Neutron Detection

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- Applications of Neutron
- Neutron Detection
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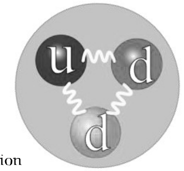
Neutrons?



- Discovered in 1932 by Chadwick
- **No electric charge**
  - Very week electromagnetic interaction
  - Neutrons penetrate matter easily, no direct atomic ionization ("highly penetrating, indirectly ionizing radiation")
  - Neutrons are able to interact directly with the atom nucleus and not electron cloud
- Interact only by strong nuclear interaction
  - Nuclear force is very short ranged, thus neutrons have to pass close to a nucleus to be able to interact
  - Because of small size of the nucleus in relation to the atom, neutrons have low probability of interaction and **long travelling distances** (several cm without interaction) in matter
- Mass = 939.56 MeV/c^2
  - Slightly heavier than proton
- Lifetime = 886.7 s ($n \rightarrow p + e^- + \overline{\nu}$)
  - Free neutrons (unstable) undergo beta decay

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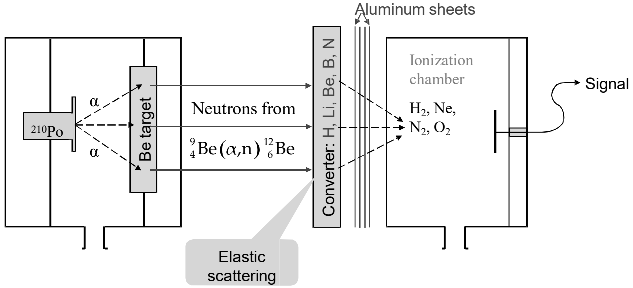
Neutron Discovery

- In 1930 W. Bothe and H. Becker in Germany: α → light elements (Be, B, Li); unussually strong penetrating radiation was produced.
- In 1932 Irène Joliot-Curie and Frédéric Joliot in Paris: (unknown) radiation → paraffin or any other hydrogen containing compound it ejected protons of very high energy.
- Finally (later in 1932) the physicist James Chadwick in England performed a series of experiments showing that the gamma ray hypothesis was untenable.

Neutron Discovery

Sir James Chadwick, 1932 (Nobel prize - 1935)



Mme I. Curie and F. Joliot had an idea: $^{9}_{4}\mathrm{Be} + ^{4}_{2}\mathrm{He} \rightarrow ^{13}_{6}\mathrm{C} + \gamma$

Chadwick guessed: $^{9}_{4}\mathrm{Be} + ^{4}_{2}\mathrm{He} \rightarrow ^{12}_{6}\mathrm{C} + ^{1}_{0}\mathrm{n} + \gamma$

Categorization of Neutrons

- **Energy ranges**
  - Ultracold: $E < 10^{-6}$ eV
  - **Cold** neutrons: 10^{-6} eV ~ 0.005 eV
  - **Thermal** neutrons: 0.002 eV ~ 0.5 eV
  - **Epithermal** (resonance) neutrons: 0.5 eV ~ 1 keV
  - Cadmium threshold: ~0.5 (with higher energy pass through 1 mm of Cd)
  - Slow neutrons: E < 1 keV
  - Neutrons with middle energies: 1 keV ~ 0.5 MeV
  - **Fast** neutrons: 0.5 MeV ~ 20 MeV
  - Neutrons with high energies: 20 MeV ~ 0.1 GeV
  - Relativistic neutrons: 0.1 GeV ~ 10 GeV
  - Ultrarelativistic neutrons: E > 10GeV

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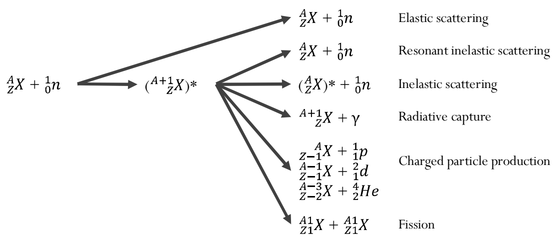
Neutron Interactions with Matter



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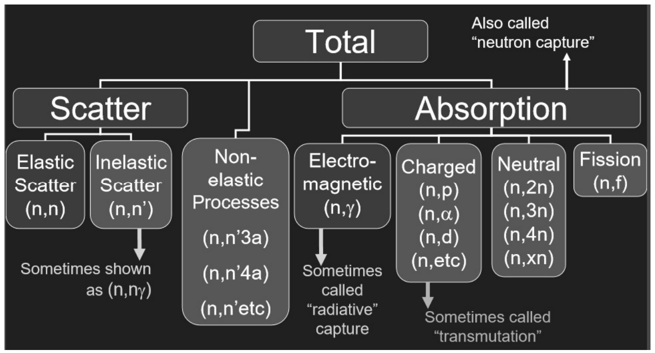
Neutron Interaction with Matter



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Neutron Interaction with Matter

- Nuclear interactions of neutrons
  - No electric charge → no direct atomic ionization
    → only collisions and reactions with nuclei
    → 10-6 times weaker absorption than charged particles
  - Characteristic secondary nuclear radiation/products
    → Always heavy charged particles (not electrons as in the case of γ interaction)
    - γ-rays (n,r) / charged particles (n,p),(n,α) / neutrons (n,n'),(n,2n') / fission fragments (n,f) / spallation reactions
- Dominant reactions depend on available neutron energy:
  - E_n ~ 1/40 eV (=$k_B T$) slow diffusion, <u>absorption</u>
  - E_n < 10 MeV <u>Elastic scattering</u>, <u>absorption</u>, nuclear excitation
  - E_n > 10 MeV <u>Elastic/inelastic scattering</u>, various nuclear reactions, secondary charged reaction products

Typical fate of neutrons: fast neutron → thermal neutron → capture (absorption)

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Neutron Interactions: Scattering

- Elastic Scattering (n,n)
  - More likely in low Z materials with neutrons at lower energies
  - Momentum and kinetic energy of the system is conserved
  - No energy transfer to nuclear excitation
  - Target nucleus can gain the amount of the kinetic energy that neutron lose
  - Most frequent process used for kinetic energy decreasing (moderation) of neutrons
- Inelastic Scattering (n,n')
  - More likely in high Z materials with neutrons at higher energies
  - Part of the neutron kinetic energy is used for excitation of the target nucleus: target nucleus is left in an excited state
  - Recoiled neutron has much lower energy than the initial kinetic energy
  - The excited nucleus decays by gamma rays (inelastic gamma rays): endothermic reaction

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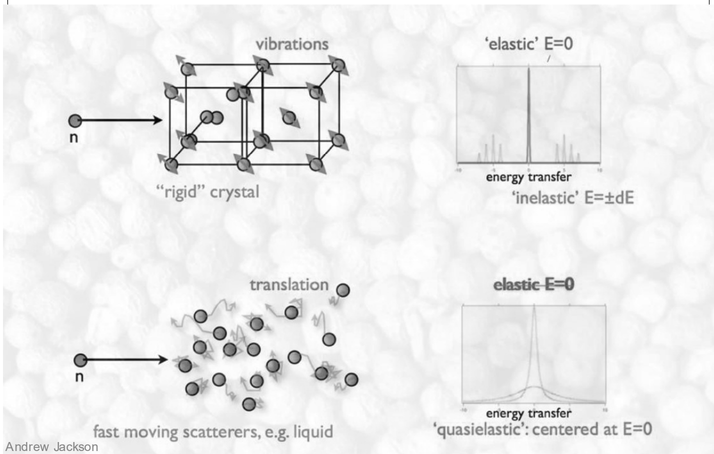
Elastic and Inelastic



Elastic Scattering

- Elastic scattering is the most likely interaction between (relatively lower energy) fast neutrons and low Z material
- Energy and direction of neutron is altered
- No intermediate excitation of recoil nucleus
- Dominant energy loss process at intermediate to high energies (mostly for E_n < 10 MeV)
- If target nucleus is light, neutron loses much energy
  →very effective slowing down process
  - Graphite and heavy water is commonly used
- Process responsible for neutron moderation ("slowing down")

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Elastic Scattering

- Physics
  - Neutron with lab velocity v, energy E_n scatters randomly off target nucleus of mass number A at rest in lab.
  - The energy of the recoil nucleus
    - Center-of-mass system

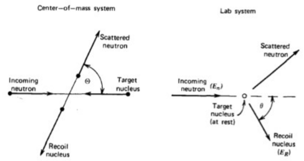
$$E_R = \frac{2A}{(1+A)^2}(1 - cos\Theta)E_n$$

    - Lab system

$$E_R = \frac{4A}{(1+A)^2}(cos^2\theta)E_n$$

    - maximum energy

$$E_R = \frac{4A}{(1+A)^2}E_n \;\; (\theta = 0)$$



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Elastic Scattering

- Physics
  - The Maximum energy of the recoil nucleus $E_R = \frac{4A}{(1+A)^2}E_n \;\; (\theta = 0)$
  - For direct head-on collisions, nuclei with lower mass are more effective on a "per collision" basis for slowing down neutrons

Target Nucleus	E_R/E_n
^{1}H	1
^{2}H	8/9=0.889
^{3}He	3/4=0.750
^{4}He	16/25=0.640
^{12}C	48/169=0.284
^{16}O	64/289=0.221

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Inelastic Scattering

- It becomes possible for neutrons above several MeV (mostly for $E_n >$ 10 MeV), to transfer sufficient energy to the target nucleus(usually heavy material) to induce an excited nuclear state
- The neutron strikes a nucleus and form a compound nucleus which is unstable and emits a neutron and gamma-ray
- Inelastically scattered neutrons typically lose large fractions of their initial energy
- Secondary radiation is produced as the target nucleus returns to its ground state
- Total energy of outgoing neutron is much less than the energy of the incoming neutron
  - Part of original kinetic energy (more than threshold energy of target nucleus) is used to excite compound nucleus

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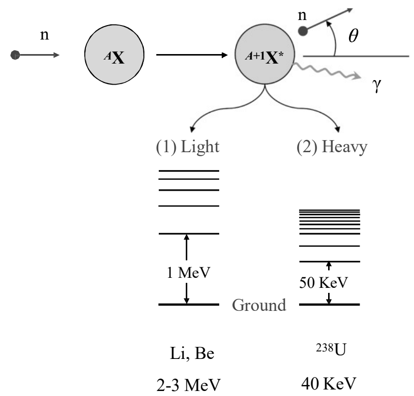
Inelastic Scattering



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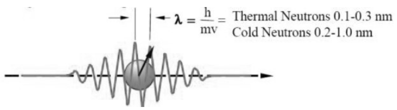
Diffraction

- Quantum mechanics: neutron exhibits wave-like properties
- The wavelength is defined by the de Broglie relation
- Thermal/cold neutrons have wavelengths on the order of crystal lattice spacing
- Neutrons are therefore a natural complement to X-rays in condensed matter physics. They are sensitive to magnetic distributions, not charge distributions



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Neutron Interactions: Absorption

- Radiative Capture (n,γ)
  - After the target nucleus captures a neutron, the formed nucleus is usually left in an excited state
  - The excitation energy is released by gamma ray emission (capture gamma rays): exothermic reaction
- Fission (n,f)
  - Neutron is absorbed in a nucleus which consequently splits into two part
  - Principal source of energy in nuclear reactors
- Charged Particles Production (n,α), (n,p)
  - Neutron is absorbed and a new particle can be ejected from the highly excited nucleus: endothermic or exothermic reaction
- Neutron Production Reactions (n,2n), (n,3n)
  - Endothermic reactions extracting neutrons from a nucleus

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Radiative Capture

- Radiative capture reactions involve absorption of neutrons followed by emission of gamma rays
  - Different elements release different characteristic radiation, which determines chemical composition of material and is very useful in many fields related to mineral exploration and security
- This reaction is most likely with thermal neutrons
- Very important in radiation protection and reactor physics: certain nuclides have very large capture cross sections (resonances) at low energies
- Therefore, neutron shielding usually includes a material to slow down neutrons and a material to then absorb the slow neutrons
- Important capture nuclides include Boron, Cadmium and Gadolinium

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Radiative Capture

- Compound nucleus is formed by absorption of the interacting neutron, which leads to one of the possible reactions
- The compound nucleus formation is the typical mode of neutron interaction except for potential scattering
- Compound nucleus formation
  - Excitation energy 6 – 10 MeV (close to binding energy)
  - 10^{-16} s of decay time
  - Prompt gamma-ray emission in $10^{-9} – 10^{-12}$ s until ground state is reached
  - Delayed radiation if ground state is not stable (α, β, …)

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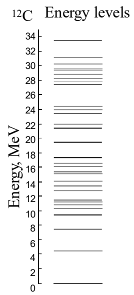
Compound Nucleus Formation

To explain nuclear reactions, Niels Bohr proposed in 1936 a two-stage model comprising the formation of a relatively long-lived intermediate nucleus and its subsequent decay.

$$^{56}_{26}\mathrm{Fe} + \mathrm{n} \rightarrow \left(^{57}_{26}\mathrm{Fe}\right)^{*} \rightarrow \begin{cases} ^{56}_{26}\mathrm{Fe} + \mathrm{n} & \text{(elastic scattering)} \\ ^{56}_{26}\mathrm{Fe} + \mathrm{n}' & \text{(inelastic scattering)} \\ ^{56}_{26}\mathrm{Fe} + \gamma & \text{(radiative capture)} \\ ^{56}_{26}\mathrm{Fe} + 2\mathrm{n} & \text{(n,2n) reaction} \end{cases}$$



Cross-sections of nuclear reactions exhibit maxima at certain incident neutron energies. The maxima are called resonances.

Fission

- A heavy nucleus is split into two more more smaller nuclei
- Discovered by Hahn, Strassmann, Frisch and Meitner in 1939
- Because the nuclear binding energies of these smaller nuclei (fission products with Z around 100) are larger than the binding energies of heavy nuclei, nuclear fission is associated with large releases of energy
- Many heavy nuclei are fissionable but Uranium, Plutonium and Thorium are the most important fissile nuclides in the nuclear fuel cycle
- By-products of fission include neutrons, photons and other radiation types, which leads to the concepts of neutron multiplication and chain reaction
- First sustained chain reaction in 1942 in Chicago (CP-1) led by Fermi

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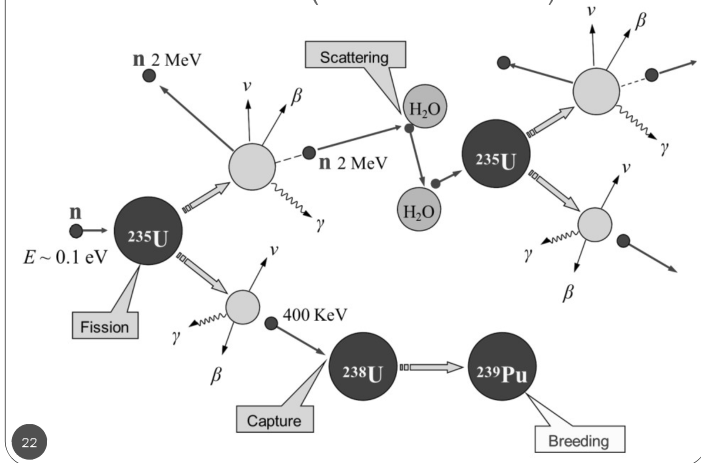
Fission Reaction (Chain Reaction)



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Charged Particles Production

- Other neutron absorption processes exist, such as proton or alpha particle emission
- As a rule, (n,p), (n,α) are endothermic and do not occur below some threshold energy
  - However there are some important exothermic reactions in light nuclei : ^{10}B(n,α)^{7}Li, ^{6}Li(n,α)^{3}H
- Boron is the most common element added to low Z materials in neutron shielding

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Neutron Cross Section

- Cross sections are used to quantitatively describe the probability of interactions between neutrons and matter (target nucleus)
- Cross section depends on neutron energy
- **Microscopic** cross section, σ is defined **per nucleus** for each type of interaction (unit: dimensions of area)
  - Probability of reaction between neutron and an individual particle or nucleus(i.e. ^{235}U)
  - Common unit for neutron cross section is the barn (10^{-24} cm^2)
- **Macroscopic** cross section, Σ is probability **per unit path length** that a particular type of interaction will occur (unit: cm^{-1})
  - Probability of interaction between neutron and some bulk material(i.e. concrete)
  - Σ = Nσ (N=number of nuclei per unit volume)

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Neutron Cross Section

- Each of neutron interactions is characterized by its cross section (Xsec)
  - σ_s (Σ_s): microscopic(macroscopic) scattering Xsec
    - σ_e (Σ_e): microscopic(macroscopic) elastic scattering Xsec
    - σ_i (Σ_i): microscopic(macroscopic) inelastic scattering Xsec
  - σ_a (Σ_a): microscopic(macroscopic) absorption Xsec
    - σ_γ (Σ_γ): microscopic(macroscopic) radiative capture cross section
    - σ_f (Σ_f): microscopic(macroscopic) fission cross section
- Total Xsec σ_t is sum of cross sections of all possible interactions
  - $\sigma_t = \sigma_s + \sigma_a = (\sigma_e + \sigma_i) + (\sigma_\gamma + \sigma_f + \sigma_p + \sigma_\alpha + \ldots)$

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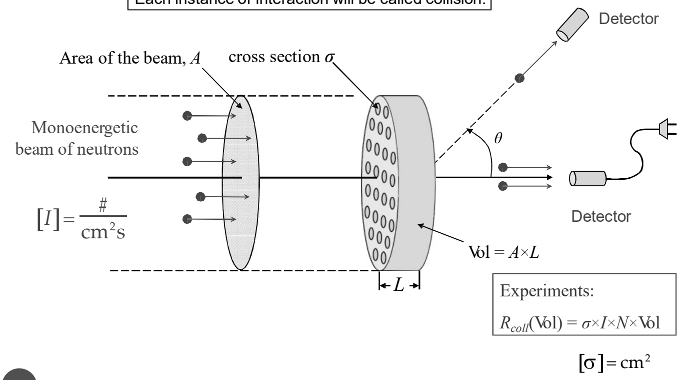
Definition of Cross-Section

Each instance of interaction will be called collision.



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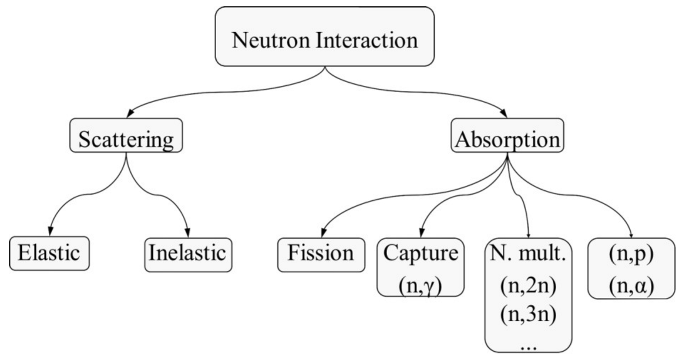
Types of Neutron Interaction



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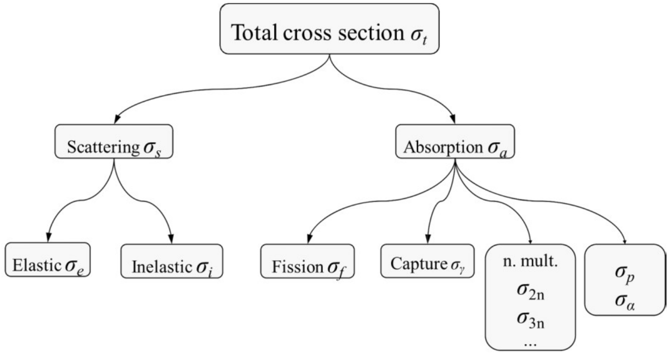
Neutron Interaction Cross-sections



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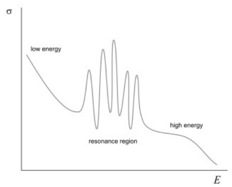
Neutron Cross Section

- Cross section shape
  - Low energy region
    - Absorption cross section for most nuclei in the low energy region varies as a function of $\frac{1}{\sqrt{E}} \sim \frac{1}{v}$
    - "1/v region"
  - Resonance region
    - "Resonance peaks" for neutrons of certain energies closely matches the discrete, quantum energy level of the compound nucleus
    - The resonance happens when the binding energy plus the kinetic energy of the neutron are exactly equal to the amount required to raise a compound nucleus from its ground state to a quantum level
  - High energy region



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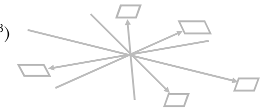
Neutron Cross Section – Neutron Flux

- Flux is the total number of neutrons passing through a surface (perpendicular to each neutron direction) per area per time
- Neutron flux $\Phi = nv$
  - n: neutron density (number of neutrons per cm^3)
    v: speed of neutrons (cm per s)
    Φ: neutron flux (neutrons per cm^2 per s)
- Reaction rate $F = \Sigma_t \Phi = N\Phi\, \sigma_{total}$
- Neutron intensity I: vector quantity, number of neutrons crossing through some arbitrary cross-sectional unit area in a single direction per unit time
  Neutron flux Φ: scalar quantity, number of neutrons crossing through some arbitrary cross-sectional unit area in all direction per unit time



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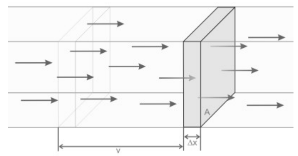
Calculation of Reaction Rate

- Monoenergetic beam of neutrons that impinges on a thin target of thickness Δx and area A (one layer of nuclei)
- Intensity of the beam (number of neutrons per cm^2/sec): $I = nv$
  - n: neutron density (number of neutrons per cm^3), v: velocity of neutrons)
- Number of collision per second in the target = $\sigma INA\Delta x$
  - N: number density of the target, A: irradiated area, σ: microscopic cross-section



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Calculation of Reaction Rate

- Number of collision per second in the target = $\sigma I N A \Delta x$
  - $NA\Delta x$: total number of nuclei in the target
    IA: number of neutrons per second which reach the target
- Relative probability of interaction of arbitrary neutron in the beam
  - $\frac{\sigma I N A \Delta x}{IA} = \left(\frac{\sigma}{A}\right)(NA\Delta x)$
- Number of collision per target volume
  - $F = \frac{\sigma I N A \Delta x}{A \Delta x} = \sigma I N$ (Collision density)
- Collision density is generally called reaction rate

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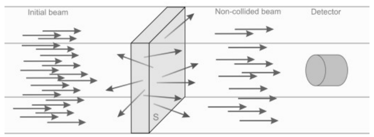
Neutron Cross Section: Neutron Attenuation

- A neutron beam with intensity I_0 that collides with a target of thickness X
- $I(x)$ is the intensity of a beam of neutrons, which did not collide while travelling a distance x
- While covering a distance dx the intensity will decrease, because of collision in that path



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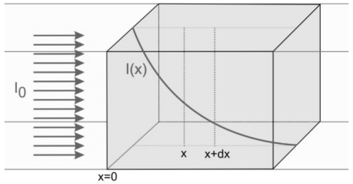
Neutron Cross Section: Neutron Attenuation



- $-dI(x) = \mu I(x)\,dx$
  $I(x) = I(0)e^{-\mu x}$
  - μ: linear attenuation coefficient
  - Intensity of the beam that has not collided decreases exponentially with distance inside target
- The linear attenuation coefficient varies with the density of the absorber, even with the same absorber material
- Thus, mass attenuation coefficient μ/ρ is much more widely used
  - ρ: density of the medium

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Attenuation of Neutrons

Number of neutrons survived traveling distance x.

$$-\frac{dI(x)/I(x)}{dx} = \Sigma_t \qquad I(x) = I(0)e^{-\Sigma_t x} \qquad \Pr\{\text{No collision}\} = \frac{I(x)}{I(0)} = e^{-\Sigma_t x}$$

$-dI(x)$ Number of neutrons that survived penetrating the distance x and made their first collision within dx.

$-\frac{dI(x)}{I(x)}$ = Probability of the first collision within dx = $\Sigma_t dx$

Σ_t = Probability of some interaction per unit path length

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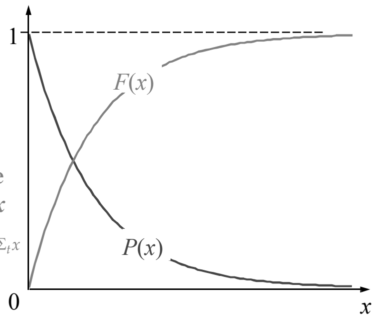
Probability of Uncollided Flight

Probability of uncollided flight within distance *x*

$P(x) = e^{-\Sigma_t x}$

Probability of at least one collision within distance *x*

$F(x) = 1 - P(x) = 1 - e^{-\Sigma_t x}$



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Neutron Cross Section: Mean Free Path

- Mean Free Path λ is the average path length in material between two collisions
- Intensity of neutrons $I(x) = I(0)e^{-\Sigma_t x}$
- Σ_t dx: probability that an uncollided neutron will interact in next dx
- $e^{-\Sigma_t x}$: probability that neutron will survive up to x without collision
- $p(x)dx = e^{-\Sigma_t x} \times \Sigma_t dx$: probability function that a neutron will have its first collision at distance dx in neighborhood of x
- Mean free path $\lambda = \int_0^\infty xp(x)dx = \Sigma_t \int_0^\infty xe^{-\Sigma_t x}dx = \frac{1}{\Sigma_t}$
- λ is one the order of 1 cm or less for slow neutrons while it may be tens of centimeters for fast neutrons

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Application of Neutrons

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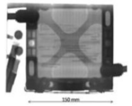
Applications of Neutron Science

Charge neutral

Deeply penetraTng

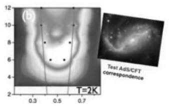


Li moTon in fuel cells

Help build electric cars

S=1/2 spin

Directly probe magneTsm

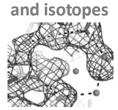


Solve the puzzle of High-Tc superconducTvity

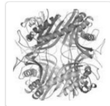
Efficient high speed trains

Nuclear sca_ering

SensiTve to light elements and isotopes



AcTve sites in proteins

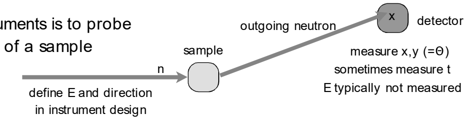


Be_er drugs

Probing length scales and dynamics

Neutrons as a Probe

- The purpose of the instruments is to probe with neutrons some aspect of a sample



- Very generically, this can be divided into elastic and inelastic categories
  - elastic: gives information on where atoms are
  - inelastic: gives information on what atoms do (ie move)
- This is measuring the cross sections:

elastic

$$\frac{d\sigma}{d\Omega}(\lambda, 2\theta, \psi)$$

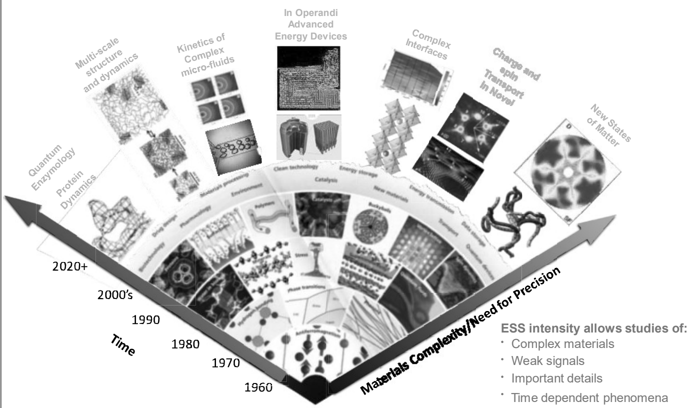
- cross section / scattering probability into a solid angle, as a function of wavelength, scattering angle and aximuthal angle

inelastic

$$\frac{d^2\sigma}{d\Omega dE}(\lambda_{in}, \lambda_{sc}, 2\theta, \psi)$$

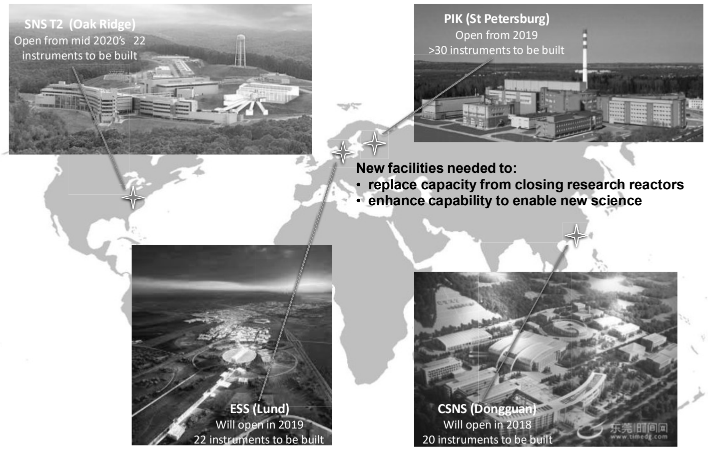
- double differential cross section / scattering probability into a solid angle, as a function of wavelength, scattered wavelength scattering angle and aximuthal angle

Application of Neutron Science



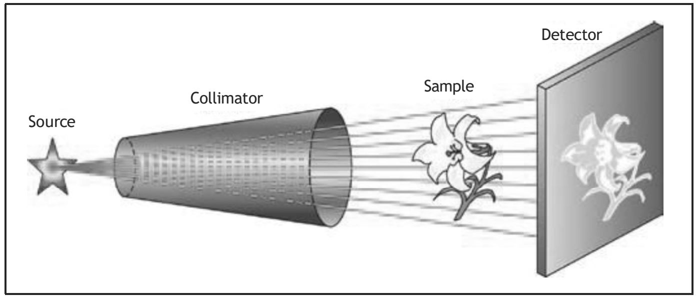
ESS intensity allows studies of:
- Complex materials
- Weak signals
- Important details
- Time dependent phenomena

Major Neutron Research Facilities

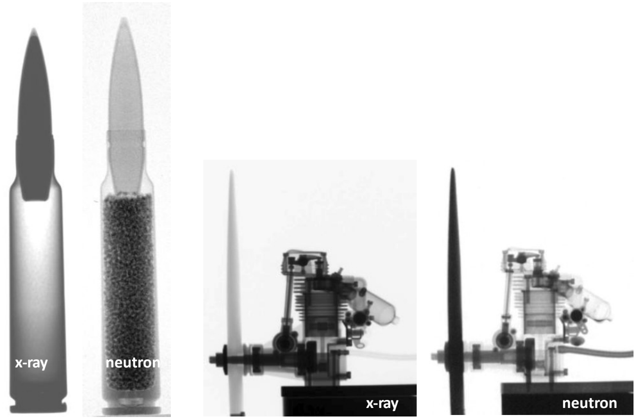


New facilities needed to:
- **replace capacity from closing research reactors**
- **enhance capability to enable new science**

Neutron Imaging



Neutron Radiography



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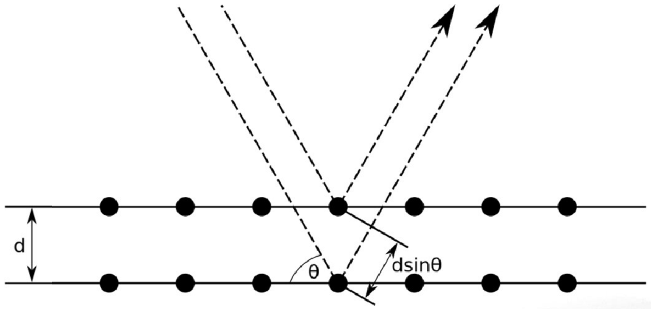
Diffraction

EUROPEAN SPALLATION SOURCE

Sizes probed = "atomic structures" = 0.1 nm - 10 nm



Position and intensity of diffraction peaks gives atomic positions

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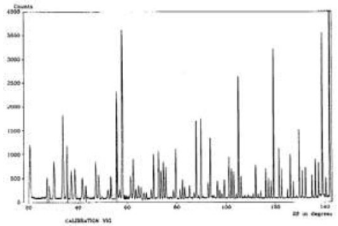
Purpose of the Instrument

EUROPEAN SPALLATION SOURCE

Basically, in some form, you want to measure Br agg's equation

$$n\lambda = 2d \sin\theta$$



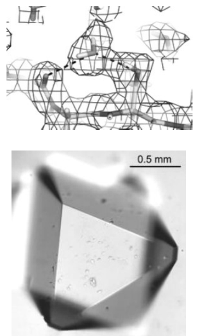
Define the neutron wavelength with your instrument design

Detectors allow you to measure theta

It means that you can calculate "d"

EUROPEAN SPALLATION SOURCE

Macromolecular Crystallography with neutrons

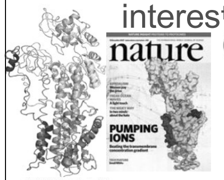


Oksanen, E *et al. J. R. Soc. Interface* **2009**, *6 Suppl 5*, S599-610.

☺ Hydrogens are visible
☺ No radiation damage
☹ Large crystals needed
☹ Data collection tak
es weeks
☹ Few instruments availabl
e

Esko Oksanen, European Spallation Source

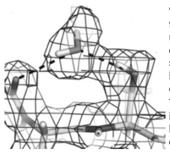
The community is interested



"Structural Biology / crystallography is one of the research areas where Europe is a global leader, and with the investment in ESS, Europe and the Danish researchers will get access to the world's best and most advanced research facilities in the area. The possibility of retaining the leadership position in structural biology and crystallography is present, and thus also the potential to leverage its position to promote bio-based innovation." The Danish Association of the Pharmaceutical Industry (Lif)Contribution to the Danish INNO+ policy

Hartmut Michel - Facts

"There is no alternative to neutron crystallography in order to uniquely identify the location of protons, which is of particular importance when dealing with proton translocating proteins" H. Michel, MPI of Biophysics



"-One of our first questions using the ESS MX beamline will be to understand the protonation states of the reactive site aspartate acids, which will help us understand the mechanism and so lead to novel drug molecules. This, given the size of the protein, is impossible with any current technology." A. Goldman, University of Leeds

Venkatraman Ramakrishnan - Facts

"Hydrogens represent nearly half of the atoms in biomolecules. Hydrogen bonding and proton transfer play a critical role in biological structure and in catalytic mechanisms." V. Ramakrishnan, University of Cambridge

Esko Oksanen, European Spallation Source

Neutron Sources

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Neutron Source

- Common sources of neutrons
  - Nuclear reactors
  - Nuclear fusion sources (D-T generators)
  - Accelerator-based sources (spallation)
  - Radioactive decay (^{252}Cf)
  - (α,n), (γ,n) sources
- Also produced from other radiation types through secondary nuclear reactions
- In radiation protection, neutron shielding is also not straightforward: one needs to use a material with a high probability of absorbing neutrons (concrete, paraffin, borated water or borated polyethylene)

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Classification of Neutron Sources

- Radioisotope neutron sources (small, portable, reliable, low-cost, no maintenance)
  - Fission source based on ^{252}Cf (overwhelming favourite)
  - (α,n) sources
  - (γ,n) sources
- Nuclear reactors
- Accelerator-based neutron sources
  - proton and deuterium bombardment
  - electron bombardment and photo-nuclear reactions
  - Spallation neutron sources

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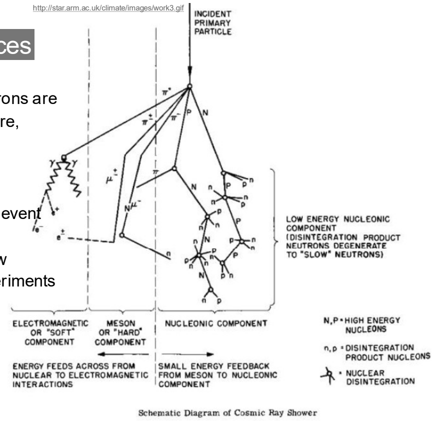
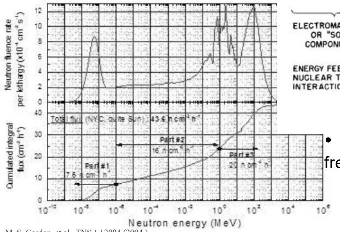
Natural Sources

- Natural sources of unbound neutrons are spallation processes in the atmosphere, fusion in stars and natural fission
- Example: cosmic neutrons in the atmosphere
- Of interest for as can cause single event upsets in chips
- Neutrons may be signature for new physics in various underground experiments

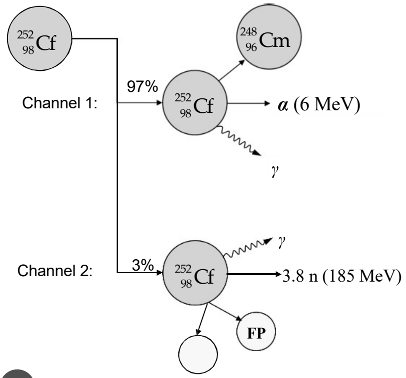




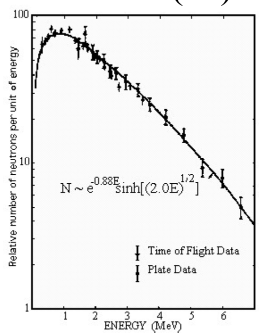
M. S. Gordon, et al., TNS 1 12004 (2004.)

- Whilst of interest in themselves, none of them are frequent enough to be used a probe

Californium Source



$$N \sim e^{-0.88E} \sinh\left(\sqrt{2E}\right)$$



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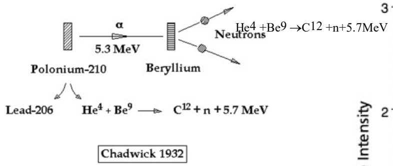
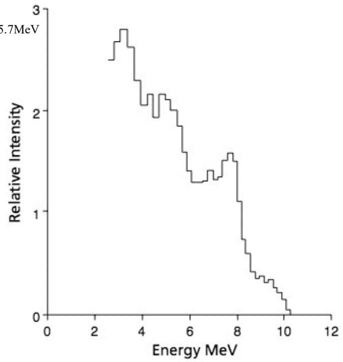
Radioactive Sources

- Neutron was discovered by the (alpha, n) reaction, where in some lighter elements the last neutron is weakly bound, and released when alpha particle is incident





- Americium-241 commonly used
- typical number 6. 10^7 n/s for 1 TBq
- Details of neutron production depend upon geometry of sources
- ca. 40% of neutrons below 1 MeV

- Some isotopes undergo spontaneous fission
- eg Cf-252, 3.1% of decays, average of 3.7 neutrons per fission.
- Cf-252 not naturally occurring
- Abstract N33-8 this week

Radioisotope Sources

Source	Half-life	Reaction	Neutron yield ($n \times s^{-1} \times g^{-1}$)	Neutron energy (MeV)
^{124}Sb-Be stibium-antimony	60.9 d	(γ,n)	2.7×10^{9}	0.024
^{140}La-Be	40.2 h	(γ,n)	10^{7}	2.0
^{210}Po-Be	138 d	(α,n)	1.28×10^{10}	4.3
^{241}Am-Be	458 y	(α,n)	1.0×10^{7}	~4
^{226}Ra-Be	1620 y	(α,n)	1.3×10^{7}	~4
^{227}Ac-Be	21.8 y	(α,n)	1.1×10^{9}	~4
^{239}Pu-Be	24400 y	(α,n)	10^{9}	~4
^{228}Th-Be	1.91 y	(α,n)	1.7×10^{10}	~4
^{252}Cf	2.65 y	fission	2.3×10^{12}	2.3

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Fission, Fusion, Spallation

Nuclear Reaction	Energy (MeV)	Number of neutrons per particle or event	Heat deposition (MeV/n)
T (d,n)	0.2	8×10^{-5} n/d	2500
W (e,n)	35	1.7×10^{-2} n/e	2000
Be (d,n)	15	1.2×10^{-2} n/d	1200
Fission ^{235}U (n,f)	2.2	2.5 n/fission	80
Fusion (T,d)	~1	1 n/fusion	17
Pb spallation	~ GeV	20 n/p	23
^{235}U spallation	~ GeV	40 n/p	50

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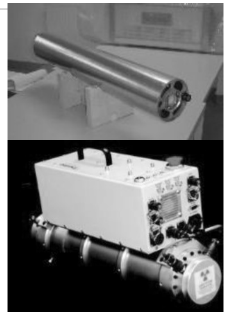
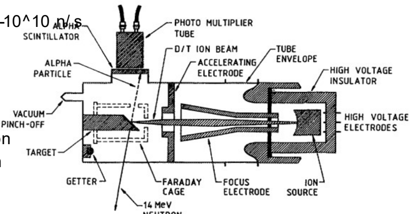
Neutron Generator

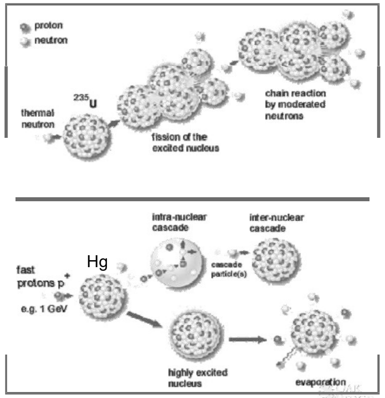


- Use Deuterium-Deuterium or Deuterium-Tritium fusion
- Small accelerator arrangement of few 100 keV
- DD:
  - 50%: $_1D^2 + {}_1D^2 \rightarrow {}_2He^3$ (0.82 MeV) + $_0n^1$ (2.45 MeV)
  - 50%: $_1D^2 + {}_1D^2 \rightarrow {}_1T^3$ (1.01 MeV)+ $_1p^1$(3.02 MeV)
- DD:
  - $_1D^2 + {}_1T^3 \rightarrow {}_2He^4$ (3.5 MeV) + $_0n^1$ (14.1 MeV)
- Many generators available commercially
- eg SODERN, NSD-GRADEL fusion, Thermoscientific
- fluxes typically in the range 10^6 -10^10 n/s
- Abstract N62-7 this week
- Fusion Facilities may not be neutron facilities, but they are facilities which produce lots of neutrons



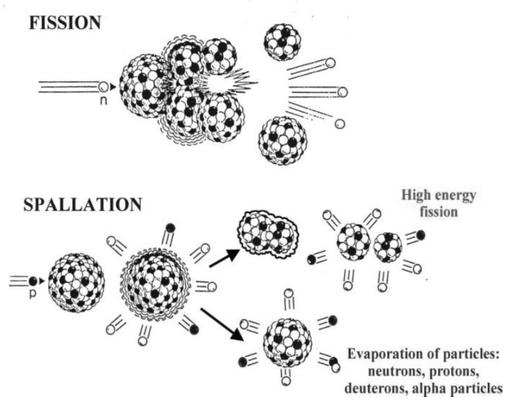
Fission and Spallation

- Processes very different
- Fission results in light and heavy debris
- Spallation results in debris close to that of target
- Neutron yield:
  - Fission: 2.5n / fission. 1 needed to substain criticality
  - Spallation: very energy dependent. Typically ca. 10.
- Heat:
  - Fission: ca. 160 MeV/ neutron
  - Spallation: ca. 25 MeV/ neutron



Phil Ferguson

Fission and Spallation



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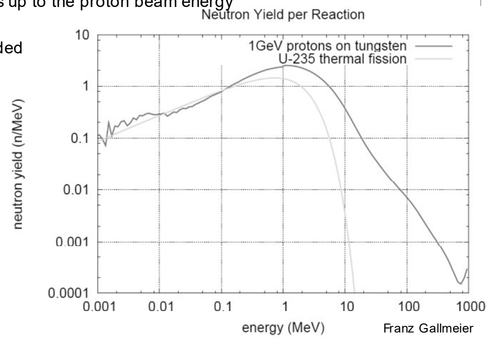
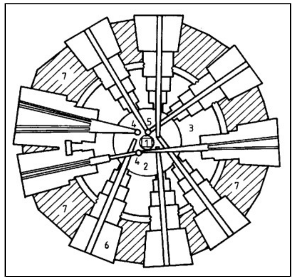
Fission and Spallation Source Spectrum

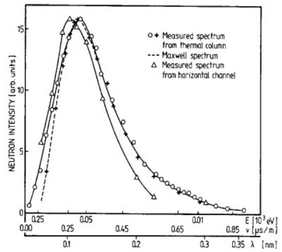
- Energy spectrum very different
- Spallation yields neutrons up to the proton beam energy
- Significant shielding needed



Nuclear Reactors: HFL (High Flux Reactor in Grenoble)



1. Core. 2 Heavy water reflector. 3 Light water pool. 4 Cold source. 5 Hot source. 6 Horizontal channel. 7 Concrete shield.



Thermal neutron spectrum of HFR

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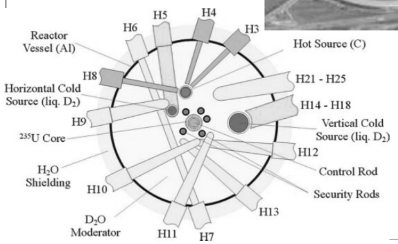
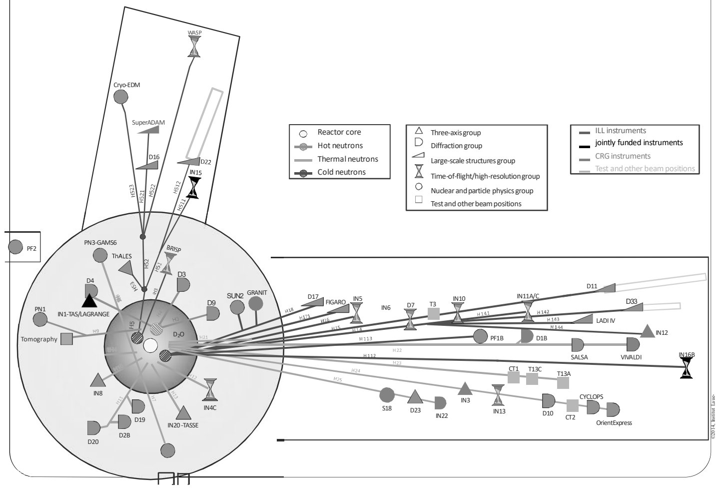
Institut Laue-Langevin, Grenoble

- World's leading research reactor
- Came into operation in 1971
- 58 MW thermal power
- Most intense continuous neutron flux in the moderator region:
- 1.5 . 10^15 n/ cm^2/ s
- ca. 600 papers/ year



https://www.ill.eu/



Spallation Sources

https://neutrons.ornl.gov/sns

SNS (Oak Ridge, USA)

https://www.j-parc.jp/index-e.html

J-PARC (Tokai Japan)

Instantaneous power on target (e.g. 1 MW at 60 Hz, i.e. 17 kj in ~1 μs pulses on target): **17 x**
→ **Pressure wave: 300 bar**

Reaches limits of technology

Charged Particle Bombardment

These neutron sources are based on electro static accelerators or cyclotrons.

$^{7}Li(p,n)^{7}Be$
$^{3}H(d,n)^{3}He$
$^{2}H(d,n)^{3}He$
$^{3}H(d,n)^{4}He$
$^{1}H(t,n)^{3}He$
$^{1}H(^{7}Li,n)^{7}Be$

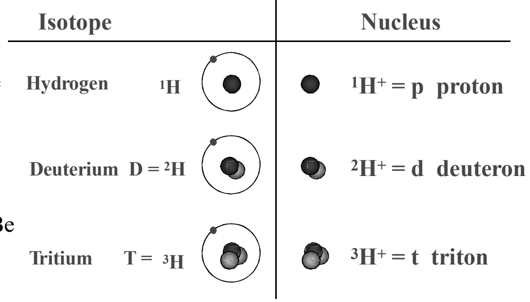


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Photo Neutron Sources

$$^{2}_{1}H + \gamma \rightarrow ^{1}_{1}H + ^{1}_{0}n \qquad Q = -2.226\ \text{MeV}$$
$$^{9}_{4}Be + \gamma \rightarrow ^{8}_{4}Be + ^{1}_{0}n \qquad Q = -1.666\ \text{MeV}$$

All other target particles have much higher binding energy

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Comparison of Neutron Sources

Fast neutrons produced / joule heat deposited:

Fission reactors:	~ 10^9	(in ~ 50 liter volume)
→ Spallation:	~ 10^{10}	(in ~ 2 liter volume)
Fusion:	~ 1.5×10^{10}	(in ~ 2 liter volume)
	(but neutron slowing down efficiency reduced by ~20 times)	
Photo neutrons:	~ 10^9	(in ~ 0.01 liter volume)
→ Nuclear reaction (p, Be):	~ 10^8	(in ~ 0.001 liter volume)
Laser induced fusion:	~ 10^4	(in ~ 10^{-9} liter volume)

Spallation: most favorable for the foreseeable future (neutrons/€)
Compact source: lowest cost / facility

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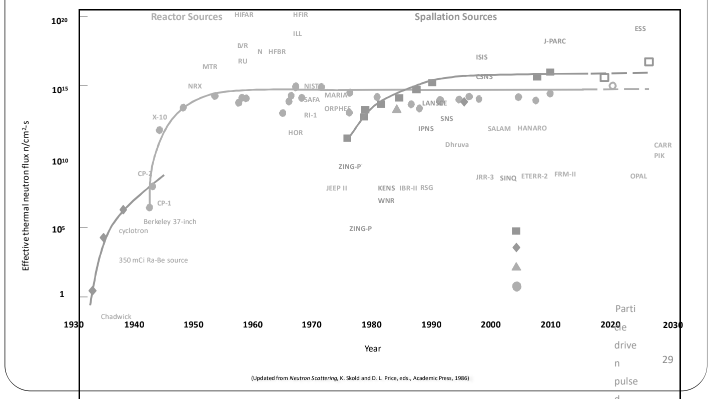
Neutron facilities – reactors and particle driven

EUROPEAN SPALLATION SOURCE



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Neutron Detection

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Neutron Detection

- Conversion of incident neutron into secondary charged particles
- Direct detection of **charged particle**
- Relative probabilities of different interaction change with E_n
  - slow neutrons ($E_n < 0.5$ eV)
    - neutron-induced reactions creating secondary radiation (γ, α, p, fission product) with sufficient energy
    - elastic scattering is not favorable since little energy is given to the nucles to be detected
  - fast neutron
    - scattering probability becomes greater and large energy is transferred in one collision

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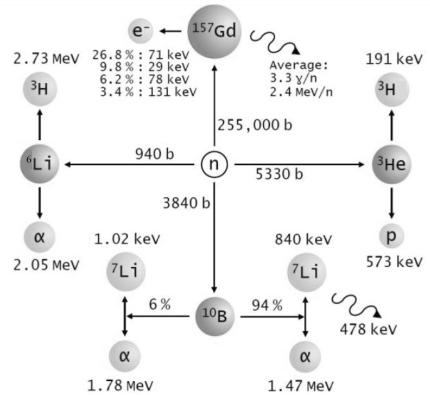
Neutron Interactions



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Key Mechanisms for Neutron Detection

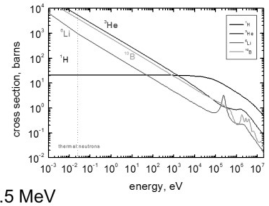
*** Thermal neutrons**

$^{6}Li + {}^{1}n \rightarrow \alpha + {}^{3}H + 4.8$ MeV

$^{3}He + {}^{1}n \rightarrow {}^{1}p + {}^{3}H + 0.77$ MeV

$^{10}B + {}^{1}n \rightarrow \alpha + {}^{7}Li + 2.3$ MeV

$^{157}Gd + {}^{1}n \rightarrow {}^{158}Gd^{*} \rightarrow {}^{158}Gd^{*} + \gamma's + e's + 8.5$ MeV

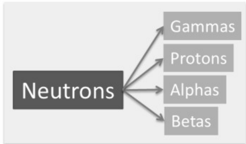


*** Fast neutrons**

$^{1}H + {}^{1}n \rightarrow {}^{1}p + {}^{1}n'$

$^{35}Cl + {}^{1}n \rightarrow {}^{35}S + {}^{1}p + 0.617$ MeV

$^{23}Na + {}^{1}n \rightarrow {}^{23}Ne + {}^{1}p + ?$ MeV



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Neutron Detection by Materials

- Thermal Neutron – Neutron Counting
  - Boron Reaction Based:
    - BF_3 proportional tube
    - B-lined proportional tube
    - B loaded Scintillators
    - B Converters
  - Li-Based: more frequently used in fast neutron detection
    - LiI(Eu) Scintillator
    - Li-loaded Scintillators
    - Li-glass scintillator
    - Li-layered semiconductor detector
  - He-3
  - Silicon Carbide Detectors (4H-SiC)

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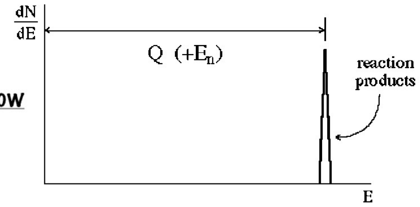
Slow Neutron Detection

Need <u>exoenergetic</u> (positive Q) reactions to provide energetic reaction products



Useful Reactions in <u>Slow</u> Neutron Detection

$^{10}B\,(n,\alpha)\,{}^{7}Li$

$^{6}Li\,(n,\alpha)\,{}^{3}H$

$^{3}He\,(n,p)\,{}^{3}H$

$(n, fission)$

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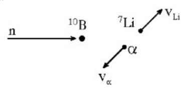
Boron Reaction Based Detectors

The ^{10}B (n,α) Reaction

$$^{10}B + n \rightarrow \begin{cases} {}^{7}Li + \alpha & \quad 2.792 \\ {}^{7}Li^{*} + \alpha \quad 94\% & \quad 2.310 \end{cases} \qquad Q\ (MeV)$$

19.8% abundant



$$\left[{}^{10}B\ (n,\alpha)\ {}^{7}Li^{*}\right]$$

- Conservation of energy:

$$E_{Li} + E_{\alpha} = Q = 2.31\ MeV$$

- Conservation of momentum:

$$m_{Li}v_{Li} = m_{\alpha}v_{\alpha}$$
$$\sqrt{2m_{Li}E_{Li}} = \sqrt{2m_{\alpha}E_{\alpha}}$$
$$E_{\alpha} = \frac{m_{Li}}{m_{\alpha}}E_{Li}$$
$$\Downarrow$$
$$E_{Li} = 0.84\ MeV \qquad E_{\alpha} = 1.47\ MeV$$

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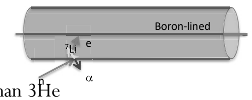
Boron Reaction Based Detectors



- Neutrons captured by ^{10}B yields α + ^{7}Li
- BF_3
  - Equivalent or better gamma discrimination than 3He
  - Cross-section ~70% that of ^{3}He
  - Operates at low pressure (~1 atmosphere) for reasonable HV
  - Requires multiple tubes for ^{3}He replacement
  - BF3 is corrosive (hazardous gas): shipping regulations
- Boron-lined proportional tubes
  - Thin layer on tube wall to collect reaction products in proportional gas
  - Surface area limited, lower (<0.5) efficiency per tube than BF_3
  - Requires configuration with many tubes
  - Safe, operates at low pressure (~1 atmosphere)

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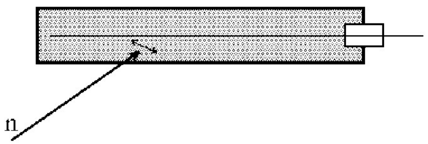
The BF_3 Tube



1. Typical BF_3 pressure < 1 atm
2. Typical HV: 2000 - 3000V
3. Usual ^{10}B enrichment of 96%

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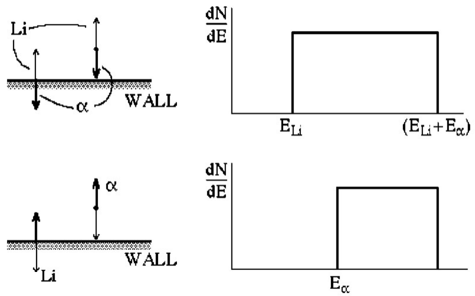
The “Wall” Effect (Gas Detectors)



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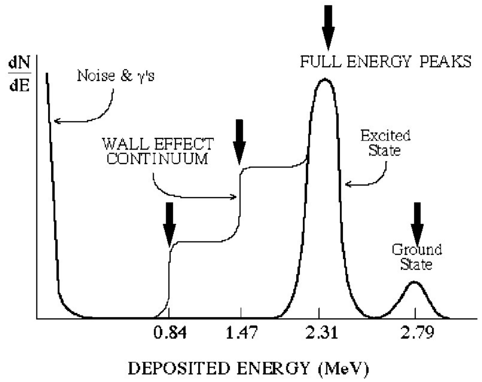
The BF_3 Tube: Pulse Height Spectrum



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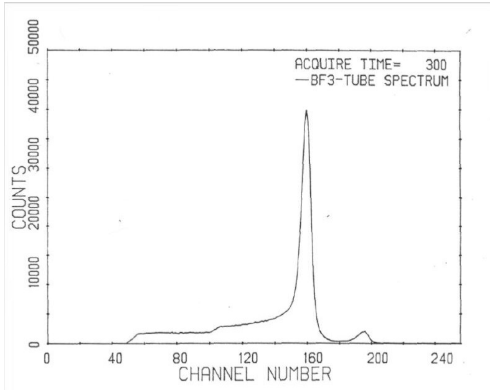
The BF_3 Tube: Measured Spectrum



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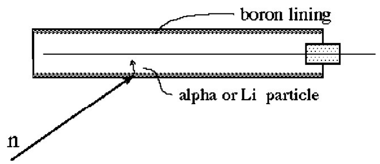
Boron-lined Proportional Tube



- Conventional proportional gas
  - BF3 gas dissociates in high gamma fluences
- Detection efficiency limited by boron thickness
- Used in high gamma backgrounds and temperatures
  - reactor instrumentation, spent fuel monitoring and assay

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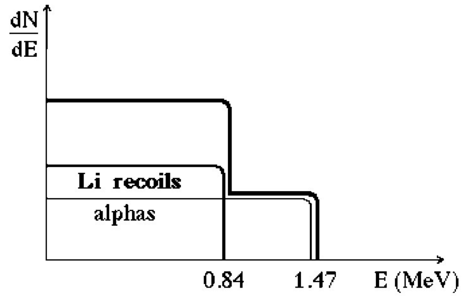
The Pulse Height Spectrum



No plateau on counting curve

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Other Reactions

Reaction	Q(MeV)
$^{6}Li + n \rightarrow {}^{3}H + \alpha$ (7.4% abundant)	4.78
$^{3}He + n \rightarrow {}^{3}H + p$ (trace)	0.765
$X(n, \text{fission})$	~ 200

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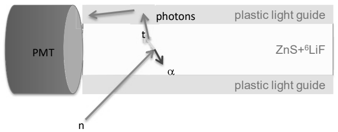
^{6}Li/ZnS Based Detectors

- Neutron capture by the ^{6}Li yields $\alpha + {}^3H$
- Glass fibers
  - ^{6}Li-enriched lithium silicate glass fibers doped with cerium which fluoresces (Bliss et al. 1995, PNNL)
  - Good efficiency (per unit surface area or neutron module)
  - Gamma-ray sensitive: discrimination with PSD
- Coated wavelength shifting paddles/fibers
  - ZnS scintillator material mixed with ^{6}Li coating
  - Good efficiency (per unit surface area or neutron module)
  - Coating gamma-ray sensitive: Good discrimination with PSD



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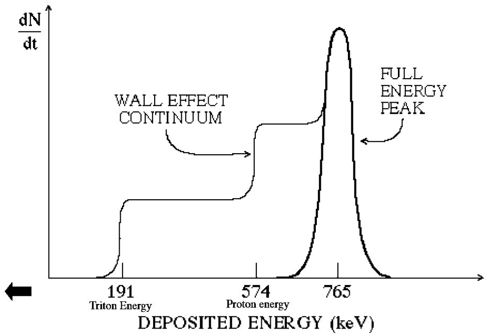
He-3 Pulse Height Spectrum



Wall effect more prominent due to range of particles with Z=1
He3 counters need shielding from gammas when gammas more than a few mR/hr

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Fission Chambers

- Neutron-induced Fission:
  - Fission energy possessed by fission fragments
  - Large Energy (~180 MeV): Good discrimination
  - Ionization Chamber with Frisch Grid – coated with fissile deposit
  - Pulse mode (non-reactor) and Current mode (reactor flux)
- Fissile deposition thickness affects the pulse height spectrum
  - Double-hump distribution → Distortion (Wall-effect-like)
- Range of energetic particles: ~ few cm (typical dimension)
- Alpha radioactivity: alpha induced pulse rates

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Comparison for Thermal N Detection

Reaction	Q (MeV)	Common Form	Adv	Dis adv
B-10(n, alpha)	2.31	BF3, boron lined proportional	Good gamma discrim.	Wall effect
Li-6 (n, alpha)	4.78	solid scint. LiI (Eu), glass	no wall	solid -> poorer gamma discrim.
He-3(n,p)	0.764	prop. gas	good prop. gas, high P	discr. low Q, wall effect
Fissile(n,f)	~200	lined ion chamber	good dscrim.	SNM

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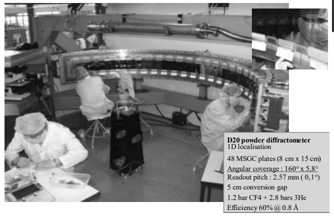
10/15/2020

Neutron Detectors in a large system

- Helium-3 Tubes most common
- Typically 3-20 bar Helium-3
- 8mm-50mm diameter common
- Using a resistive wire, position resolution along the wire of ca. 1% possible
- See abstract N13-2 this week

Curved 1D MSGC for the D20 Powder Diffractometer (2000)



- First micro pattern gaseous detectors was MSGC invented by A Oed at the ILL
- A. Oed, NIM A 471 (1988) 351
- Rate and resolution advantages
- Helium-3 MSGCs in operation

Neutron Detection by Materials

- Fast Neutrons – Counting and Spectroscopy
  - Counters based on neutron moderation: REM ball counter
  - Detectors based on fast neutron-induced reactions
    - Measure (Q + neutron energy deposited)

      $^{6}Li + {}^{1}n \rightarrow \alpha + {}^{3}H + 4.8$ MeV

      $^{3}He + {}^{1}n \rightarrow {}^{1}p + {}^{3}H + 0.77$ MeV

      $^{10}B + {}^{1}n \rightarrow \alpha + {}^{7}Li + 2.3$ MeV
  - Detectors utilizing fast neutron scattering
    - Measure recoil energy of nucleus (proton, deuteron, helium, carbon, …)
      - Proton Recoil Scintillator
      - Gas Recoil Proportional Tube
      - Proton Recoil Telescope
      - Capture-gated spectrometer

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Fast Neutron Detection and Spectroscopy

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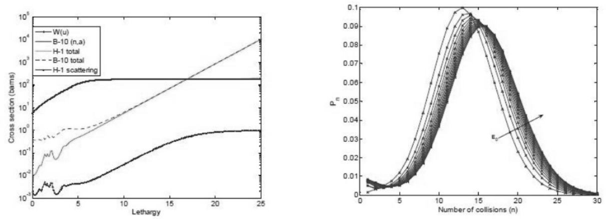
Different approaches to fast neutron detection

- counters based on neutron moderation detectors
- based on fast neutron-based reactions detectors
- utilizing fast neutron scattering



Source: Pozzi, University of Michigan

19 Course 2.3

Moderated Neutron Detectors

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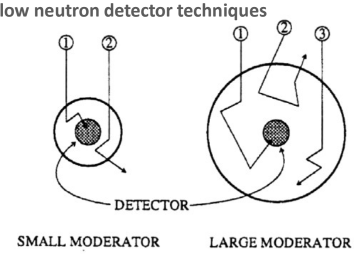
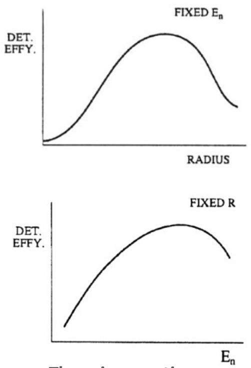
Slow down (moderate) neutrons until observable by slow neutron detector techniques

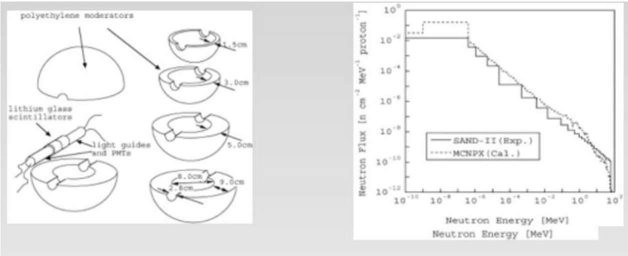




There is an optimum amount of moderation

20 Course 2.3

Fast n Detection Based on Moderation

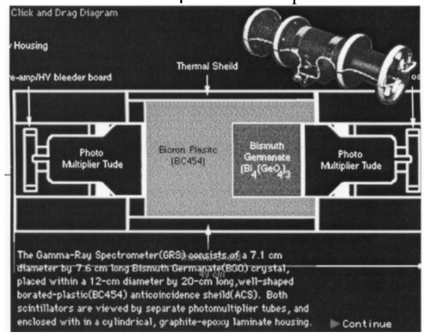


Development of multi-moderator neutron spectrometer using a pair of ^{6}Li and ^{7}Li glass scintillators, NIMA, 460(2-3), March 2001

92

Fast n Spectrometer – $^{10}B(n, \alpha)^7Li^*$

- Measure total E deposit in borated plastic scintillator
  $E_{neutron} = E_{deposit} - Q^*$
- Gate on coincident γ from Li* captured in BGO (γ-discrimination)



Lunar Prospector

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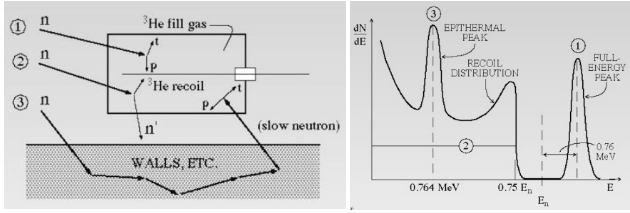
$^6Li(n, \alpha)^3H$ and $^3He(n, p)^3H$ Reactions

- Li Glass Scintillator: nonlinear and small light output, not for spectroscopy, useful for timing (~100 ns)
- LiI(Eu) Scintillator: nonlinear output limits resolution ~20%
- Li sandwich spectrometer: coincidence mode to reduce BG



^{3}He Proportional Counter

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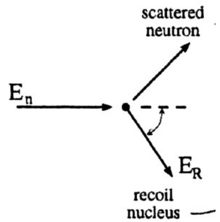
Fast Neutron Measurements Based on Elastic Scattering

Pacific Northwest National Laboratory

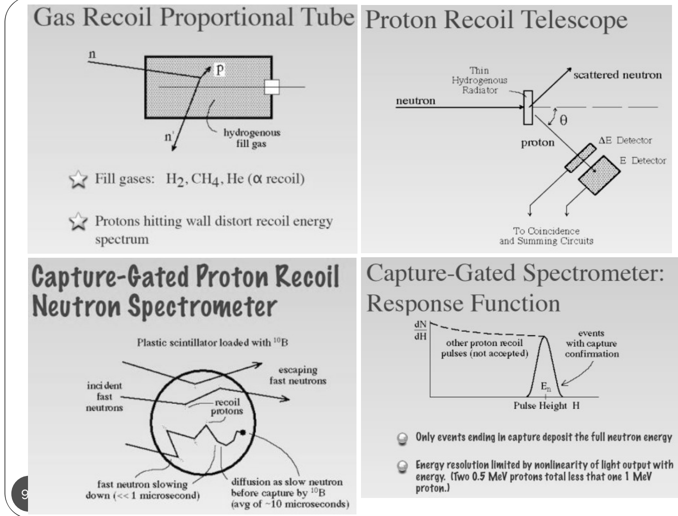
Neutron collision transfers part of its kinetic energy to nucleus of detector atom , forming a recoil nucleus that is now "visible" as an ionizing particle.

When the atom consists of hydrogen, this process forms a recoil proton. Then it is possible to transfer up to the full neutron energy in a single elastic scattering collision.

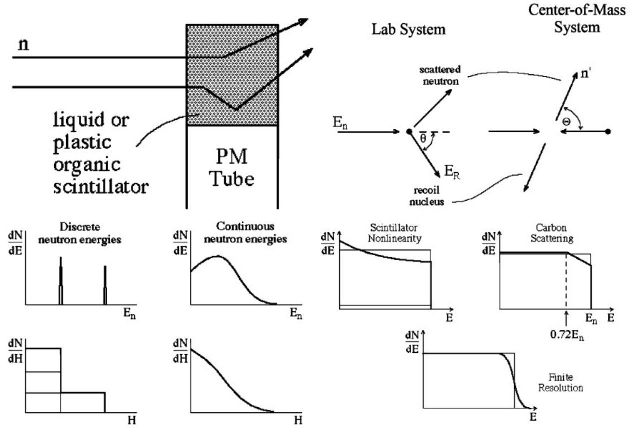


A	$E_{R,max}$
1 (H)	E_n
3 (^{3}He)	$0.75E_n$
12 (C)	$0.28E_n$

22 Course 2.3



Proton Recoil Scintillators



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Detectors Utilizing Fast Neutron Scattering

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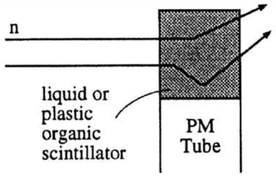
1. **Proton recoil scintillator**
   high (10 – 50%) detection efficiency, complex response function, gamma rejection by pulse shape discriminati on



Detectors Utilizing Fast Neutron Scattering

Pacific Northwest NATIONAL LABORATORY

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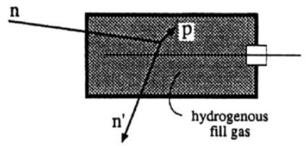
2. **Gas recoil proportional tube**
   low (.01 - .1%) detection efficiency, can be simpler response function, gamma rejection by amplitude



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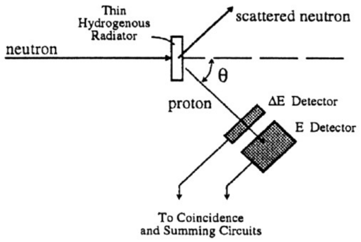
   very low (~ .001%) detection effi ciency, usable only in beam geo metry, simple peak response fun ction



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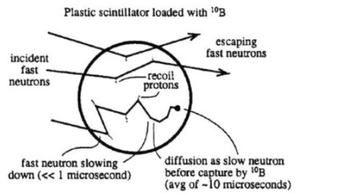
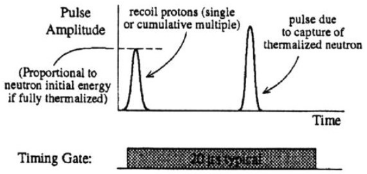
4. **Capture-gated spectrometer**
   modest (few %) detection efficiency, simple peak response function





Detectors Utilizing Fast Neutron Scattering

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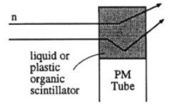

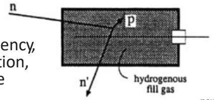

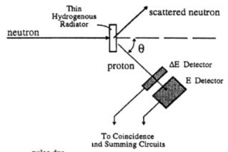

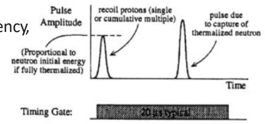

Multiplicity/Coincidence Counting

Considerations for multiplicity/coincidence counting

Sample mass: sets scale for event rate

Multiplication: when a neutron from spontaneous fission [or (α,n) reaction] induces another fission in the sample; geometry and mass are important

(α,n) rate: observed single neutron rate is a combination of detecting single fission neutron and detecting neutrons from (α,n) processes

Coincidence counting gives two out of three

- need to make assumptions about third parameter

Multiplicity counting enables determination of all three values

10/15/2020

Recent Developments in Neutron Detection

- Material Developments
  - Organic Scintillators
  - Inorganic Scintillators
  - Semiconductor Devices
- Detection Methods
  - Pulse Shape Discrimination
  - Position-sensing and Imaging
  - Spectrum Deconvolution
  - Coincidence Techniques

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Fundamentals of Neutron Detection

How Radiation Interacts with Matter

Gamma rays

- **Interactions with electrons (PE, CS, PP) -> produce fast electrons (<u>delta rays</u>)**
- **Hence a use of heavy elements**

Charged particles

- **Interactions with electrons – Coulomb**
- **Cherenkov radiation**

Neutrons (Thermal & Fast)

- **Interactions with nucleus**
- **Use of specific isotopes – produce gamma rays and <u>charged particles</u>**

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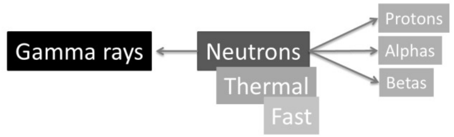
Fundamentals of Neutron Detection

What are we detecting?



<u>Detected **either** together **or** separately</u>

<u>In either case **we need to recognize what is what**!</u>

<u>(Charged) Particle Identification</u>

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Pulse Shape Discrimination (PSD)

<u>Methods (Examples)</u>

- Charge integration (~1959, Owen)
- Pole Zero type (~1959, Brooks)
- Zero-crossing (~1961)
- Gatti's Filter (1970)
- Etc.

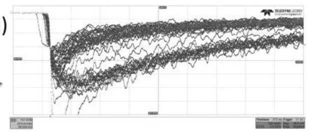
<u>Finding Parameters</u>

- Trial & Error
- Combinatorial
- Analytical

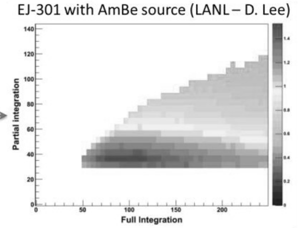
<u>Other considerations</u>

- Photodetector (Speed vs. QE)
- Digital vs. Analog (Power)
- Digital (Sampling, **Resolution**)

CLYC with Am/Be source (RMD)



EJ-301 with AmBe source (LANL – D. Lee)



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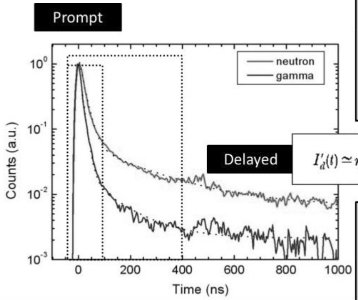
Pulse Shape Discrimination (PSD)

Scintillation kinetics



More delayed emission for particles that create higher density of excitation

$$I'_{d}(t) \simeq \eta_s \frac{N}{[1 + A \ln(1 + t/t_a)]^2 (1 + t/t_a)}$$

Through analysis of scintillation time profiles it is possible to identify particle's kind

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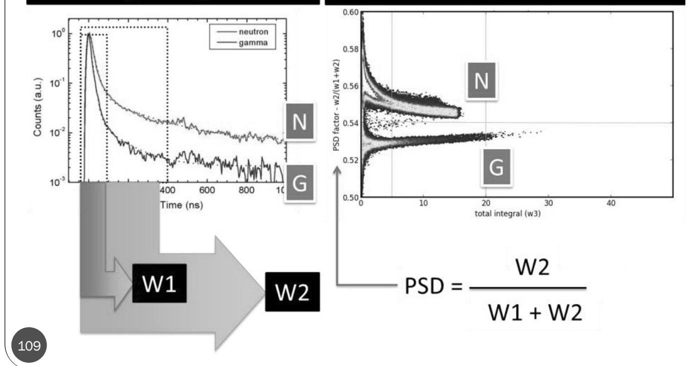
Pulse Shape Discrimination (PSD)

Achieving Particle Identification

Charge Integration | **PSD Plot**



$$PSD = \frac{W2}{W1 + W2}$$

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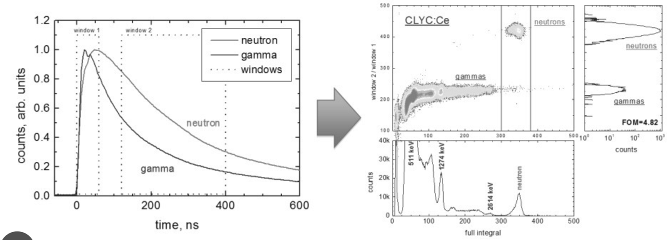
PSD-capable Scintillators

CLYC: Thermal Neutron + Gamma Rays

Cs_2LiYCl_6

- Discovered at Delft, *Combes* (1999)
- Further developed at RMD, 2003 - ????

$^{6}Li + ^{1}n \rightarrow \alpha + ^{3}H + 4.8\ MeV$



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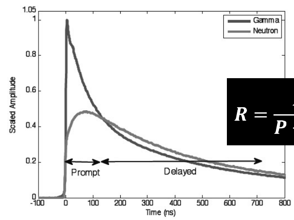
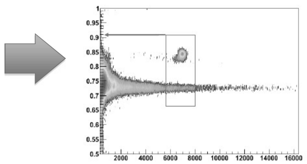
Pulse Shape Discrimination on CLYC

$^{35}Cl + ^{1}n \rightarrow ^{1}p + ^{35}S$ + energy
$^{6}Li + ^{1}n \rightarrow \alpha + ^{3}H$ + energy



$$R = \frac{D}{P + D}$$

In CLYC, relative lack of fast scintillation components for neutron-induced events



- **Form a tail-to-total ratio (PSD ratio) of pulse**
  - Delayed Integration (D) divided by sum of delayed(D) and prompt (P)
- **2D plot of PSD ratio vs. energy shows distinct regions for different particle types**
  - Gammas appear as a horizontal band with lower PSD ratio
  - Neutrons appear as a bubble with higher PSD ratio

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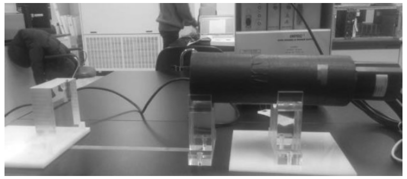
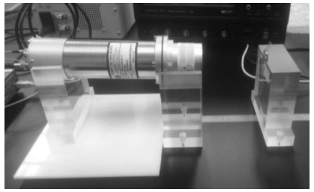
Pulse Shape Discrimination Experiment

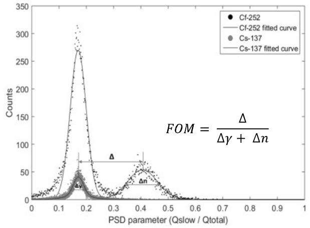


- Digital Signal Processing with a 14-bit 500 MHz Fast Digitizer (CAEN DT5730)
- PSD with Charge Comparison

Stilbene Scintillator



EJ-301 Liquid Scintillator



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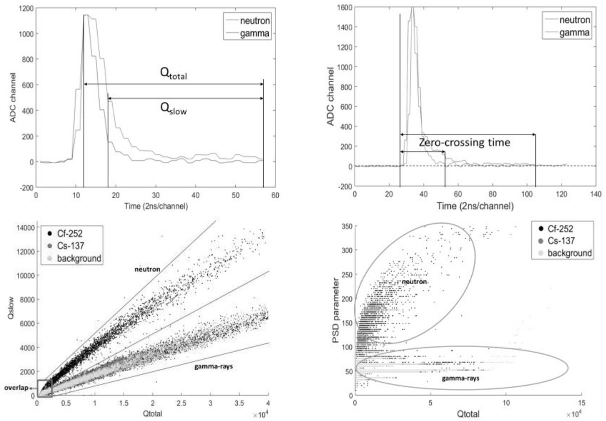
Pulse Shape Discrimination Study



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Material Development - Scintillators

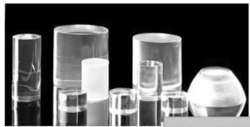
- Liquids
- Crystals
- Plastics

Organics

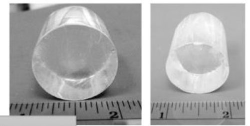
- Alkali halides
- Elpasolites
- Other

Inorganics

Plastics (RMD)



Elpasolites: CLYC & CLL(B,C) (RMD)



Phoswiches

Pulse Shape Discrimination

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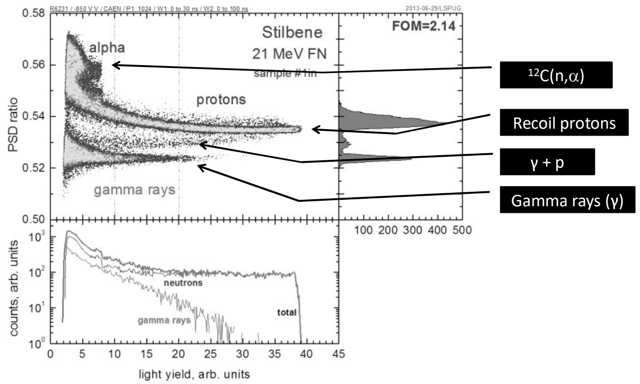
21 MeV Excitation



SORMA 2014, June 11, Jarek Glodo

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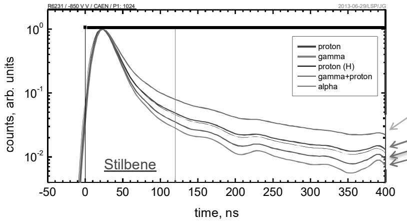
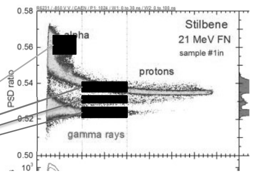
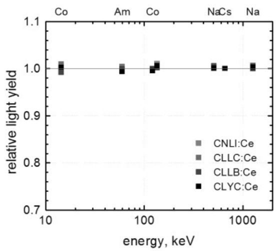
Time profiles (21 MeV neutrons)

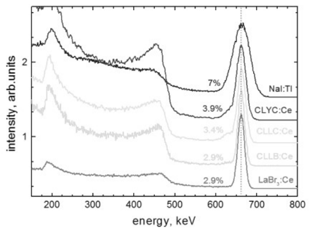
The higher density of excitation (e → p → α) the higher intensity of slow components





Elpasolites: Proportionality and Resolution





CsLiLn halide scintillator (K.S. Shah, US Patent no. 8,440,980)
Mixed cesium sodium and lithium halide scintillator compositions, US Patent no. 7,977,645
Cesium and lithium-containing quaternary compound scintillators, US Patent no. 8,415,637
Cesium and sodium-containing scintillator compositions, US Patent no. 8,242,452
Yttrium-Containing Scintillator Compositions, US Patent no. 8,153,982

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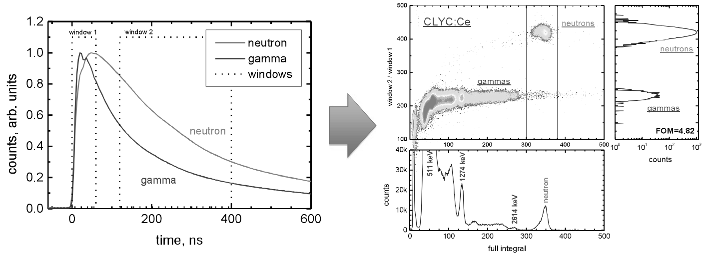
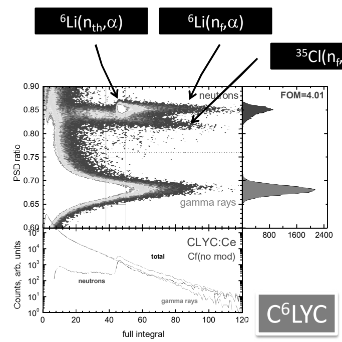
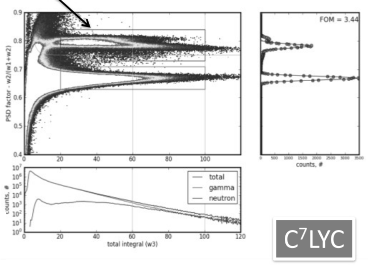


+ Fast Neutrons



No thermal peak



Discovered by BTI

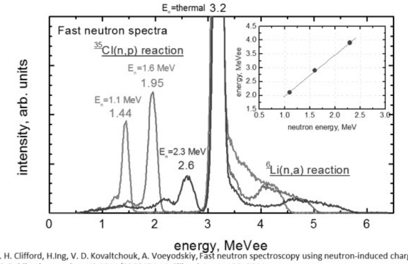
$^{35}Cl + ^1n \rightarrow ^1p + ^{35}S$ + energy

10/15/2020

CLYC: Fast Neutron Detection

$^{35}Cl + ^{1}n \rightarrow ^{1}p + ^{35}S + \text{energy}$

Proportional to ^{1}n energy



T. Achtzehn, H. R. Andrews, E. T. H. Clifford, H.Ing, V. D. Kovaltchouk, A. Voeyodskiy, Fast neutron spectroscopy using neutron-induced charged particle reactions, Application number: 13/096,228, Publication number: US 2011/0266451 A1, Filing date: Apr 28, 2011

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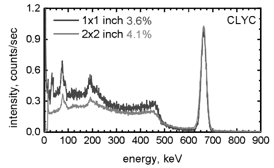
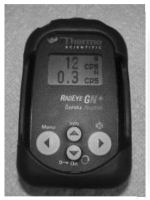
CLYC Commercialization

- CLYC product launch November 2012
  - Added to RMD web site
  - CLYC packaged with or w/o PMT
- Thermo Scientific RadEye GN+
  - Product launch January 2013
  - CLYC production at Hilger Crystals, a Dynasil Company



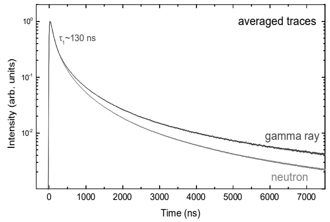


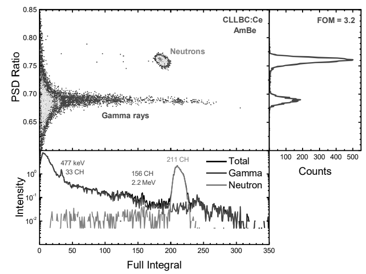
RMD A Dynasil Company *See Hilger booth for a 1.5" CLYC detector in operation* 121

Other elpasolites

Many other elpasolites show PSD capabilities:

- $Cs_2LiLaCl_6$
- Cs_2LiYBr_6
- $Cs_2LiLaBr_6$
- $Cs_2LiLa(Br,Cl)_6$

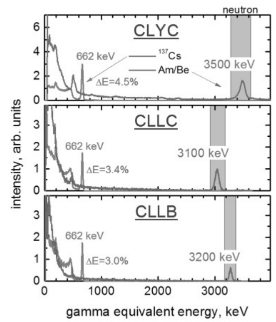


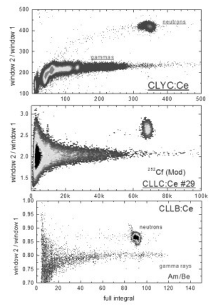


RMD A Dynasil Company SORMA 2014, June 11, Jarek Glodo 122 of 30

Thermal Neutron Detection: $Cs_2Li(Re)X_6$:Ce

$^{6}Li + ^{1}n \rightarrow \alpha + T$ 4.8 MeV





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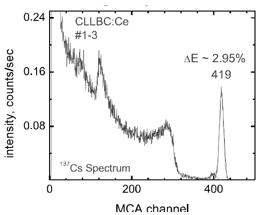
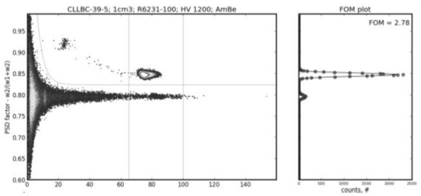
CLLBC – A Brighter Elpasolite

2 x higher light yield than CLYC leads to better energy resolution



Good PSD



Due to presence of Cl, fast neutron spectroscopy possible

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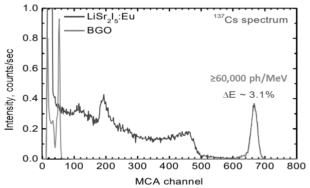
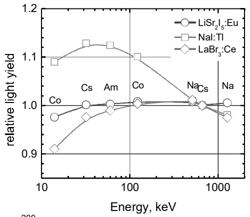
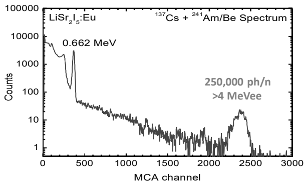
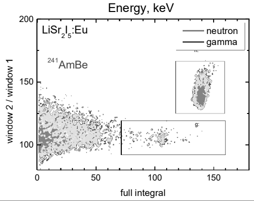
$LiSr_2I_5$:Eu (LSI) – A New G-N Scintillator









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See oral presentation 105 for new results on larger crystals

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Neutron Cross Sections (probability of interaction) - $LiInSe_2$ example

- ^{7}Li – 1.16 b while ^{6}Li – 938 b (7.5% content in the natural Li)
- Σ = macroscopic absorption cross [Rinard]
- $\Sigma = \rho N_A/M(n_1\sigma_1 + n_2\sigma_2 + n_3\sigma_3 + \ldots)$
- The mean-free path (MFP) length λ for $LiInSe_2$ is:
  - $\Sigma_{t(LISe)\text{-enriched}} = (4.5 \times 0.6022/278.84) \times (938 + 194 + 11.7 \times 2) = 9.12 + 1.89 + 0.22 = 11.23\ cm^{-1}$
  - λ = 1/ Σt = 3.58 mm; λenrich = 1/ Σt-enr = 0.893 mm
  - Coefficient of performance (P) defined as a ratio of ^{6}Li contribution to the total cross-section is:

$$P = {}^6Li/LiInSe_2 = 9.12/11.23 = 81.2\ \%$$

P. Rinard "Neutron interaction with mater" in *Passive Nondestructive Assay of Nuclear Materials*, ed. by D. Reilly *et al.*, U.S. Nuclear Regulatory Commission, NUREG/CR-5550, March 1991, p. 366.

20

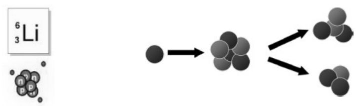
Absorbers

- ^{10}B (19.9% abundance in natural boron) has a cross-section of 3840 barns for thermal neutrons
- The ^{10}B(n, α) Li reaction results in the following reaction products

$$n + {}^{10}B \rightarrow {}^7Li^* + {}^4He \rightarrow {}^7Li + {}^4He + 0.48\ MeV\ \gamma + 2.3\ MeV \quad (94\%)$$
$$\rightarrow {}^7Li + {}^4He + 2.8\ MeV \quad (6\%)$$

- The ^{6}Li(n,α)^{3}H reaction results in the following products

$$n + {}^6Li \rightarrow {}^4He(2.05MeV) + {}^3H(2.73MeV) + 4.79\ MeV\ (Q\text{-value})$$



The Q-value of the reaction is defined as the energy liberated following the neutron capture and it can be determined from the masses of reactants and products:

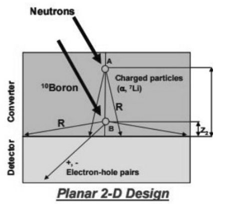
$$Q = K_{(final)} - K_{(initial)} = (m_{final} - m_{initial})c^2$$

Glenn F. Knoll, "Slow Neutron Detection Methods", chap. 14 in Radiation detection and measurement, 3rd ed. New York: John Wiley & Sons, 1989. .

40

Planar 2-D design

- Semiconductor detector (p-n structure) is coated with the converter material
- Boron-10 (^{10}B) converter has excellent microscopic absorption cross section of 3840 barns.
- Converter must be < 3.3 μm for the alpha particle (1.47 MeV) and ^{7}Li (0.84 MeV) to reach the semiconductor → detection ef ficiency is low. Most of the deposited energy doesn't reach detector → poor pulse height discrimination



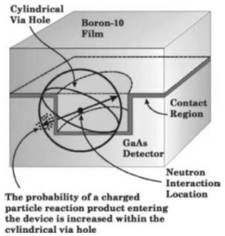
Planar 2-D Design

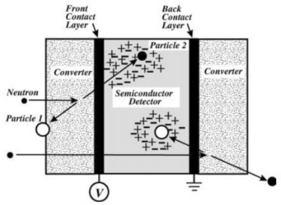
M. Wielunski, R. Schütz, E. Fantuzzi, A. Pagnamenta, W. Wahl, J. Palfálvi, P. Zombori, A. Andrasi, H. Stadtmann and C.H.. Schmitzer, "Study of the sensitivity of neutron sensors consisting of a converter plus Si charged-particle detector," Nuclear

Instruments and Methods in Physics Research A, 517, pp. 240-253, 2004.

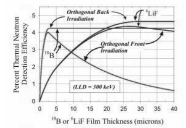
Improved designs

- Double coated
- Cylindrical holes - increase the neutron detection efficiency from the 2-D semi conductor devices (2-5%) towards 70%.





D. S. McGregor *et al.*, "Designs for Thin-Film-Coated Semiconductor Thermal Neutron Detectors," Nuclear Science Symposium Conference Record, (IEEE 2001), p. 2454.

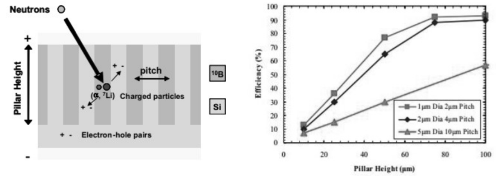


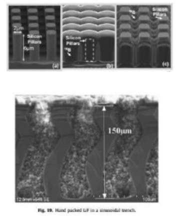
D. S. McGregor *et al.*, "Design Considerations for Thin Film Coated Semiconductor Thermal Neutron Detectors - I: Basics Regarding Alpha Particle Emitting Neutron Reactive Films", Nucl. Instrum. Methods Phys. Res. A **500**, 272 (2003).

10/15/2020

Microstructured semiconductor neutron detectors detector design

- Simulations of thermal neutron capture efficiency versus pillar height for several pillar diameters and pitches for neutron-converter (10B) and the detect or material (silicon)

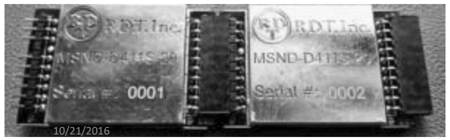




- J. Shultis and D. Mcgregor, Efficiencies of coated and perforated semiconductor neutron detectors, IEEE Trans. Nucl. Sci. 5 3 (2006) 1659.
- High-efficiency neutron detectors and methods of making same, US Patent 7164138 B2 (Filing date: Oct 29, 2003, Inventors: Douglas S. McGregor, Raymond Klann)
- S.L. Bellinger, R.G. Fronk, W.J. McNeil, T.J. Sobering, D.S. McGregor, IEEE Transactions on Nuclear Science 59 (2012) 167 [*dual stack of 1-cm2 devices, demonstrated 42% efficiency*]
- Douglas S. McGregor , Steven L. Bellinger, J. Kenneth Shultis, Present status of microstructured semiconductor neutron detectors, Journal of Crystal Growth 379 (2013) 99–110 [4 cm2 area, 32.4% efficiency)

Domino® Neutron Detector by Radiat ion Detection Technologies, Inc

- The Micro-structured Semiconductor Neutron Dete ctor (MSND®) technology developed at Semiconduc tor Materials and Radiological Technologies (S.M.A. R.T.) Laboratory at Kansas State University was imple mented in RDT Domino® with 4-cm^2 detection area and 20% to 30% thermal neutron efficiency



R&D 100 2014 Winner

2009 Winner R&D 100

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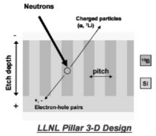
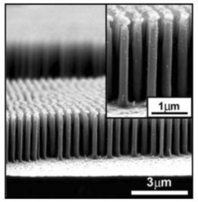
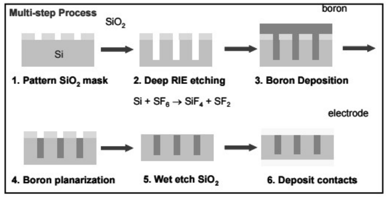
LLNL Pillar







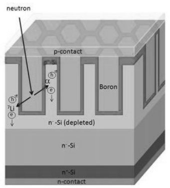
MCNP→ etch depth of 50μm and a pillar width and spacing of 2μm

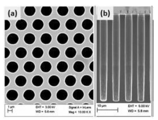
- R.J. Nikolic, C.L. Cheung, C.E. Reinhardt, and T.F. Wang, “Roadmap for High Efficiency Solid State Neutron Detectors,” SPIE - International Sympos ium on Integrated Optoelectronic Devices, Phot onics West, Optoelectronic Devices: Physics, Fa brication and Application II, Boston, MA, Oct. 2 005.

	^{3}He Tube	LLNL 3-D Pillar detector
Efficiency (fieldable)	< 30 %	70%
Cost	$ 4K	$ 0.2K
Required voltage	1000 V	< 10 V
Size (probe)	(5 ϕ) cm x 10 cm	1 x 1 x 0.1 cm^3
Weight (includes power)	700 g	10 g
Fieldability	Microphonics, HV, air transport	Not commercially available

Self-powered micro-structured

- micro-structured silicon diodes with deep h exagonal holes filled with natural boron as a conversion material for thermal neutron det ection applications
- intrinsic thermal neutron detection efficie ncy of 4.5% +/- 0.5%, and gamma to neutr on sensitivity of (1.1 +/- 0.1) 10^{-5}
- Monte-Carlo simulation predicts a maximum efficiency of 45% for such devices filled with 95% enriched ^{10}boron.





R. Dahal, K. C. Huang, J. Clinton, N. LiCausi, J.-Q. Lu, Y. Danon, and I. Bhat, Self- po wered micro-structured solid state neutron detector with very low leakage current and high efficiency, APPLIED PHYSICS LETTERS 100, (2012) 243507

Diamond for Neutron Detection

- Bandgap of diamond is 5.5 eV this leads to negligible dark current noise
- Radiation hardness
- High mobility of free charges-> Fast response
- Fast neutrons are detected directly in the bulk of the intrinsic diamond layer through the ^{12}C (n,α) ^{9}Be and ^{12}C (n,n')$^{12}C^*$ reactions.

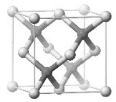
- The produced ^9Be and α ions have a total energy:

  $E_\alpha + B_e = E_n – 5.7$ MeV
  where E_n is the energy of the impinging neutron.

- To detect both fast and thermal neutrons a layer of ^6LiF or ^{10}B

- S. F. Kozlov, E. A. Konorova, and I. A. Kuznetsov, IEEE Trans. Nucl. Sci. NS-24, 235 (1977).
- V. D. Kovalchuk, V. I. Trotsik, and V. D. Kovalchuk, Nucl. Instrum. Methods Phys. Res. A 351, 590 (1994).
- M. Pillon, M. Angelone, A.V. Krasilnikov, Nucl. Instr. and Meth. Phys. Res. B 101 (1995) 473.
- P. Bergonzo et al., Nucl. Instrum. Methods Phys. Res. A 476, 694 (2002).
- G. J. Schmid, J. A. Koch, R. A. Lerche, and M. J. Moran, Nucl. Instrum. Methods Phys. Res. A 527, 554 (2004).

Cubic boron phosphide (BP)



- Bandgap: 2.1 eV, low Z_{ave} → low background from gamma-ray interactions
- B-10 (21% abundance)
- ^{10}B(n, α)^7Li
- A ^{10}BP film only 200 μm thick will absorb over 95% of incident thermal neutrons [Kumashiro]
- The largest single crystals (5 x 5 x 3 mm^3) were obtai ned by high pressure flux method [Kumashiro]

Y. Kumashiro, T. Yao, S. Gonda, J. Crystal Growth 70 (1984) 507
Y. Kumashiro, Y. Okada, S. Misawa, T. Koshiro. Proc. 10th Int. Conf. Chemical Vapor Deposition, vol. 87-88 (1897) 813

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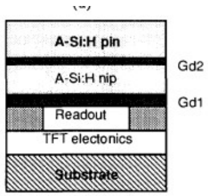
Amorphous Silicon Pixel Detectors

n + ^{155}Gd → Gd* → γ-ray spectrum → conversion electron spectrum
n + ^{157}Gd → Gd* → γ-ray spectrum → conversion electron spectrum

- A multi-layer a-Si:H based thermal neutron detec tor was designed, fabricated and simulated by M onte Carlo method.
- The detector consists of two a-Si:H pin detectors prepared by plasma enhanced chemical vapor de position (PECVD) and interfaced with coated laye rs of Gd, as a thermal neutron converter.
- Simulation: Optimum detector consisting of 2 Gd films with thicknesses of 2 and 4 µm, sand wiched properly with two layers of sufficiently thick (30µm) amorphous silicon diodes
- The detectors had an intrinsic efficiency of a bout 42% (63% possible with enriched Gd)
- Pixel size as low as 30µm



High Efficiency Neutron Sensitive Amorphous Silicon Pixel Detectors A. Mireshghi, G. Cho, J.S. Drewery, W.S. Hong, T. Jing, H. Lee, S.N. Kaplan, and V. Perez-Mendes, IEEE TRANSACTIONS ON NUCLEAR SCIENCE, VOL. 41, NO. 4, AUGUST 1994, p. 915

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LISe™: A High-Efficiency Thermal Neutron Detector

- Technology was originally developed by B&W Y-12 (Ashley Stowe and Zane Bell) and Fisk University (Arnold Burger)
- LISe is designed to be used in handheld nuclear non proliferation and homeland security applications to find fissile materials. Currently being transferred to commercialization by Radiation Monitoring Devices, Inc.

2013 R&D 100 Awards winner

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Semiconductor detectors based on bulk crystal containing B or Li

- Such material from which 1 cm² wafers can be produced has been realized for the first time in the form of $^{6}LiInSe_2$ in which collectable charge from the ^{6}Li(n,t) reaction indicates a neutron event. In the following we will cover the neutron and gamma responses of $^{6}LiInSe_2$

- Zane Bell, Arnold Burger, Semiconductor radiation detector, US Patent 20070080301 A1 Publication date : Apr 12, 2007
- Tupitsyn, E.; Bhattacharya, P.; Rowe, E.; Matei, L.; Groza, M.; Wiggins, B.; Burger, A.; Stowe, A, Single crystal of LiInSe2 semiconductor for neutron detector. Appl. Phys. Lett. **101**, 202101 (2012)
- Zane W. Bell; A. Burger; Liviu Matei; Michael Groza; Ashley Stowe; Joshua Tower; Alireza Kargar; Huicong Hong Proc. SPIE 9593, Hard X-Ray, Neutron detection with $LiInSe_2$, SPIE Gamma-Ray, and Neutron Detector Physics XVII, 95930D (26 August 2015); doi:10.1117/12.2189418
- Ashley C. Stowe, Arnold Burger, Michael Groza, Bulk semiconducting scintillator device for radiation detection, US Patent no. 20140209805 A1, Publication date: Jul 31, 2014

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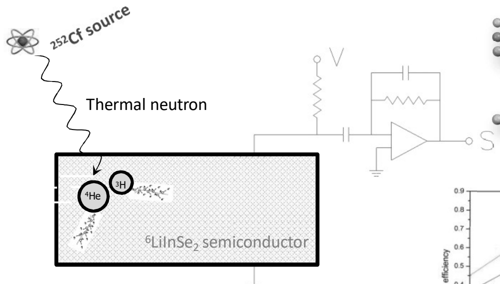

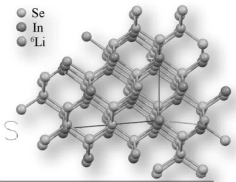

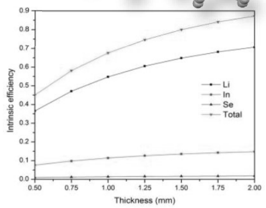

^{6}Li(n,α)^{3}H –> [Q=4.78 MeV, E(^{3}H) = 2.73 MeV, E(α) = 2.05 MeV]

$$=940\frac{\lambda}{1.8}\text{ barns}$$

For thermal neutrons, λ=1.8Å

1 barn =10^{-24} cm², or ~cross sectional area of a uranium nucleus

←Pixels and cathode contacts deposited

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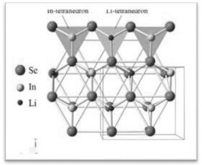
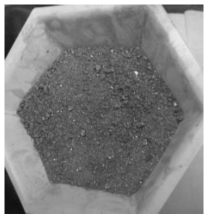
Summary of Li-chalcogenides

Crystal	$LiGaTe_2$	$LiGaSe_2$	$^6LiInSe_2$
Band gap	2.41 eV	2.9 eV - 3.4 eV	2.1eV (red) 2.8 eV (yellow)
Structure	Chalcopyrite	Chalcopyrite	Orthorhombic
Synthesis	LiGa + Te	LiGa + Se	LiIn + Se
Growth crucible	Glassy carbon	Glassy carbon/quartz	PBN/quartz
Grown crystal	Polycrystalline, unstable in air	Large crystal	Large crystal
Resistivity	~10^7 Ω-cm	~10^{10} Ω-cm	~10^{11} Ω-cm
Photo response	No	No	Yes
α–detection	No	No	Yes
γ–detection	No	No	Yes
neutron detection	No	No	Yes

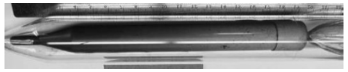
Material Synthesis



- Lithium is highly reactive metal
- Create the stoichiometric composition of I-III-VI_2 in two steps – 1: Li+Ga ⟹ 2: LiGa+Se
- Choosing of the crucible material: CC Quartz/Glassy carbon/PBN



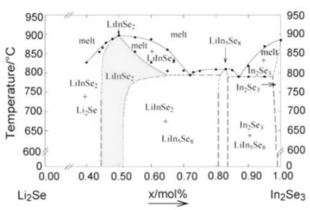
Material Synthesis challenges



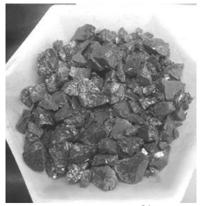
- Lithium is highly reactive metal
- Create the stoichiometric composition of I-III-VI_2 in two steps – 1: Li+Ga 2: LiGa+Se
- Choosing of the crucible material: CC Quartz/Glassy carbon/PBN

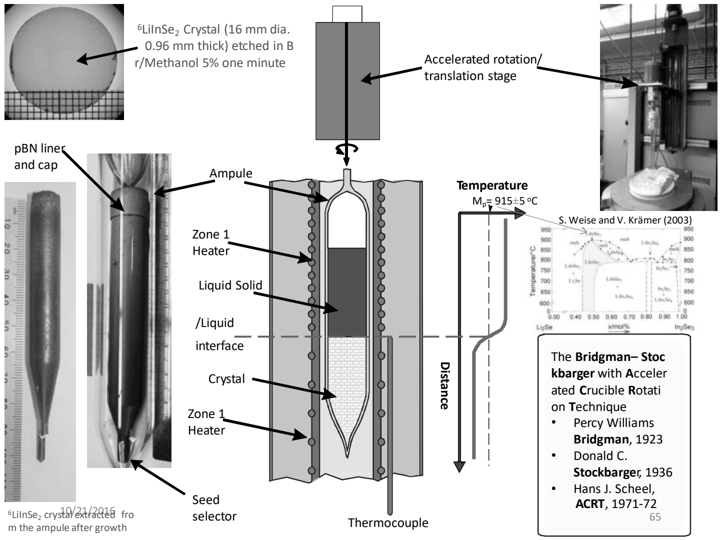
+5% Li

Phase diagram



stoichiometric



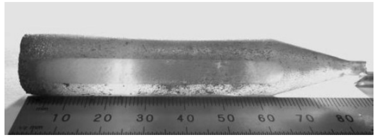


The **Bridgman– Stockbarger** with **A**ccelerated **C**rucible **R**otation **T**echnique

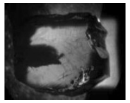
- Percy Williams **Bridgman**, 1923
- Donald C. **Stockbarger**, 1936
- Hans J. Scheel, **ACRT**, 1971-72

Crystal Growth

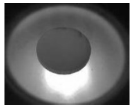
$LiGaTe_2$ **$LiGaSe_2$**



- Synthesis produced non-uniform material.
- Compound is unstable in air.
- Many attempts yielded 3-4 mm size crystalline material at best.



- Growth procedure was devel oped and standardized.
- Crystals are transparent, bright yellow/orange.
- 15 mm diameter crystals are readily grown.



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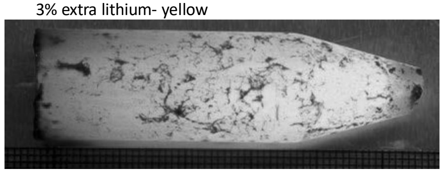
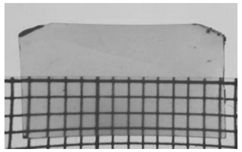
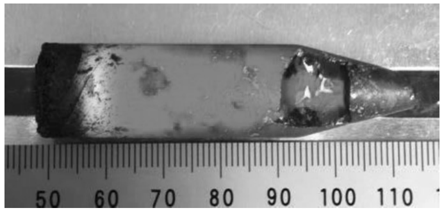
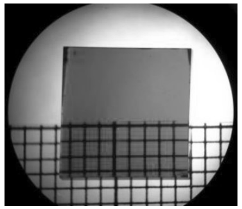
$^6LiInSe_2$ grown ingots

slight variation of lithium content may affect on color of the crystal
1% extra lithium - red
3% extra lithium- yellow









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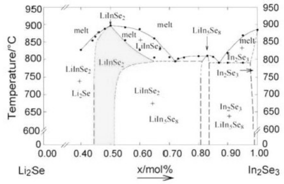
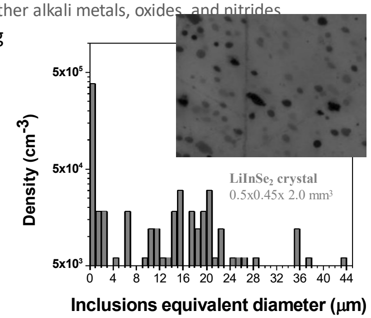
Defect Analysis and optimizing mu-tau

- Defects are observed in the crystal which act as charge trapping sites.
- What are defects?
  - Starting material impurity precipitates
    - Purifying Li metal will remove other alkali metals, oxides, and nitrides
  - Impurity phases/ excess starting material precipitates
    - Control synthesis stoichiometry excess Li or Se and phases.





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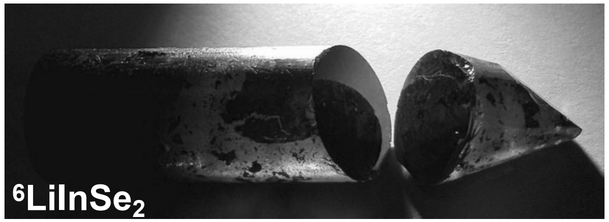
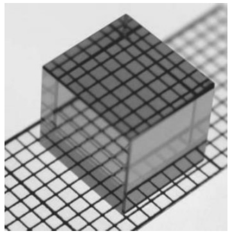
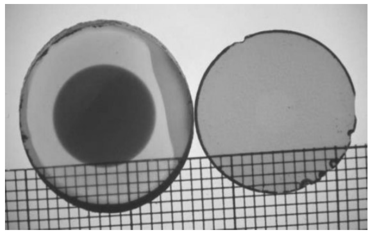
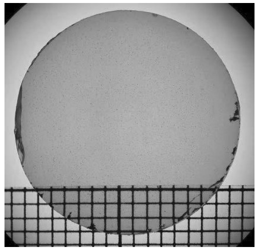




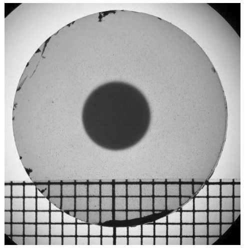


10/21/2016 Ruler is under the wafer, left is w/Au contact 69

Detector fabrication

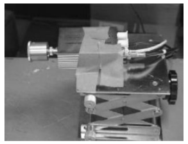


$^{6}LiInSe_2$ Crystal (16 mm dia. 0.96 mm thick) etched in Br/Methanol 5% one minute

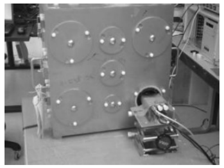


Gold contact applied by s puttering (33 nm thick)

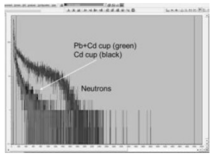
$^{6}LiInSe_2$ detector



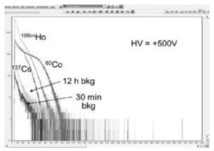
$^{6}LiInSe_2$ detector+ PA



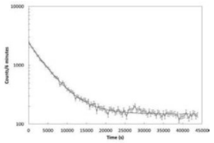
Detector in AmLi neutron source.



Neutron Response



Gamma Response



^{116m}In activation, half-life is 52.4 minutes

50% efficiency demonstrated in 1 mm thick detector, consistent with simulations. The density of Li in the crystal is sufficient to insure 95% efficiency in 3.4 mm thick wafers.

Zane W. Bell, A. Burger, Liviu Matei, Michael Groza, Ashley Stowe, Joshua Tower, Alireza Kargar, Huicong Hong, Neutron Detection in $LiInSe_2$, Proc. SPIE 9593, Hard X-Ray, Gamma-Ray, and Neutron Detector Physics XVII, 9593 0D (26 August 2015)

$LiInSe_2$ developed at Fisk and RMD, Inc

- Despite the fact that the absorption cross-sections for ^{6}Li or ^{10}B are modest in comparison to ^{157}Gd, usually < 0.2 cm of enriched semiconductor material (e.g. $LiInSe_2$) is enough to stop >75% of neutrons.

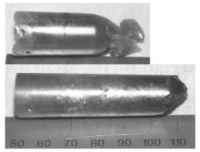


Figure 1. Crystal grown of 35 mm (top) and 55mm long ingots with ∅16mm diameter, both with natural Li.

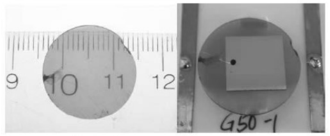
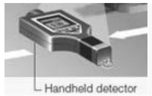


Figure 4. $LiInSe_2$ ∅16 mm wafer (left); assembled detector with 300/300 Å Cr/Au contacts deposited through thermal evaporation (right).



Four planar $LiInSe_2$ detectors each with area of 1 cm^2 will be able to provide very compact and robust substitution of the ^{3}He tubes neutron detection module for a number of handheld radiation measurement instruments on the market.

A. Gueorguiev, J. P. Tower, H. Hong, K. S. Shah, A. Burger, **"Semiconductor neutron detector"**, *(Invited Paper)*, Proceedings of SPIE, Hard X-Ray, Gamma-Ray and Neutron Detector Physics XVIII, San Diego, CA. August 29, 2016.

Reference

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- J. Frýbort, L. Heraltová, "Interaction of Neutrons with Matter", Chapter 2, Nuclear Reactor Theory (10/03/2014)
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