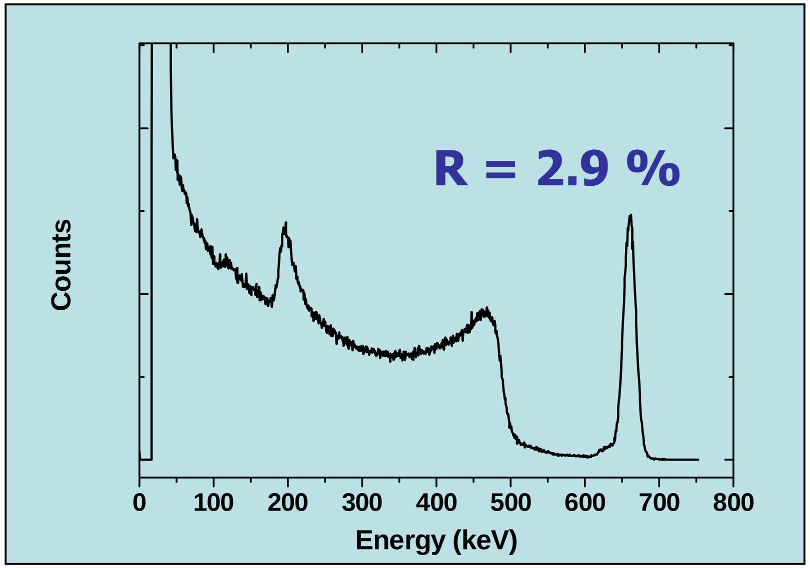



$Lu_2SiO_5:Ce^{3+}$
τ=42 ns
ρ=7.4 g/cm^3
25000 ph/MeV
8% @ 662 keV
NaI:Tl
τ=230 ns
ρ=3.7 g/cm^3
40000 ph/MeV
6.5% @ 662 keV
Intensity (a.u.)
0
200
400
600
800
1000
Time (ns)

Scintillator performances

Pulse-height spectra (662 keV gamma rays)





NaI:Tl

43. 10^3 ph/MeV

230 ns

3.7 g/cm^3

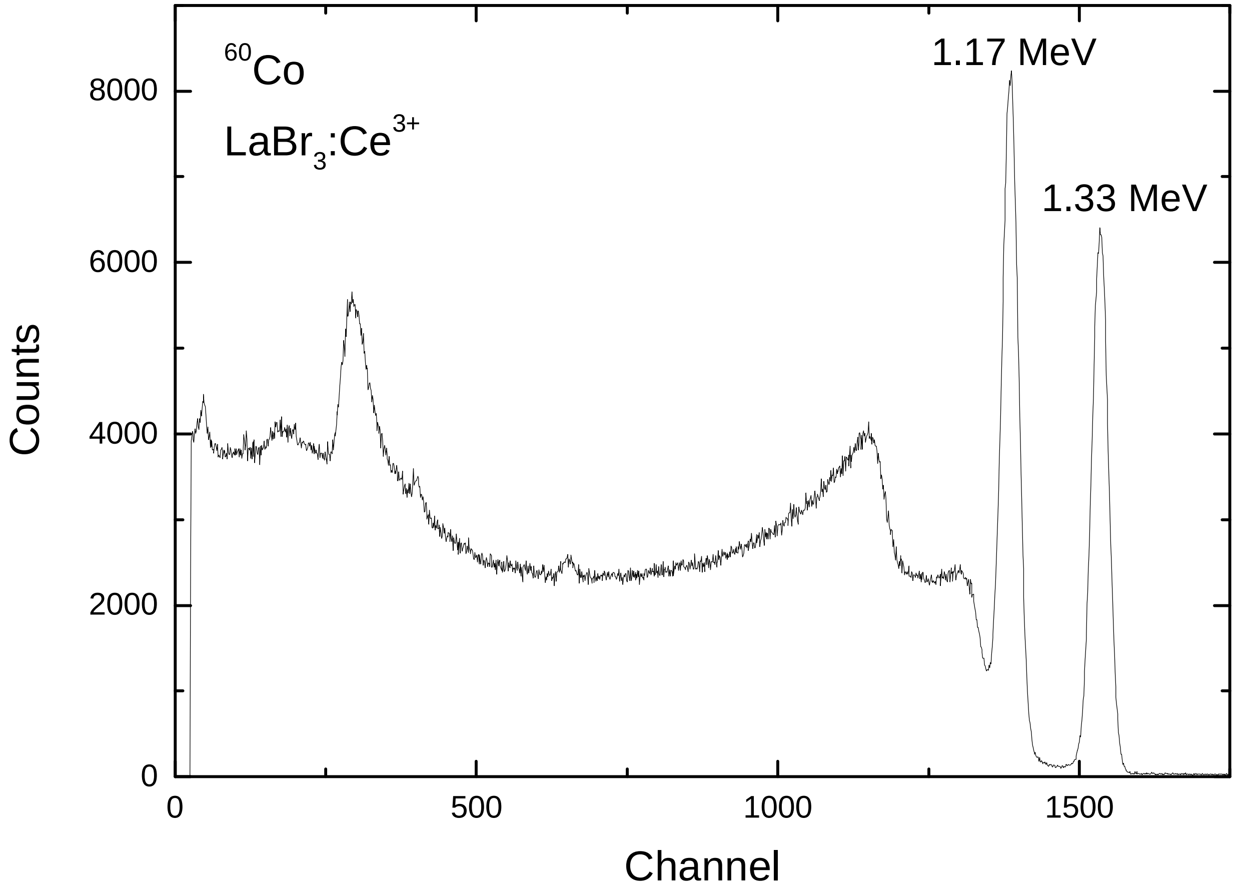
$LaBr_3$:Ce

61. 10^3 ph/MeV

18 ns

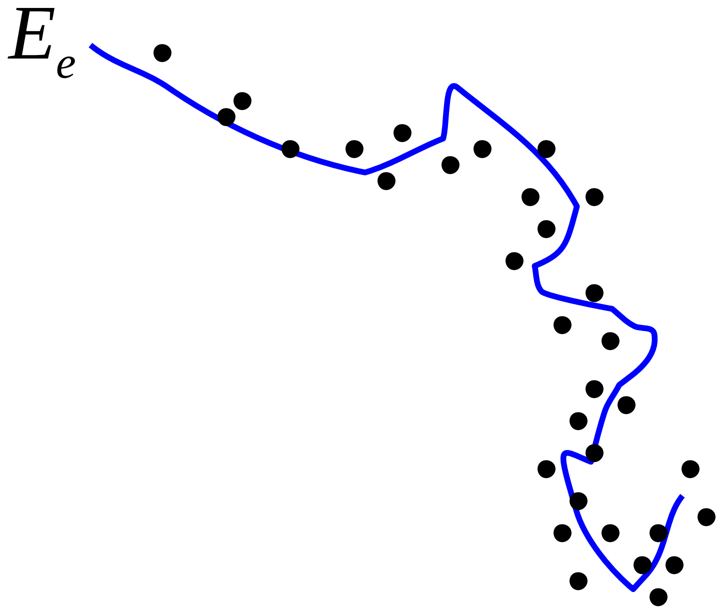
5.3 g/cm^3

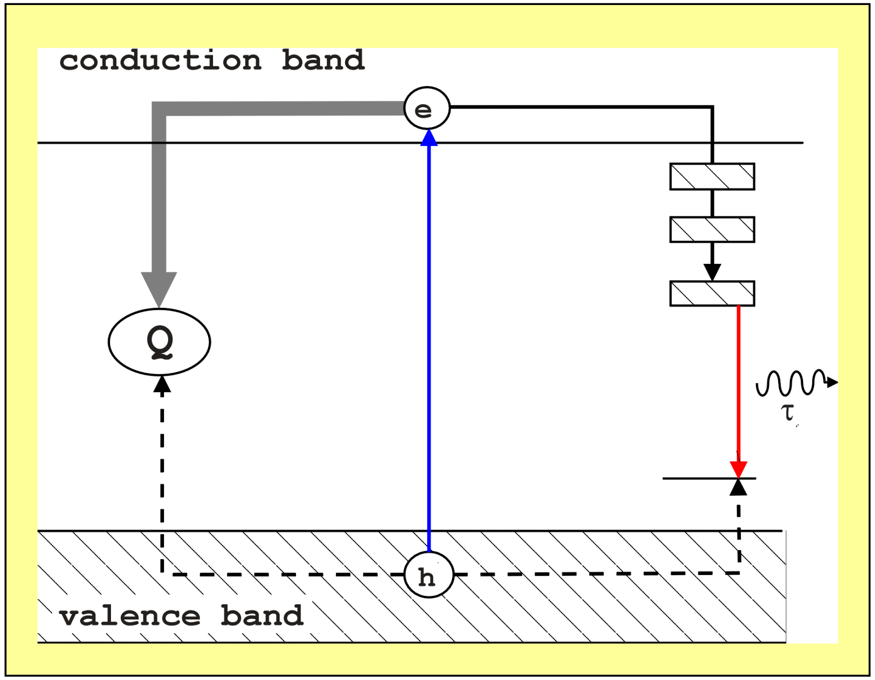
^{60}Co spectrum measured with prototype $LaBr_3$:Ce scintillator



Speed of impurity activated scintillators

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





Radiative decay rate

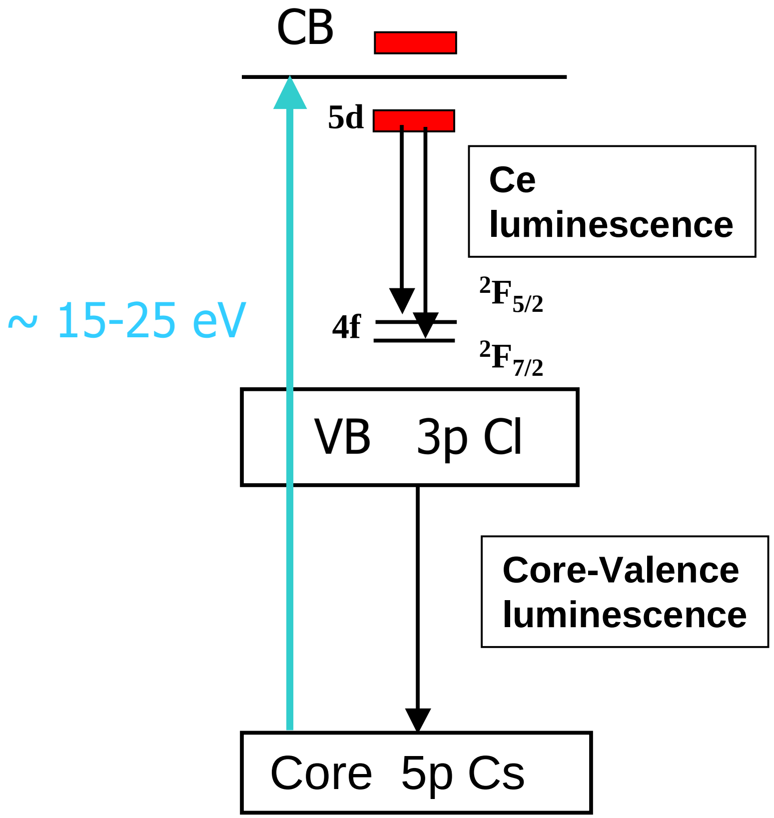
$$\Gamma_\nu = \frac{1}{\tau_\nu} \propto \frac{n}{\lambda^3}\left(\frac{n^2+2}{3}\right)^2 \sum_f \left|\langle f|\mu|i\rangle\right|^2$$

- 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
  - $Yb^{2+} + h_{VB} \rightarrow Yb^{3+}$

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

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

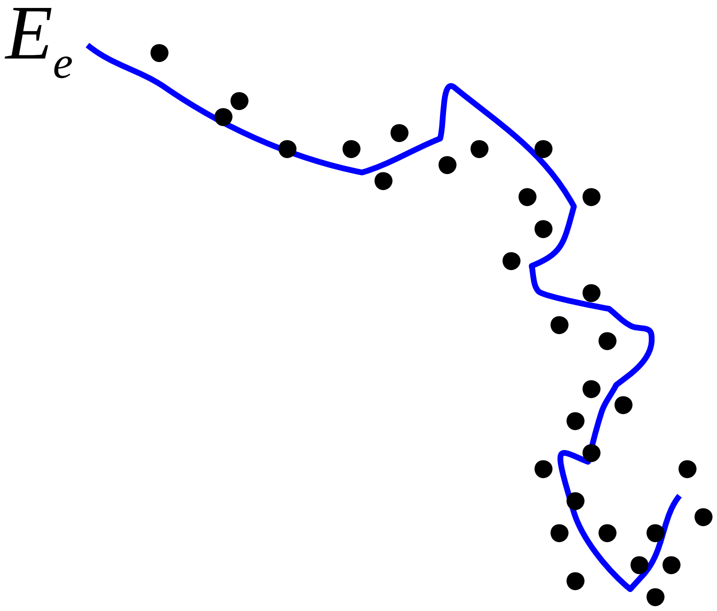


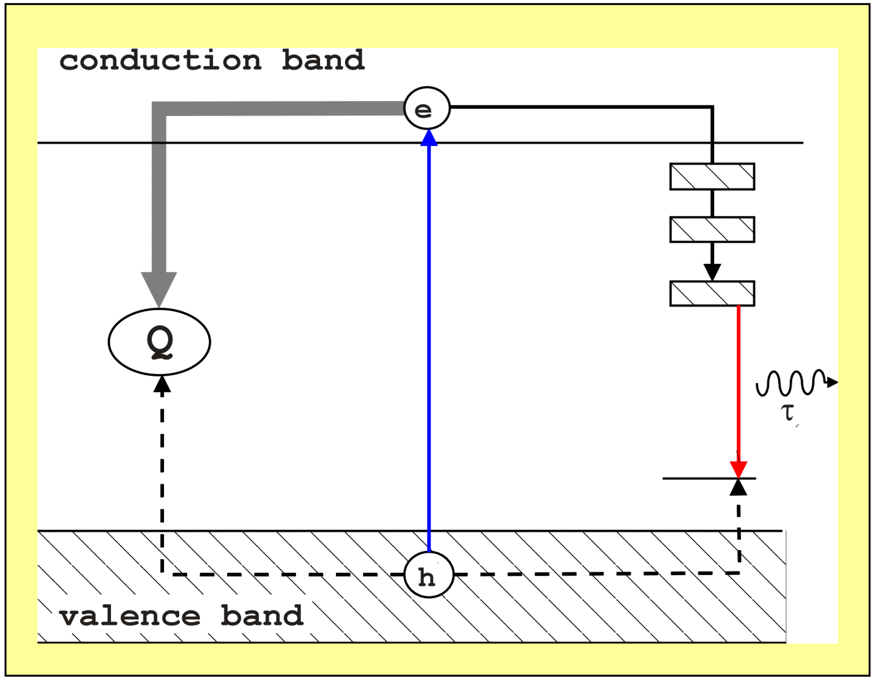
Properties of CVL materials

- 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

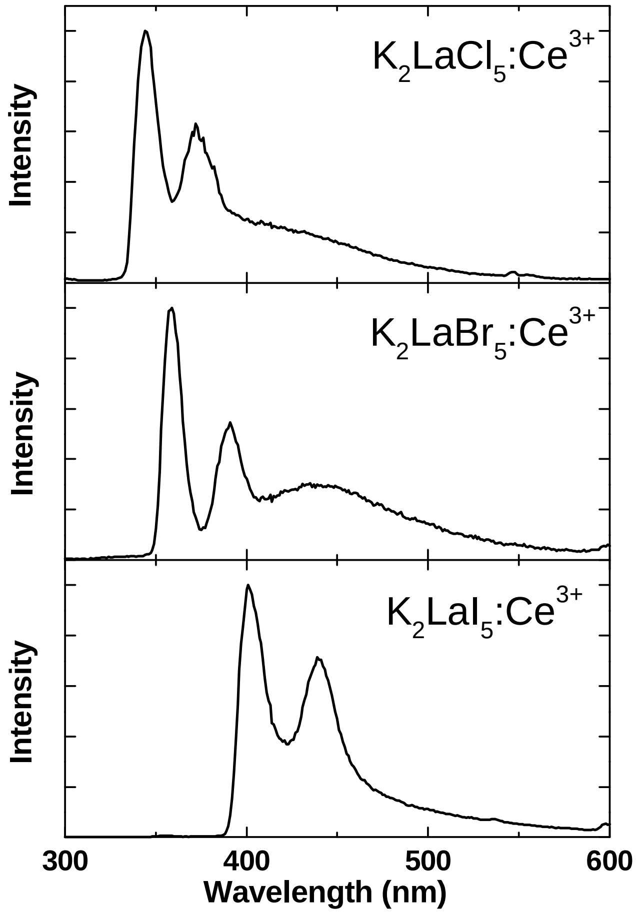
Efficiency of impurity activated scintillators

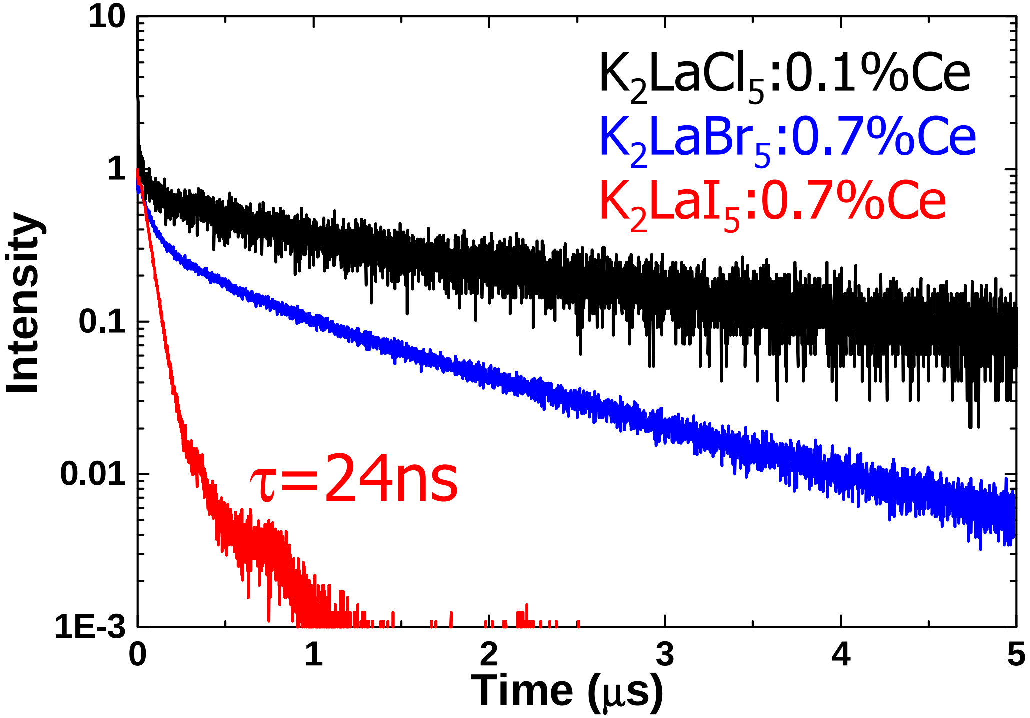
$$N_{dph} = f\left(N_{eh}, S, Q, \eta_{lc}, \eta_{pd}\right)$$



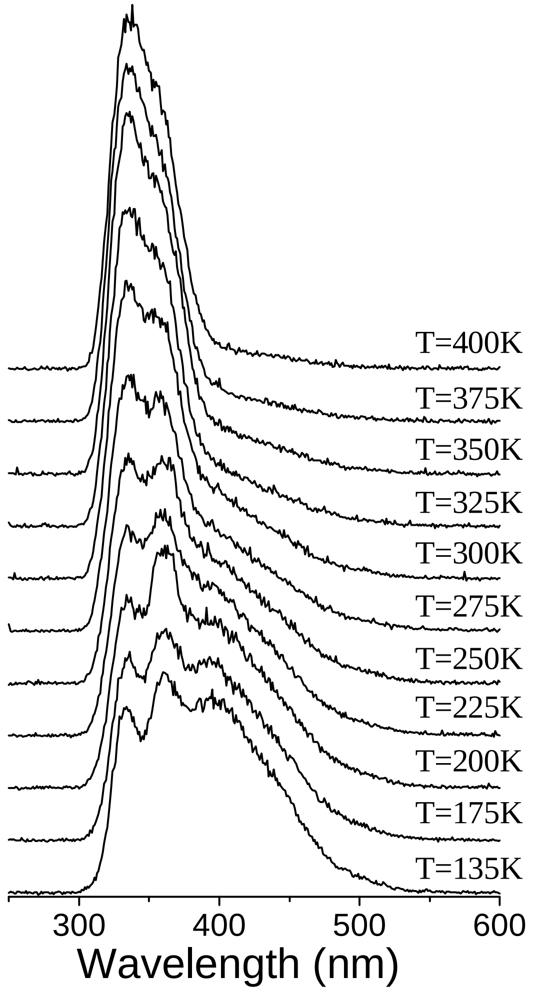


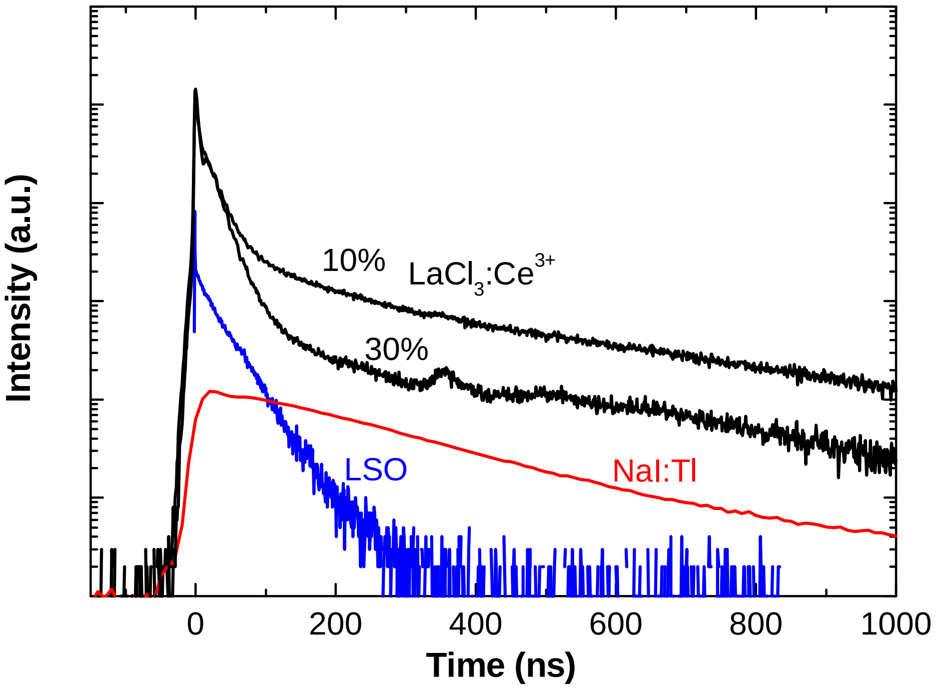
Intrinsic emission and slow transfer





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

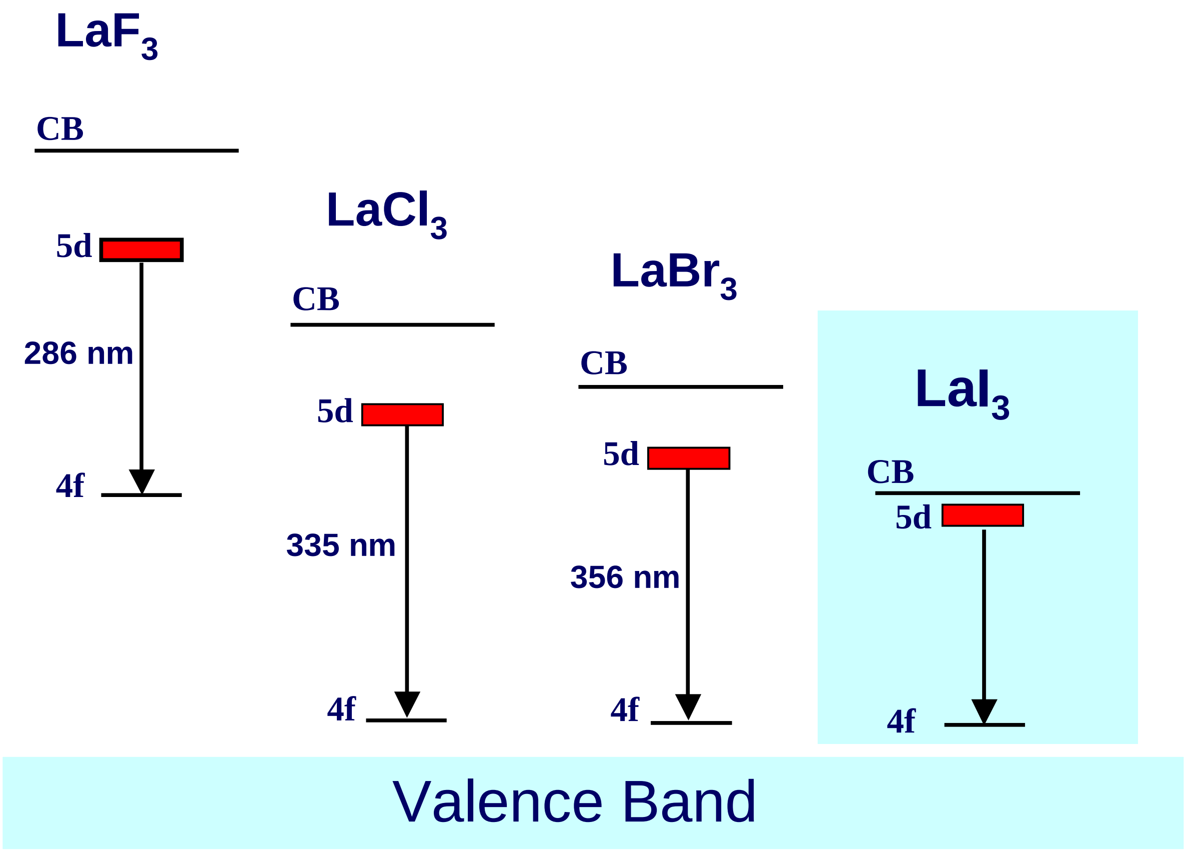




The LaX_3:Ce series

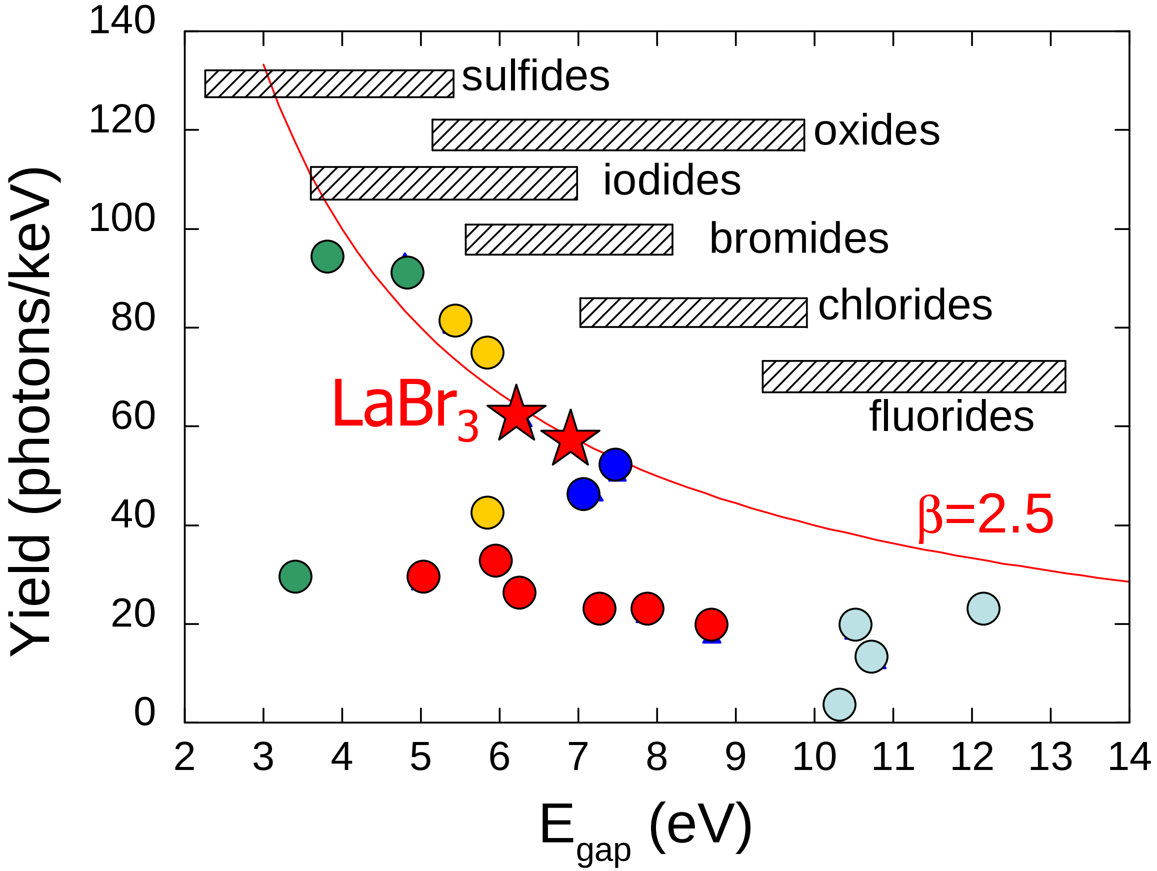
Number of thermalised electron-hole pairs

$\propto \quad E_\gamma / 2.5\, E_g$



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

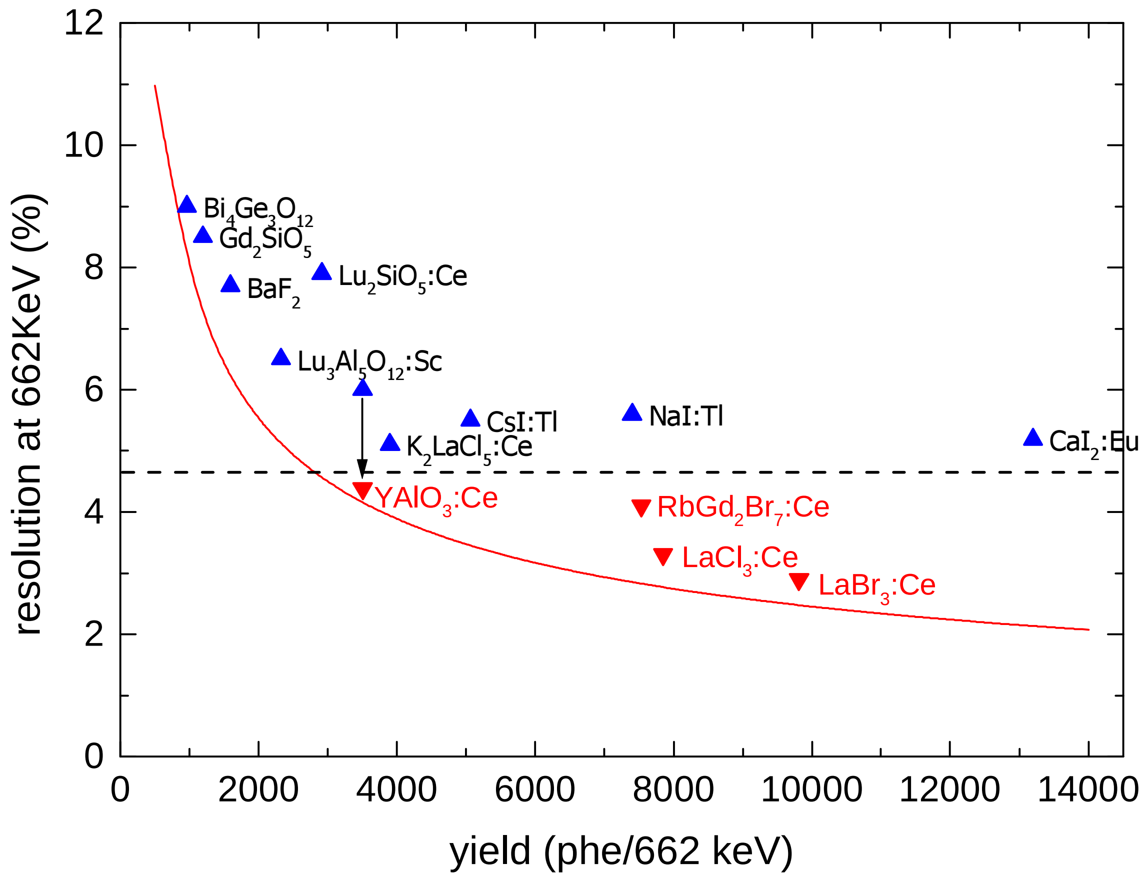
Limits to the light output



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

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

Limits to the energy resolution



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

Progress of past five years

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

Scintillator requirements for the ELOISATRON calorimeter

- Bunch spacing: $\tau_b < 20$ ns
- requirement: $\boxed{\tau_r = f\left(\tau_i, \tau_t, \tau_v, \tau_{lc}, \tau_{pd}\right) < \tau_b / 4 = 5ns}$
- 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

ELOISATRON calorimeter speed considerations

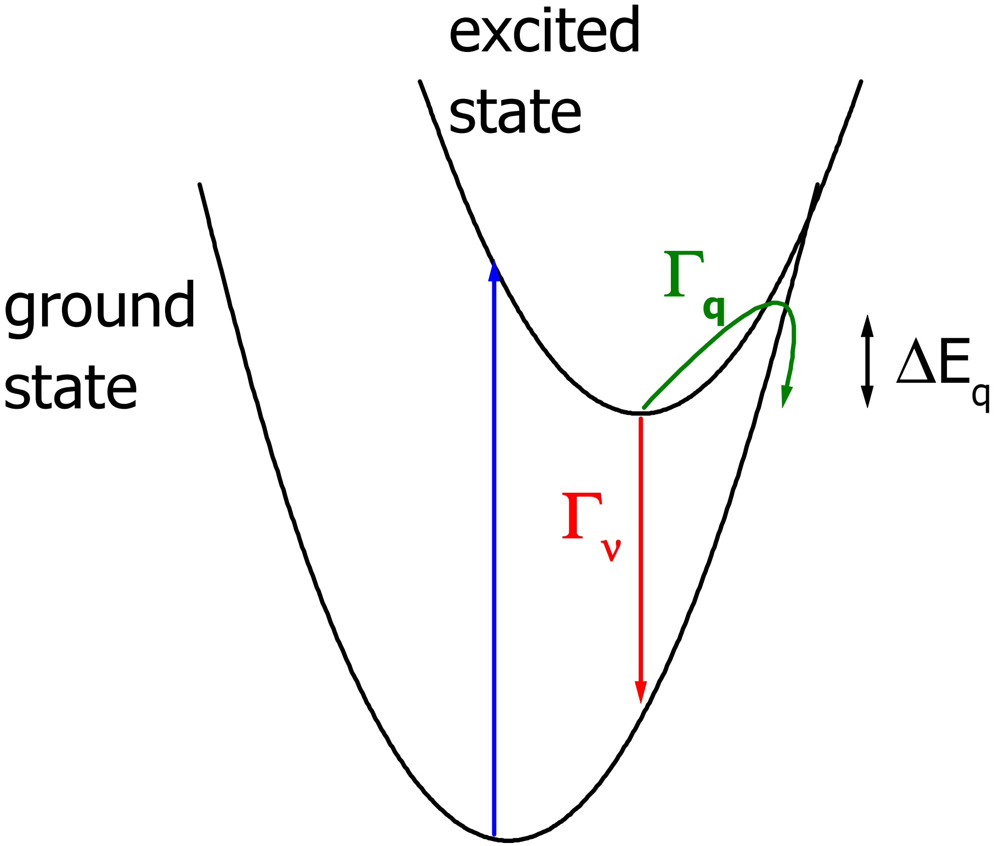
- Ce^{3+} is too slow
- Pr^{3+} and Nd^{3+} are faster, but in addition slow 4f→4f emission. Emission at too high energy for read-out
- CVL is fast (ns) but limited to too low density (halide) materials
- Remaining option
    - quenched luminescence

Internal luminescence quenching

$$\Gamma = \frac{1}{\tau} = \Gamma_\nu + \Gamma_q$$

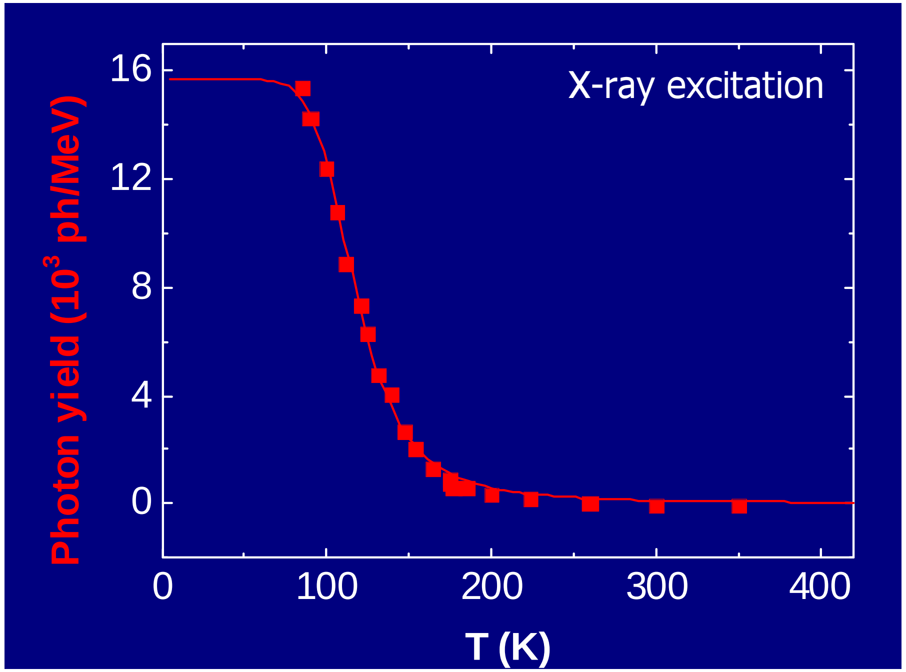
$$\Gamma_q(T) = \Gamma_0 \exp\left(\frac{-\Delta E_q}{k_B T}\right)$$

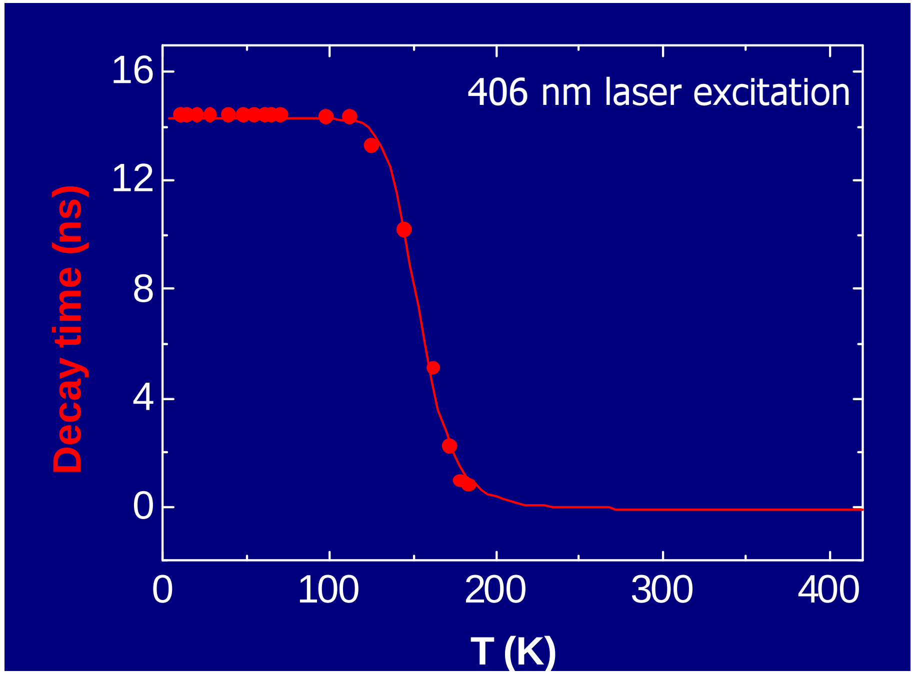
$$I = \frac{\Gamma_\nu}{\Gamma_\nu + \Gamma_q}$$



LaI_3:Ce; quenching via the conduction band?

T evolution





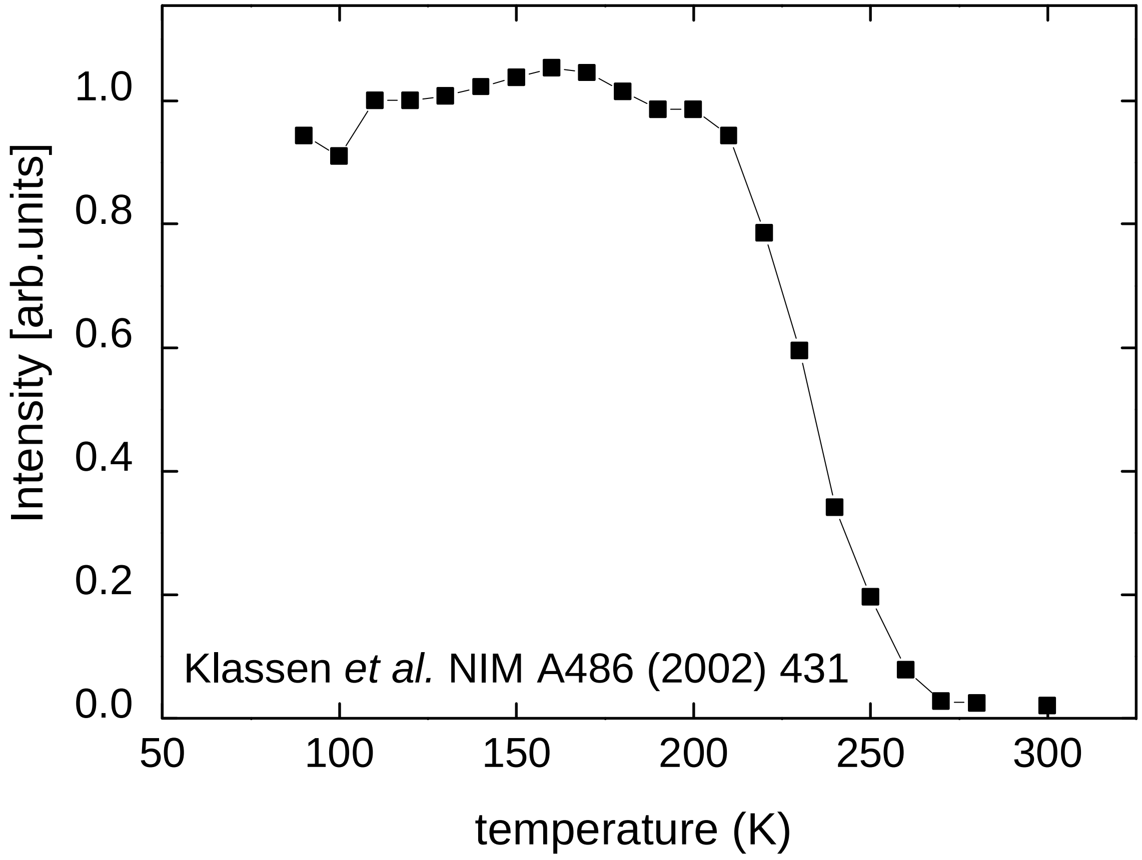
$$I(T) = \frac{I_0}{1 + \frac{\Gamma_0}{\Gamma_\nu} \exp\left(-\frac{\Delta E_q}{kT} \right)}$$

➡ $\Delta E_q = 0.08$ eV

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

➡ $\Delta E_q = 0.20$ eV

Quenching of $PbWO_4$ emission



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

-
-
-

Conclusion

crystal quality is more important than light yield

N.B. 1 photon/MeV→10^6 photons/TeV

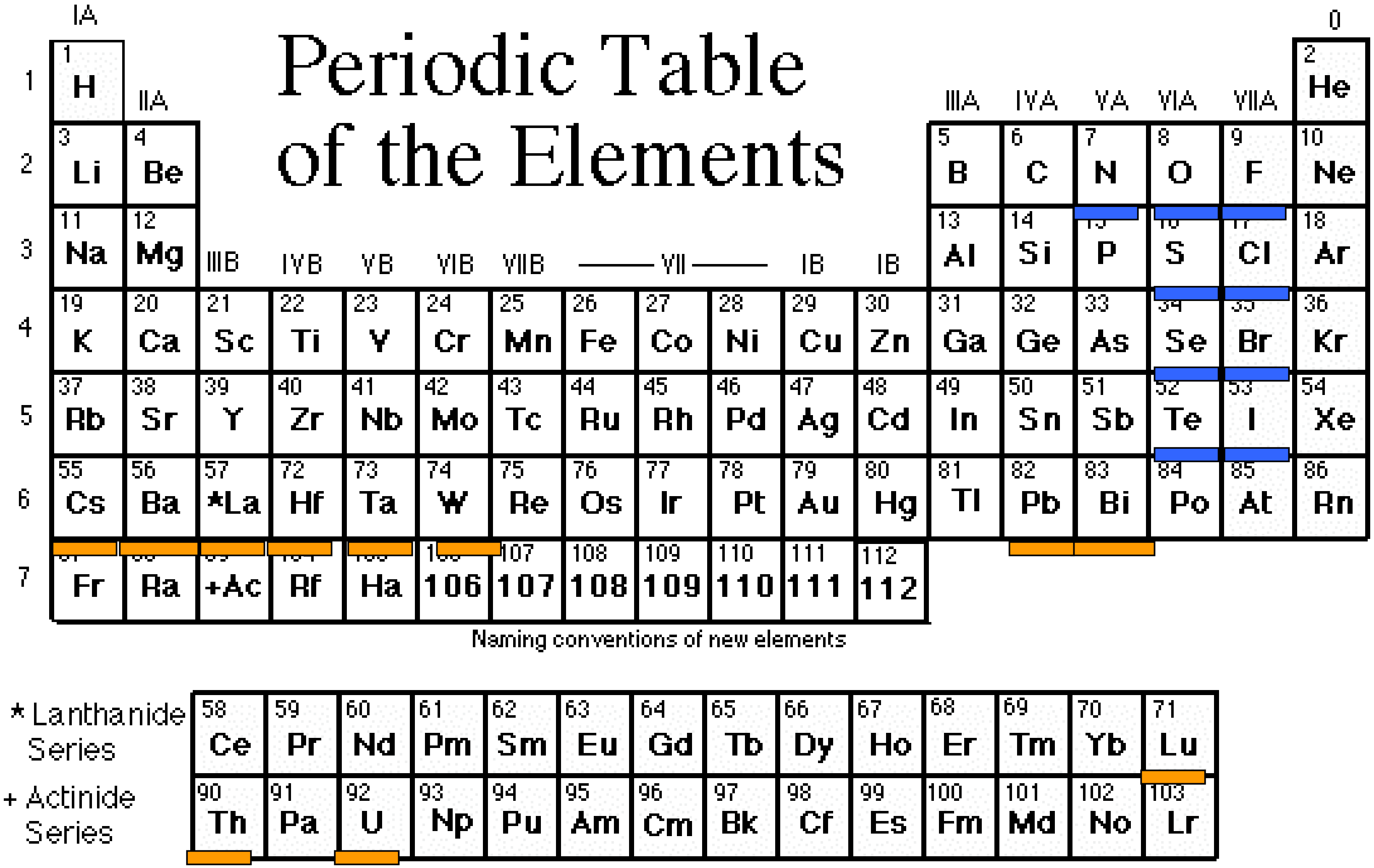
light output can be traded off against faster decay

- temperature fluctuations

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

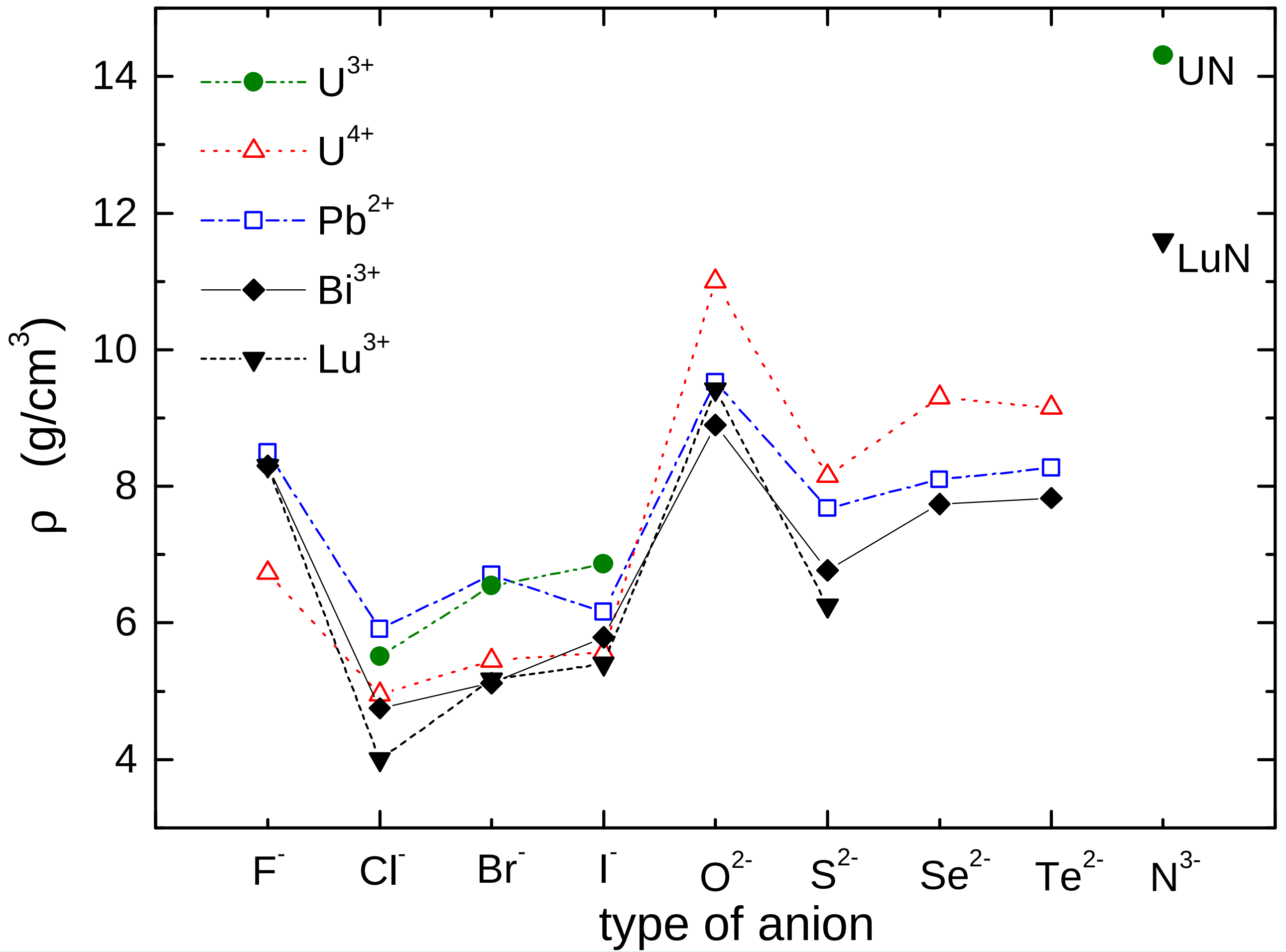


	IA	IIA	IIIB	IVB	VB	VIB	VIIB	—— VII ——			IB	IB	IIIA	IVA	VA	VIA	VIIA	0
1	1 H																	2 He
2	3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne
3	11 Na	12 Mg											13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
4	19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr
5	37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe
6	55 Cs	56 Ba	57 *La	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
7	87 Fr	88 Ra	89 +Ac	104 Rf	105 Ha	106 106	107 107	108 108	109 109	110 110	111 111	112 112						

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

Density of binary compounds

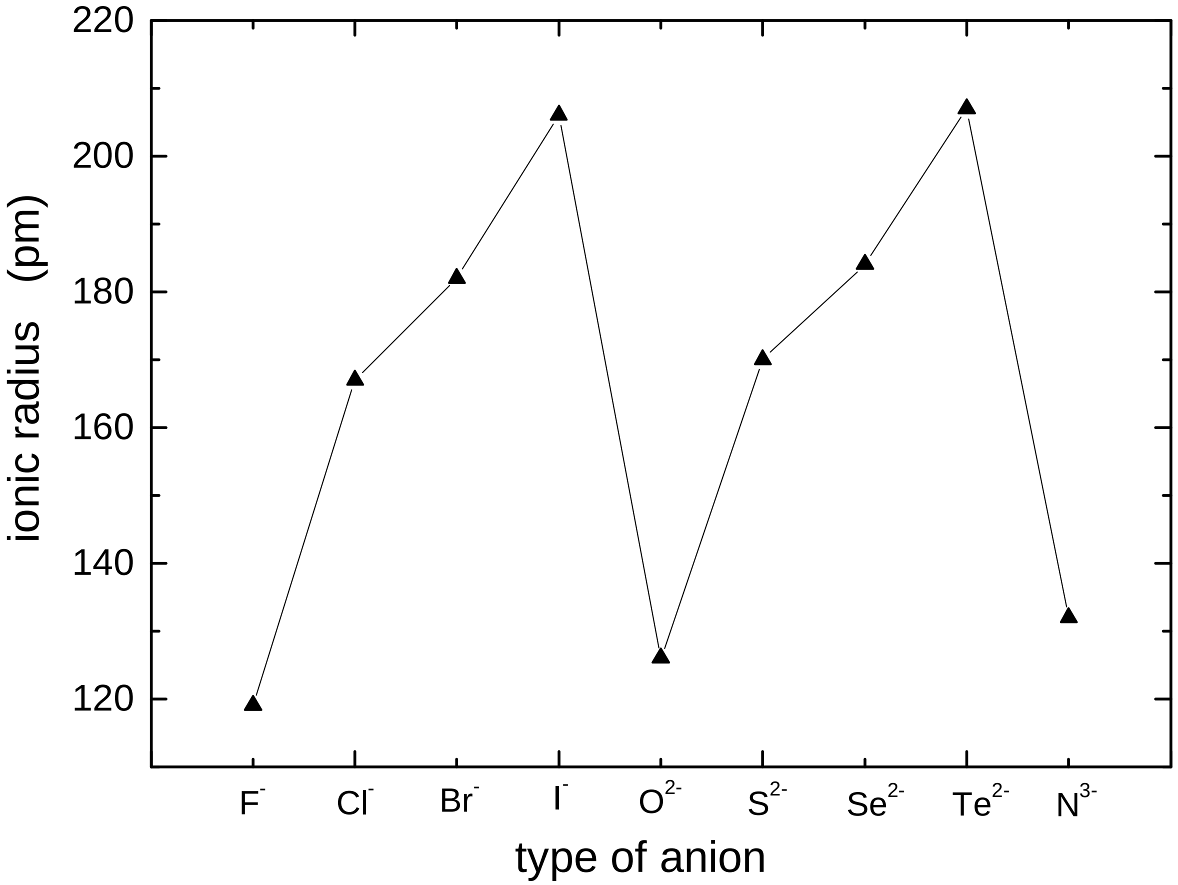


$F > Cl \le Br \le I$

$O > S \le Se \le Te$

$N > O > F$

Ionic radii of anions in inorganic compounds

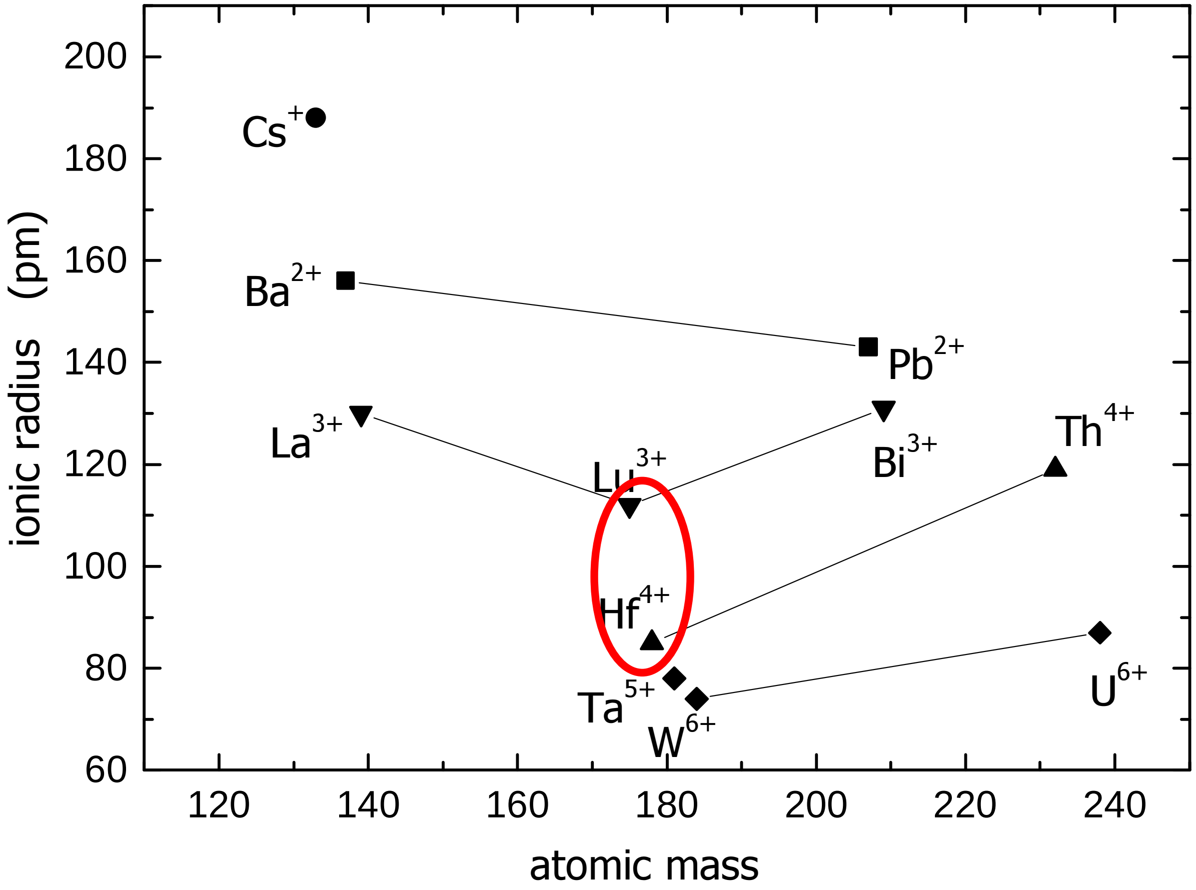


What compound can do the job?

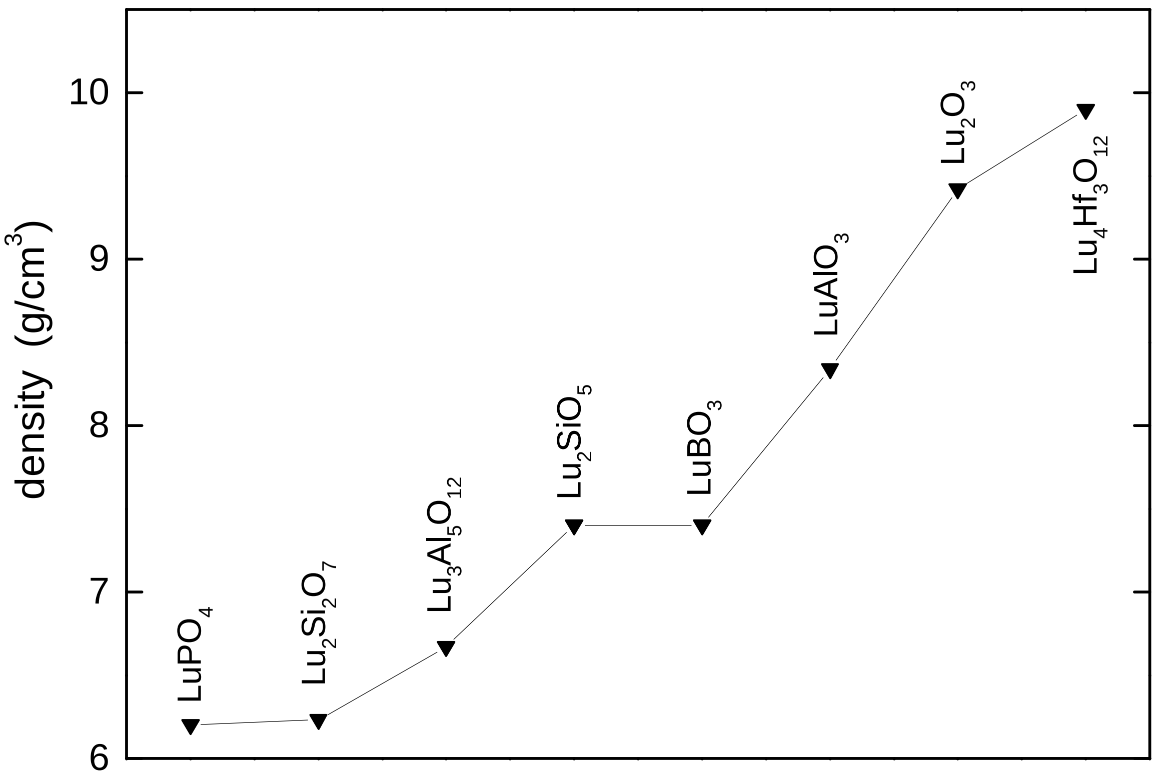
- High density of fluorides, oxides, and nitrides is related with small ionic radius
- Density $LuF_3 < Lu_2O_3 < LuN$ because of higher charge of anion → larger cation/anion ratio
- Nitride compounds is not a feasible option for calorimeter

Conclusion
The highest density compounds must be found amongst the oxides

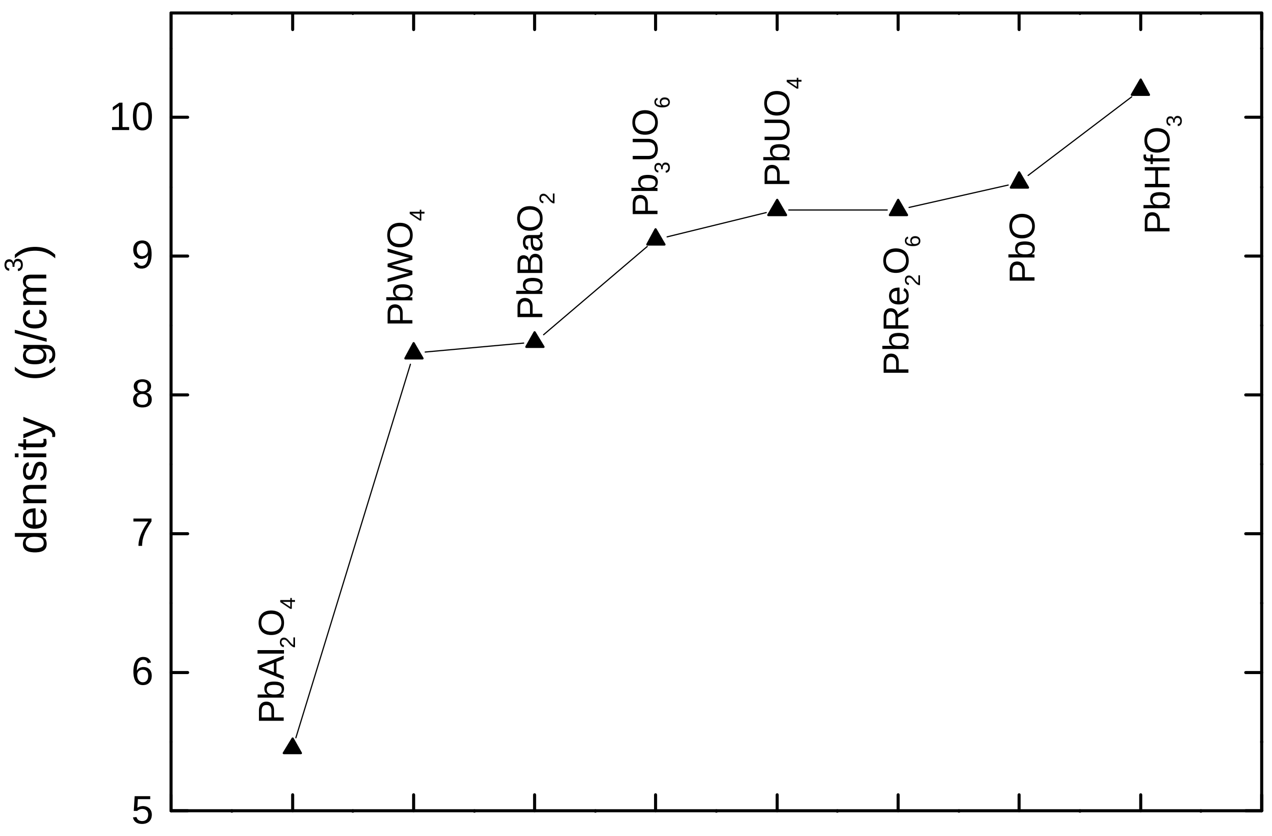
Ionic radii of high atomic number cations



Density of ternary Lu-based compounds



Density of ternary Pb-based compounds



Future prospects and directions for research

- Ultimate density of 10 g/cm^3 is feasible
- to obtain $\tau_\nu < 4$ns, a quenched luminescence mechanism is needed
- impurity activation may introduce problems with
    - transfer time
    - concentration gradients →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