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Semiconductor Scintillators and Three-Dimensional Integration

U.S. DEPARTMENT OF HOMELAND SECURITY

critical needs:

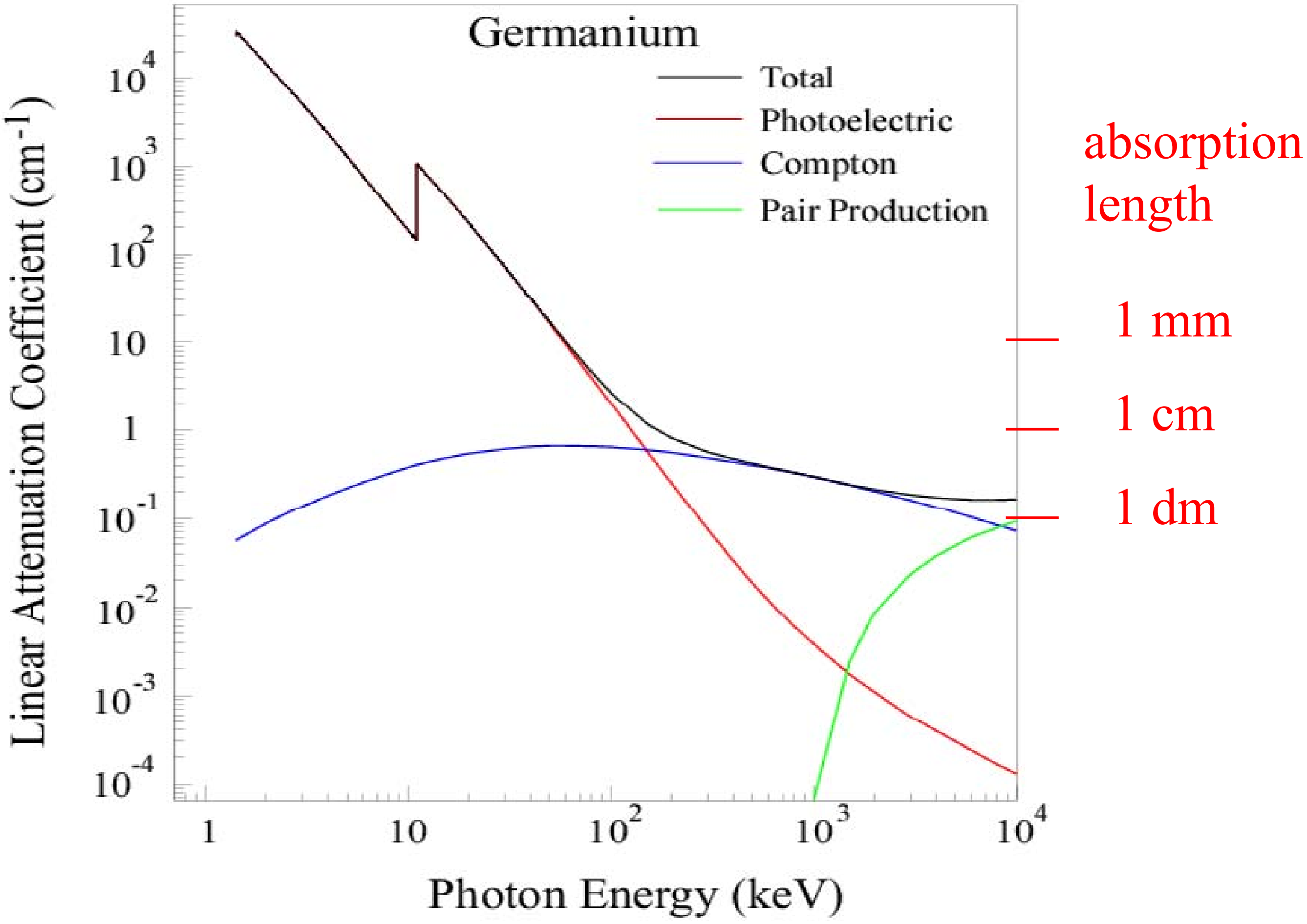
- Isotope identification

  spectroscopic energy resolution

- Direction to source

  angular resolution

X-ray (γ-ray) attenuation



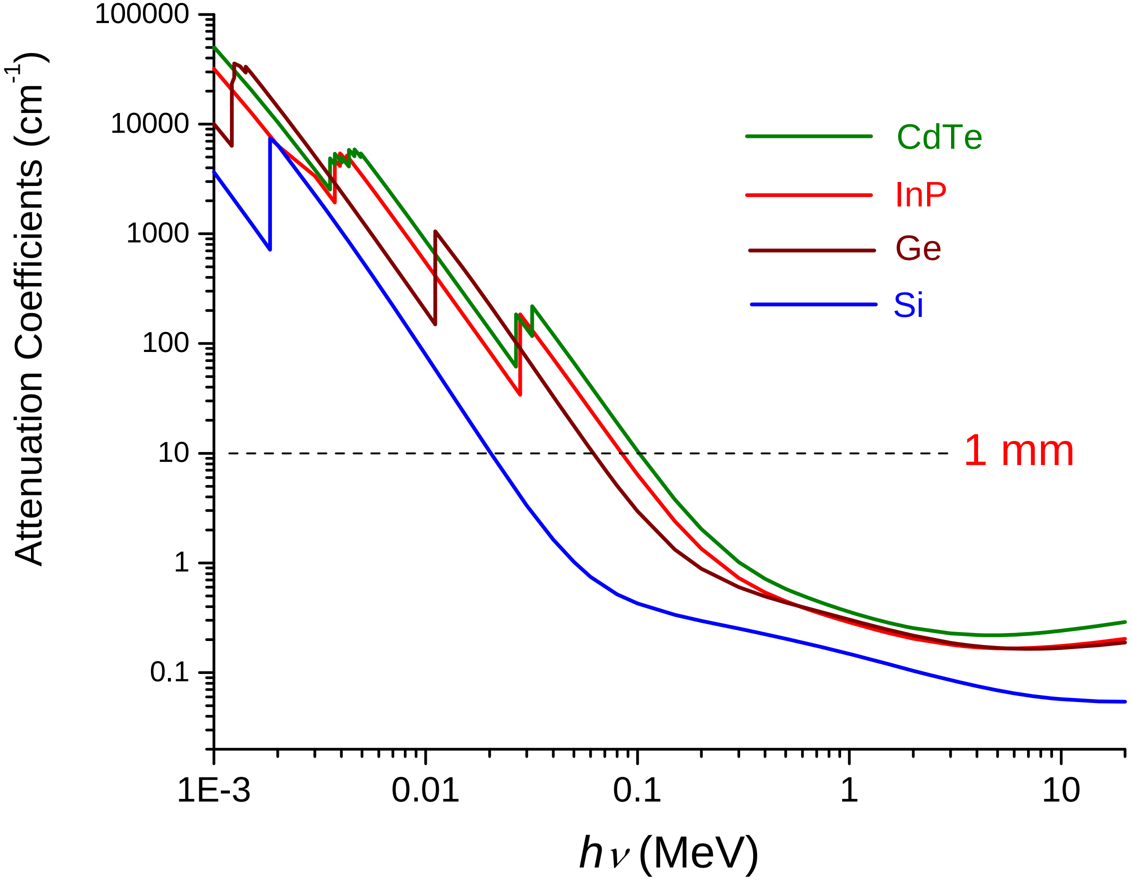
absorption length

1 mm

1 cm

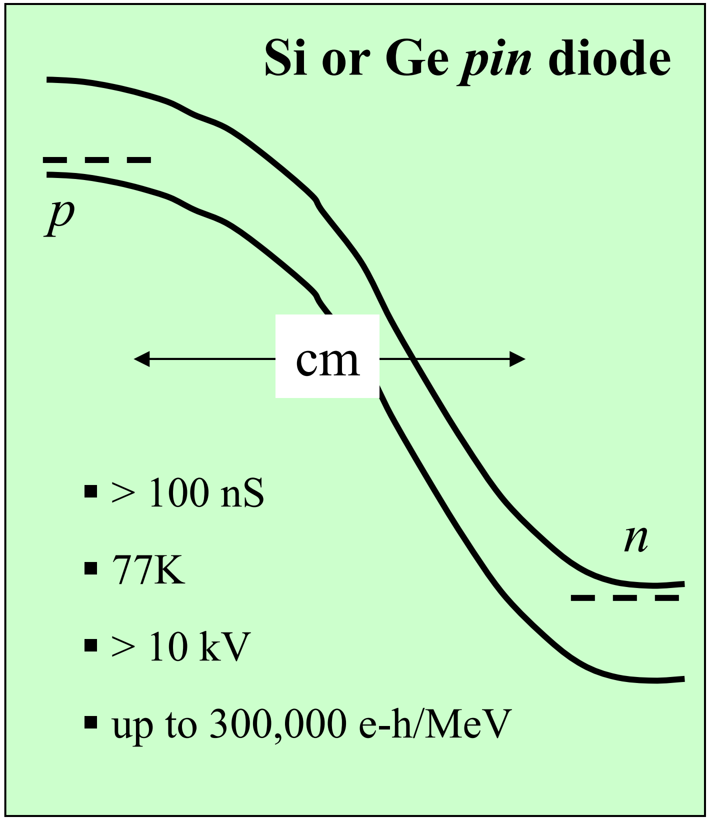
1 dm

X-ray (γ-ray) attenuation by materials



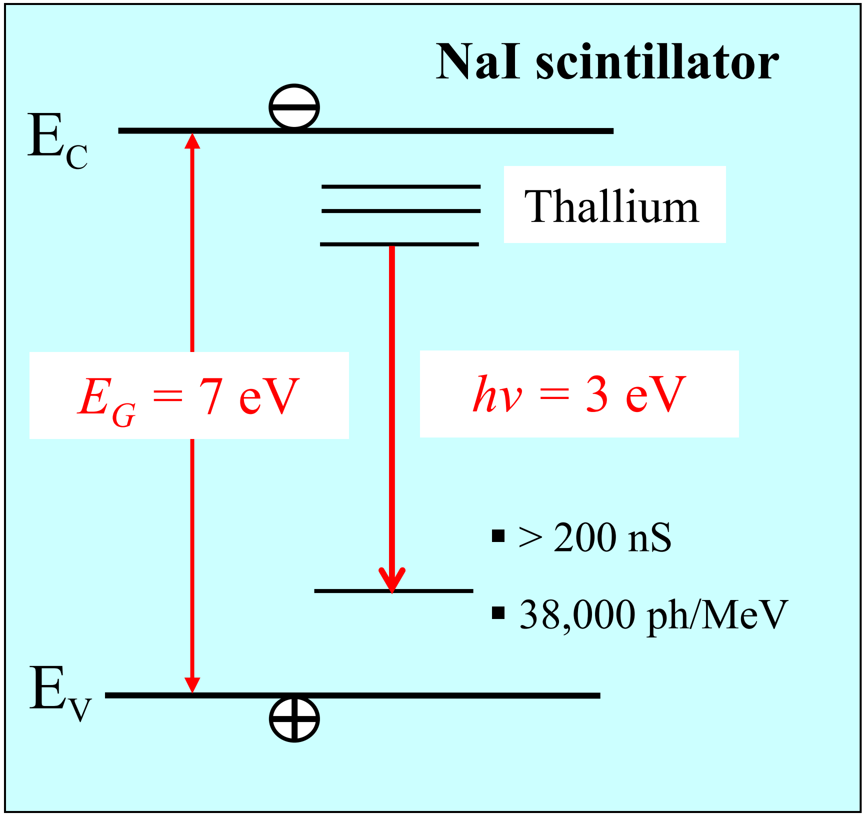
Element	Z
Si	14×2
Ga/As	31/33
Ge	32×2
In/P	49/15
Cd/Te	48/52

Semiconductors and scintillators



Si or Ge *pin* diode

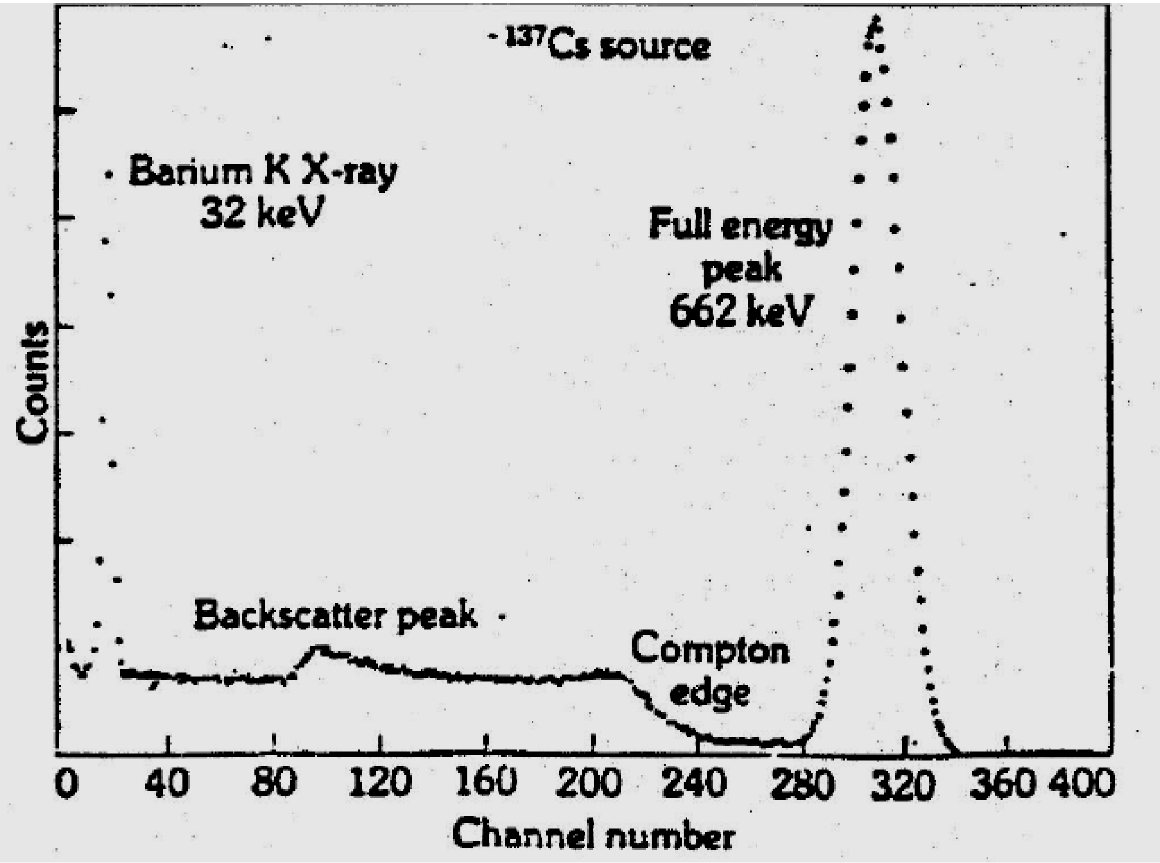
- > 100 nS
- 77K
- > 10 kV
- up to 300,000 e-h/MeV



NaI scintillator

- > 200 nS
- 38,000 ph/MeV

Gamma spectroscopy



One measures the number N of electrons and holes produced by incident gamma particle

$$N \sim E_\gamma$$

That number fluctuates.

If the statistics of N were Poisson,

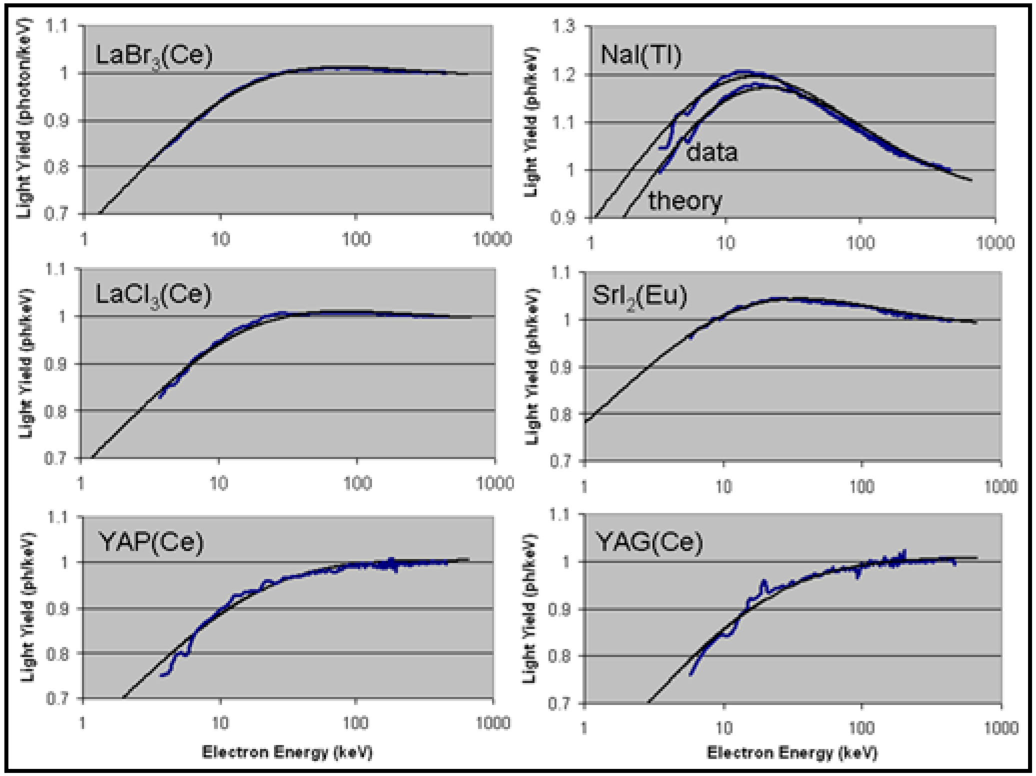
$$\text{var}\,(N) = N$$

but due to correlations
(in semiconductors)

$$\text{var}\,(N) = F\,N$$

the Fano factor, $F \approx 0.1$

Non-proportionality in scintillators



S. A. Payne *et al* (LLNL group), preprint, 2009

Need: $\boxed{N \sim E_\gamma}$ but ...

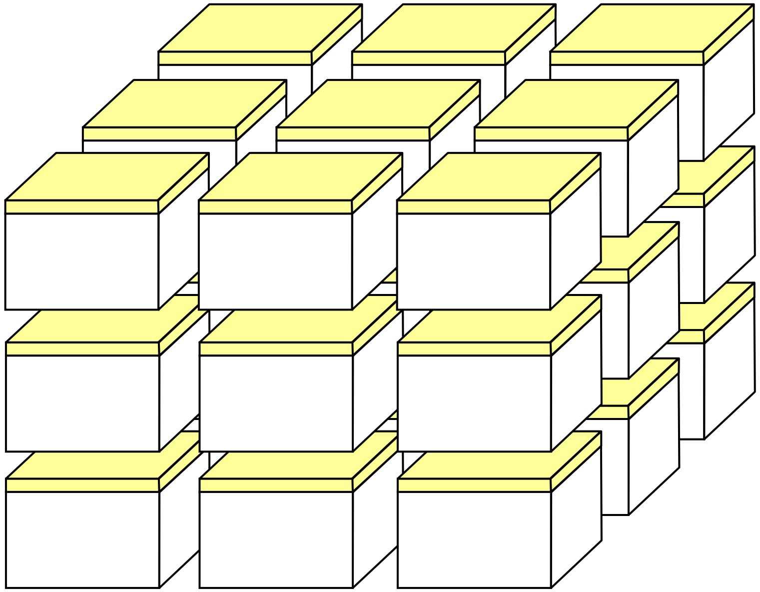
Luminescence in dielectric scintillators is controlled by reactions nonlinear in N (exciton formation, Auger, etc.)

This is one of the reasons γ spectroscopy with scintillators is not as accurate as it is with semiconductor (diodes).

In semiconductor scintillators, every reaction on the way to luminescence is *linear* in the N of *minority carriers*

Expect *no* non-proportionality effects!

3D scintillator array



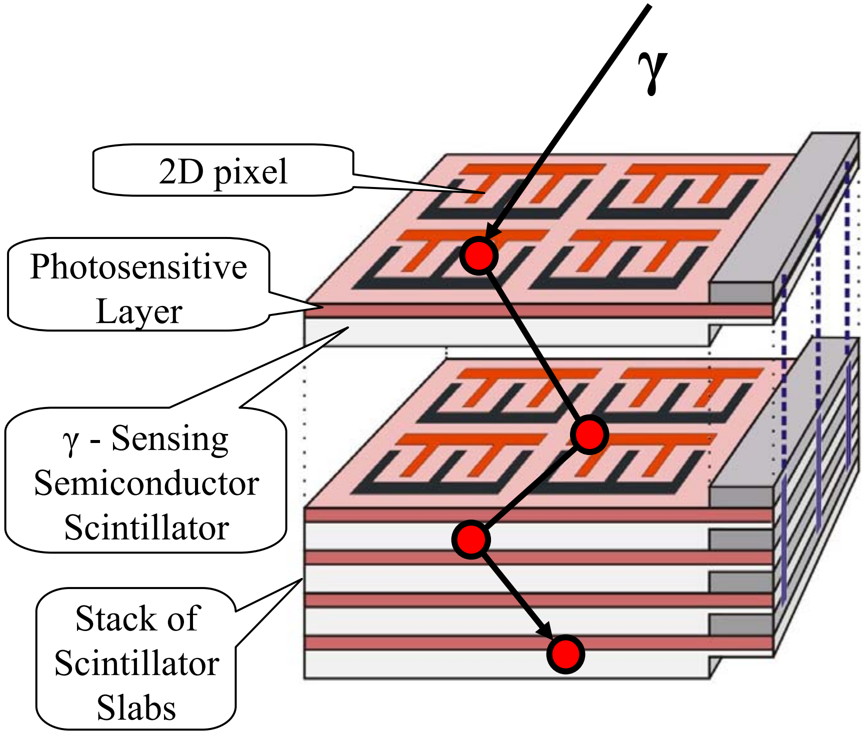
Semiconductor scintillators, each endowed with its own photoreceiver

10 × 10 × 10 array contemplated

Enables *both* isotope discrimination and determination of the direction to source

A different way of determining E_γ (unlike γ spectroscopy)

3D pixellation of response to a single γ photon



Upon analog-to-digital conversion each unit reports not a 1 ns pulse but an information-carrying signal:

- where ionization occurred
- time of the event
- amplitude of the event

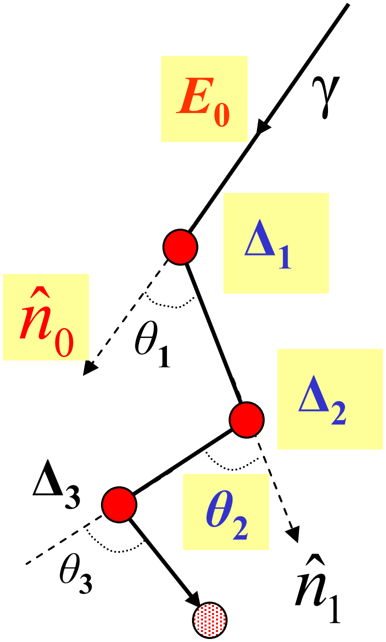
Compton “telescope”

Compton kinematics:

$$\cos\theta_i = 1 + E_{i-1}^{-1} - E_i^{-1}$$
$$\Delta_i = E_{i-1} - E_i$$

two equations at each interaction site *i*

(in units of $m_e c^2 = 511\,\text{keV}$)



The energy E₀ of the incident γ-photon

$$E_0 = \Delta_1 + \frac{\Delta_2}{2} + \frac{1}{2}\left(\Delta_2^2 + \frac{4\Delta_2}{1-\cos\theta_2}\right)$$

The incident direction

$$\cos\theta_1 = 1 + \left(\frac{1}{E_0} - \frac{1}{E_1}\right)$$

What is needed

The triad:

- Semiconductor scintillator

  "transparent" to its own luminescence

- Integrated (optically tight) surface photoreceiver system

  of slightly smaller bandgap

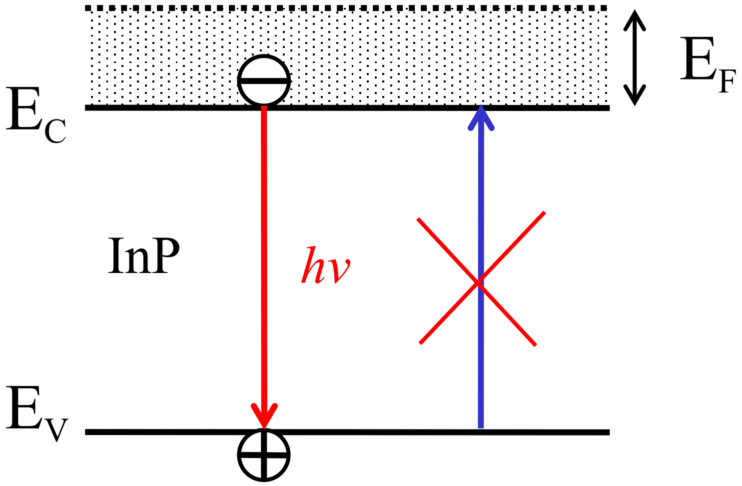
- Readout ASIC

  customized to the photoreceiver system

I will focus on the first two legs of the triad

Semiconductor scintillator: transparency

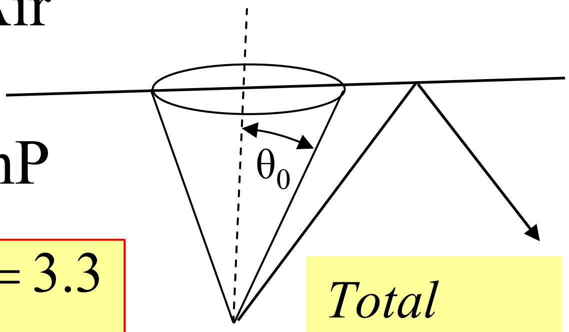
- *Need material transparent to its own fundamental light emission*
- *Photons must be delivered to the surface*



- Moss-Burstein shift
- Photon re-absorption suppressed
- Radiative decay time ≈ 10^{-9} s

"Conventional" transparency

Need for optically-tight photoreceiver



$n = 3.3$

$\sin\theta_0 = 1/n$

Total Internal Reflection

escaping fraction of photons

$$\frac{1}{4\pi}\int_0^{\theta_0}\sin\theta\, d\varphi d\theta = \sin^2\left(\theta_0/2\right) \approx 0.023$$

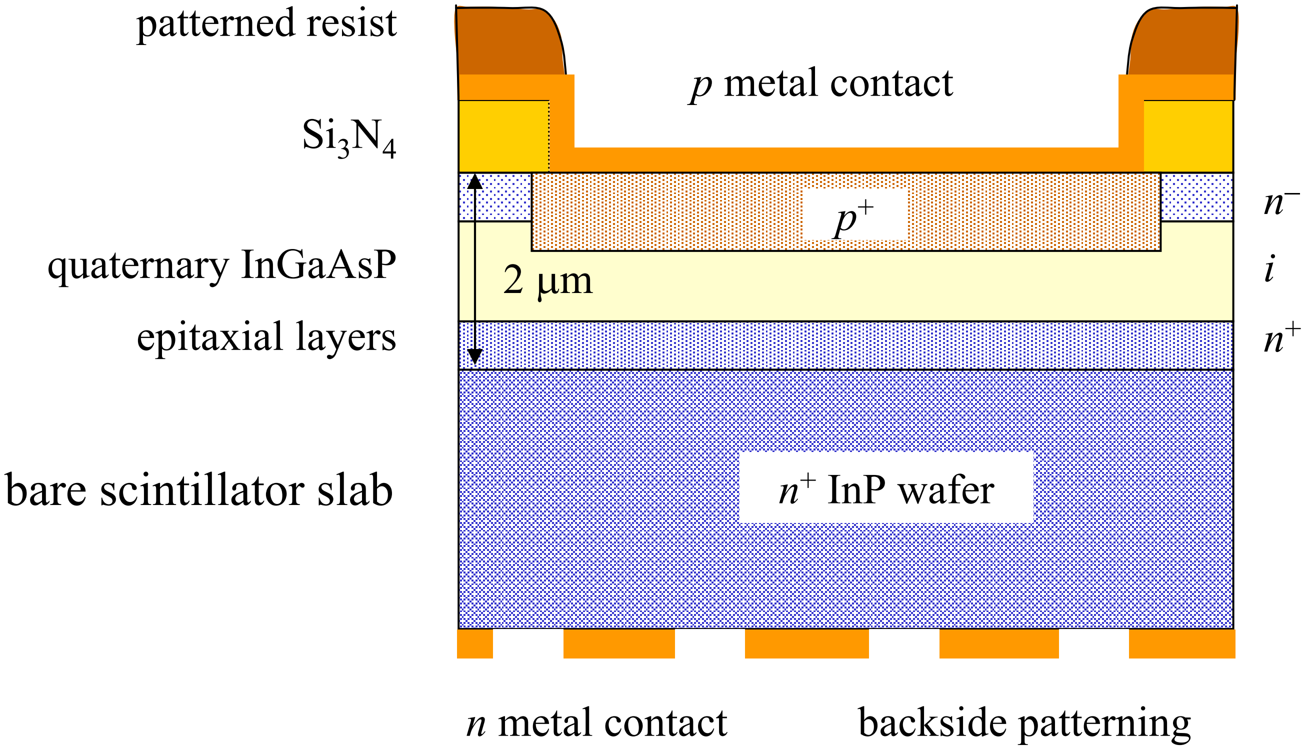
2% ☹ *for free-space detectors*

Optically-tight integrated detectors collect the entire scintillating radiation

Epitaxial InGaAsP diodes on InP

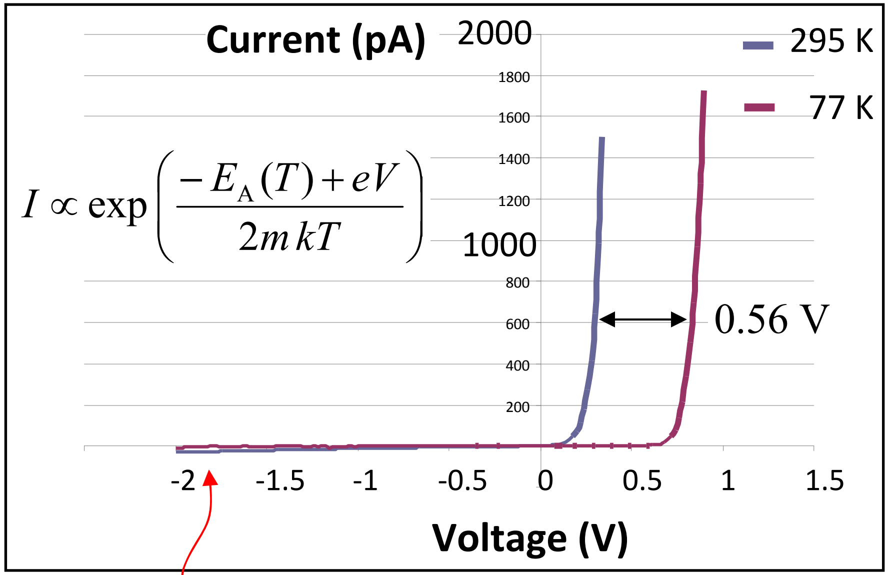
Epitaxially integrated *pin* diode

Sarnoff – SBU collaboration



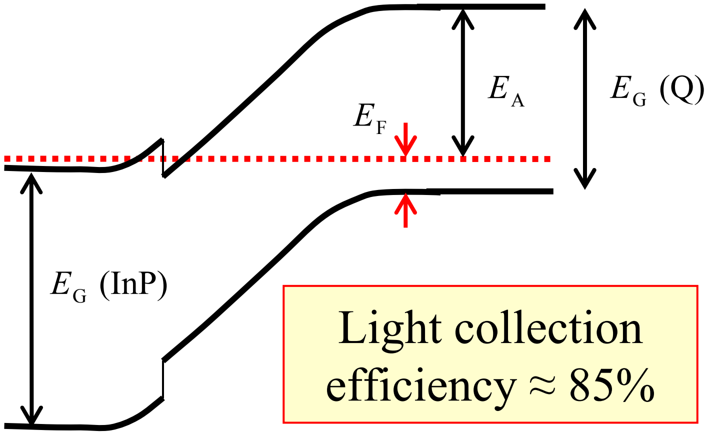
Characteristics of quaternary epi diodes

IV Characteristics of Diodes at 300 and 77 K



$$I \propto \exp\left(\frac{-E_A(T)+eV}{2m\,kT}\right)$$

< 10 pA at 300 K
(1 pA in best diodes)



Light collection efficiency ≈ 85%

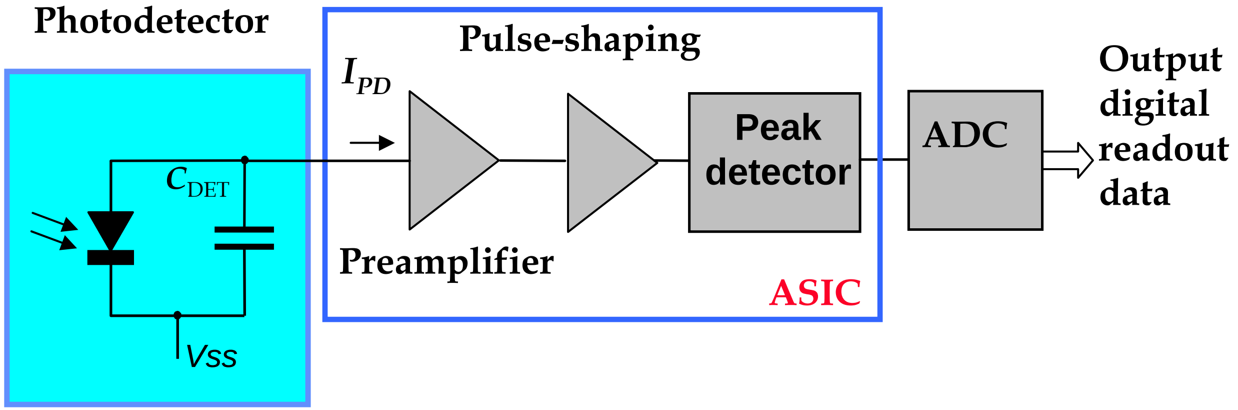
$$E_A(300K) \approx 1\,\text{eV (expt)}$$

$$E_G(\text{Q; 300K}) = 1.24\,\text{eV (designed)}$$

$$E_F = 0.23\,\text{eV (for } n \approx 10^{15}\,\text{cm}^{-3}\text{)}$$

as determined by *CV* profiling
both estimate jibe !

Read-Out Circuits



$$\mathrm{ENC}^2 = (C_{\mathrm{DET}} + C_{\mathrm{MOS}})^2 \left[\frac{a_{\mathrm{th}}}{\tau} \cdot \frac{8kT}{3g_{\mathrm{m}}} + \frac{a_f\, K_f}{C_{\mathrm{MOS}} \cdot f} \right] + a_{\mathrm{sh}}\, \tau\, 2qI_{\mathrm{LK}}$$

ENC – currently: 3×10^3

Next generation: $< 10^3$

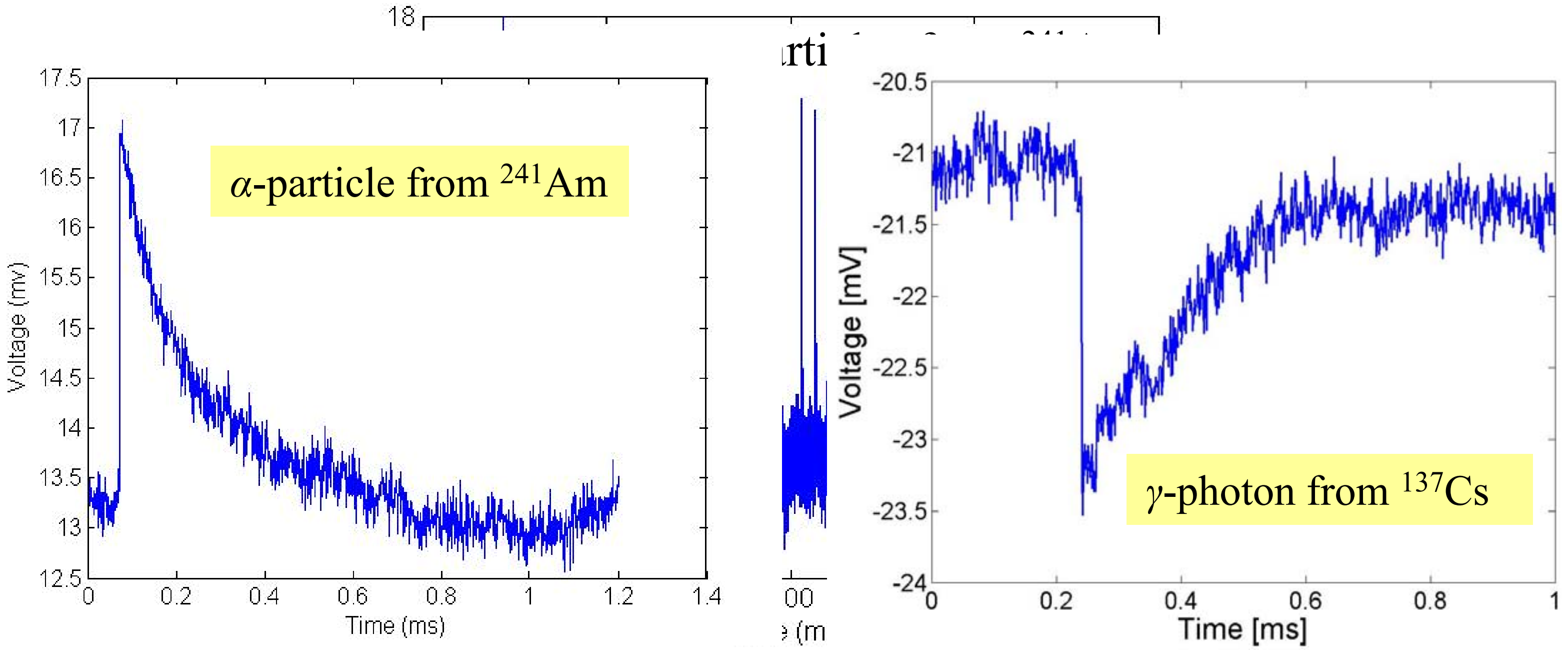
thermal noise

☹ high $C_{\mathrm{DET}} \approx 50$ pF

shot noise

☺ low $I_{\mathrm{LK}} \approx 10$ pA

Single-quanta response



α-particle from ^{241}Am

γ-photon from ^{137}Cs

Train of scintillator pulses recorded as voltage waveform in the read-out circuit

Making semiconductor transparent

… to its own fundamental luminescence

- Moss-Burstein shift

  Success "mixed"
- Photon recycling

  Nearly ideal *non-transparent* scintillator
- Subband luminescence centers

  E.g., Yb^{3+} luminescent ion in InP (1μm emission)
- Impregnated "guest-host" structures

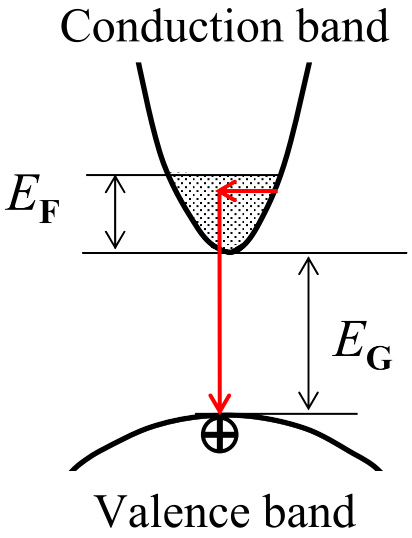
  Non-layered two-phase random systems

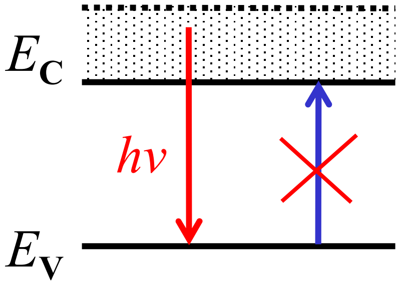
… new ideas are welcome

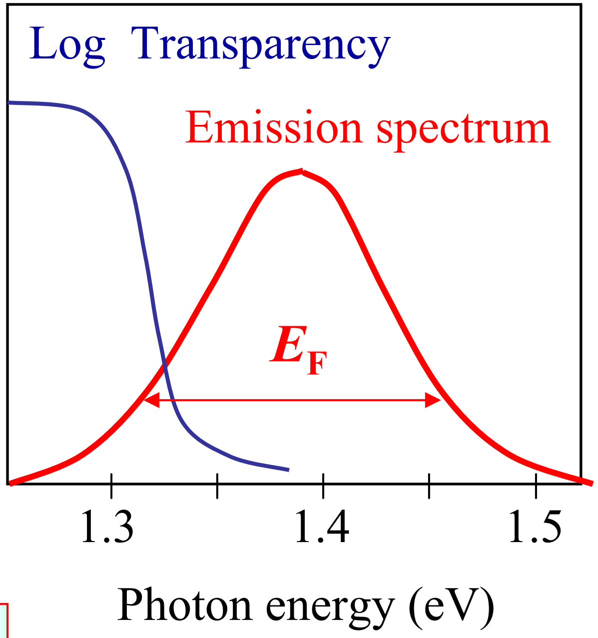
Transparency: theory *vs.* reality

no free
lunch
☹

Momentum non-conservation in heavily-doped InP

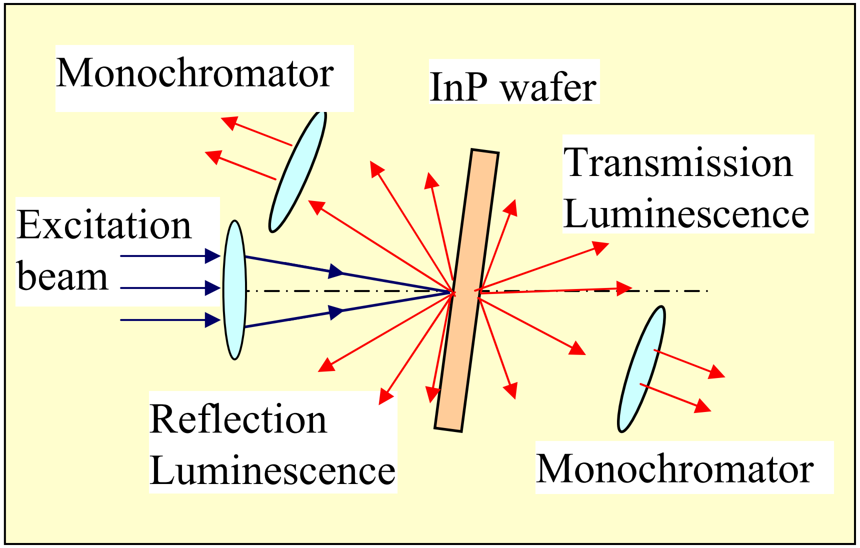






"Average" (over the emission spectrum) photon mean free path is about 0.1 mm

Luminescence Experiments



Monochromator

InP wafer

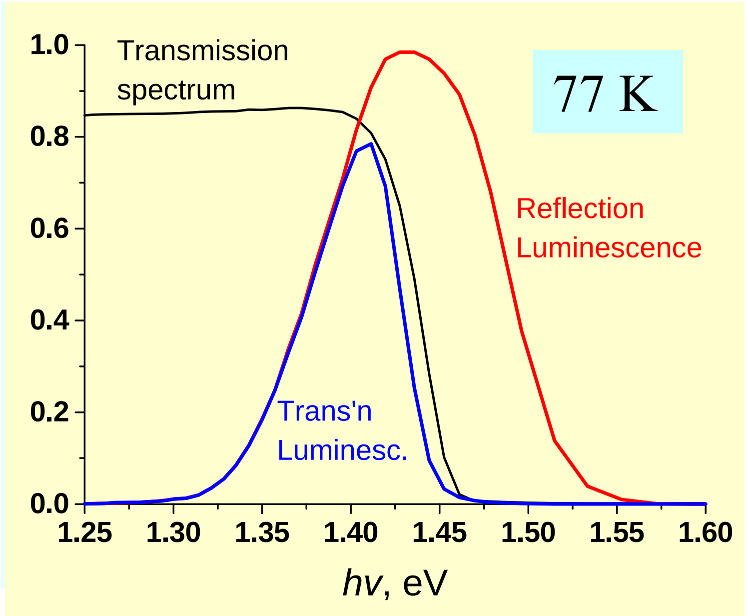
Transmission Luminescence

Excitation beam

Reflection Luminescence

Monochromator

InP, $N_D=6.3\times10^{18}$ cm^{-3} (S)

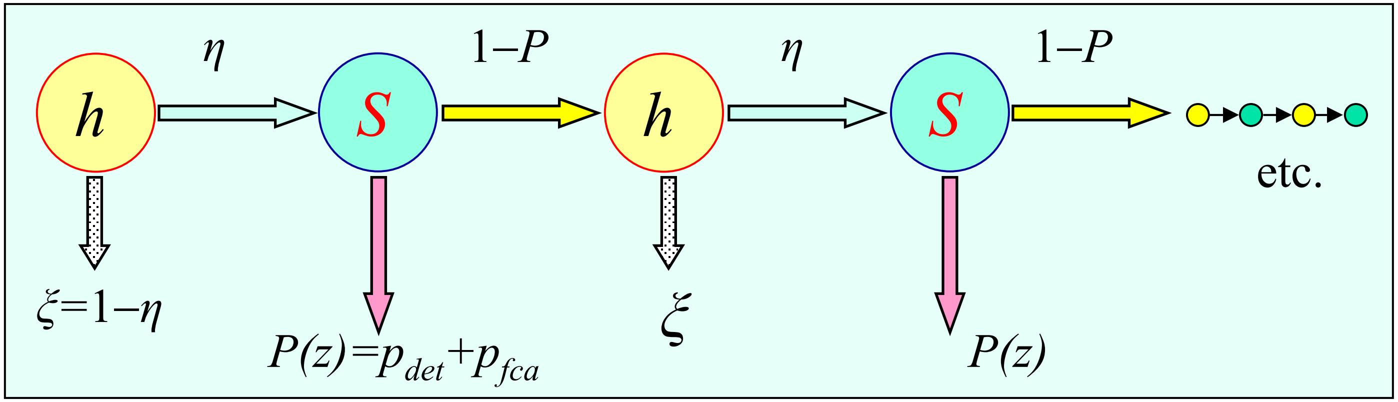


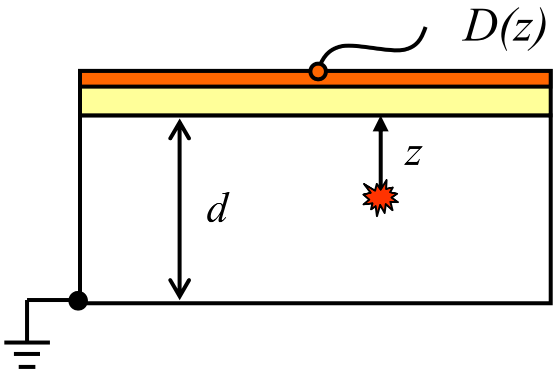
Heavily doped n-type InP wafers from Nikko materials (ACROTEC)

Transport of Holes/Photons

- In a direct-gap *n*-type semiconductor, an event of interband absorption does not finish off the photon:
  - It emerges as a new hole and then again a new photon (spectrum)
- Holes and photons inside are “interchangeable” entities
  - As a photon, its lifetime is limited by free-carrier absorption (rate ~ *n*). Key parameter — *residual absorption coefficient* μ_r
  - As a hole, its lifetime is limited by non-radiative processes, (rate ~ n^2). Key parameter — *radiative efficiency* η
- Photon “recycling”
  - a.k.a. photon-assisted diffusion of holes

Photon recycling





$$D(z) = \eta\, p_{\mathrm{det}} \times \sum_{n=0} \left[\eta\left(1-P\right)\right]^{n} = \frac{\eta\, p_{\mathrm{det}}(z)}{\xi + \eta\, P(z)}$$

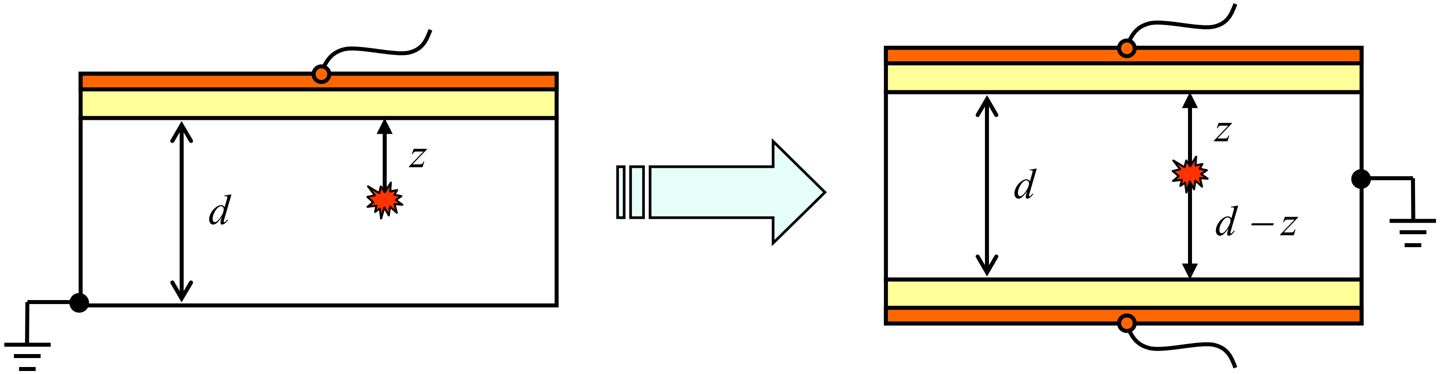
where $\eta \approx 0.9$ is the radiative efficiency

and $\xi=1-\eta$

Problem with InP and possible remedy

The signal dependence on the position *D(z)* presents a fundamental problem for the intended application:

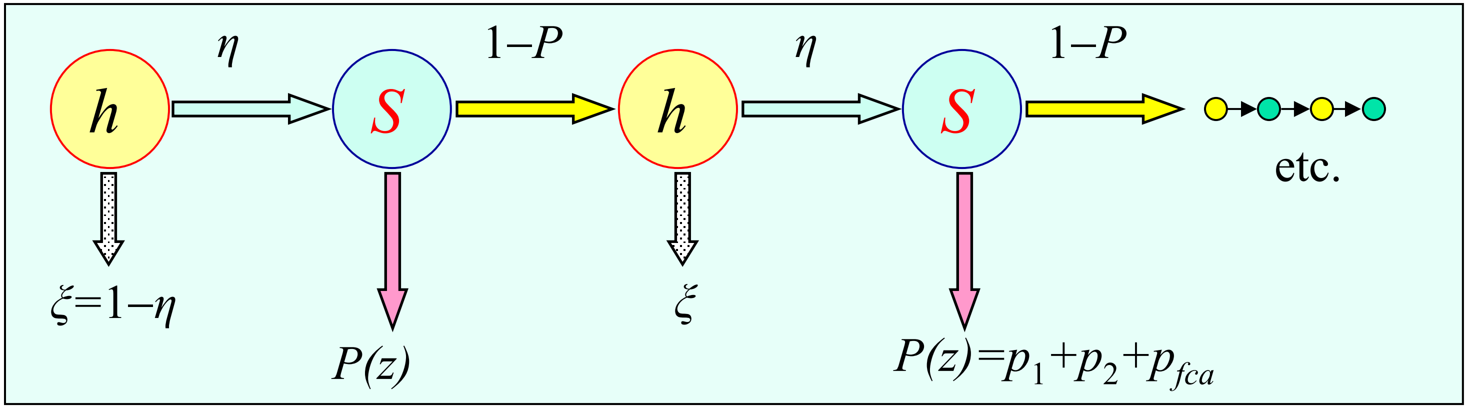
- ☹ How do we distinguish large faraway event from a smaller event closer to the detector surface?
- 😐 Can we correct for the *z* dependence?
- ☺ Double-sided epi detector



if we know *z*, we can correct for the attenuation

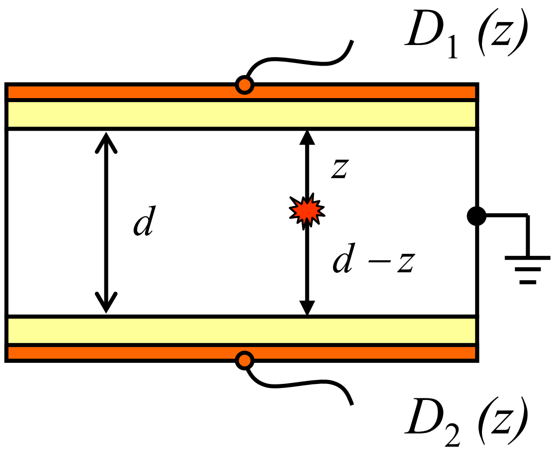
J. H. Abeles & S. Luryi
US Pat. Prov. Appl. (May 2009)

Photon recycling with 2-sided detector



$$D_1(z) = \frac{\eta\, p_1(z)}{\xi + \eta\, P(z)}$$

$$D_2(z) = \frac{\eta\, p_2(z)}{\xi + \eta\, P(z)}$$



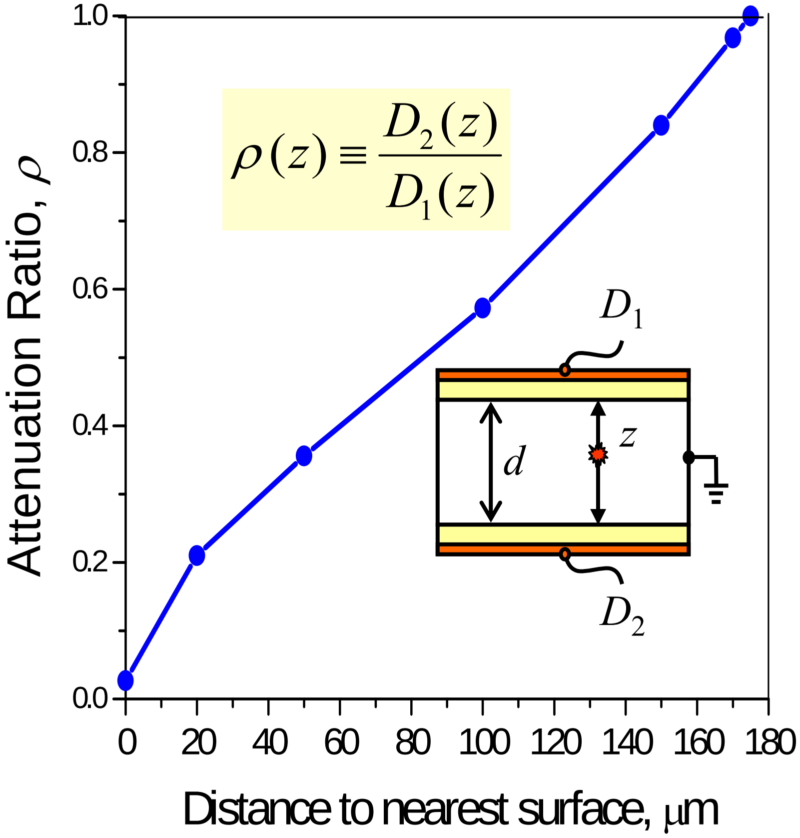
Calculated attenuation ratio

$$D_1(z) = \frac{\eta p_1(z)}{\xi + \eta P(z)}$$

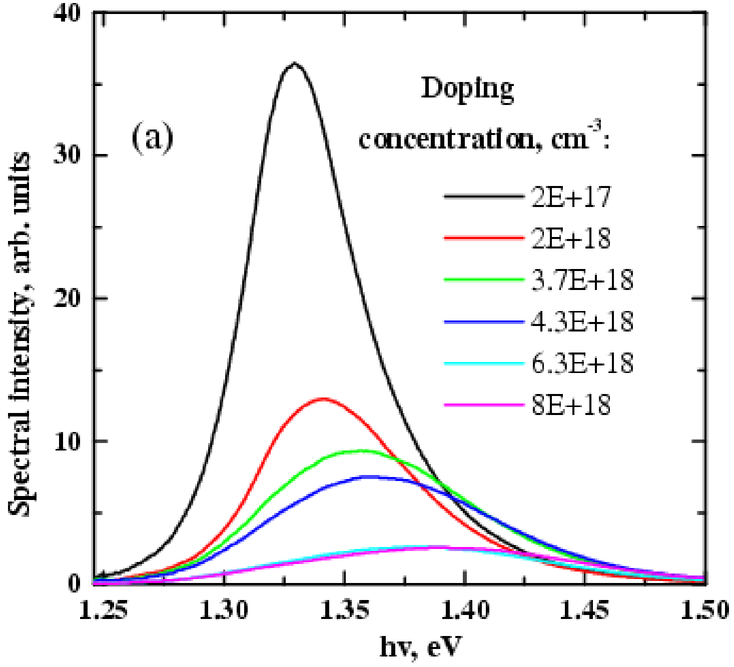
$$D_2(z) = \frac{\eta p_2(z)}{\xi + \eta P(z)}$$

$$\frac{D_1(z)}{D_2(z)} = \frac{p_1(z)}{p_2(z)}$$

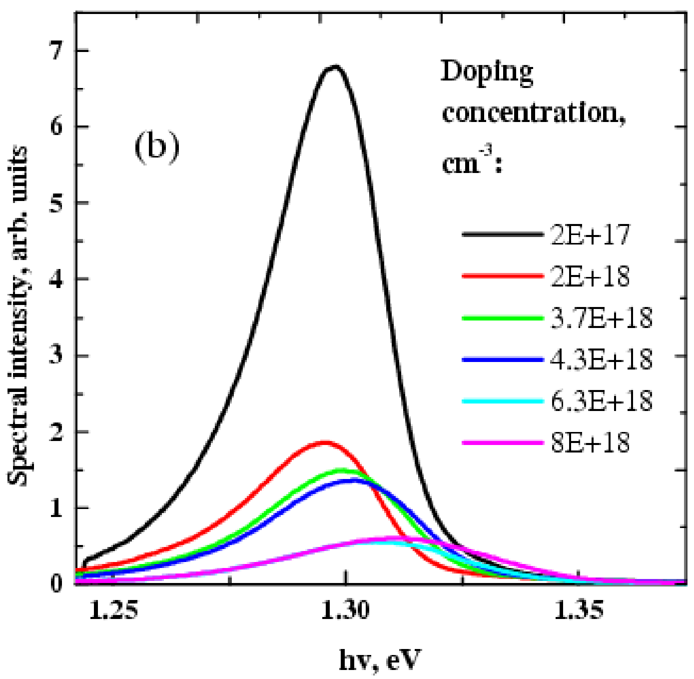
ratio of single-pass probabilities



Lightly doped InP: bright luminescence

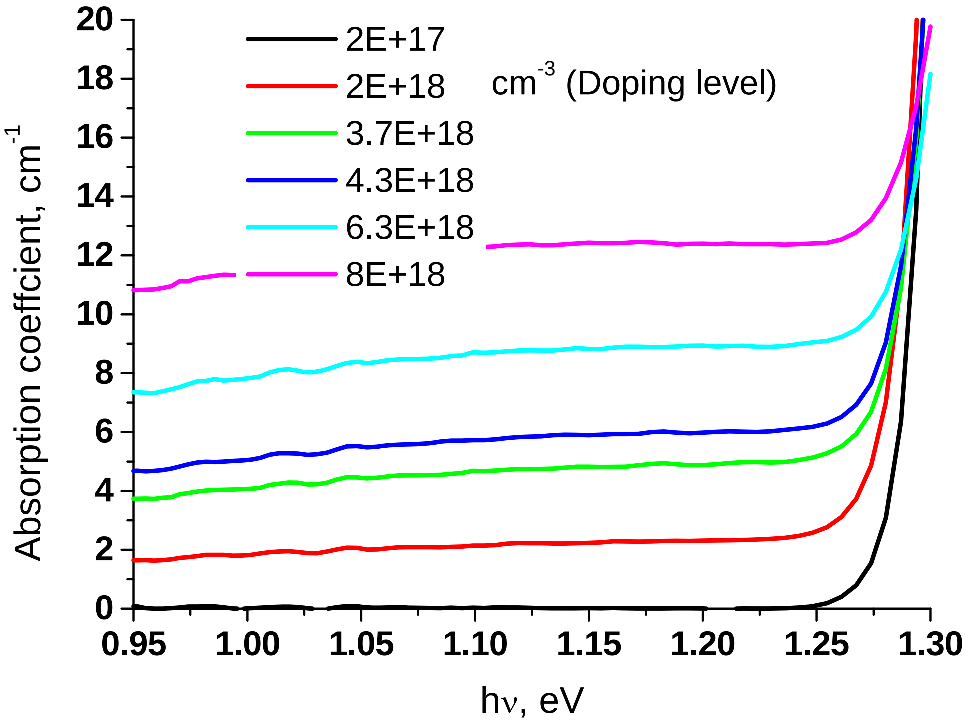


Reflection geometry



Transmission geometry

Lightly doped InP: low free-carrier abs’n

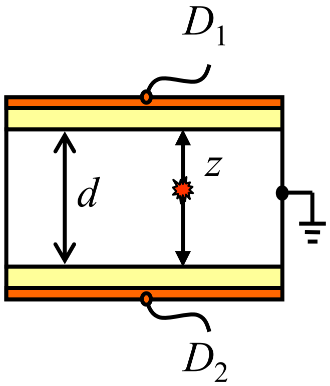


For $N_D = 2\times10^{17}$ cm^{-3}, the measured $\alpha_{FCA} < 0.1$ cm^{-1}

Ideal *non-transparent* scintillator

$$D_1(z) = \frac{\eta\, p_1(z)}{\xi + \eta\, P(z)}$$

$$D_2(z) = \frac{\eta\, p_2(z)}{\xi + \eta\, P(z)}$$



Based on photon recycling in low-doped (2×10^{17} cm^{-3}) InP, where:

$\eta \approx$ 98-99% *and* FCA negligible

$$D_1(z) + D_2(z) = \frac{\eta\left[p_1(z) + p_2(z)\right]}{\xi + \eta\, P(z)}$$

$$\text{PCE} \;= \frac{\eta\left[p_1(z) + p_2(z)\right]}{(1-\eta) + \eta\left[p_1(z) + p_2(z) + p_{FCA}\right]}$$

for $\eta \approx$ 98%, PCE =91%

for $\eta \approx$ 99%, PCE =95%

Unusual situation: semiconductor is opaque, in the conventional sense, but photon collection efficiency approaches unity. No losses!

Pause for reflection

- Two-sided detection pinpoints the *z*-coordinate precisely because of distance-dependent attenuation of single-pass photons
  - Would not work in a *transparent* scintillator (except for the factor of 2)
- Suppose we find semiconductor with *much* higher PCE?
  - Is there advantage in 2-sided detection (apart from the factor of 2)?
  - Precision of z determination is no longer ± d, hence can make thick voxels!
- Search for better PCE
  - Must be accompanied by due regard to leg 2 of the triad (integrated PD)
  - Going beyond InP needs ideas for optically tight integration

Two-phase semiconductor structures

Random non-planar inclusions: "impregnations"

Laye

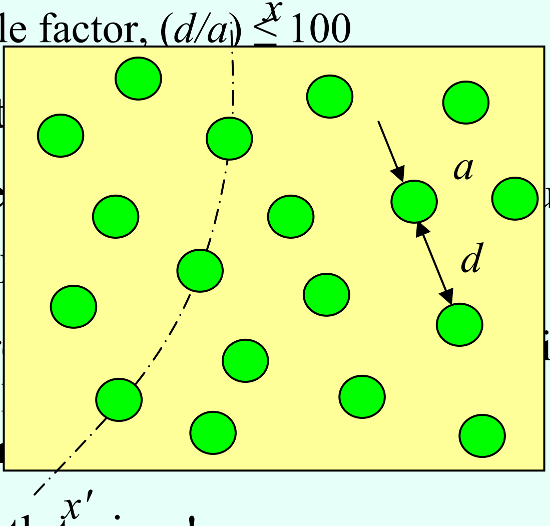
Transparency enhancement by the duty cycle factor, $(d/a) \leq 100$

Difficult

Nee ur)

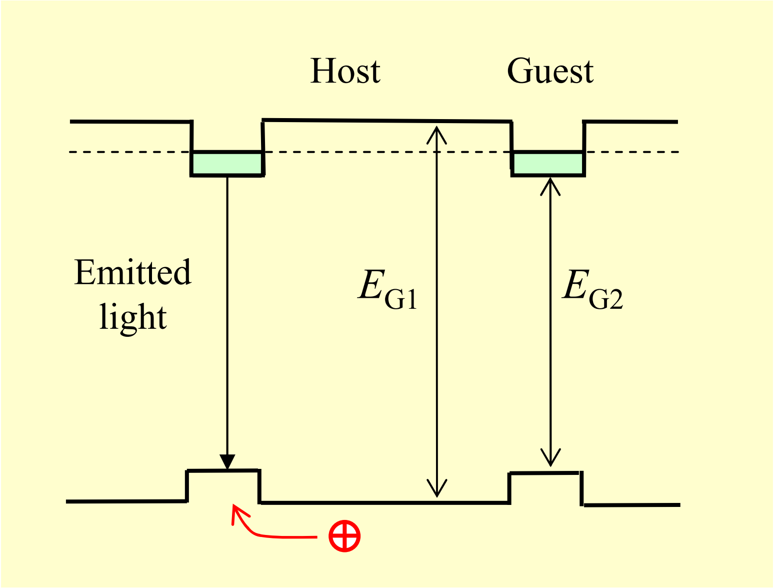
HV

Har in a thic eve



Still worth trying !

Any volunte



holes still migrate to the nearest inclusion

han lifetime

nited

- Transparency gain scales as ratio of volumes $(d/a)^3$
- Lattice-matching requirement is removed

Making a bulk impregnated structure ...

- Direct growth with interruptions and self-organization

  e.g. InGaAs quantum dots on GaAs substrate

- Phase separation via spinodal decomposition

  e.g. VPE grown InGaN shows subband emission (Shur)

- Transparent ceramics techniques (LLNL)

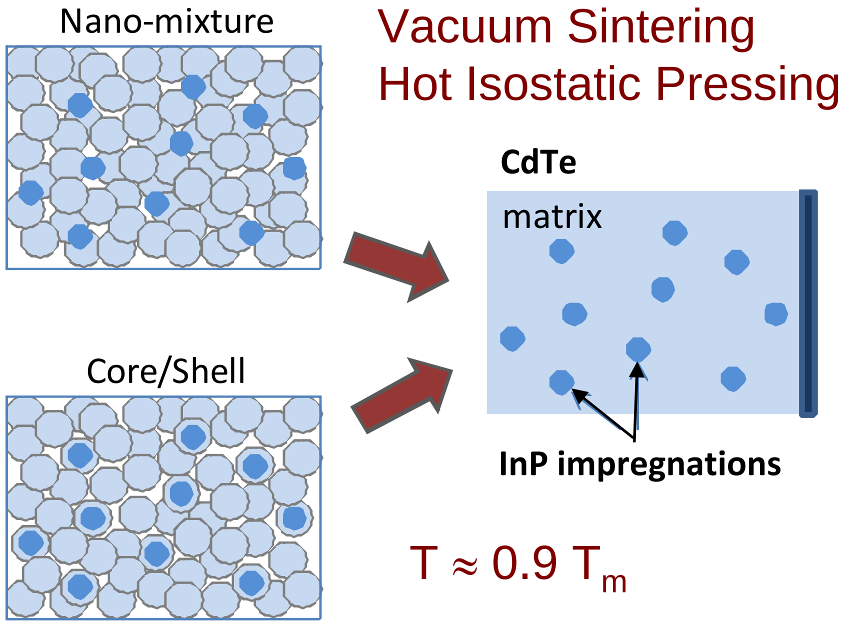
  Consolidation of nano-dot powders (e.g. ZnSe) by HIP

  Two-phase ceramic, e.g. InP guest in CdTe matrix

... any other ideas ?

Two-phase transparent ceramics

Vacuum Sintering
Hot Isostatic Pressing (HIP)



$T \approx 0.9\ T_m$

impregnations must have smaller bandgap but higher melting point

Mater.	E_G (eV)	T_m (ºC)
ZnS	3.68	1,850
ZnSe	2.822	1,100
ZnTe	2.394	1,240
CdS	2.50	1,750
CdSe	1.714	1,350
CdTe	1.474	1,041
InP	1.344	1,060
GaAs	1.424	1,240

Dots	Matrix
CdSe	ZnTe
CdSe	ZnSe
ZnTe	ZnSe
CdS	ZnSe
InP	CdTe

Summary

- Semiconductor scintillators – enabling technology
  - Compact and portable Compton telescope
  - 3D array of units, each endowed with integrated photoreceiver
  - Optically tight integration, two-sided photodiode
- Semiconductors “transparent” to own luminescence
  - Direct-gap semiconductors with photon recycling
  - Semiconductors activated with luminescent centers
  - Composite “impregnated” systems

Sponsors

Semiconductor high-energy radiation detector with excellent isotope identification and directional capability (DHS)

Three-Dimensionally Pixellated Semiconductor Scintillator (DHS/NSF)

Energy and Spatial Correlation Effects in the Energy Resolution of Semiconductor Radiation Detectors (DTRA)

Going beyond InP... e.g., GaAs

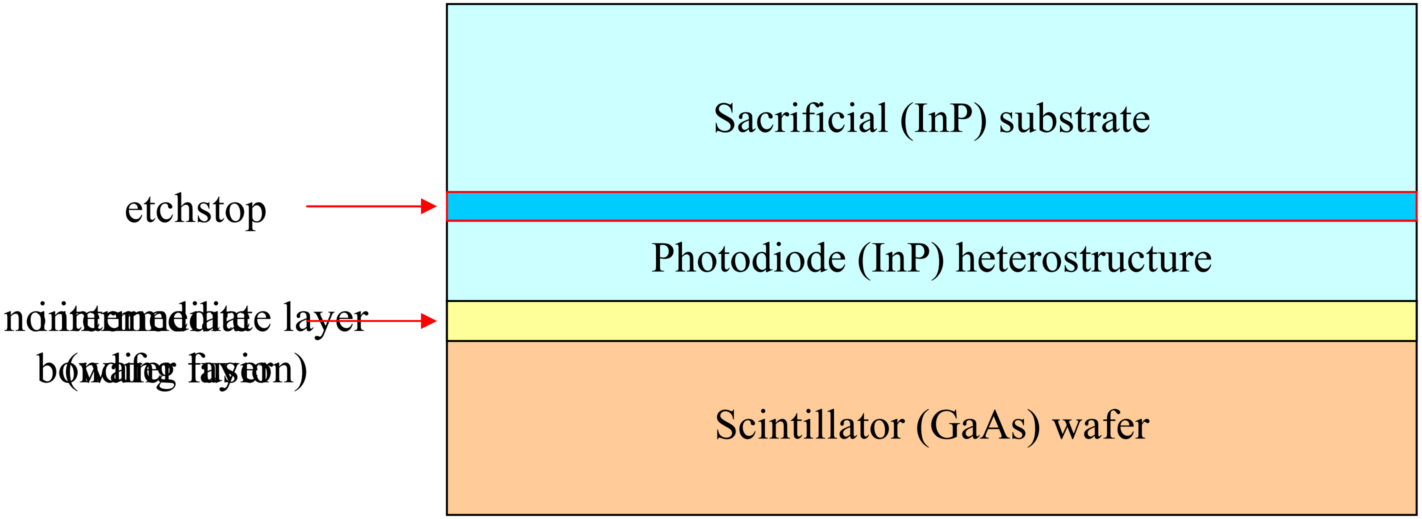
GaAs may be better than InP, but

- **GaAs pros**
  - Higher radiative recombination coefficient, while nonradiative similar

$$\boxed{B_{\mathrm{GaAs}} \approx 7B_{\mathrm{InP}}} \qquad \boxed{C_{\mathrm{GaAs}} \approx C_{\mathrm{InP}}}$$

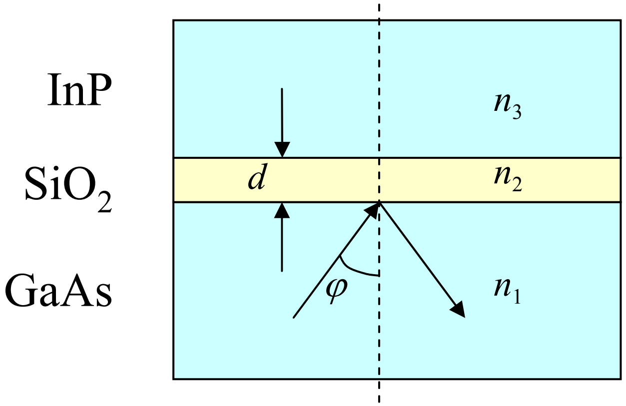
  - Higher bandgap (good for low-noise photodetection at room temperature)
  - May be cleaner, mature technology
- **GaAs cons**
  - No proven *epitaxial* detector
- **Wafer fusion technology may come to rescue**

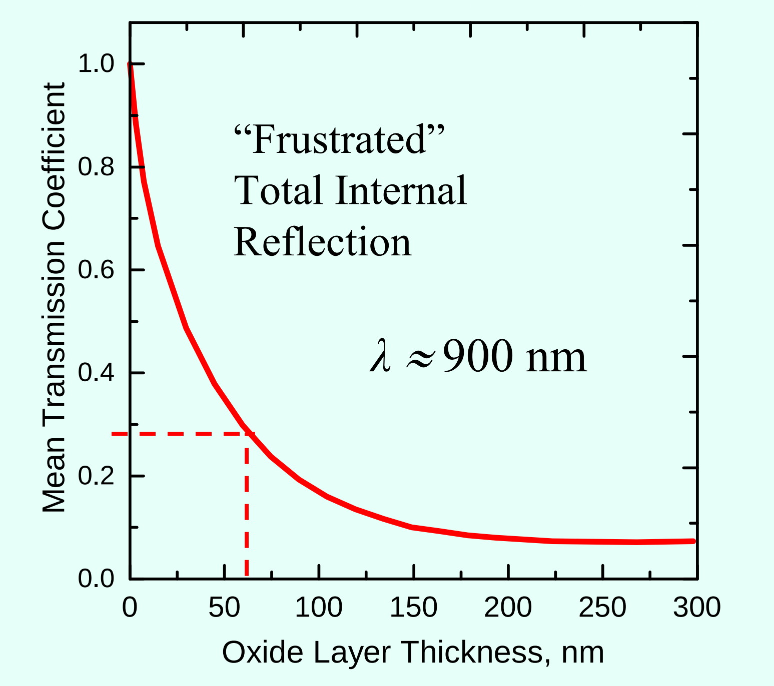
Wafer bonding *versus* wafer fusion



What's wrong with an intermediate layer ?

NOTHING *if it is really thin*

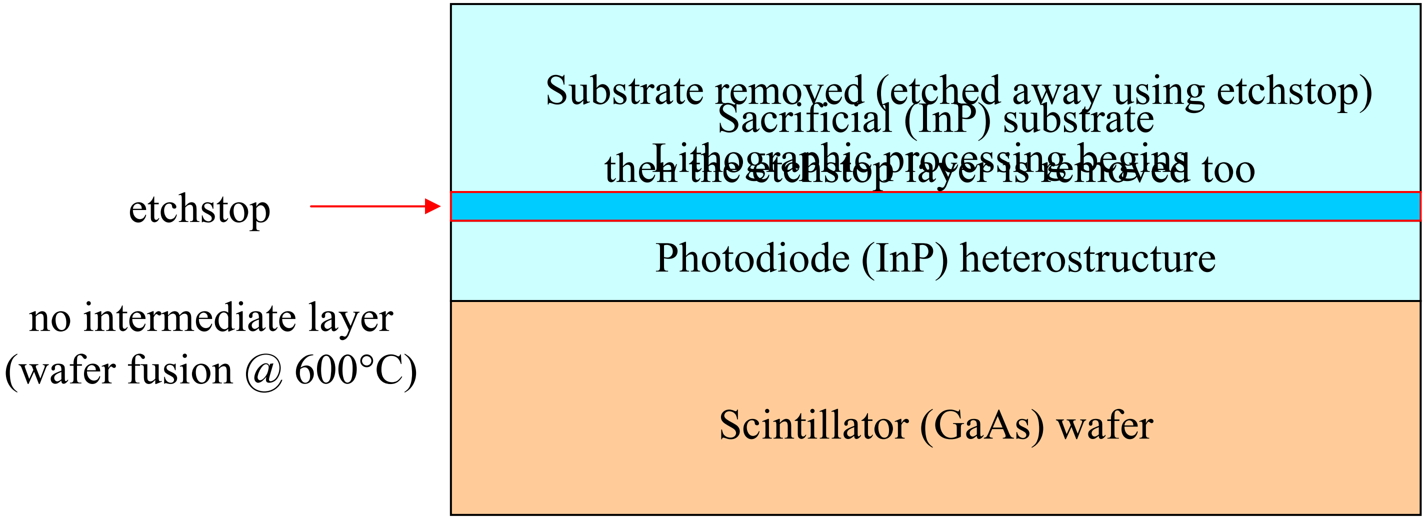




BUT *naive comparison of the optical thickness with wavelength does not work for lower-index layers*

even for 60 nm thick oxide layer, over 70% of GaAs scintillation will not go through

Photodiode integration by wafer fusion



Substrate removed (etched away using etchstop)

Sacrificial (InP) substrate

then the etchstop layer is removed too

Lithographic processing begins

etchstop

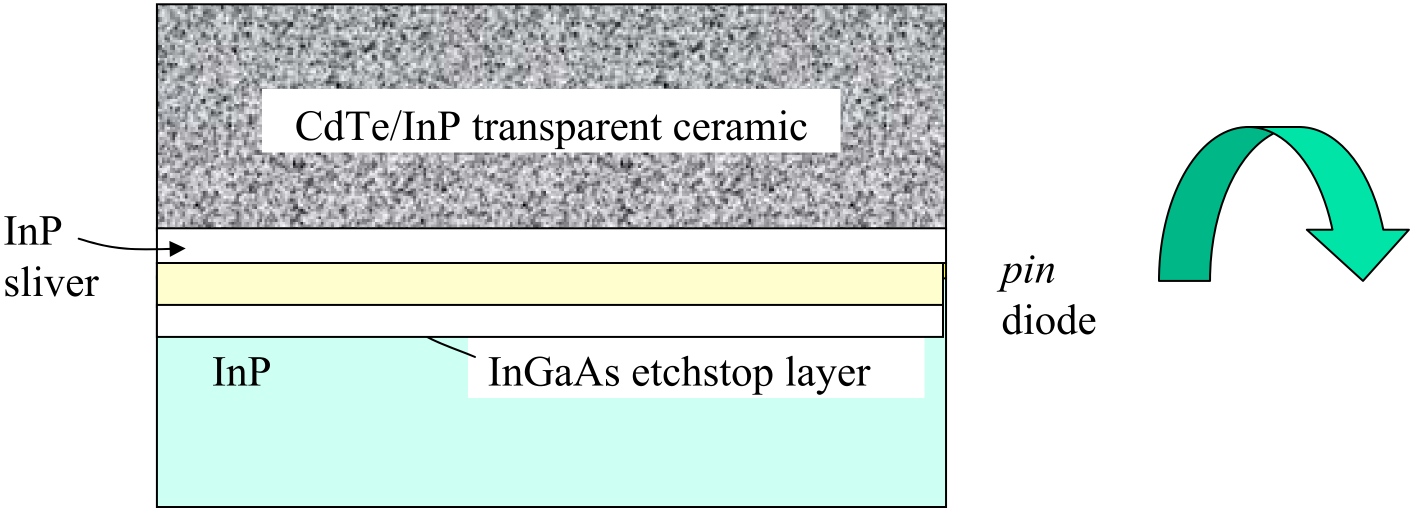
Photodiode (InP) heterostructure

no intermediate layer
(wafer fusion @ 600°C)

Scintillator (GaAs) wafer

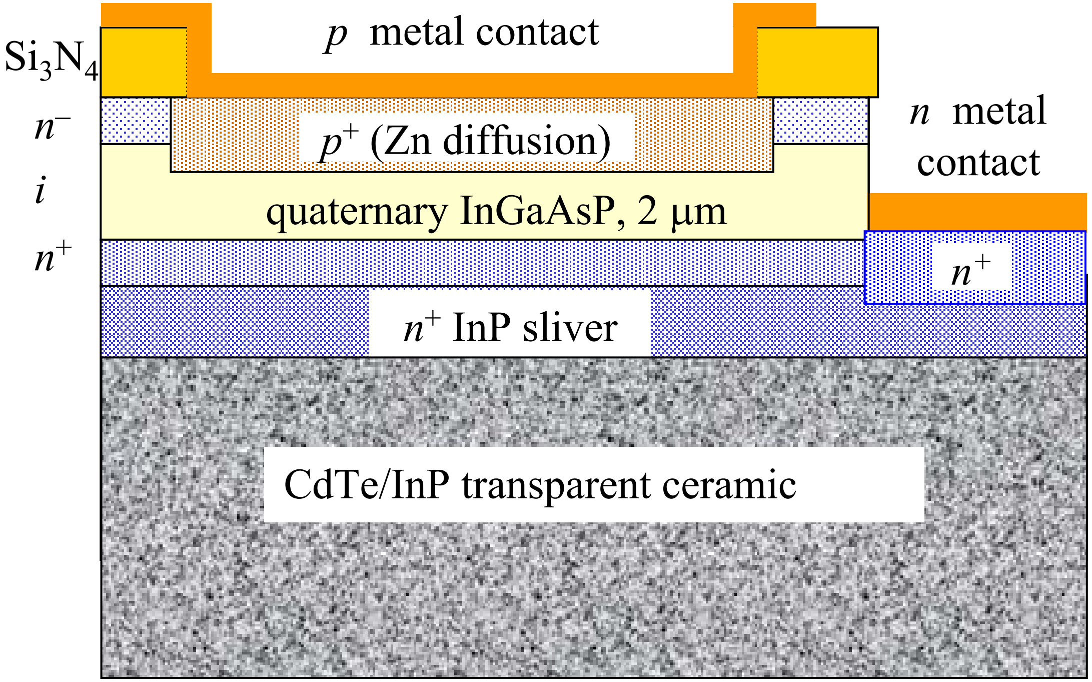
Optically tight photodiode integration

3. Etchstop layer goes away too ..



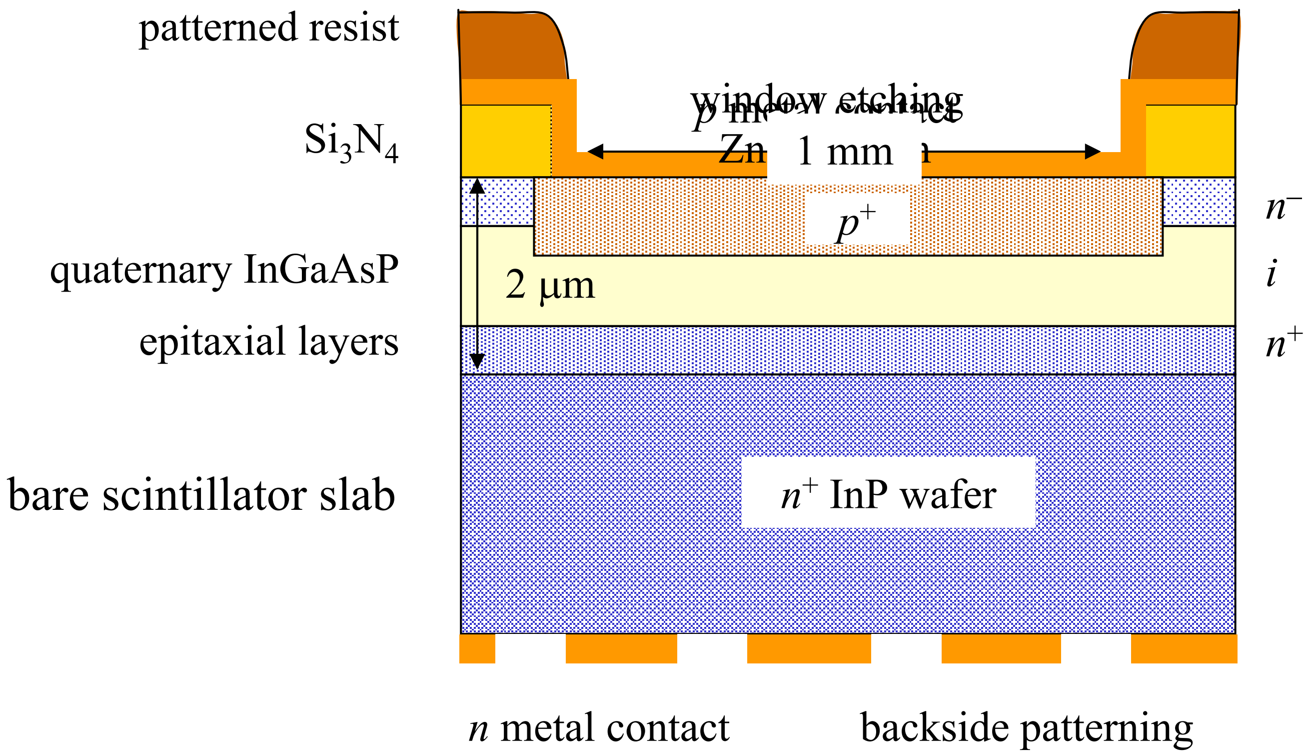
Optically tight photodiode integration

6. ... and looking at it again

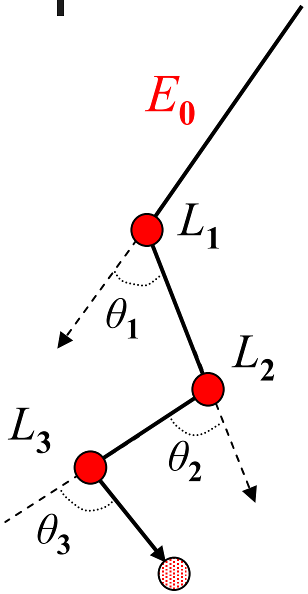


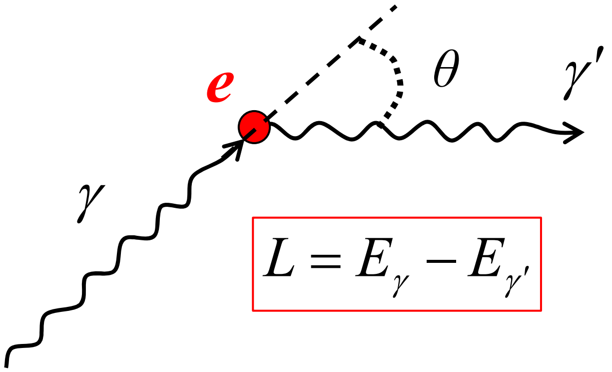
Epitaxial *pin* diode fabrication

Sarnoff – SBU collaboration



Compton Scattering





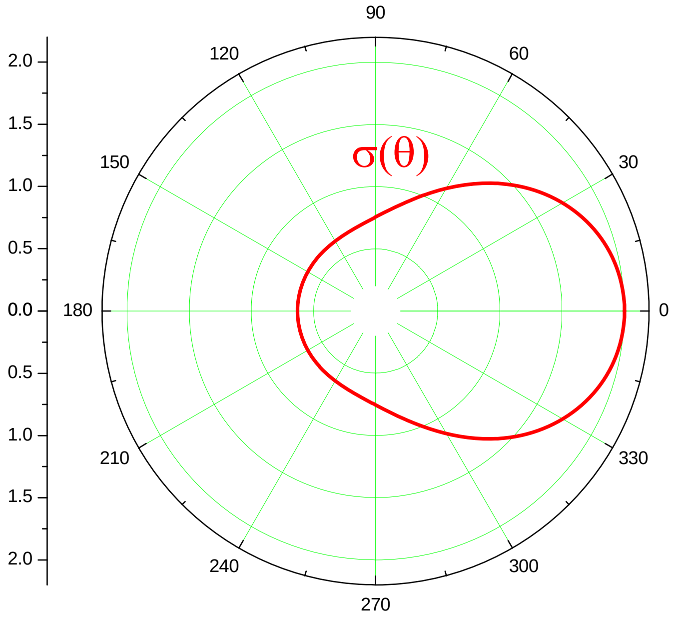
$$L = E_\gamma - E_{\gamma'}$$

$$m_e c^2 = 511\,\text{KeV}$$

$$E_\gamma = 662\,\text{KeV}\ \ (\text{Cs}^{137})$$

kinematics (Compton):

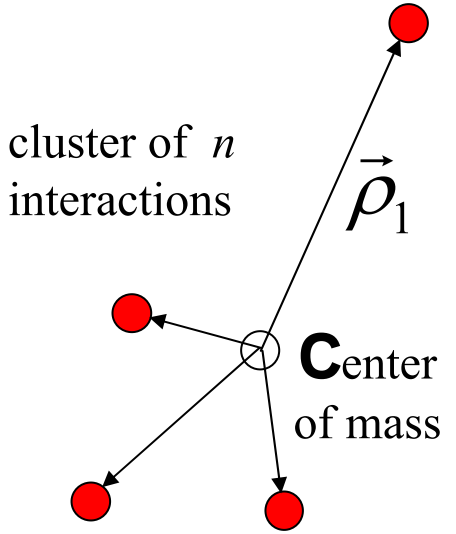
$$\cos\theta = 1 + \frac{m_e c^2}{E_\gamma} - \frac{m_e c^2}{E_{\gamma'}}$$



dynamics (Klein-Nishina):

$$\sigma(\theta) = \sigma_0 \left(\frac{E_{\gamma'}}{E_\gamma}\right)^2 \left(\frac{E_{\gamma'}}{E_\gamma} + \frac{E_\gamma}{E_{\gamma'}} - \sin^2(\theta)\right)$$

Direction to source



cluster of n interactions

$\vec{\rho}_1$

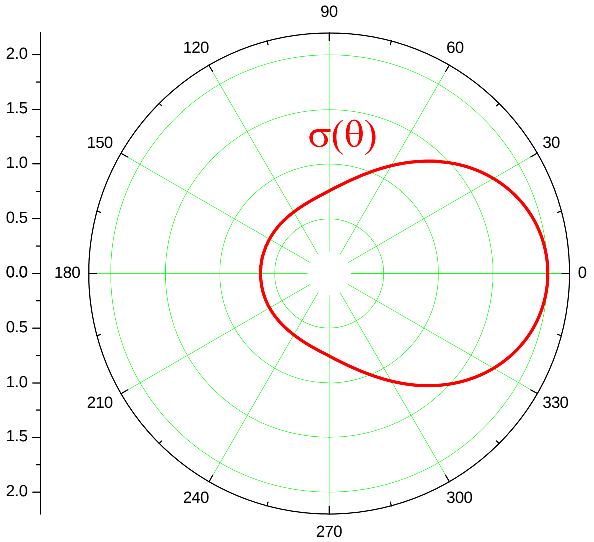
Center of mass

dynamics (Klein-Nishina formula):

$$\boxed{\sigma(\theta_i)=\sigma_0\left(\frac{E_i}{E_{i-1}}\right)^2\left(\frac{E_i}{E_{i-1}}+\frac{E_{i-1}}{E_i}-\sin^2(\theta_i)\right)}$$

anisotropic scattering cross-section

662 keV:



$$\left\langle\vec{\rho}_1\right\rangle_n\equiv\frac{1}{N_n}\sum_{j=1}^{N_n}\vec{\rho}_1^{\,(j)}=\left(np_n-1\right)\vec{\rho}+\frac{\vec{\delta}}{\sqrt{N_n}}$$