Introduction to scintillators

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17 November, 2003

1. Luminescence, fluorescence, scintillation, phosphorescence, etc.
2. Scintillation mechanism
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4. Main characteristics
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    Decay,
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5. Comparison between inorganic and organic scintillators

1. Luminescence, scintillation, phosphorescence, etc. Scintillators

Luminescence is the basic notion-----one of the four phenomena giving light emission

(1) thermal radiation----black-body radiation, etc.
(2) bremsstrahlung
(3) Cherenkov radiation
(4) luminescence----electronic states of atoms are excited by external energy, and the excitation energy is converted to EM radiation above thermal radiation, usually in the visible, UV, IR range.
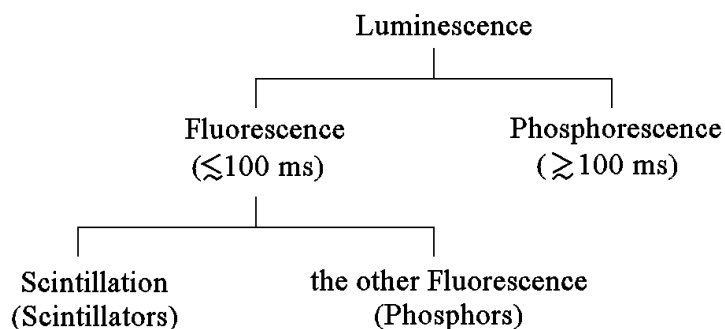
Luminescence excited by different external energies:
  photo-luminescene   ---by UV photons, etc.
  radio-   "   "   ---by radiation like X-, γ-, α-, β-rays, etc.
  cathode- "   "   ---by electron beam,
  electro- "   "   ---by electric field, (examples: LED)

Fluorescence ~ Luminescence during excitation (not rigorous)
Phosphorescence ~   "   " after excitation

Scintillation ~ Fluorescence due to radiation

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                          Luminescence
                               |
              ┌────────────────┴────────────────┐
         Fluorescence                     Phosphorescence
         (≲100 ms)                          (≳100 ms)
              |
       ┌──────┴──────┐
   Scintillation      the other Fluorescence
   (Scintillators)          (Phosphors)
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Scintillators:
Inorganic: crystals, ceramics, glasses, noble liquid, noble gasses,
Organic: crystals, liquid, plastic

2. Scintillation mechanism

Basic processes:
   excitation
   →thermal relaxation to the bottom of excited state
   →emission

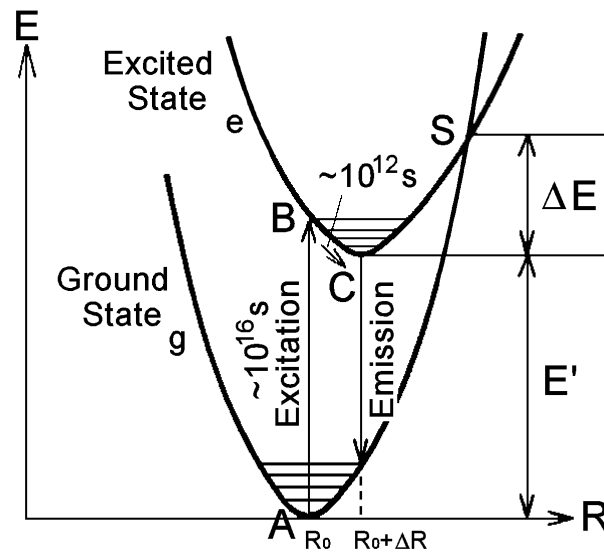
Fig.
luminescence in the configurational coordinate diagram.
R=inter-atomic distance between ground state of ligand atom and the
excited state of luminescence centre atom

Four types of luminescence in inorganic scintillators
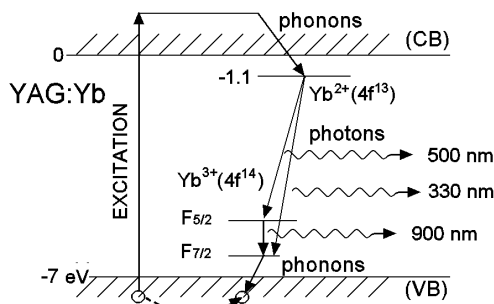
(1) Exciton luminescence: BGO, …

Ionization/excitation by radiation creates unbound e-h pairs or bound e-h pairs called excitons. Excitons can move rather freely in crystals, caught at impurities, defects, and so on, and the STE (self-trapped excitons) gives luminescence upon radiative recombination.

phonons
0 ───── (CB)
5d ──── GSO:Ce^{3+}
Ce^{3+}
photon ──→ ~440 nm
Excitation
4f
-6.2 eV phonons
(VB)
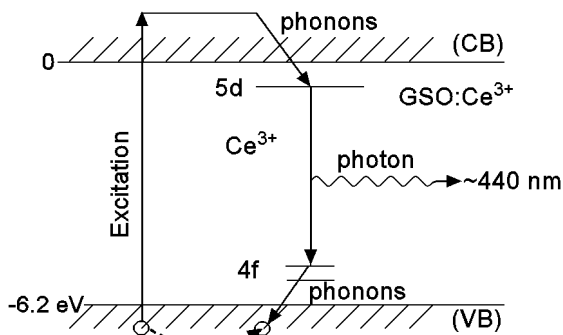
(2) Dopant luminescence: GSO:Ce, …
Radiative recombination of STE at dopant (activator) ions.
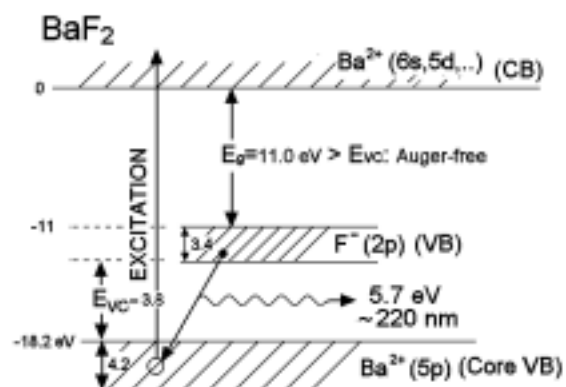
(3) Charge-transfer luminescence

Belongs to exciton luminescence. Due to charge transfer where initial and final states are different, selection rules for EM transition are loosened, thereby enhancing transition probability.

phonons
0 ───── (CB)
-1.1 Yb^{2+}(4f^{13})
YAG:Yb
photons
Yb^{3+}(4f^{14}) ──→ 500 nm
──→ 330 nm
F$_{5/2}$ ──→ 900 nm
F$_{7/2}$
-7 eV phonons
(VB)
EXCITATION

(4) CVL (Core-valence luminescence, Cross luminescence)
After excitation of the core-valence electron, an electron in the valence band recombines with the resultant hole radiatively. To avoid Auger process, $E_{VC} < E_g$ is necessary. BaF$_2$, CsF, LiF,.....

>small temperature dependence
>fast decay
>LY less than 10% of NaI:Tl

BaF$_2$
0 ──── Ba^{2+} (6s,5d,...) (CB)
E_g=11.0 eV > Evc: Auger-free
-11
EXCITATION
3.4 F$^-$(2p) (VB)
5.7 eV
~220 nm
E_{VC}=3.6
-18.2 eV
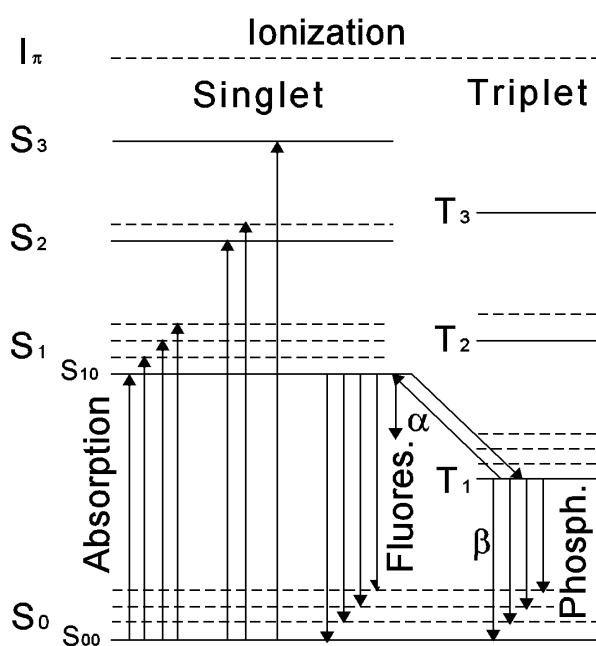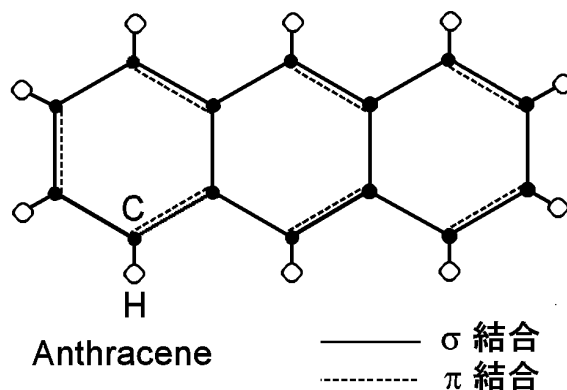4.2 Ba^{2+} (5p) (Core VB)

Scintillation mechanism in organic scintillators

Radiative transition of π electrons:

π electrons = one of the four outer shell electrons (2p) of each
C-atom in benzene-like ring.
loosely bound to each C-atom, and unlocalized to the
benzene-ring.
Luminescence occurs at each molecule level, and therefore is similar
among crystals, liquids, and plastics.

Anthracene

—————— σ 結合
------------ π 結合

I_π Ionization

Singlet   Triplet

S_3

S_2 T_3

S_1 T_2

S_{10}

T_1

Absorption   Fluores. α β Phosph.

S_0 S_{00}

3. Scintillation efficiency

Scintillation efficiency η can be defined as

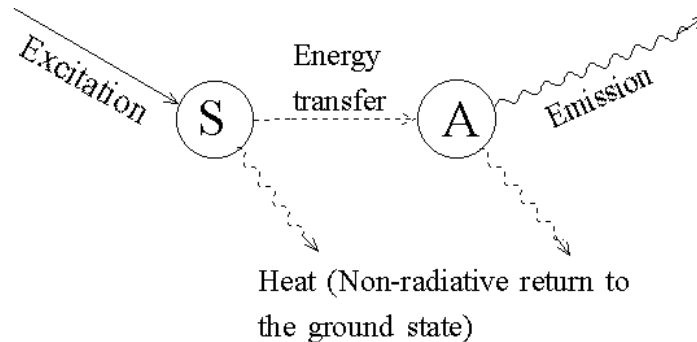$$\eta = \frac{energy\ of\ scintillation\ light}{enrgy\ deposit} = \frac{h\nu_e}{w_{e-h}}Tq_A$$

where

   $h\nu_e$ = average energy of emitted photon
   w_{e-h}=energy required to create one electron-hole pair,
   T=energy transfer efficiency from excited ion to luminescence centre
   q_A= quantum efficiency of luminescence centre

η_{max} can be estimated for T = 1 and q_A =1.
Writing the w_{e-h} to E_g(bandgap) ratio as β = w_{e-h}/ E_g, we have

$$\eta_{max} = \frac{h\nu_e}{w_{e-h}} = \frac{h\nu_e}{\beta E_g}$$

β is known to be 3~7, and typically ~3 for NaI, CsI, ZnS, GaP, etc.

NaI:Tl; $h\nu_e$=3.02 eV, E_g = 5.9 eV, η_{max} ~ 17%, η(exp)=13~8 % at RT.
      η(exp)=13 % gives 0.13 x 1MeV/ $h\nu_e$ ~ 40,000 ph/MeV
NaI(pure): η(exp) at 80K~ 2 x (NaI:Tl at RT)=26~16%.
ZnS:Ag; $h\nu_e$=2.75 eV, E_g = 3.8 eV, η_{max} ~ 25%, η(exp) ~ 21%.
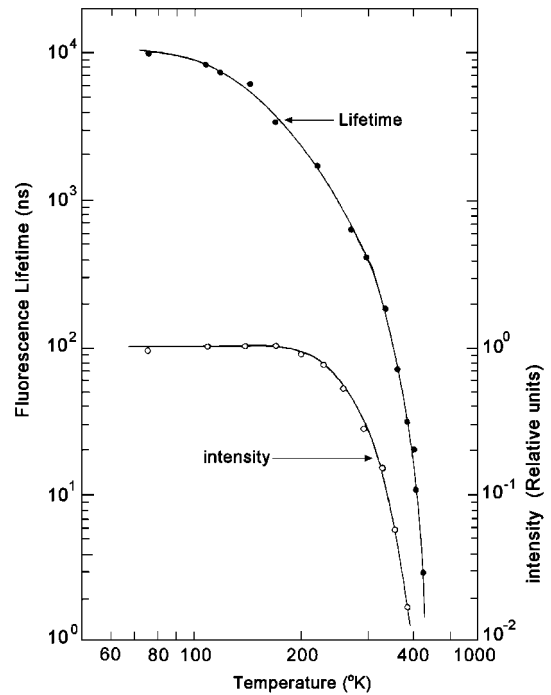Anthracene; $h\nu_e$=2.63 eV, η(exp) ~ 4%. (1.5 x10^4 ph/MeV)

Absolute scintillation efficiency is not large! Most of deposited energy is lost by phonons.

4. Main characteristics

4-1. Quenching

(1) thermal (temperature) quenching

As temperature rises, intensity decreases and decay becomes faster as seen in a typical example of BGO
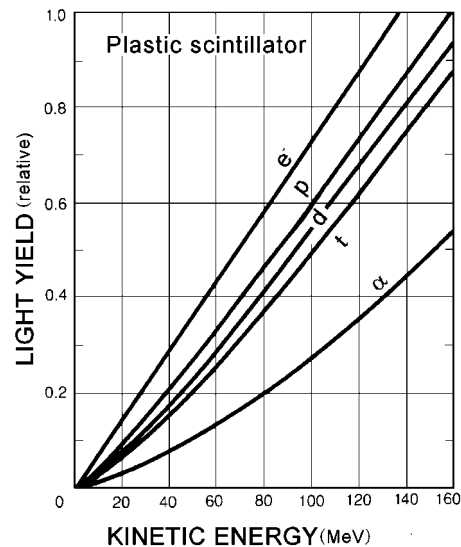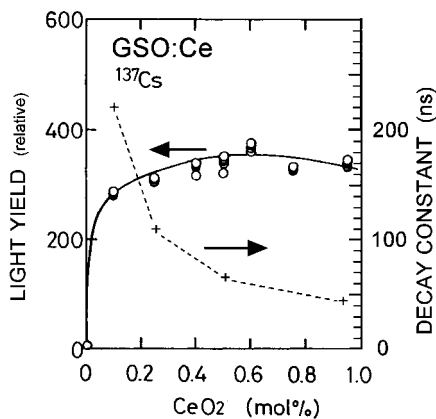
(2) impurity quenching

As the impurity increases, intensity decreases, and decay may become faster or may not be much affected.

(3) concentration quenching

As the activator concentration increases, intensity has a maximum while decay continues to be faster.

(4) ionization quenching: important in organic scintillators

When dE/dx is large, the dense ionized ions quench the excited π-electrons.

4-2. Decay

There are three causes for afterglow:
(1) Natural afterglow--$\rightarrow$fluorescence decay time
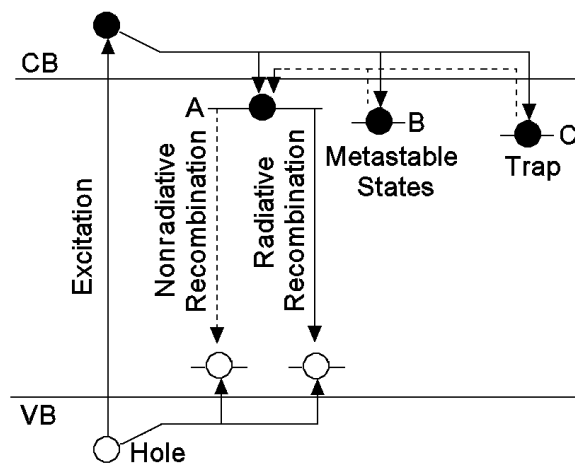
$\qquad I(t) = I(0) \exp(-t/\tau)$

$\qquad \tau = (P_{nr} + P_r)^{-1}$

$\qquad P_r$ = radiative transition probability to lower states

$\qquad P_{nr}$ = nonradiative transition probability

(2) Afterglow due to metasatble states or traps (impurities, defects, ..)

$\qquad$ --$\rightarrow$phosphorescence

(3) Speed of energy transfer, and
$\qquad$ Number of luminescence centres
affect the simple exponential decay.

4-3. Particle discrimination   (Pulse shape discrimination)

Possible for some inorganic and most of organic scintillators.

Inorganic scintillators: a typical example is BaF_2 in which both STE (slow) and CVL(fast) luminescences occur.   Since large energy transfer in collision is necessary to excite CVL, electrons have CVL, while heavy particles (α-rays) not.
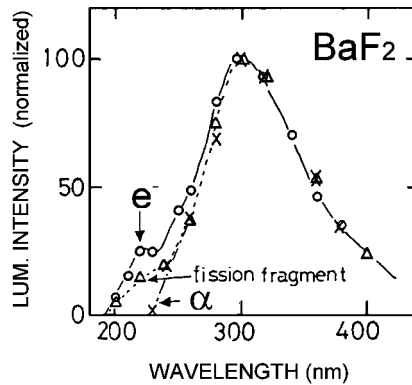
In organic scintillators, both fast and slow components exist.   Only the fast component suffers from ionization quenching, being less for heavy particles with large dE/dx.   Decay is slower for heavier particles.
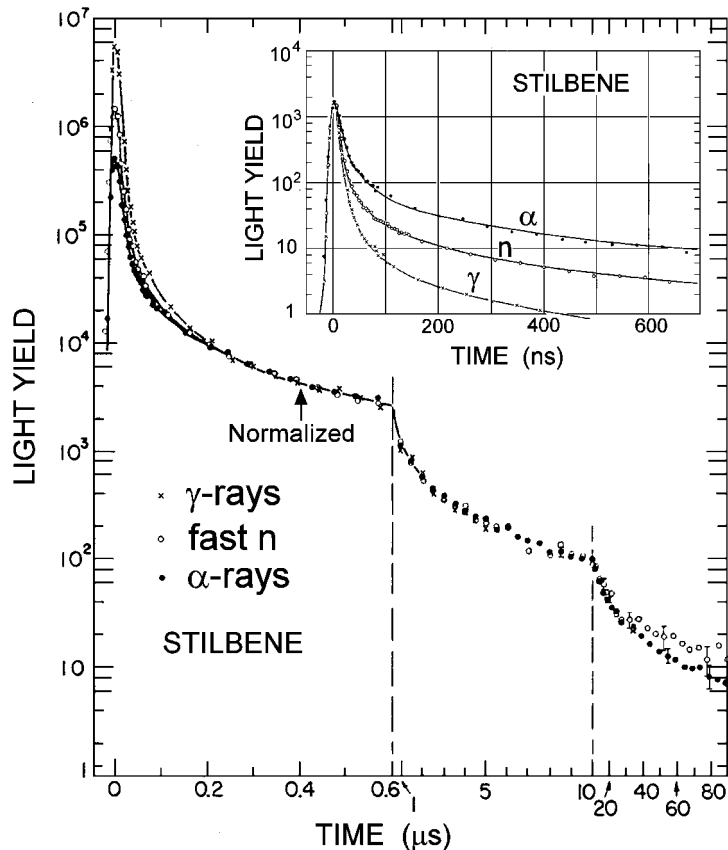
4-4.   Radiation hardness

There are significant differences between inorganic (crystal) scintillators and organic (crystals, liquid, plastic) scintillators.

	Inorganic crystals	Organic scintillators
damages	mainly in colouring (transmittance)	mainly in scintillation efficiency. Colouring to some extent in plastics
γ-rays versus hadrons	larger damage for hadrons 10-100 times	similar in magnitude
recovery in time	yes (spontaneous, thermal, UV annealing)	almost absent
Radiation hardness	depends largely on crystals	similar level at 10^{4-5} Gy

The differences can be explained by
        existence (or lack) of large-scale regularity in inorganic crystals (or organic scintillators), which could restore the equilibrium positions of ions after disturbance due to irradiation.

5. Comparison between inorganic and organic scintillators

items	inorganic	organic
scintillation mechanism	Excitons trapped at luminescence centres recombine radiatively.	Excited π-electrons in benzene ring recombine with holes radiatively.
scintillation efficiency	scatters widely in 0.13 (NaI:Tl) ~ 0.001 (PWO)	almost the same: 0.02-0.04
ionization quenching	small (due to restoring force from crystal periodicity)	large
nonradiative transition or thermal quenching	large	small (radiative transition is large and fast)
decay of fast fluorescence component	10~100 times slower than in organic scintillators	fast(2~30 ns)
temperature dependence	large	small
radiation damage	mainly in transmittance. spontaneous, UV, and thermal annealings may occur.	mainly in scintillation efficiency. hardly recovers in time.
density	can be large; 8.28 g/cm^3 (PbWO$_4$)	~1 g/cm^3 consisting of C and H.
γ-ray detection	one of the aims	hardly used
particle discrimination from pulse shape	feasible only in some scintillators	fast/slow ratio is larger for e- and γ-rays than for α-rays.