New properties and prospects for hot intraband luminescence

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ISMART, Minsk, October 11, 2018

10-ps challenge

Goal:

Increase scintillation time resolution to **10 ps**

Applications:

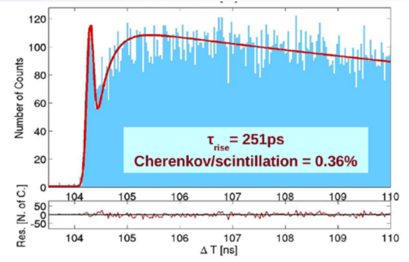
- Time-of-flight positron emission tomography (TOF-PET)
- High-energy physics (pile-up events discrimination)
- X-ray imaging at GHz frame rate (XFEL, rapid processes, ...)
- Space telescopes: miniature time-of-flight neutron detectors

FAST
Fast Advanced Scintillator Timing

Prompt photons concept for high-resolution TOF-PET

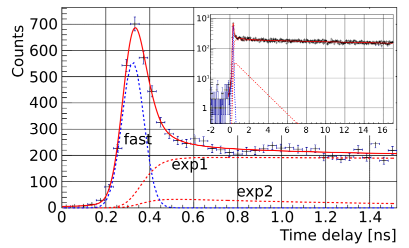
Scintillation under 511 keV γ-excitation (small crystals)

LuAG:Pr
CTR **205 ps** (2x2x10 mm^3)



[1] S. Gundacker et al, Phys. Med. Biol. 61 (2016) 2802

BGO
CTR **330 ps** (3x3x20 mm^3)



[2] S E Brunner and D R Schaart Phys. Med. Biol. 62 (2017) 4421

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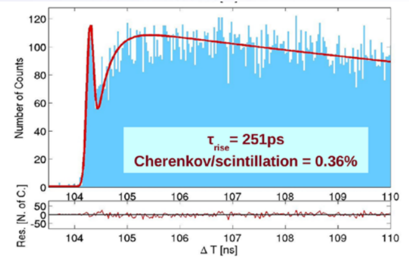

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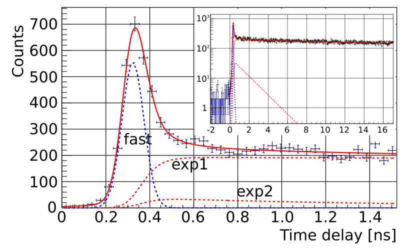

Conventional scintillation record
LYSO:Ce,Ca: CTR **100 ps** (2x2x10 mm^3)

FAST — Fast Advanced Scintillator Timing

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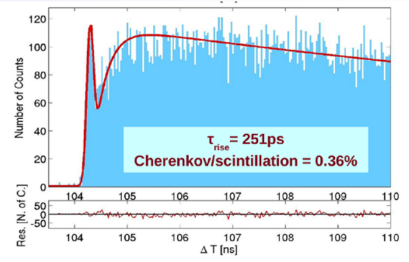

BGO

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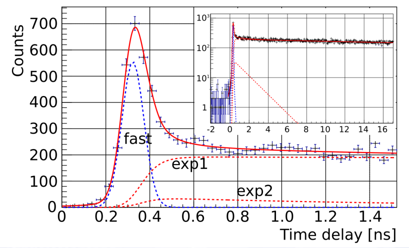
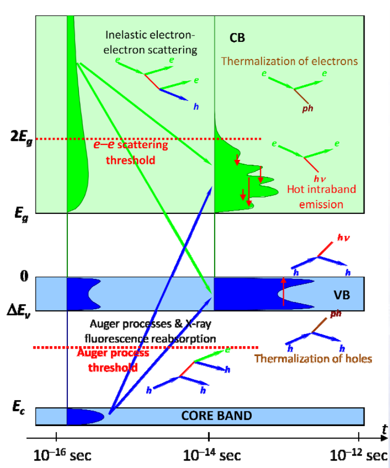


Conventional scintillation record

LYSO:Ce,Ca: CTR **100 ps** (2x2x10 mm^3)

Need more prompt photons!

Intraband luminescence (IBL) mechanism



The mechanism of IBL (drawing by A. Vasil'ev)

- Decay 10^{-12} s
- Broad structureless spectrum
- Yield determined by branching ratio (D. Vaisburd,1980s):

$$\eta = \frac{P_{rad}}{P_{nonrad}} \approx \frac{\tau_{nonrad}}{\tau_{rad}} \approx$$

$$\approx \frac{1ps}{10ns} \approx 10^{-4} \to 10 \ ph/MeV$$

The experimental facts about IBL in insulators

- Observed under excitation by (sub)nanosecond pulses of electrons and laser radiation

First publication:

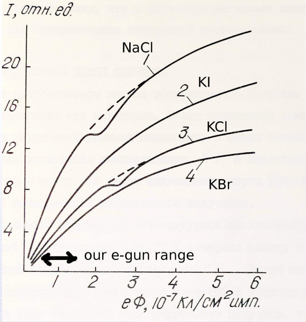
D.I. Vaisburd et al. Izv. Nauk AS USSR. Phys. Ser. 38/6 (1974) 1281-1284

The experimental facts about IBL in insulators

- Observed under excitation by (sub)nanosecond pulses of electrons and laser radiation
- No excitation density threshold found

First publication:

D.I. Vaisburd et al. Izv. Nauk AS USSR. Phys. Ser. 38/6 (1974) 1281-1284



Excitation pulse density dependence of IBL

D. I. Vaisburd et al, *High-energy solid-state electronics*, Izdatel'stvo Nauka, Novosibirsk, 1982. In Russian.

The experimental facts about IBL in insulators

- Very fast decay: $\tau < 10$ ps (R. Deich), must be around 10^{-12} s.

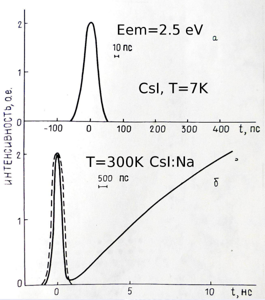


Figure: The IBL time shape at 2.5 eV of CsI and CsI:Na from Deich thesis and [2]. Excitation by electron beam, pulse width 50 ps

[2] R. Deich and M. Abdrakhmanov, NIM B 65 (1992) 525–529

The experimental facts about IBL in insulators

- Alkali halides (D. Vaisburd):
  - Structureless spectrum, terminated by fundamental absorption
  - Independence on temperature and impurity content
  - Quantum yield in KI about 10^{-4}

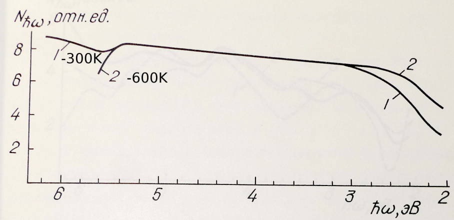


Рис. I.4. Спектры люминесценции кристалла RbCl при наносекундном возбуждении плотным потоком электронов: I – 300 К, 2 – 600 К

The IBL spectra of RbCl, excited by electron beam (3ns)

D. I. Vaisburd et al, *High-energy solid-state electronics*, Izdatel'stvo Nauka, Novosibirsk, 1982. In Russian.

The experimental facts about IBL in insulators

- Hole component spectrum
  reflects valence band structure, terminated by its width
- In alkali halides the e-IBL is 20–60 times more intense
- In wide-gap oxides and some other materials the h-IBL is dominating (A. Lushchik, F. Savihhin et al.)

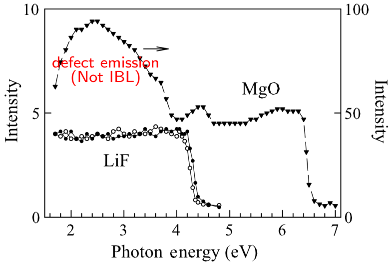


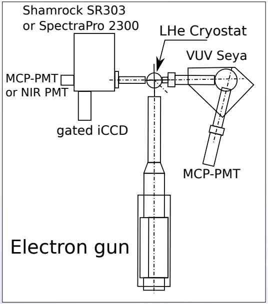
Fig. 7. The spectra of intraband luminescence ($\tau < 2$ ns) measured for LiF at 80 (○○) or 300 K (●●) and for a MgO crystal at 730 K (▼▼) under irradiation by single 300 keV electron pulses.

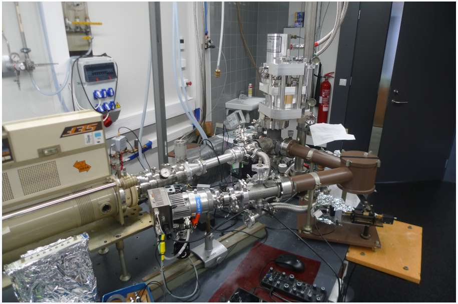
[4] A. Lushchik et al, NIM B 250 (2006) 330–336

Motivation for modern studies

- Remarkable advances in technical development:
  - pulsed excitation sources: fs lasers, ps x-ray and electrons, FEL
  - fast light detectors: MCP-PMT, SPAD and SiPM, Hybrid detectors, Streak cameras
- New application challenges: 10ps TOF-PET, GHz Xray cameras, future LHC pileup correction
- The search for materials with higher IBL yield

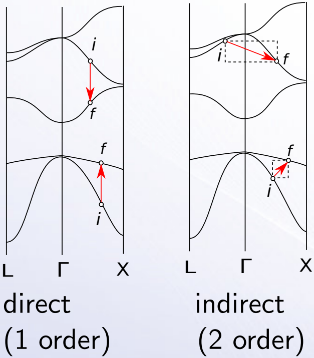
The pulse cathodoluminescence setup in Tartu





- Electron energy 100–200 keV, pulse current up to 50 A/cm^2
- **High resolution mode: 55 ps FWHM**
- Fast to slow emissions ratio (dynamic range up to seconds)
- Spectral range 0.75–10.6 eV (117–1650 nm)
- Temperature range 5.5–800 K

New spectral features



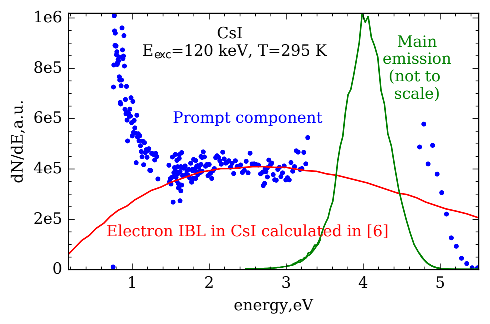
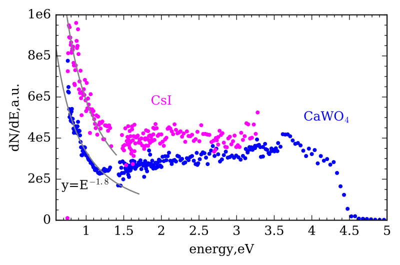


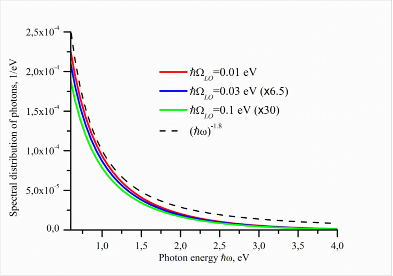
Figure: The types of radiative intraband transitions and IBL spectra of CsI

[6] D. I. Vaisburd, S. Kharitonova, Russian Physics Journal, 1997, 40:1037

Experimental and theoretical 2-order spectrum of IBL



experimental



theoretical

Figure: Spectral distribution of the number of prompt photons emitted by sample during electron pulse. Normalized arbitrarily.

S.I. Omelkov, V. Nagirnyi, A.N. Vasil'ev, M. Kirm J. Lumin., 2016, 176, 309–317
A. N. Vasil'ev, R. V. Kirkin, Physics of Wave Phenomena, 2015, 23(3): 186

FAST Fast Advanced Scintillator Timing

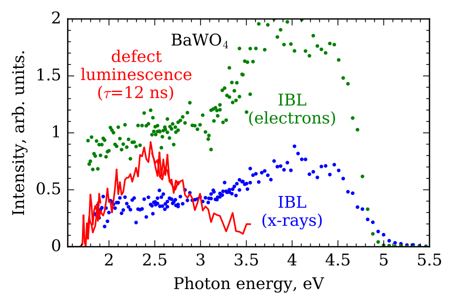
10-ps challenge

Question:

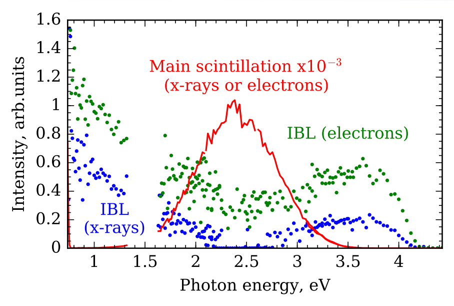
Can IBL be excited by single **x-ray** and 511-keV γ-**quanta**?

x-ray excited IBL of $Bi_4Ge_3O_{12}$ and $BaWO_4$

$BaWO_4$



BGO



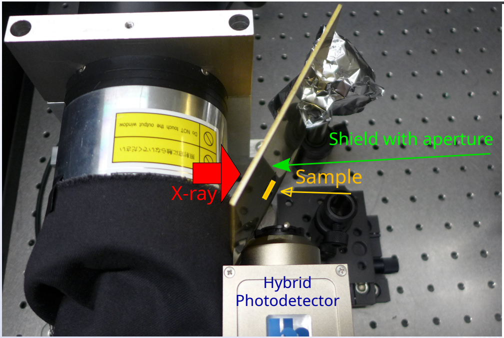
IBL spectra under electron beam and x-ray excitation, normalized to the same intensity of main scintillation band.

Electron energy: 80–110 keV; X-ray energy 10–80 keV

S.I. Omelkov et al J. Lumin., 2017, 191, 61–67

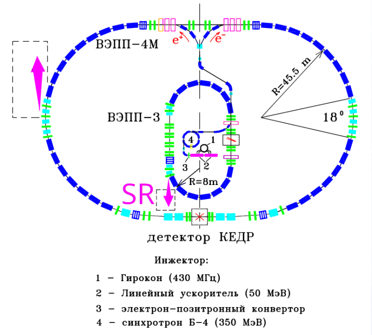
IBL under x-ray excitation: nonproportionality effects?

XRL setup in CERN
(Laser-exited xray tube)



3–40 keV max 10 keV
Low power, no spectral resolution
2 MHz repetition rate
$\sim$100 ps FWHM time resolution

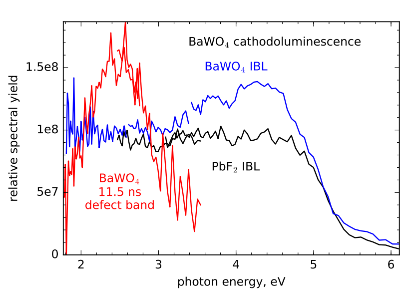
XRL setup on VEPP-3 ring
(2 GeV, Novosibirsk)



3–60 keV max 10 keV
High power, used 55 μm Cu filter 8.06 MHz repetition rate
$\sim$1 ns FWHM time resolution

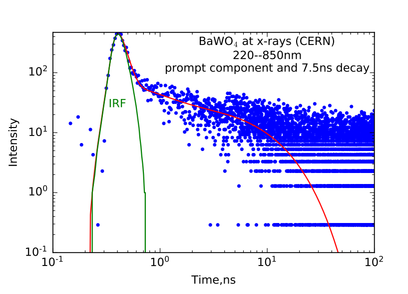
IBL in $BaWO_4$

Tartu
X-ray luminescence (10–80 keV)



Yield(IBL)=2.2×Yield(defect)

CERN
X-ray $\lesssim$ 40 keV



Yield(IBL)=0.2×Yield(defect)

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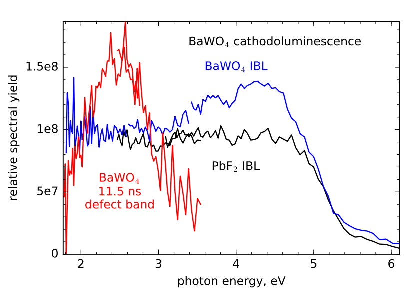
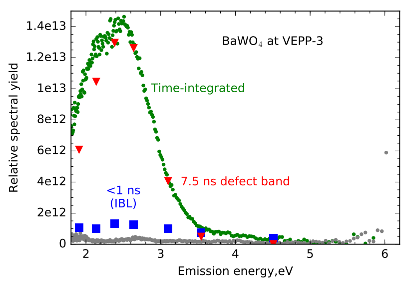


Yield(IBL)=2.2×Yield(defect)

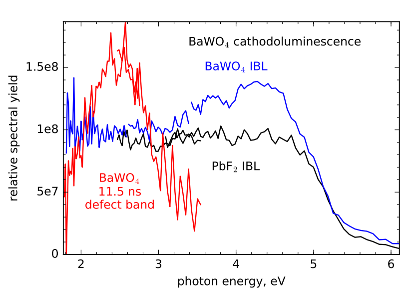
Novosibirsk
X-ray $\leq$ 60 keV (VEPP-3)



Yield(IBL)=0.3×Yield(defect)

IBL in $BaWO_4$

Tartu
X-ray luminescence (10–80 keV)



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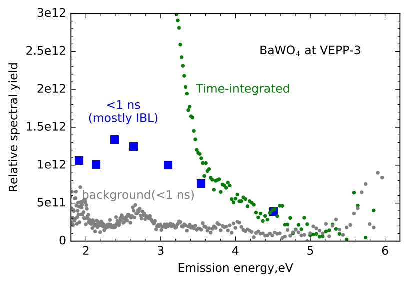


Yield(IBL)=0.3×Yield(defect)

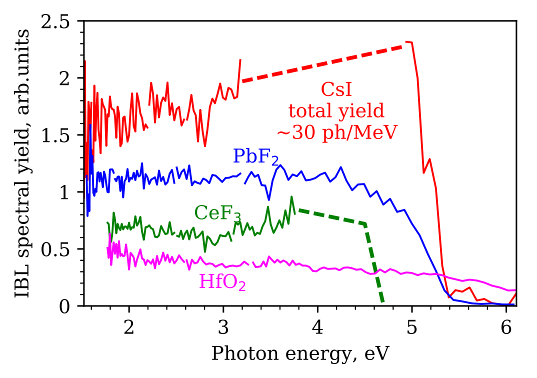
10-ps challenge

Question:

What is the brightest IBL emitter?

IBL spectral yield - binary compounds

120 keV 200 ps electron beam excitation



Highest phonon modes from Raman scattering, cm^{-1}:

- CsI: 90[1]
- PbF_2: 338[2]
- CeF_3: 396[3]
- HfO_2: 680[4]

Yield determined from spectra relative to LYSO:Ce

[1] S. Ganesan et al, *J. Phys. I*, 1965, 26(11): 639
[2] M. Dickens and M. Hutchings *J. Phys. C: Solid State Phys.*, 1978, 11: 461
[3] R.P. Bauman, S.P.S. Porto *Phys. Rev.*, 1967, 161(3): 842
[4] E. Anastassakis et al *J. Phys. Chem. Solids*, 1975, 36: 667

Radiative lifetimes depend on band structure?

$$\eta = \frac{P_{rad}}{P_{nonrad}} \approx \frac{\tau_{nonrad}}{\tau_{rad}}$$

τ_{nonrad} ← Depends on phonon spectrum

τ_{rad} ← Depends on initial and final states

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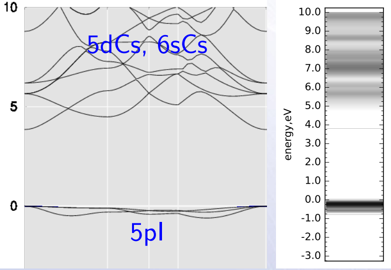
Band structure fragment for CsI from AFlowLIB.org



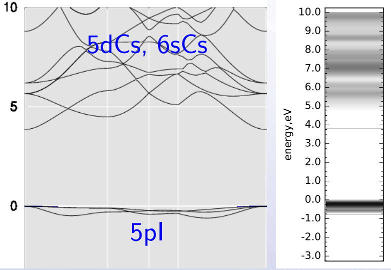
Radiative lifetimes depend on band structure?

$$\eta = \frac{P_{rad}}{P_{nonrad}} \approx \frac{\tau_{nonrad}}{\tau_{rad}}$$

Depends on phonon spectrum (τ_{nonrad})

Depends on initial and final states (τ_{rad})

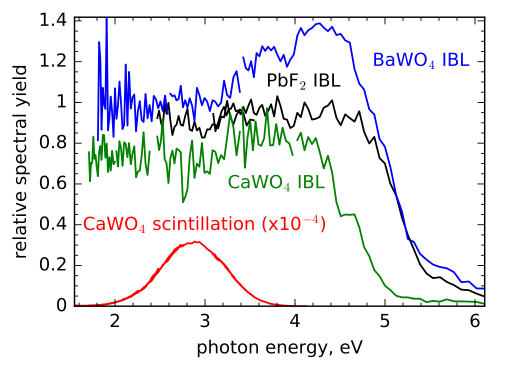
Band structure fragment for CsI from AFlowLIB.org



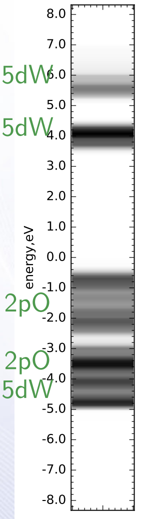
How to improve?

- Make use of hole transitions
- Diversify states (s,p,d,f)
- Increase DOS
- Include gaps in DOS

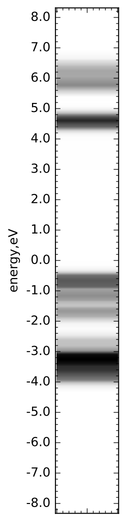
IBL and band structure: tungstates



The IBL yields:
$CaWO_4$ - 13 ph/MeV
PbF_2 - 17 ph/MeV
$BaWO_4$ - 21 ph/MeV
Band structure from [Lacomba-Perales et al J. Appl. Phys. 110 043703 (2011)]

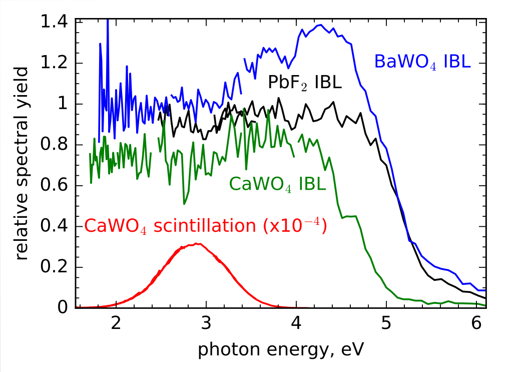


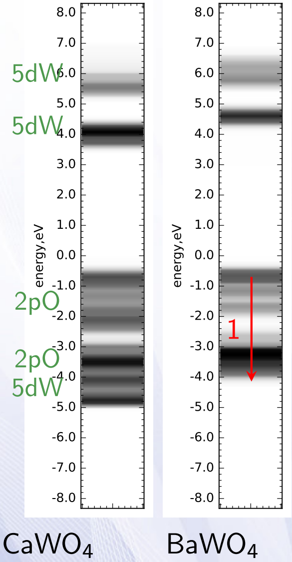
$CaWO_4$



$BaWO_4$

IBL and band structure: tungstates





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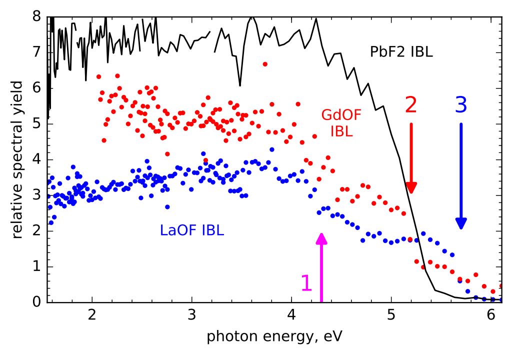
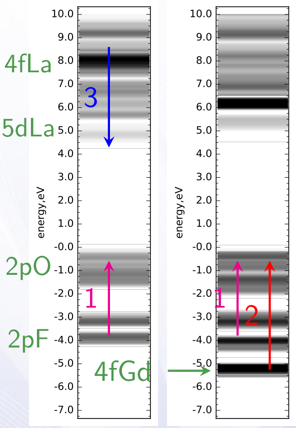
Band structure from [Lacomba-Perales et al J. Appl. Phys. 110 043703 (2011)]

IBL and band structure: rare-earth oxyfluorides

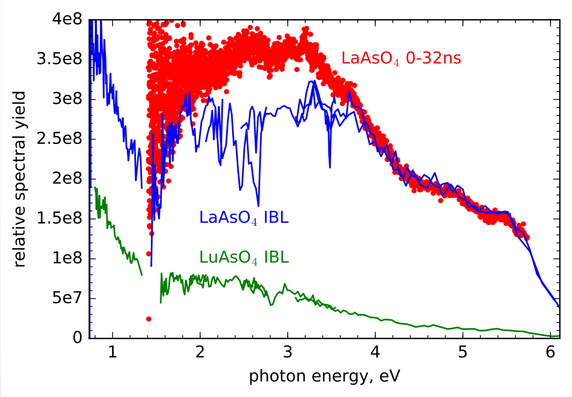


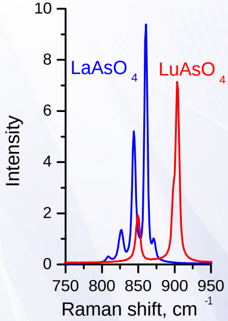
Band structure from AFlowLIB.org



LaOF GdOF

IBL and band structure: rare-earth arsenates





The IBL yields:

$LaAsO_4$≈**25–30 ph/MeV**

More on IBL and phonons: S. Omelkov et al, J.Lumin 198 (2018) p260

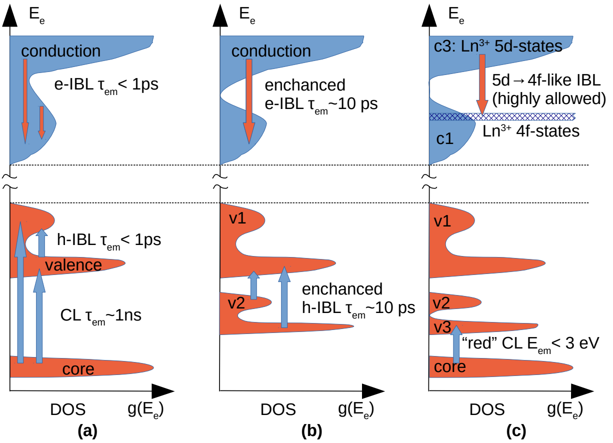
FAST
Fast Advanced Scintillator Timing

10-ps challenge

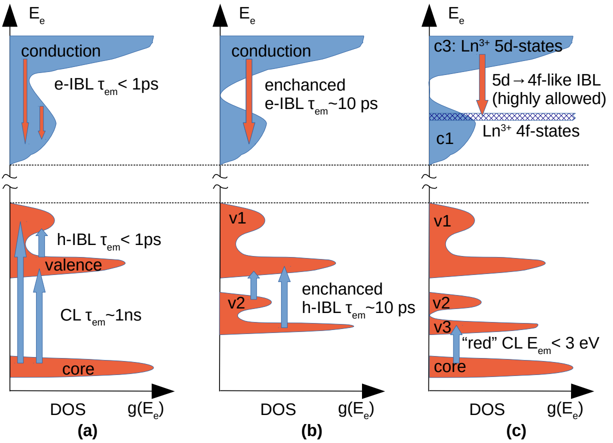
Question:

Is there a way to get more prompt photons?

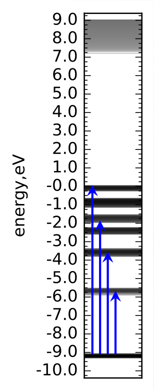
Band structure engineering



Band structure engineering

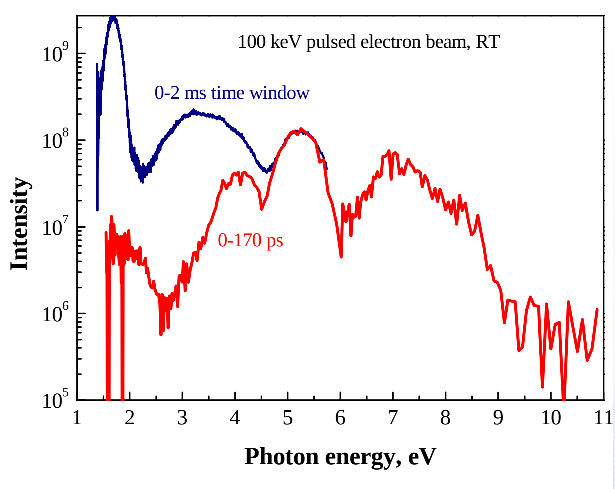


K_2SiF_6



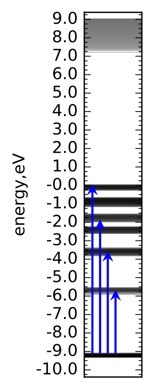
(AFlowLIB.org)

Band structure engineering



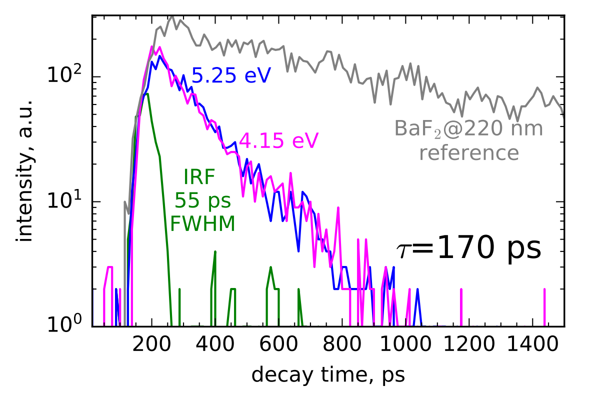
Spectrum of VUV, UV and red crossluminescence

K_2SiF_6



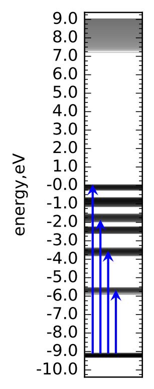
(AFlowLIB.org)

Band structure engineering



Decay of crossluminescence in K_2SiF_6

K_2SiF_6



(AFlowLIB.org)

Fast Advanced Scintillator Timing

10-ps challenge

Question:

What IBL is good for?

The prospects of hot intraband luminescence

- IBL rise time: femtoseconds
- IBL decay: $<$1ps
- IBL yield: 30 ph/MeV (possibly more)

The prospects of hot intraband luminescence

- IBL rise time: femtoseconds
- IBL decay: <1ps
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Potential time resolution on IBL: $<$**1 ps**

Problem: we need a detector with same time resolution

The prospects of hot intraband luminescence

- IBL rise time: femtoseconds
- IBL decay: $<$1ps
- IBL yield: 30 ph/MeV (possibly more)

Potential time resolution on IBL: $<$**1 ps**

Problem: we need a detector with same time resolution

SPAD approached **7.8 ps** FWHM

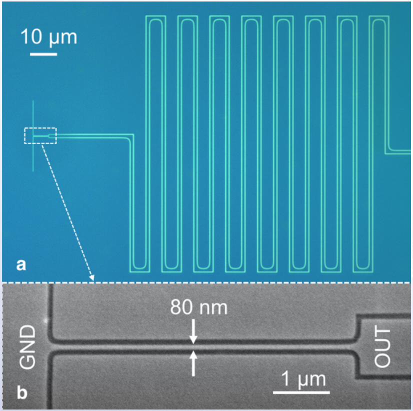
F. Nolet et al, Instruments 2018, 2, 19

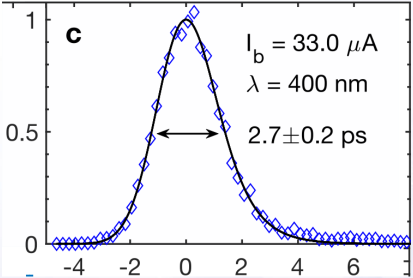
Tynode-based PMT promises $<$**10ps**

H. van der Graaf et al, NIM A 2017, 841, 148

The prospects for hot intraband luminescence

Supercondictive nanowire single-photon detectors (2018)





- **SPTR <3 ps FWHM**
- PDP ≈100%
- sensitive area 0.08x5μm^2
- cryogenic operation (0.9 K)

From arXiv:1804.06839 [physics.ins-det]

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- **Picosecond scintillation timing is possible**

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Thank you for attention