Introduction, overview and present status of inorganic scintillators

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Introduction

How to detect ionizing radiations?

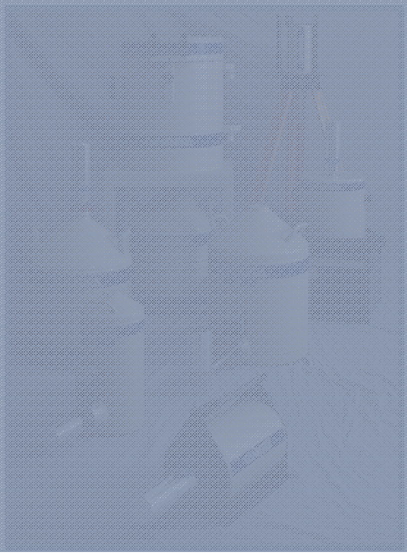
Ionizing radiations: X-ray, gamma-ray, α, neutrons, ions, electrons, VUV….

Detection requires electric pulse

Interaction radiation-matter: Ionizing -> electrons extraction

Direct charge detection

Geiger counting system, Semi-conductor



Indirect charge detection

Charges to light conversion
And light detection (PMT, CCD, SiPM…)

scintillation

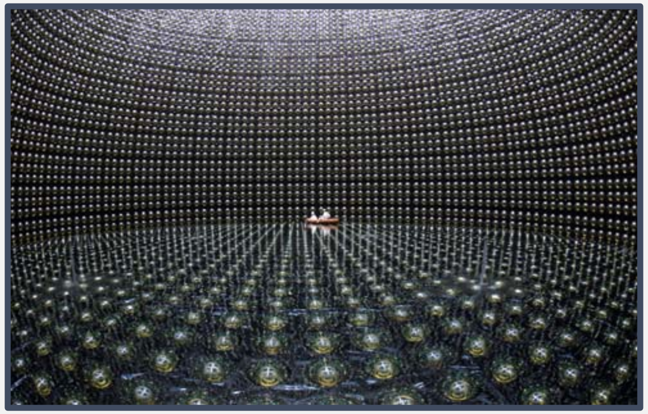
How to detect ionizing radiations?

Several scintillator classes

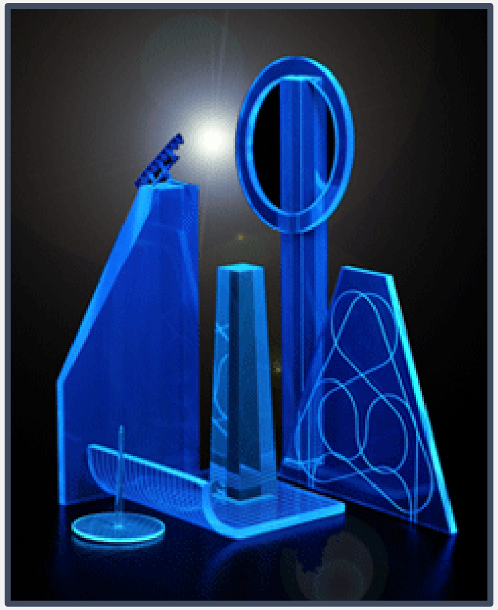
Organic solids

Liquids

Inorganic solids



Kamiokande
(neutrino detection)

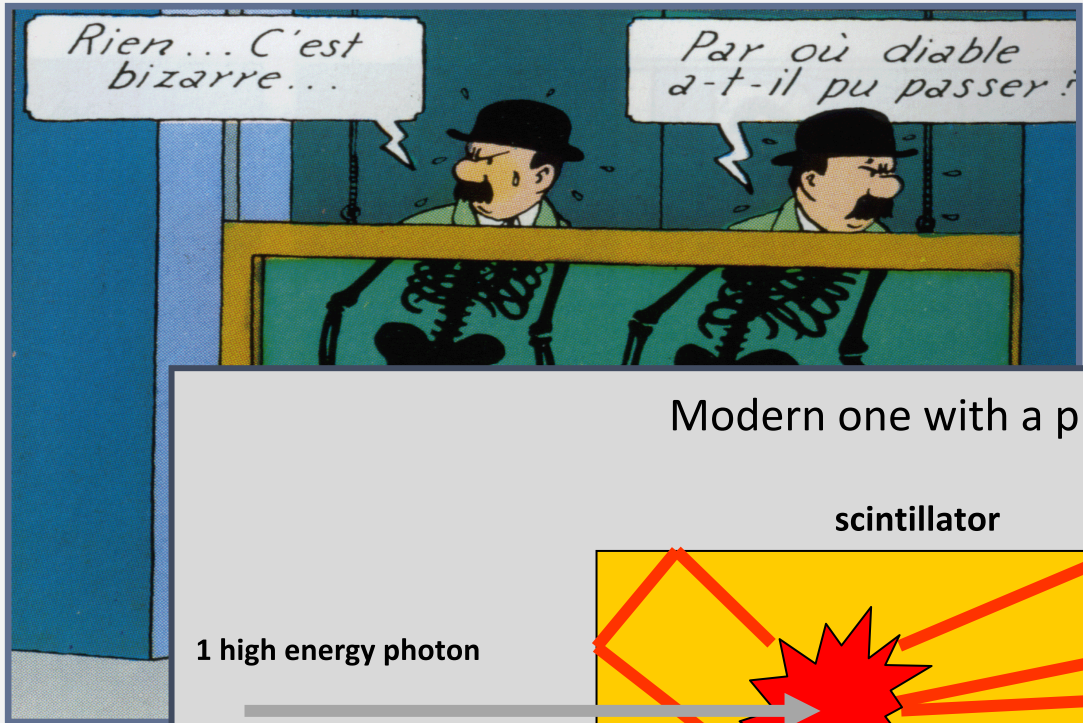


Commercial plastic Scintillators (Saint Gobain as exemple)

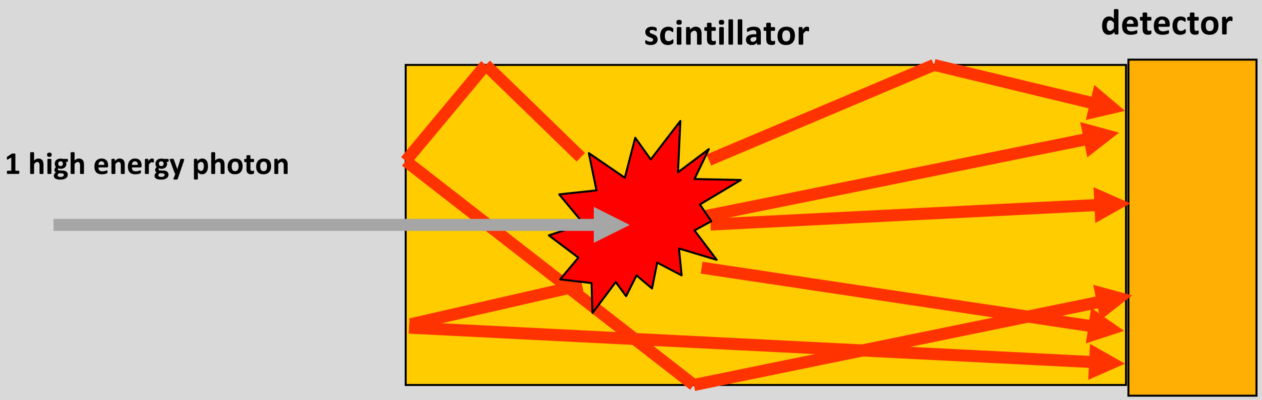
$PbWO_4$ crystals: CERN

History of scintillators connected to the detectors

Old style



Modern one with a photodetector (PM, CCD…)



1 high energy photon (several 1000 eV) → Several 1000 of photons IR, Vis ou UV (1-4 eV)

History of scintillators connected with detectors

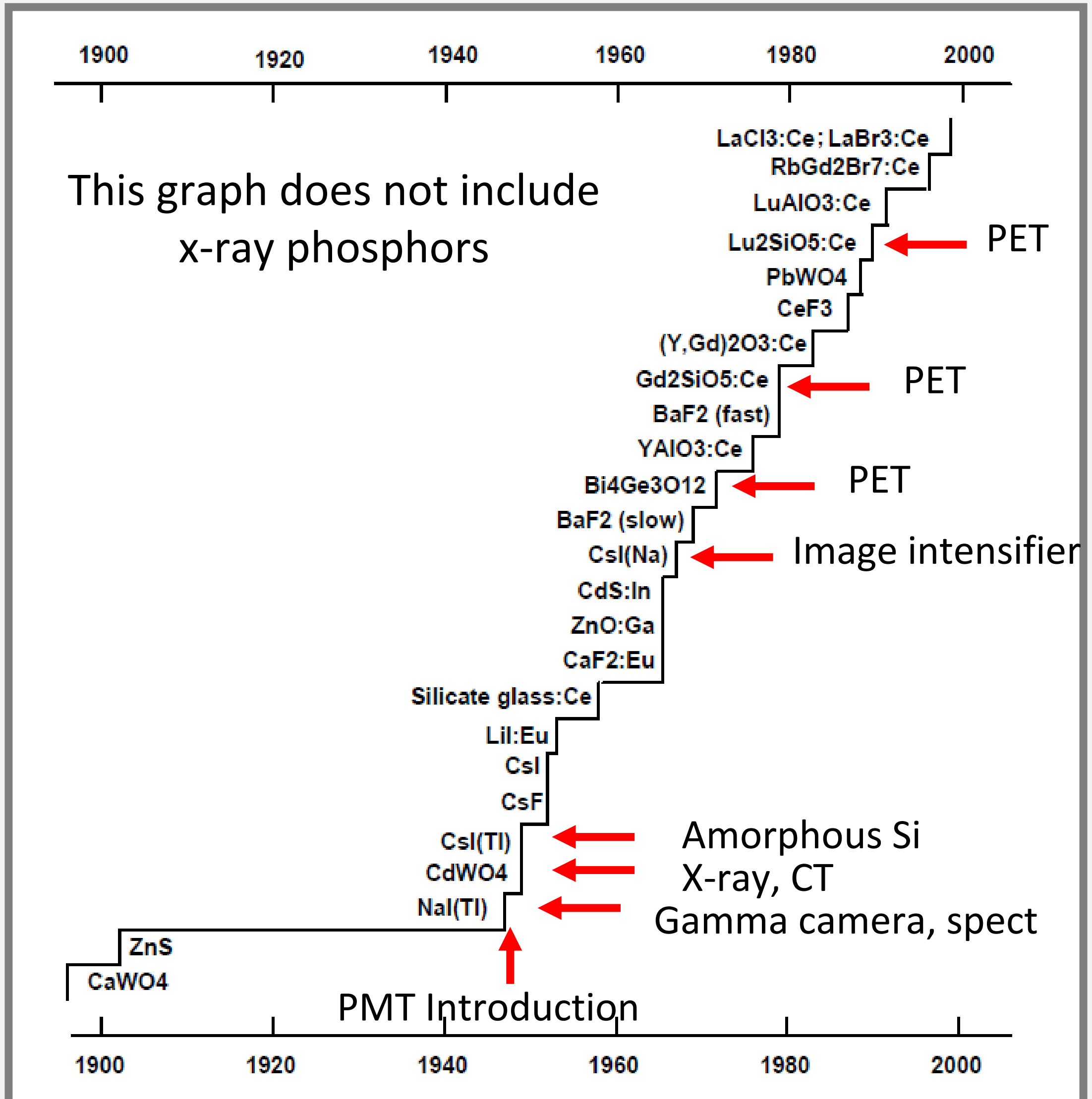


Fig. 1. History of the discovery of major inorganic scintillator materials.

M.J.Weber, Journal of Luminescence 2002

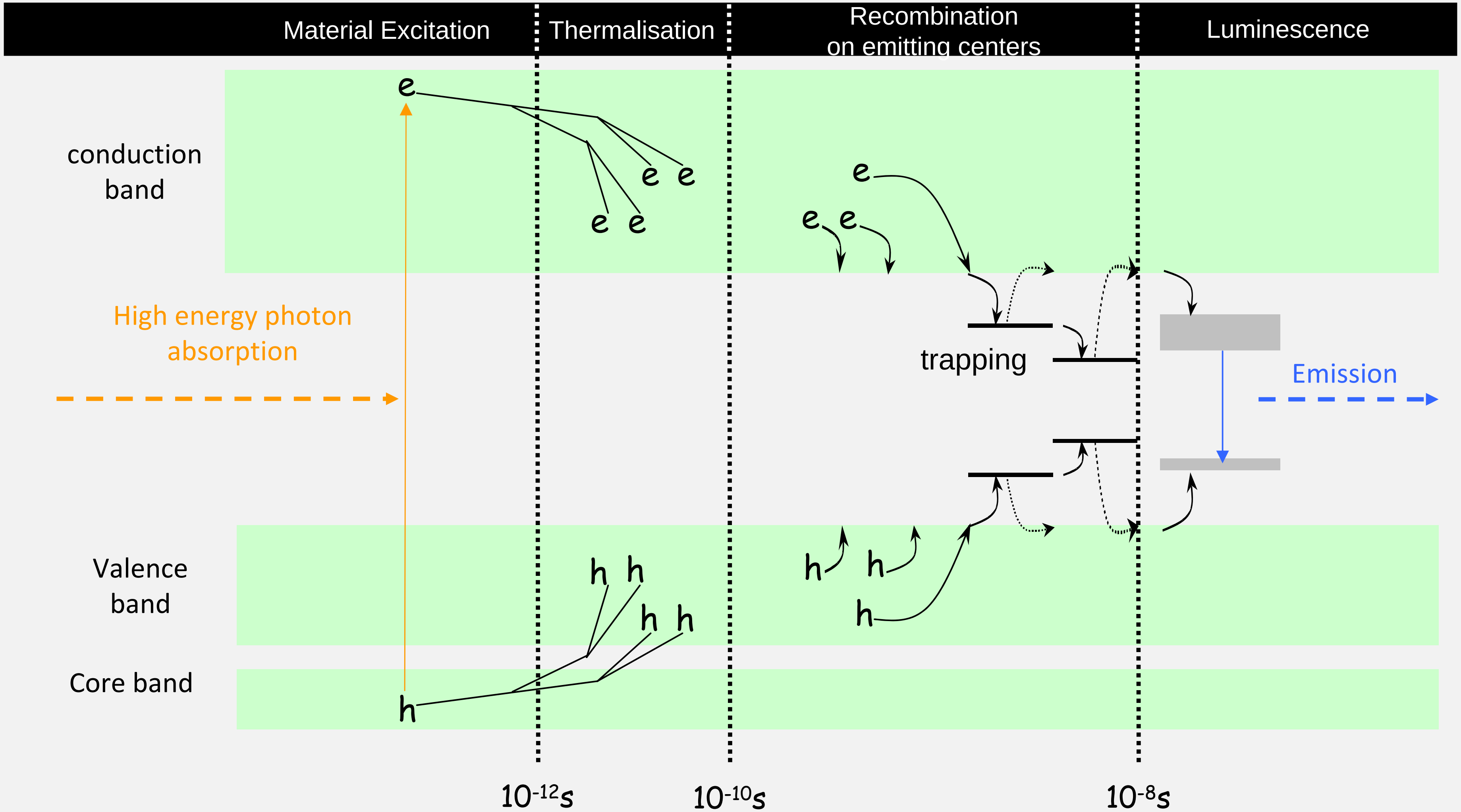
In parallel, high energy physics (L3 with BGO) and then CMS (starting program mid 80's)
Conference series on Inorganic scintillators and their applications:

SCINT
1991-93-…-2009-2011

Aim: Stimulating research on new materials

Crude processes description in 3 stages

$$\text{Scintillation yield} = \beta \cdot S \cdot Q$$



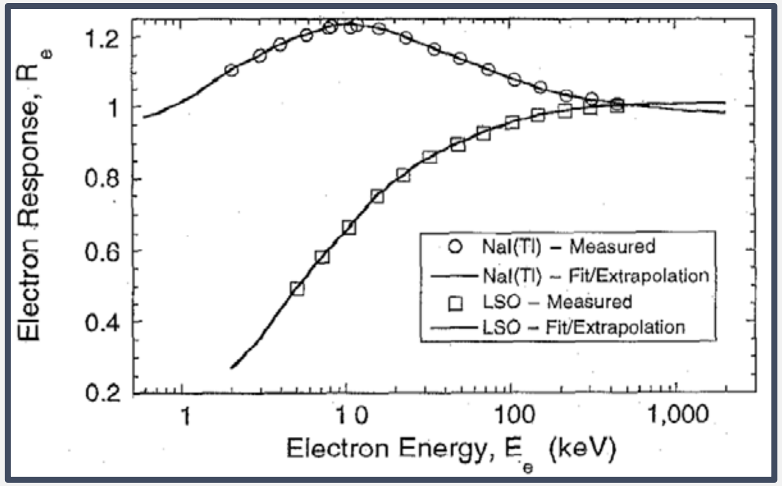
The process is more complex

Yield = β.S.Q avec β= $E_{\gamma}/(2\text{-}3 * E_G)$

The amount of emitted light should be proportional the absorbed energy particle

But some strong non linearities are observed

Under electron excitation



Valentine, IEEE 1998

Under γ-ray excitation

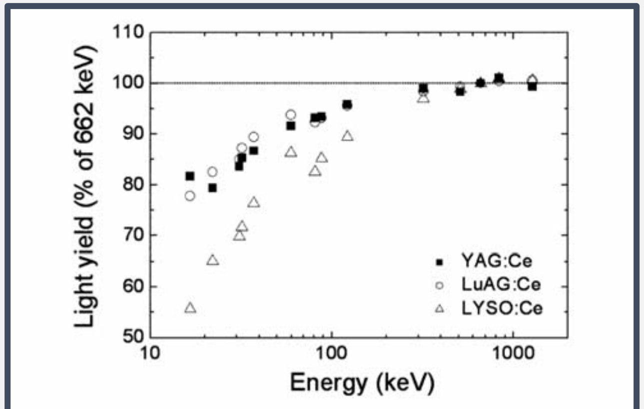


Fig. 2. Non-proportionality of the light yield as a function of γ-ray energy, measured with LuAG:Ce, YAG:Ce and LYSO:Ce crystals. Error bars are within the size of the points.

Chewpraditkul, IEEE 2009

Why so many materials ?

1 page of SCINT 1997 proceeding

Why so many materials ?

It does not exist universal scintillators:

Depending on the application, required performances as well as required shapes are strongly different.

Detection mode

Counting mode
(single event detection)

--> Fast decay

Integrating mode
(exposure time / frame)

--> afterglow?

Energy to detect?

High Energy (CERN, 10^{18} eV)

--> Density
--> Yield

Low Energy (PET, 511 10^{3} eV)

--> Yield
--> Density

Imaging PET/SPECT: localization of a known radionucleïde

Homeland security: Detection of a unknown radionucleïde

The main properties

Density

- Stopping power, Z_{eff}
- Photoelectric effect $\alpha\ Z_{eff}^4$

Scintillation yield

- Easier to detect
- Energy resolution
- Timing

Scintillation decay

- Counting rate
- Coincidences gate
- Time of flight

and: Mechanical and chemical stability, emission wavelength, cost, mass production, radio-isotopes purity, thermal stability, shaping possibilities ……

Few examples for Illustration

PET: Positron Emission Tomography

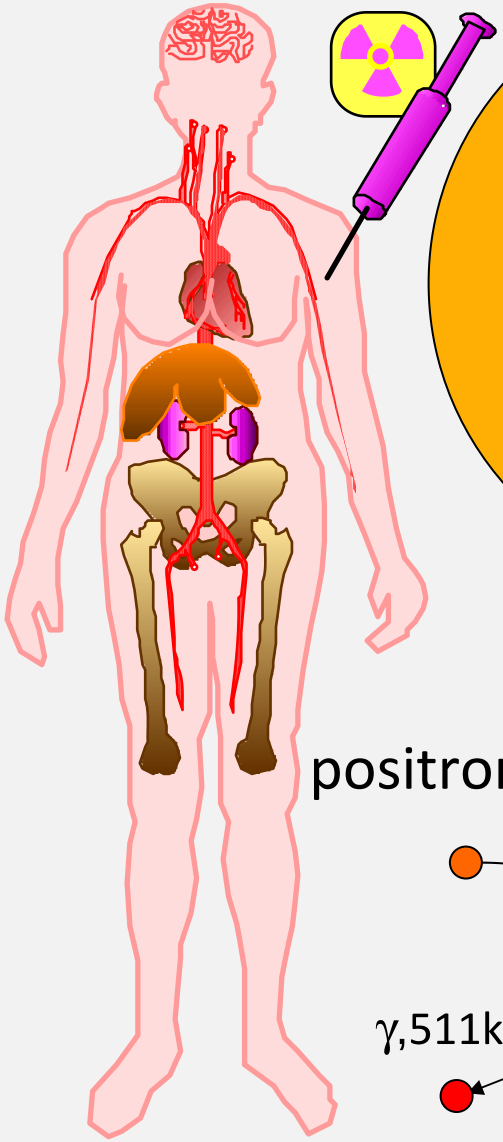
Homeland security

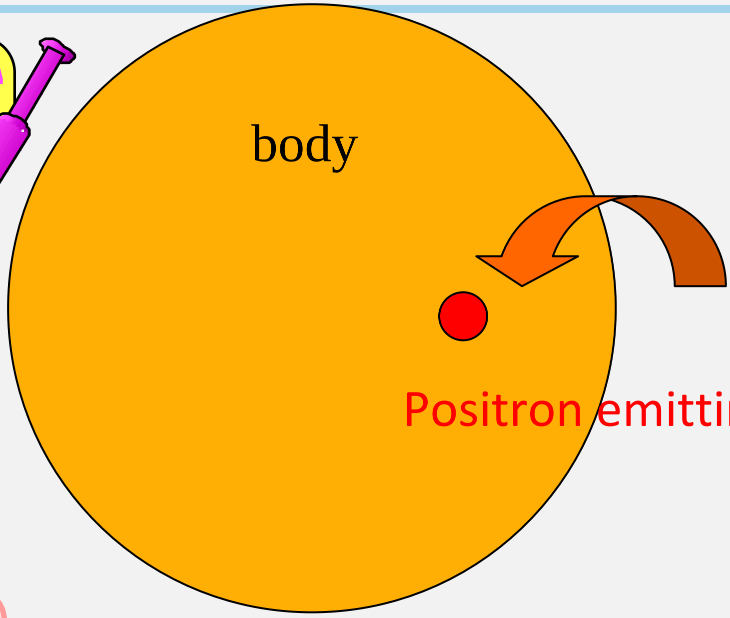
X-ray imaging for medical and high resolution imaging

Dark Matter search

Neutrino spectroscopy

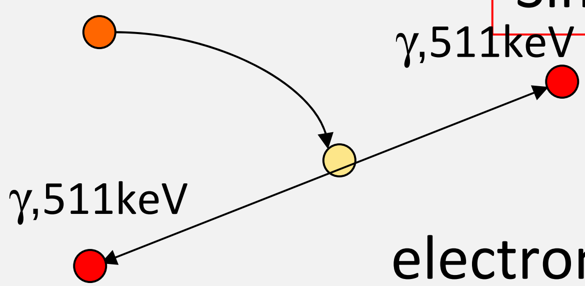
New generation of calorimeters





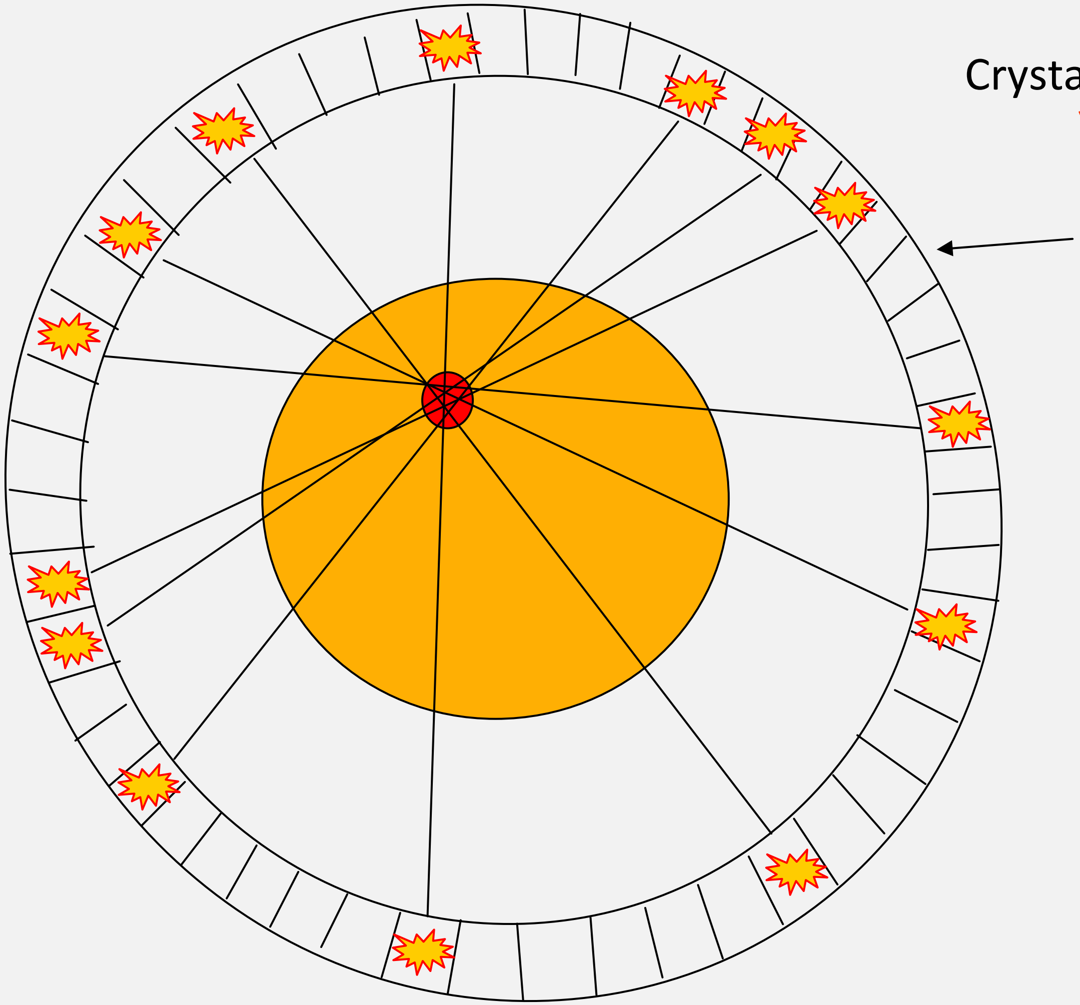
$2\ \gamma$ (511keV)

Simultaneous and opposite direction



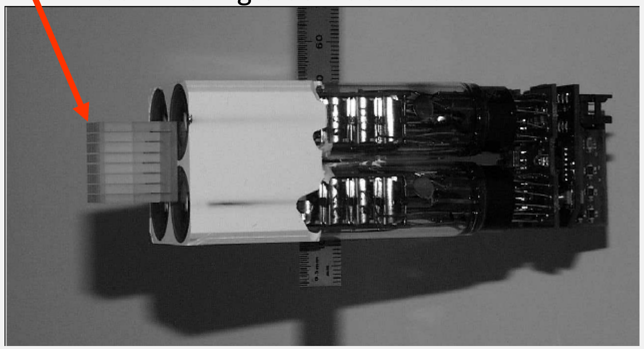
PET

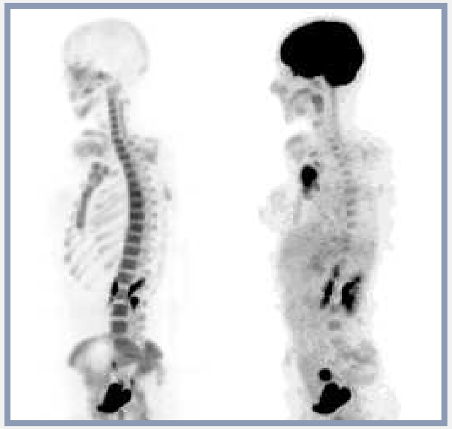
2 γ (511 keV) simultaneous, and opposite directions to detect



Crystals

scintillating bloc + detectors





PET

Main parameters:
Fast decay, scintillation yield, density

Coïncidence detection

Energy resolution

Stoping power

Before and now: $Bi_4Ge_3O_{12}$ (BGO)= 300ns, 8000ph/MeV, 7.13 g/cm^3

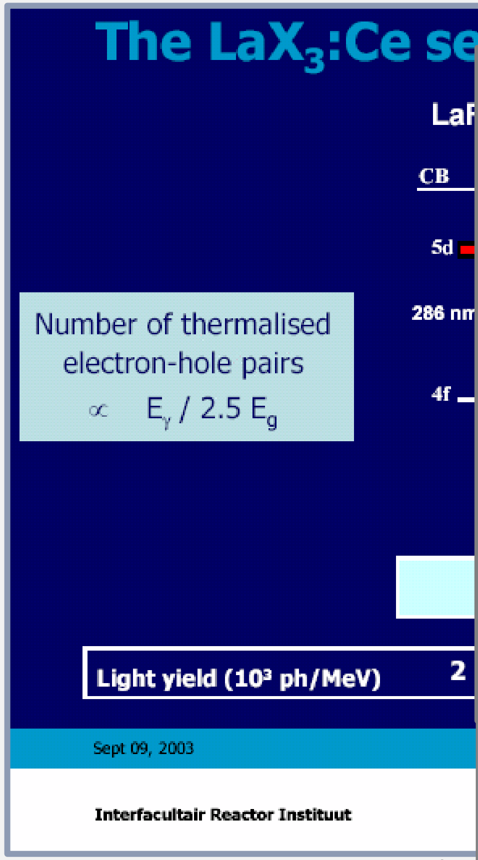
“New”: $Lu_2SiO_5:Ce^{3+}$ = 40ns, 25000ph/MeV, 7.4 g/cm^3

Easy to grow at the industrial scale

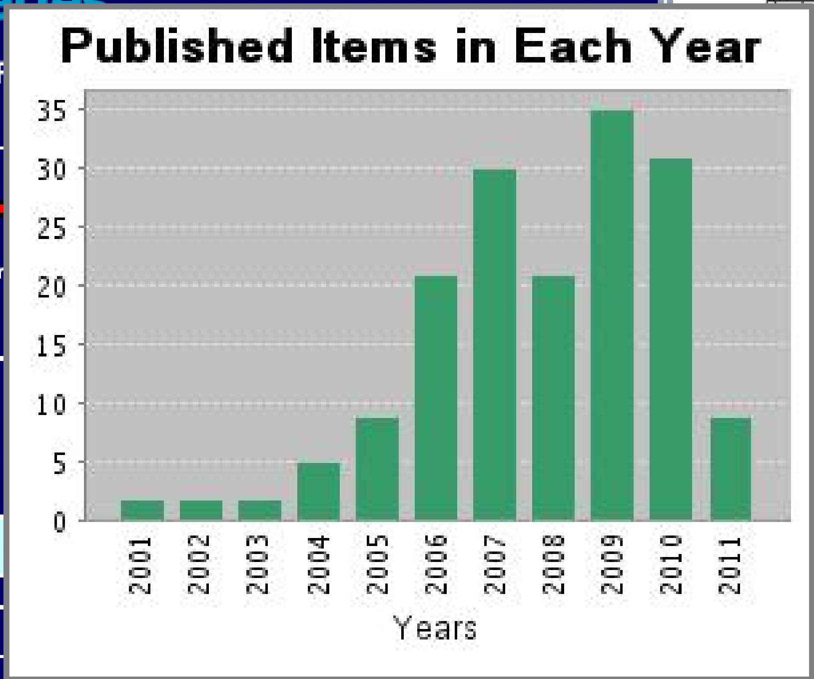
from C.Melcher, CTI

Possible evolutions

The yield depends on the forbidden band width

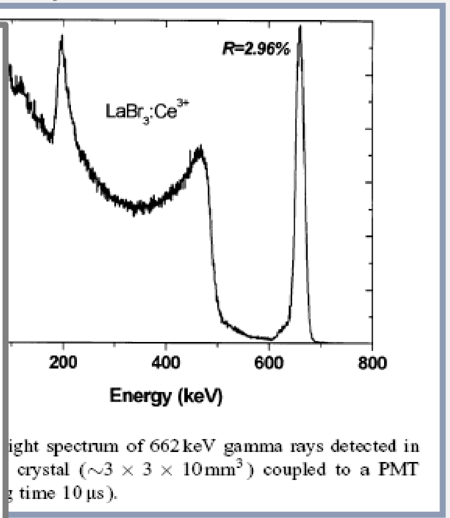


A.Bessière et al., Scint 2003

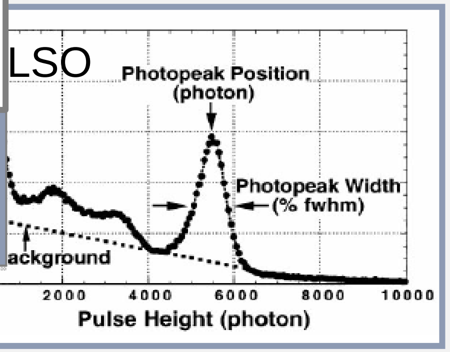


Expensive, fragile, low density
Probably for gamma spectroscopy

C.W.E. van Eijk et. al Rad Meas. 2001

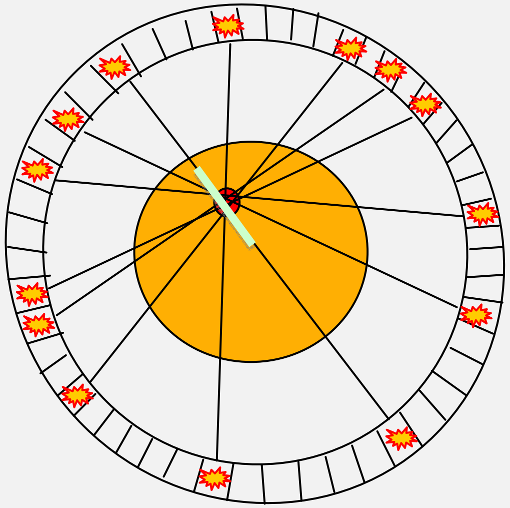


S.Huber et. al NIM 1999

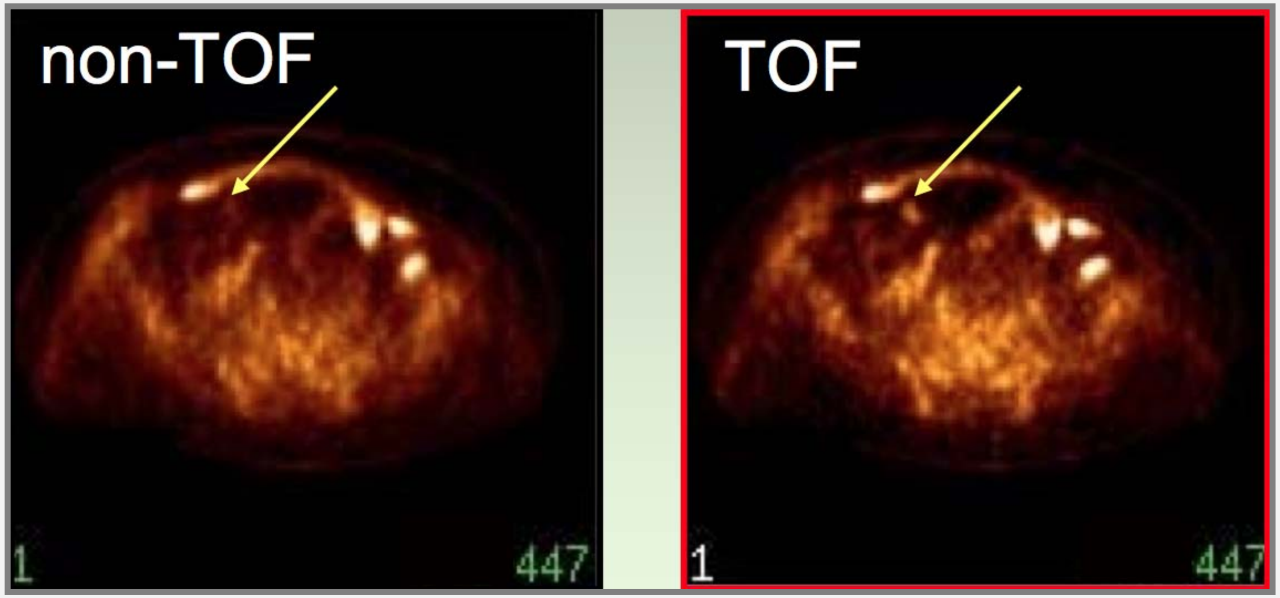


-> Saint-Gobain, BrilLance

PET activity goes toward time of fligth



Time of fligth: noise reduction



Kyba et al Scint 2007

Require time **resolution <200ps**
Depends on the whole chain
crystal-photodetector-electronic
and light collection->

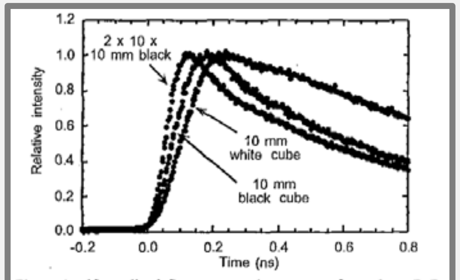


Figure 2: Normalized fluorescence time spectra from three BaF_2 crystals of different sizes and surface reflectors.

Derenzo et al, IEEE TNS 2000

Topics and applications

September 11th 2001: The US funds are redirected toward « homeland security »

Desired properties: be able to check airports, harbors, highways in terms of radio-elements detection

Criteria: energy resolution, density and decays are a bit less important

--> Huge activity on Eu^{2+} doped systems: large screening investments

combinatory chemistry

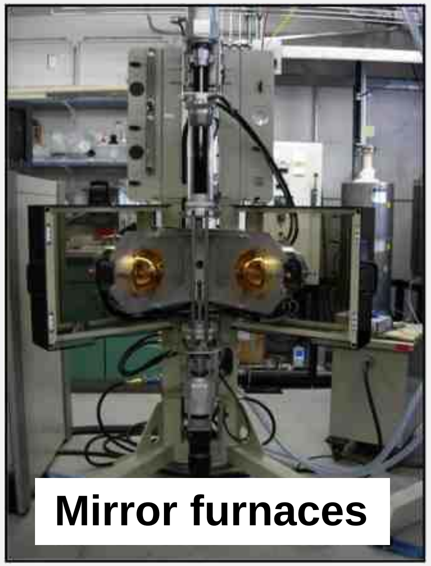
E.Bourret et al. LBNL, USA, ECS 2010

Automatic measurements

- X-ray diffraction
- X-ray induced luminescence
- Optical excitation and emission spectra
- Band gap measurement

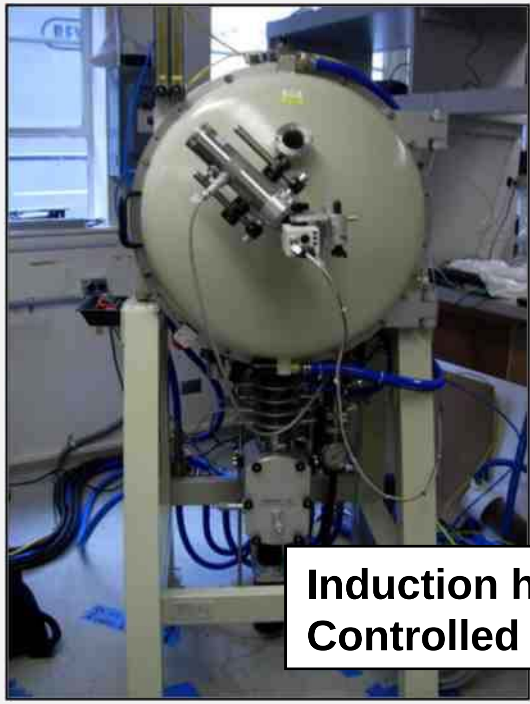
Topics and applications

Crystal growth facilities



Mirror furnaces

Bridgman furnaces



MPD/CZ

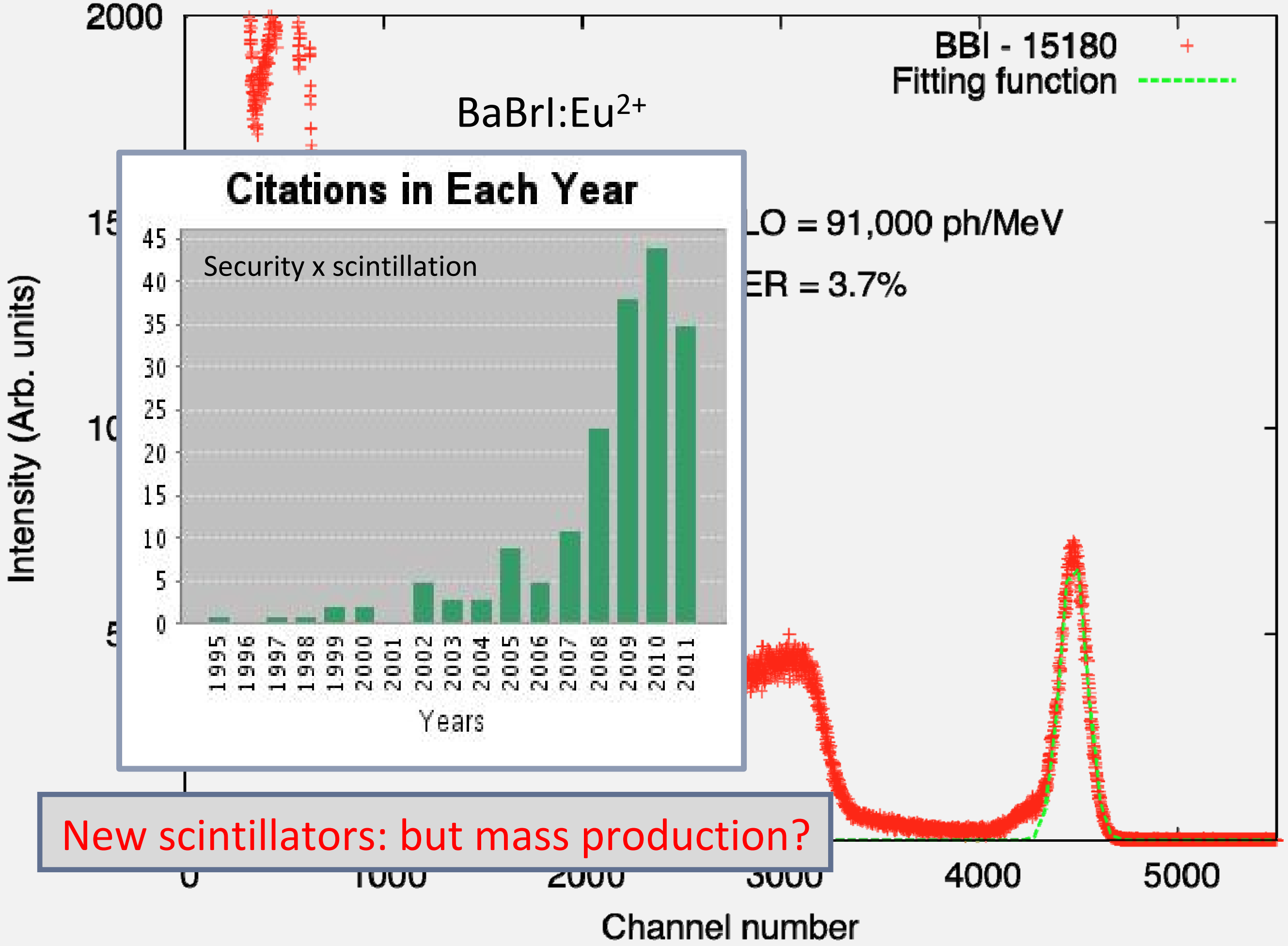
Induction heating
Controlled atmosphere

E.Bourret et al. LBNL, USA, ECS 2010

Topics and applications

Example of discovered scintillators

Crystal (1.5”diameter)

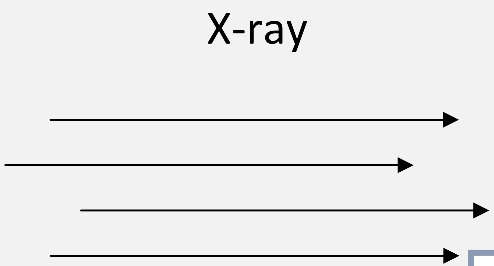


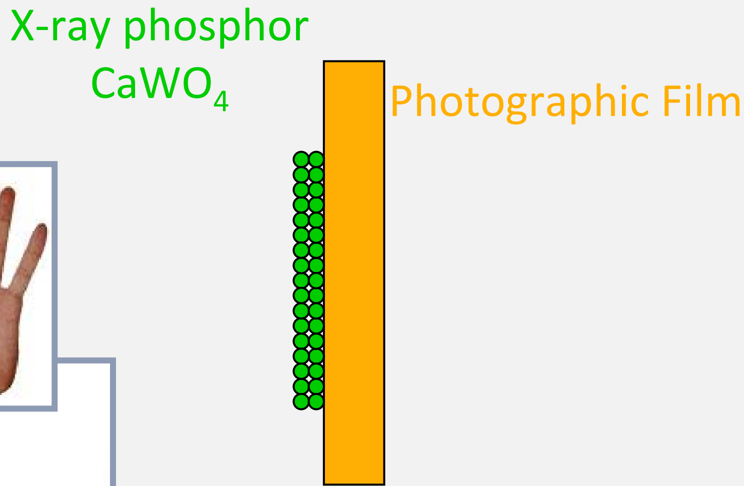
New scintillators: but mass production?

E.Bourret et al. LBNL, USA, ECS 2010

X-ray imaging

Radiography (integration technic)





--> Density, Yield

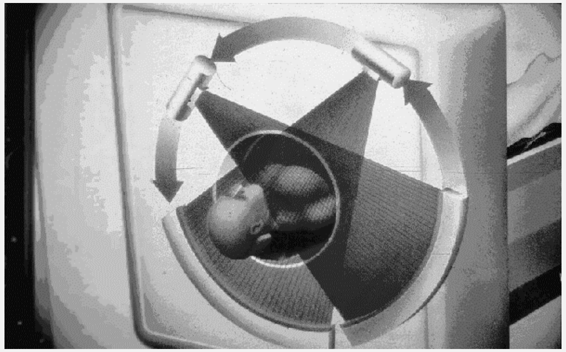
--> $CdWO_4$, $Gd_2O_2S:Tb^{3+}$

X-ray imaging

Developments

Emittor et detectors turn around the patient

scanner

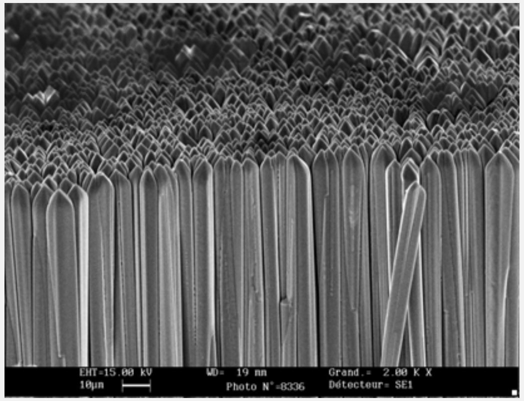


Density, yield, afterglow

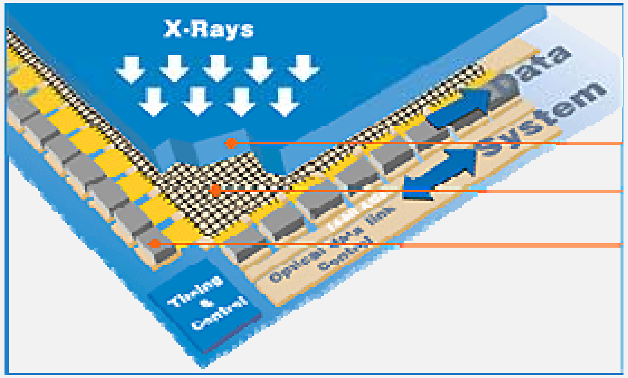
-> codopants

-> GE has still an activity on x-ray phosphors

Micro-needles of CsI:Tl
(Trixell)

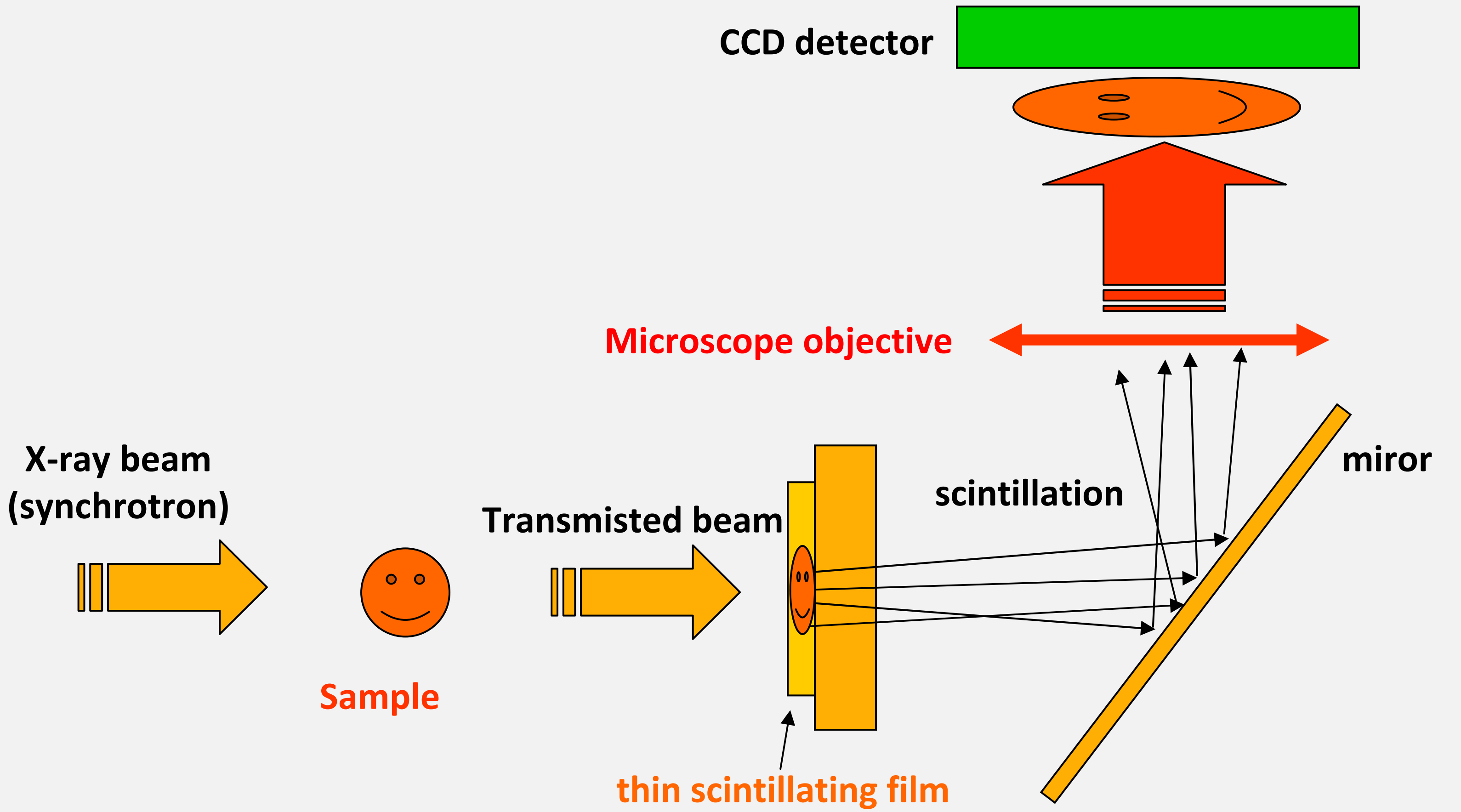


Low density are acceptable since needles act as waveguides



Digital Image , dynamics….

Toward high resolution x-ray imaging with synchrotron radiation (1pixel<1μm^2)



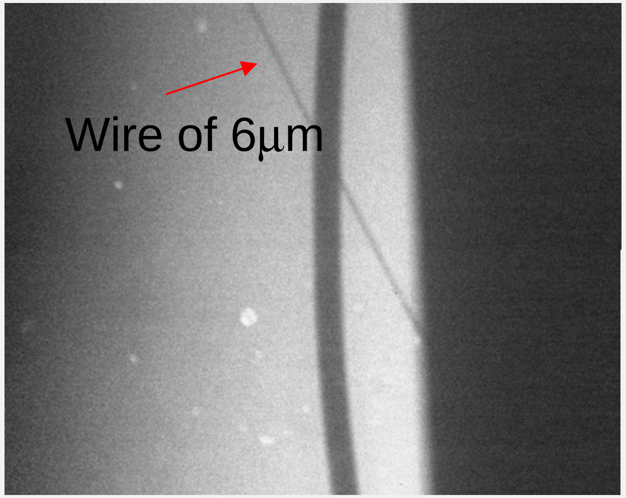
X-ray imaging

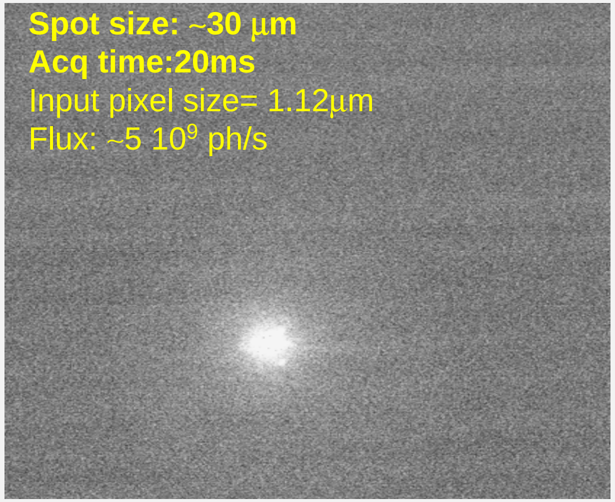
--> Thin Films: high density, yield and optical quality

Liquid Phase Epitaxy: LuAG:Eu^{3+}, GGG:Eu^{3+} (CEA LETI) (Thickness from 1μm to 25 μm)

Sol-Gel coating: Lu_2O_3 et Gd_2O_3 doped with Eu^{3+} (LPCML) (Thickness<1μm)

X-ray beam detection for monitoring





Dujardin et al. NIM A, 2005

X-ray imaging

X-ray Tomography: LPE, $LuAG:Eu^{3+}$

Paul Tafforeau
Laboratoire de Géobiologie, Biochronologie
et Paléontologie Humaine
(LGBPH) UMR CNRS 6046

Scientific collaborator at the **ESRF**
on the beamlines ID19 and ID17
European Synchrotron Radiation Facility

Increasing the absorption and preserving the image quality : multi layers with several dopant and spectral filtering

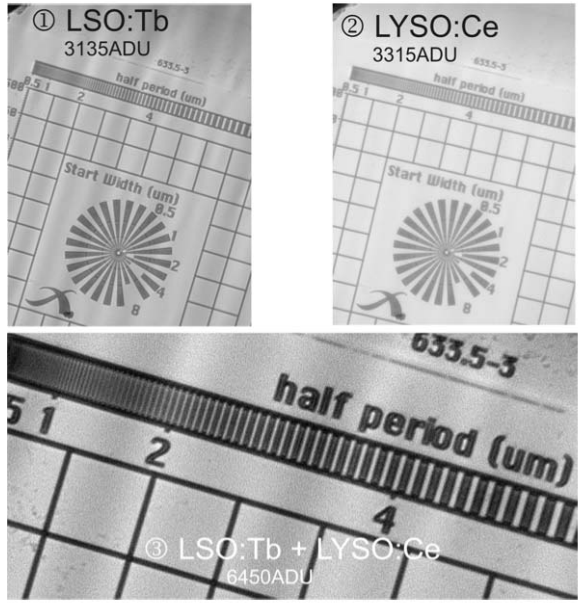


Fig. 8. Demonstration of the spatial resolution achieveable with a scintillator stack consisting of two layers ($15 \times$ magnification/NA $= 0.28$ objective). The image ③ is a summation of image ① and ② detected by selecting the different emissions of the scintillator multi-layer via optical filtering.

T. Martin et al , IEEE TNS, 2009

WIMPs detection
(Weakly Interacting Massive Particles)

to summerize

Strange displacement of galaxies -> dark matter (90%) -> WIMPs

WIMPs: resting mass of 10Gev, v=300km/s ->Ec=50keV
-> few keV deposition in matter
-> Interaction with nucleus (recoil energy)

↓

Temperature increase + scintillation

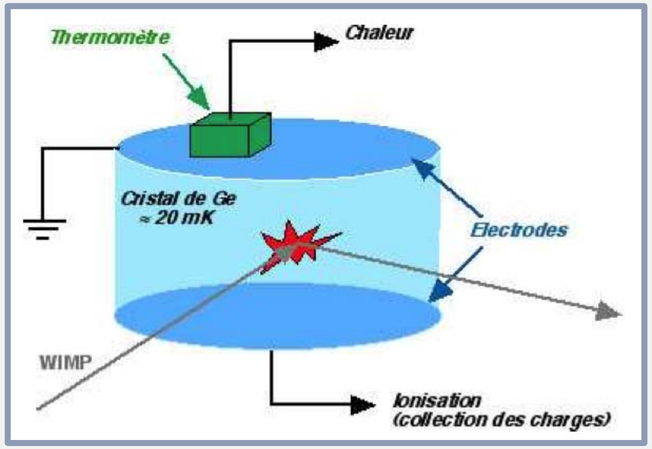
Expected counting rate<< 1count/day and per kg of detector and a energy deposition between 45 et 55 keV!!!!

Natural radioactive background of body: 100 Bq
-> competition with natural background (neutron, β, γ)

Scintillators for dark matter search

Discrimination between electromagnetic particles and (neutron, WIMP)

The ratio between photon signal/ phonon signal depends of the incident particle
Photons and β give ionization while WIMPs and neutrons interact with nucleus (recoil)



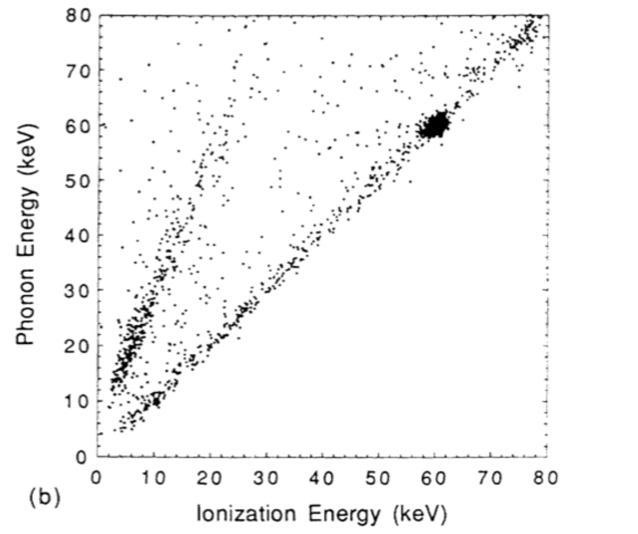


FIG. 1. Phonon and ionization equivalent energies measured for (a) 59.5 eV photons and Compton scatters of background photons. (b) The same measurement as (a) with the addition of neutrons and photons from a ^{252}Cf source.

Shutt et al, PRL 1992

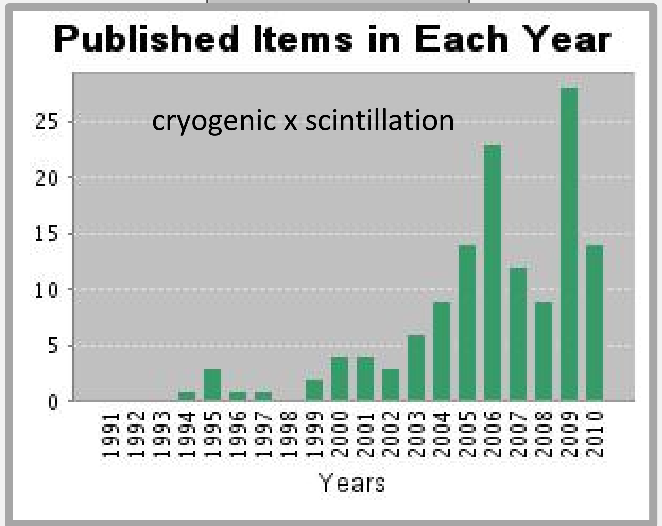
neutron / WIMP discrimination -> interaction with nucleus of various mass.
Several scintillating compositions (targets)

Scintillators for dark matter search

neutron / WIMP discrimination -> interaction with nucleus of various mass.
Several scintillating compositions (targets)

Double mesurement
(quelques mK)

Signal « lumière »



Signal « chaleur »

Combination of several detectors ->Discrimination γ, β et WIMPs, neutrons.

For the crystals:
-radio-isotope purity
-Scintillation at low T (few mK)?
- homogeneity (1 ton?)
- Al_2O_3, LiF, $CaWO_4$, BGO are under study

Angloher et al, Astroparticle Physics, 2005 et Luca M et al., NIM 2009

Several projects: Cresst, Edelweiss, Eureca…
and SUCCESS
and Interest for the double β decay

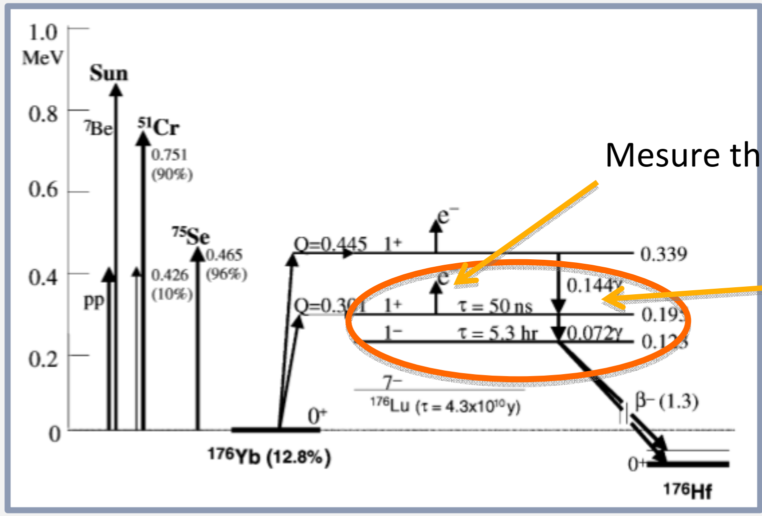
Solar Neutino Spectroscopy

The number of detected solar neutrino is << than the predicted one

Weak interaction cross section of neutrino with matter + natural background = difficult to extract true signal

-> interaction with a nucleus giving rise to clear assignement + spectroscopy

-> one of the solutions: interaction neutrino - Ytterbium



Mesure the neutrino energy

Delayed Coïncidence = neutrino signature

R.Chipaux et al NIM 2002

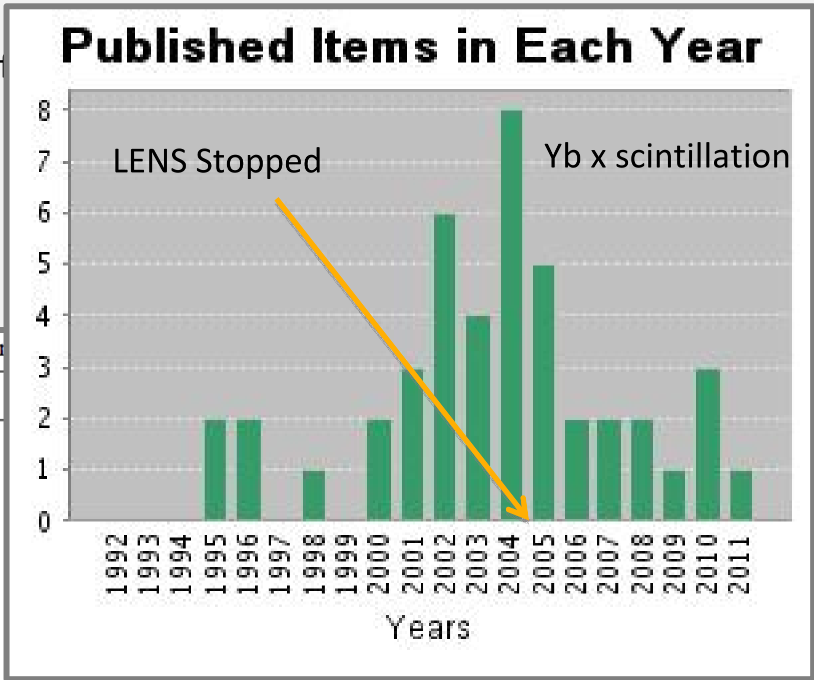
-> LENS project : Low Energy Neutrino Spectroscopy

Scintillator side: looking for high Yb loaded compounds with a fast response (delayed coïncidence et 50ns) very low radioactive background and high capapility for mass production

Yb^{3+} : known ion for it IR emission (but slow), Yb^{2+} does emit ligth but is rather complicated (P.Dorenbos, J of Phys 2003)

Yb^{3+} : gives a charge transf … t L.van Pieterson et al J.Lum 2000)

… rials



Light yield of Yb-co…

Sample	
YAG:Yb (2%)	
YAG:Yb (10%)	
YAG:Yb (15%)	
YAG:Yb (50%)	
YbAG	
YAP:Yb (8%)	
$LaYbO_3$	
$Y_3Ga_5O_{12}$:Yb (2%)	
$Y_3Ga_5O_{12}$:Yb (15%)	[illegible]
$Yb_3Ga_5O_{12}$	0.055

N.Guerassimova et al NIM 2002

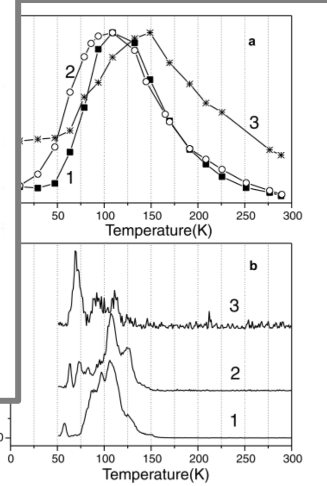


Fig. 4. Temperature dependence of the 333 nm integrated emission band intensity under X-ray excitation (a), and thermoluminescence under X-ray excitation (b) of (1) YAG:Yb(10%), (2) YAG:Yb(15%) and (3) YbAG.

N.Guerassimova et al Chem Phys Lett 2001

New concept for high energy calorimetry

New accelerators ILC: new requirements for calorimeters

New concept: fiber shaped meta-materials

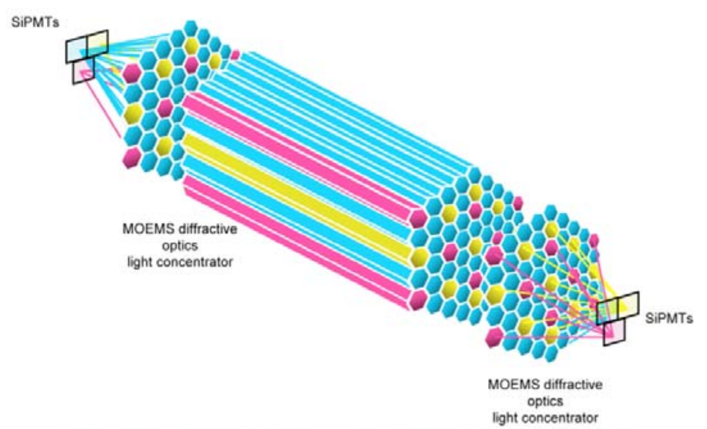


Figure 4: Principle of the metacable with 3 types of fibers and diffractive optics light concentrators

P.Lecoq, CALOR 2008 and Dujardin et. al. JAP 2010

SEE K.PAUWELS talk this morning

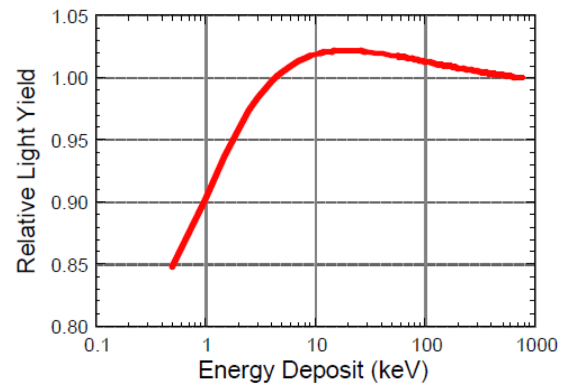
ANR INFINHI and FP7/HP2 program

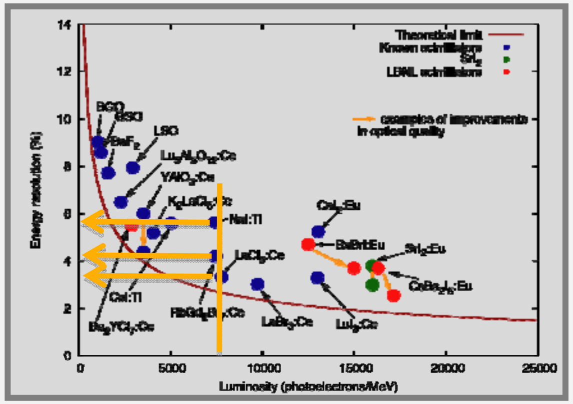
Example of fundamental studies

Yield = β.S.Q avec β= $E_{\gamma}/(2\text{-}3 * E_G)$

The amount of emitted light should be proportional the absorbed energy particle

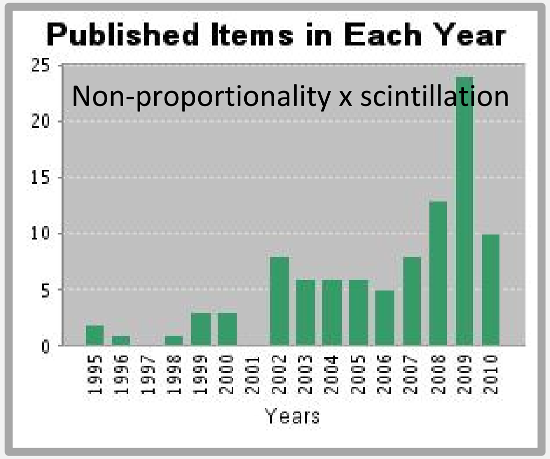
BGO	8,200 photons/MeV
NaI:TI	38,000 photons/MeV
CsI:TI	60,000 photons/MeV
LSO	28,000 photons/MeV
$LaCl_3$:Ce	50,000 photons/MeV
$LaBr_3$:Ce	63,000 photons/MeV





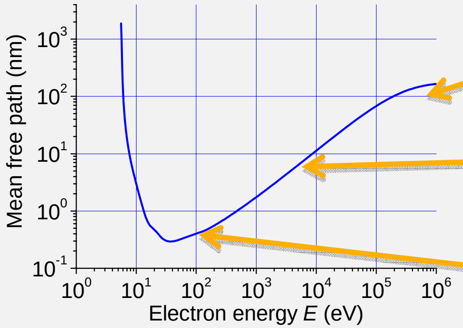
Non-linearity, non proportionnality

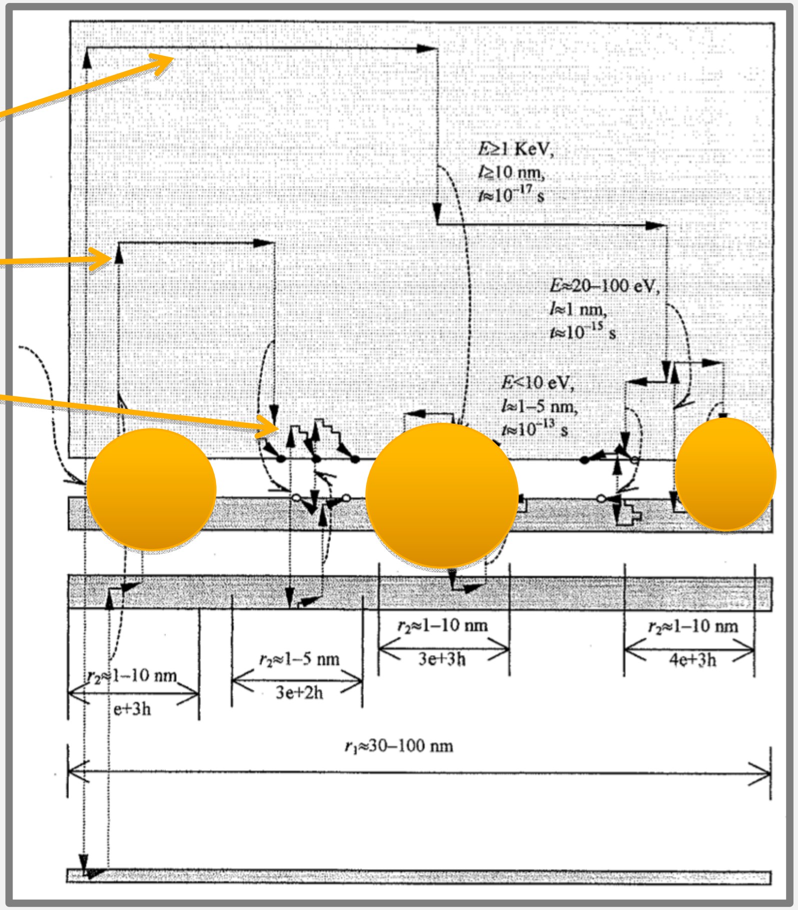
It explain why with the same ligth yield some scintillators exhibit good and bad energy resolutions



Origin: fluctuations of excitation density

Typical mean free path for electrons (NaI)





A.N. Belsky, J. of Elec. Spect. And Rel. Phen. 1996

During thermalization, the energy is deposited as separated small clusters of excitation

-> excitation Clusters

- Quenching effects
- Energy transfer toward activator process modifications
- …

SEE A.N.Vasil'ev lecture this morning

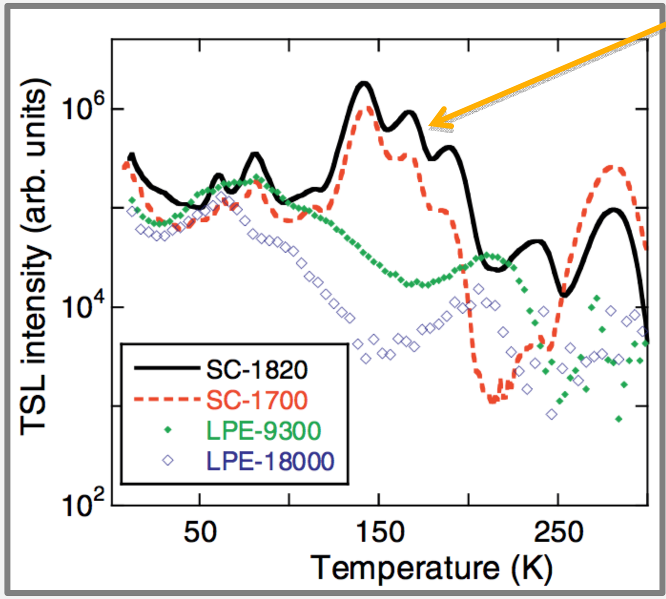
Correlation between crystal structure/ defects and performances

Example with LuAG:Ce

Comparison
single crystal (2000°C)/ LPE films (1000°C)

Thermoluminescence

Lu_{Al} antisite



M.Nikl et al, Phys. Stat. Sol. (b) 2005

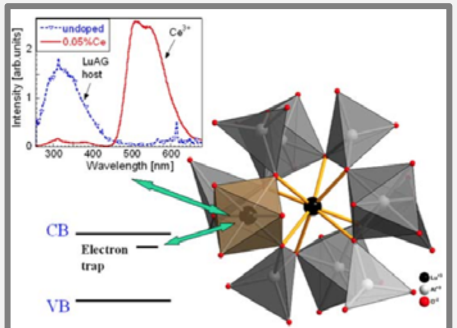


Figure 11 (online colour at: www.pss-b.com) Lu_{Al} antisite defect in LuAG structure. Resulting electron trap in the material forbidden gap is sketched on the left. Emission band within 300–350 nm due to antisite defect and its competition with that of the Ce^{3+} center can be derived from radioluminescence spectra at RT – upper left. Emission lines around 312 nm and 615 nm in the undoped sample are due to Gd^{3+} and Eu^{3+} accidental impurities, respectively.

M.Nikl et al, Phys. Stat. Sol. (b) 2008

Effects on performances

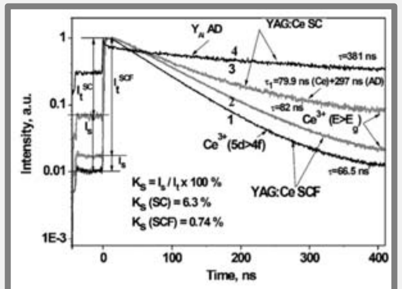
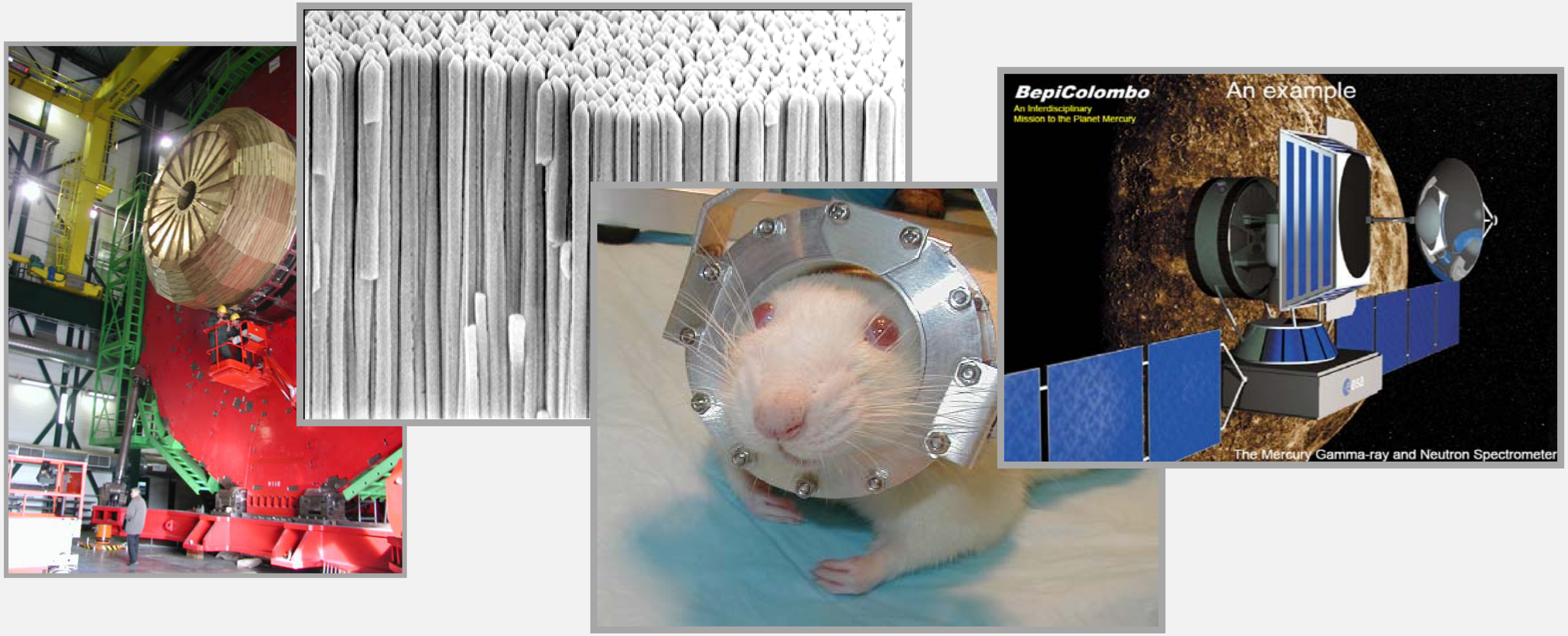


Fig. 5. Decay kinetics Ce^{3+} ion (1–3) emission in the band at 530 nm in YAG:Ce SCF (1, 2) and SC (3) at RT under excitation by SR with energies of 3.7 eV (1) and 10.5 eV (2,3). Curve 4—decay kinetics of Y_{Al} AD emission in the band at 320 nm in YAG SC.

Y.Zorenko et al, Rad. Meas. (2007)



Development and studies of materials for Scintillation applications:
Nice active area including several fields from materials science to imaging systems, medicine….

-> it has to be still developed -> SUCCESS Program

Effets de la densité d'excitation

Spectres d'excitation autour d'un seuil d'absorption X
-> modification forte du coefficient d'absorption
-> modification de la densité d'excitation

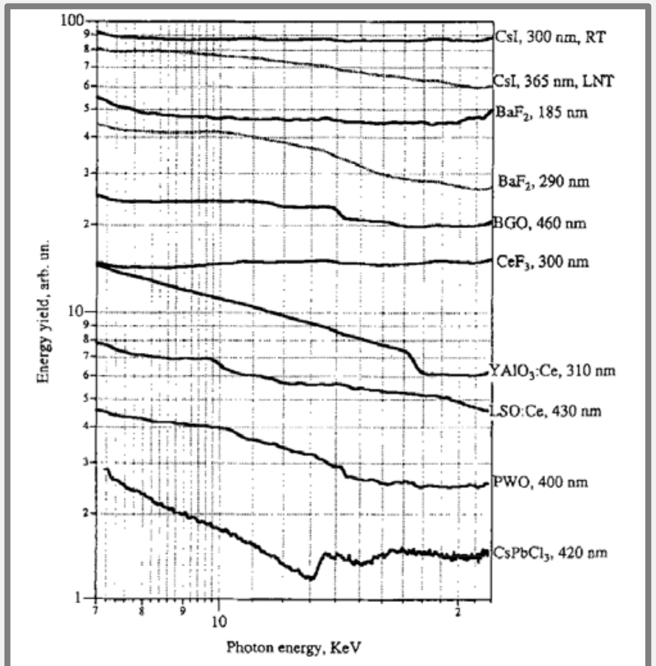
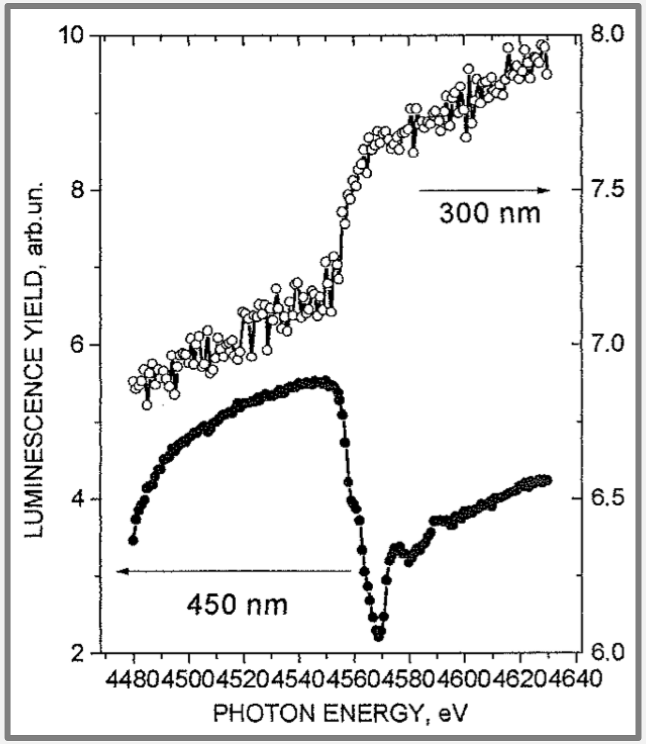


Fig. 2. Luminescence energy yield spectra of various crystals under X-ray excitation.

A.N.Belsky et al, NIM A, 1995



A.N.Belsky et al, J. Of Lum. 1997

Effets importants de la concentration d'excitations sur les processus de quenching et de transfert vers les centres luminescents

Research on Materials and applications

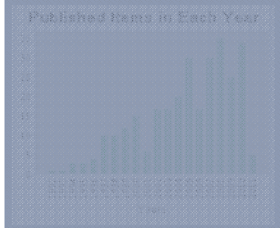
90's: A new material is needed for CMS in High energy physics (/BGO was used in L3).

$PbWO_4$ and CeF_3 appear rapidely as candidates

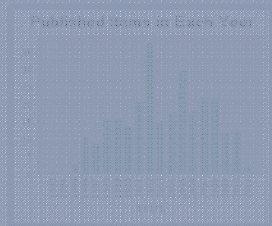
Requirements: high density, fast decay, yield, mass production capability

$PbWO_4$ won

At the same time: discovery of $LSO:Ce^{3+}$, for oil drilling, but is also of interest for PET imaging (médical = $ -> industrial interest)

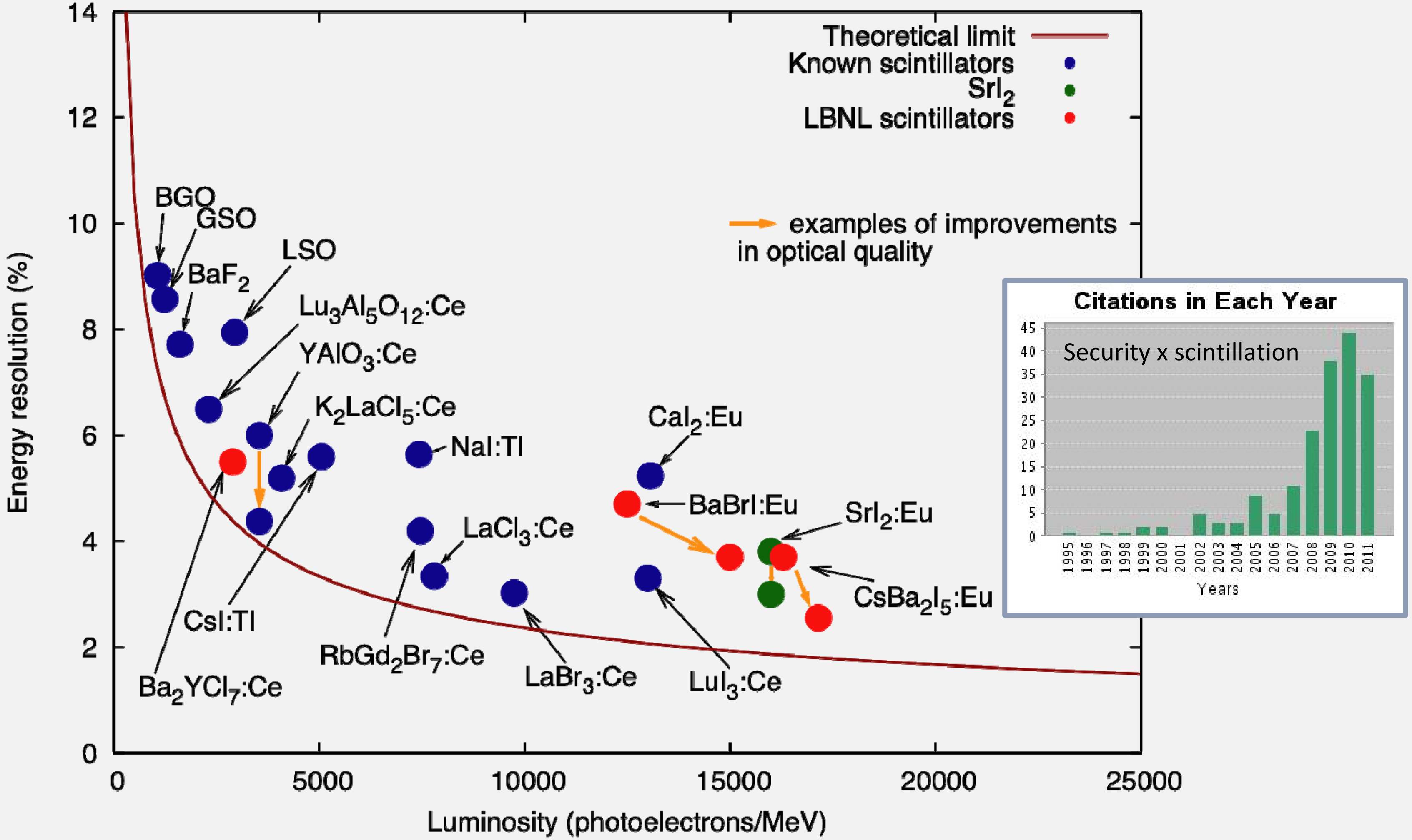


Up to 1997, activity on $PbWO_4$ and then it decreases



Topics and applications

New scintillators: but mass production?



E.Bourret et al. LBNL, USA, ECS 2010