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An Introduction to Neutrons & Neutron Diffraction Experiments

(with a slight bias towards powder diffraction ☺)

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Overview

- Properties of the Neutron
- Reactors Neutron Sources
  - Generation of Neutrons
  - Neutron Instrumentation
- Pulsed Neutron Sources
  - Generation of Neutrons
  - Neutron Instrumentation
- Neutron Experiments

Properties of the Neutron

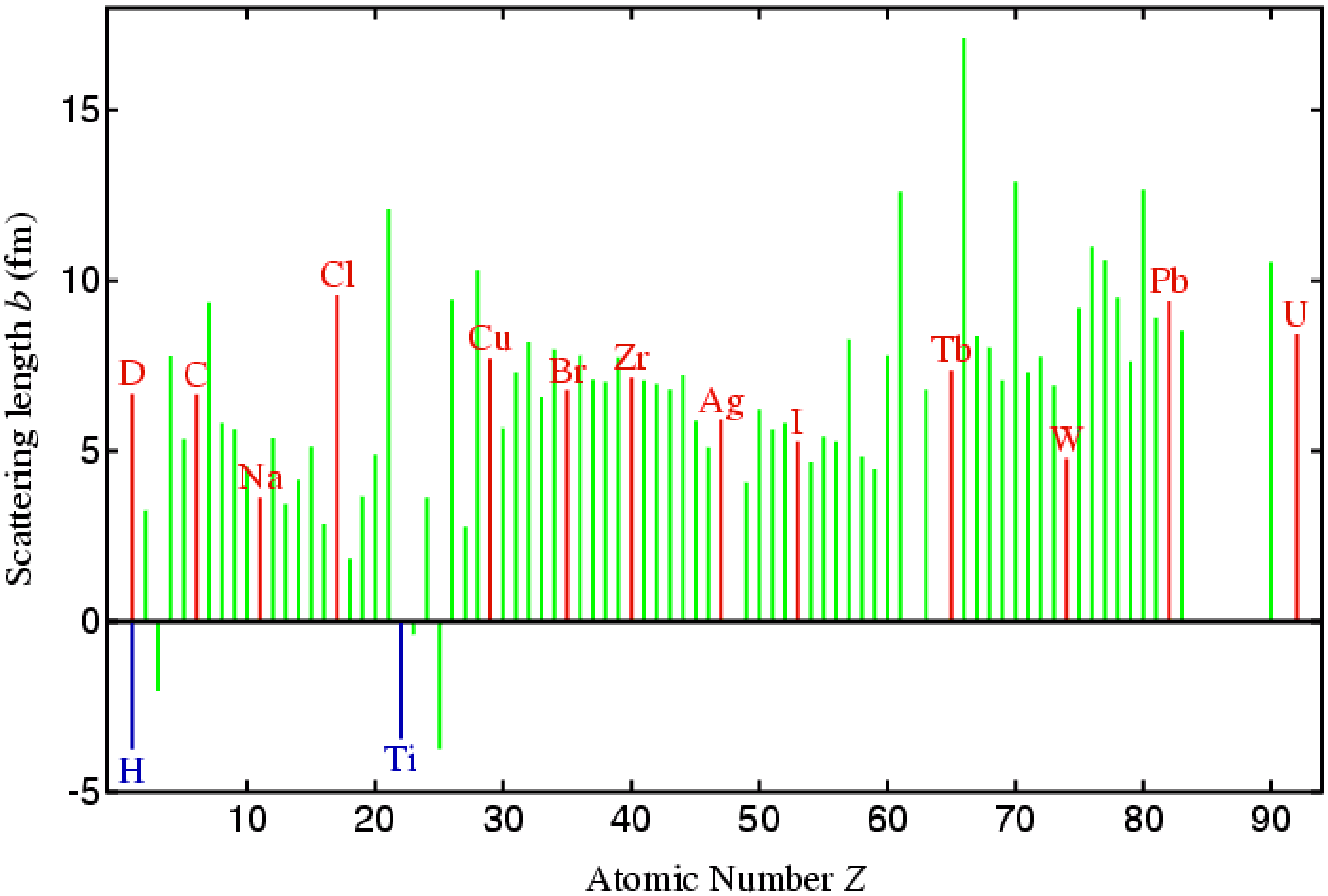
- Particle with:
  - Mass 1, Charge 0, Spin ½
    - Strong interaction with nucleus
    - Weaker interaction with unpaired electrons
      - » Magnetic crystallography
  - Kinetic Energy given by $E = \frac{1}{2}mv^2 = k_B T$
    - k_B is Boltzmann constant = 1.381×10^{-23} JK^{-1}
    - RT = 20°C (293 K) ➜ v = 2200 ms^{-1}
    - de Broglie relationship $\lambda = h / mv$ ➜ λ = 1.8 Å
      - » Thermal neutrons
      - » Properties often quoted for this wavelength

Scattering Processes

- Coherent Scattering Cross-Section, σ_c
  - Diffraction
  - Scattering length, b, where $\sigma_c = 4\pi b^2$
  - Units: b in fm (10^{-13} cm), σ in barns (10^{-24} cm^2)
- Incoherent Scattering, σ_i
  - Background
- Absorption, σ_a
  - Loss of intensity
  - Radioactive samples & γ background

Neutron Scattering-Lengths

- Dependant on the Nucleus Type



Neutron Scattering-Lengths

Element	H	D	U	Fe	Co
X-ray $f\,(0°)$ (Z)	1	1	92	26	27
Neutron b (fm)	−3.74	6.67	8.42	9.45	2.78

Element	Ba	O	V	Ti	Zr
X-ray $f\,(0°)$ (Z)	56	8	23	22	40
Neutron b (fm)	5.28	5.81	−0.38	−3.44	5.28

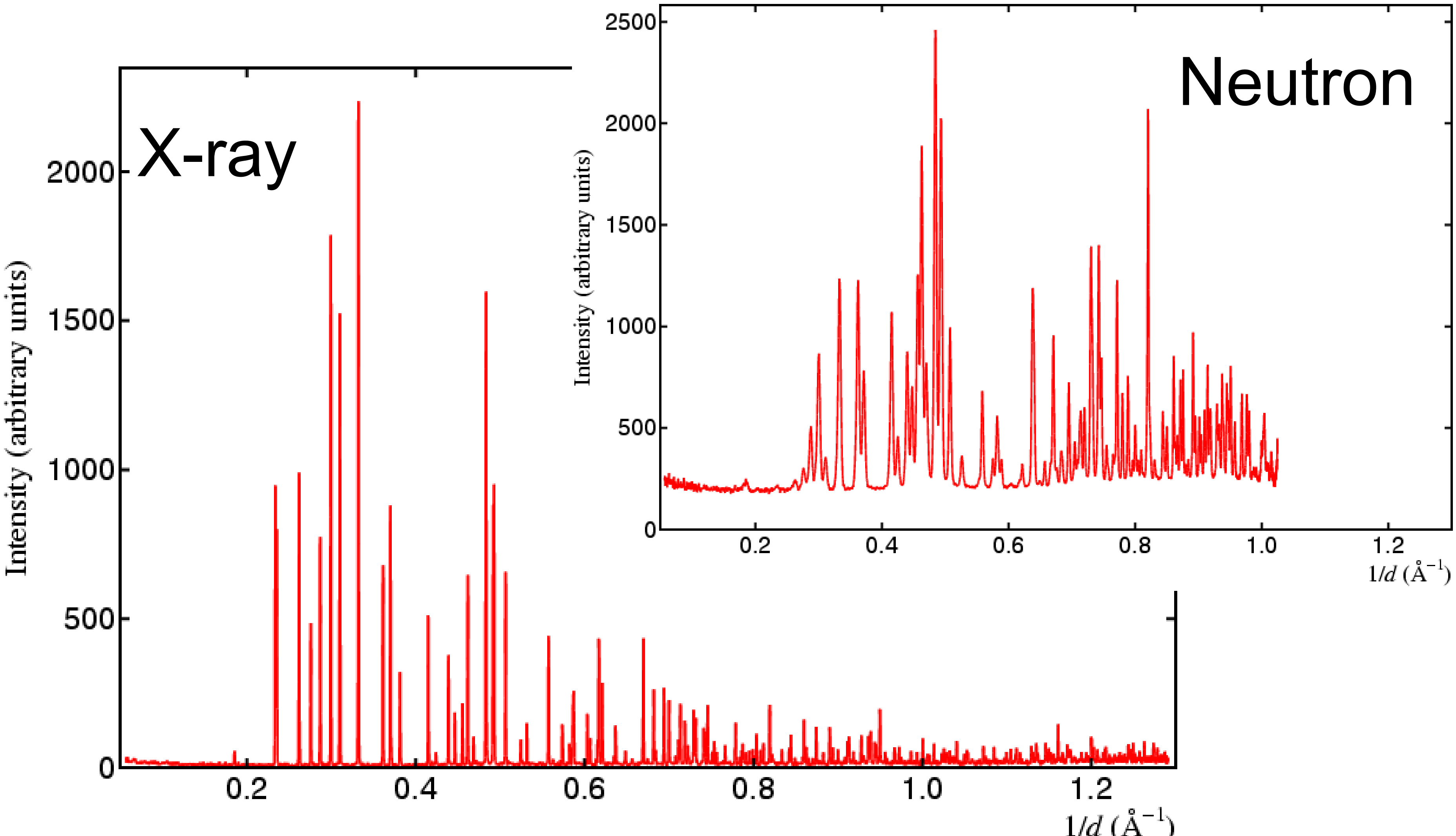
Absolute comparison: $f\,(0°) \times r_e$

where r_e classical electron radius = 2.818 fm

Scattering as a Function of 2θ

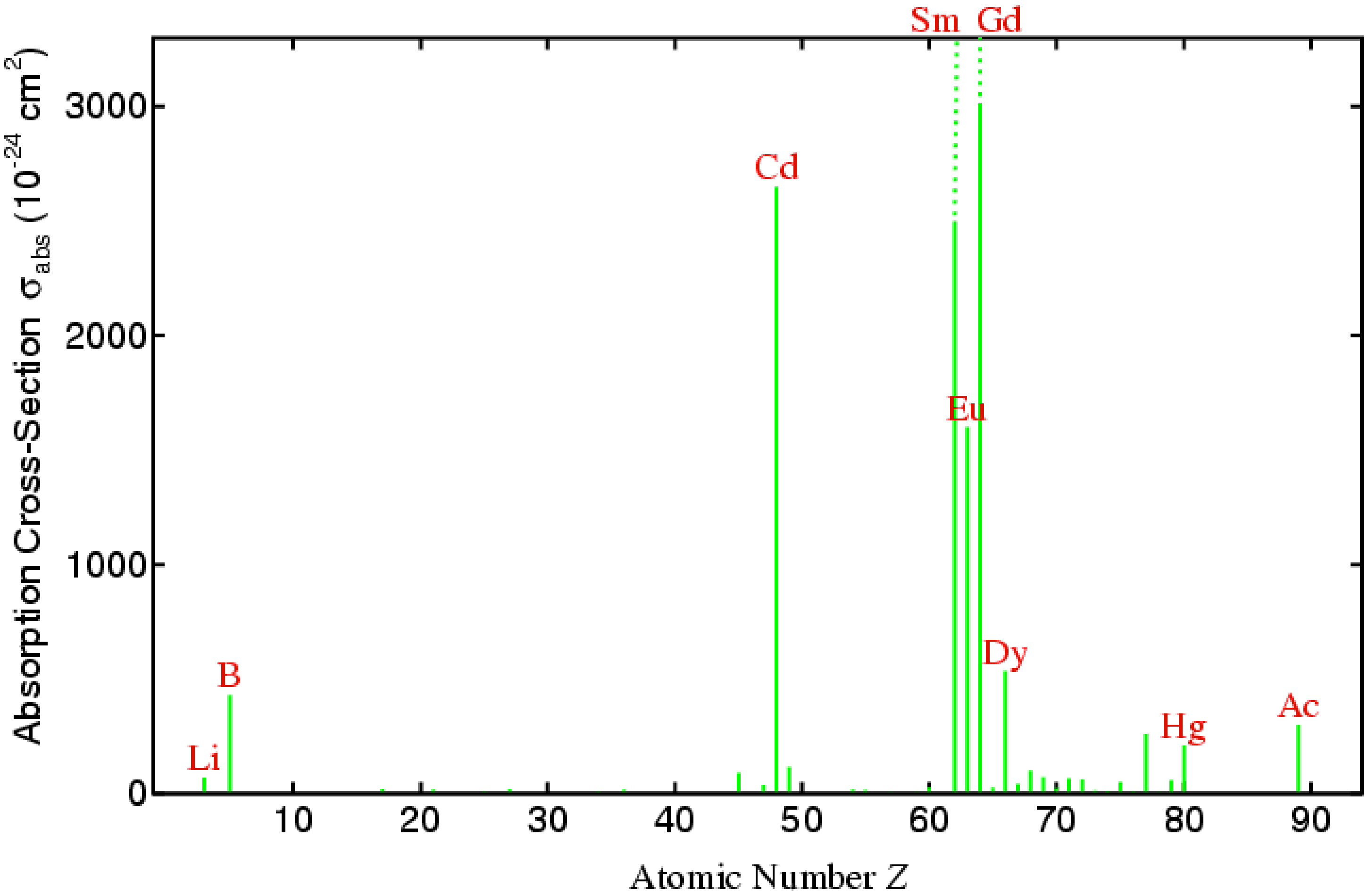
- X-ray interacts with electron cloud
  - Similar size to wavelength
  - Fourier transform of the electron cloud is the X-ray form factor (F.T. Gaussian ➜ Gaussian)
- Neutron interacts with nucleus
  - Very small (point scatterer) compared to λ
  - Fourier transform of a delta function is a horizontal line ➜ no neutron form factor
  - Interaction with unpaired electron(s)
    - Magnetic form factor

Scattering as a Function of 2θ



Absorption Cross-Section

- Relevant for Li, B, Cd plus some Lanthanides

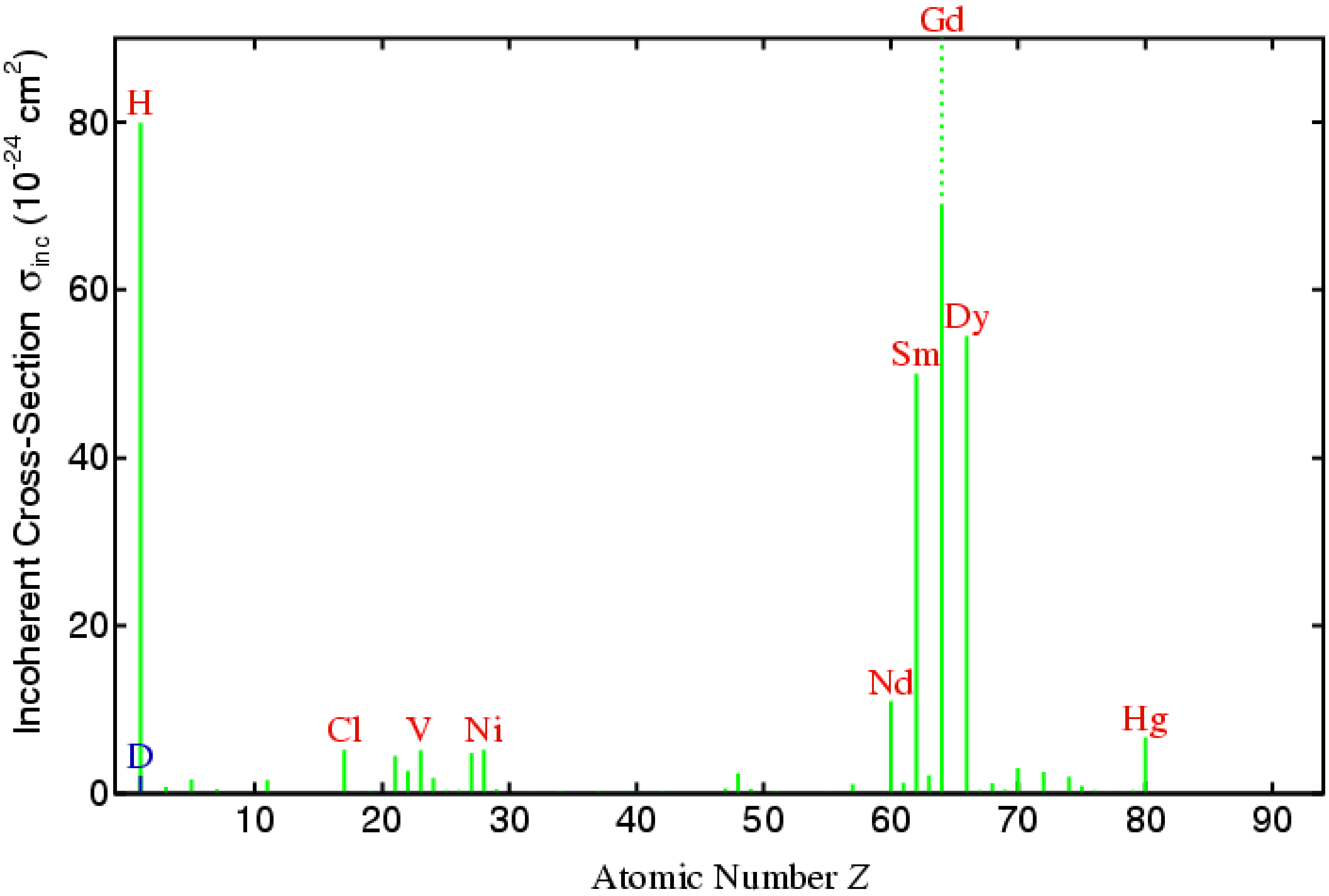


Neutron Shielding

- X-rays
  - Use Pb or thick Steel
- Neutrons
  - Use B_4C doped rubber or plastics
  - Use Cd sheet metal
    - Pliable at 1mm thickness
  - Use Gd_2O_3 paints (Soller collimators)
  - ^6Li now rarely used
    - Atomic strategic material required for military use

Incoherent Cross-Section

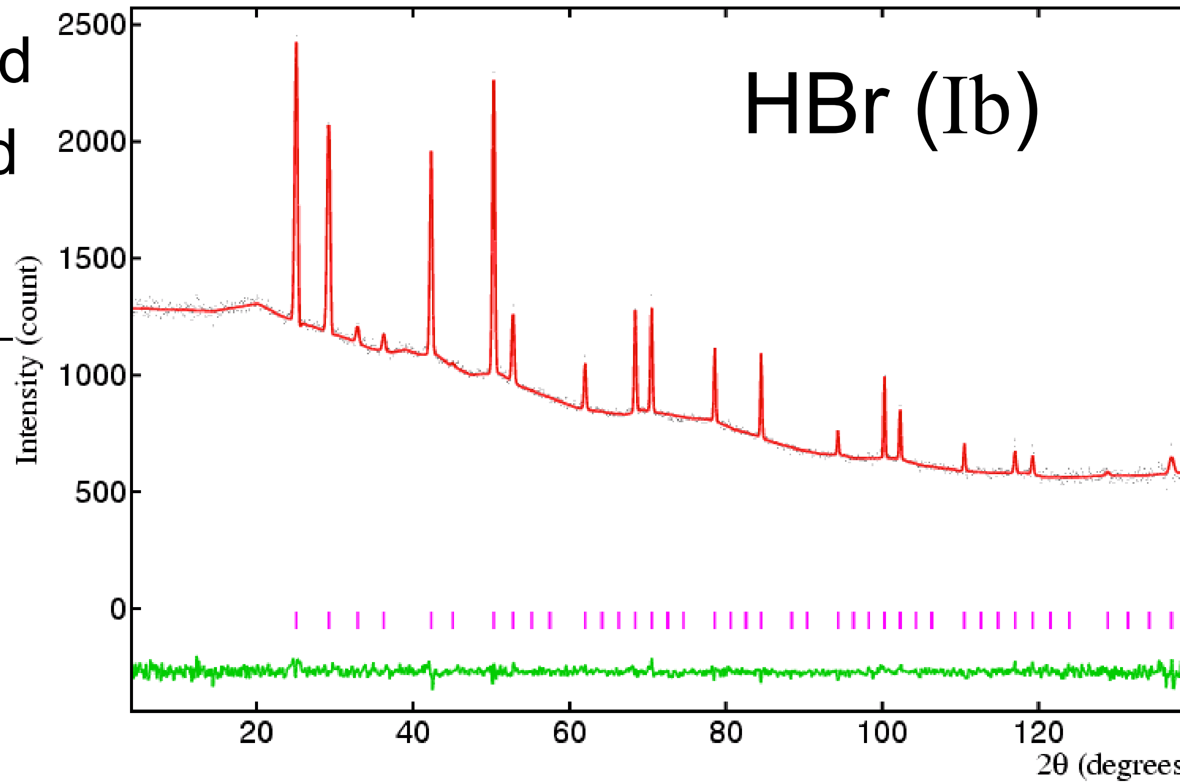
- Highly relevant for H plus some Lanthanides

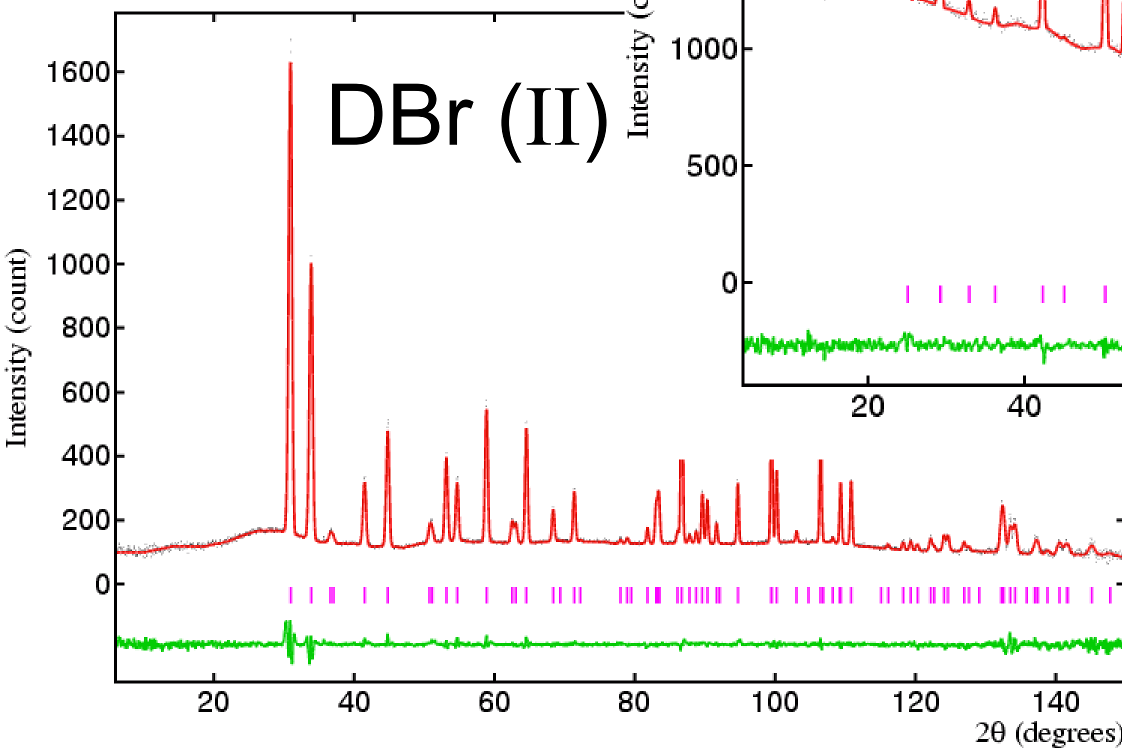


Effect of Incoherent X-Section

H ➜ High Background

D ➜ Low Background





Important for powder, but less so for single-crystal experiments

σ Comparisons

Element	σ_c	σ_i	σ_a
H	1.76	80.3	0.333
D	5.59	2.05	0.000519
V	0.0184	5.08	2.80*
Gd	29.3	150	49700
Al	1.495	0.0082	0.231

No need for special windows as aluminium can be used

Neutron Detectors

- Neutrons have no charge & non-ionizing
  - Less easy to detect than X-rays
- Detection relies on absorption by atom nucleus with simultaneous emission of γ-ray
  - Referred to as (n,γ) reaction
- Most common are proportional gas type
  - Absorbing material must absorb neutrons and be capable of existing in gaseous form
    - Limited choice of materials

Neutron Detectors

- Most common material is ^{3}He gas

$$^{3}He + {}^{1}n \rightarrow {}^{4}He + \gamma$$

  - Very expensive gas!
- Alternative is BF_3

$$^{10}BF_3 + {}^{1}n \rightarrow {}^{11}BF_3 + \gamma$$

  - Better (lower background)
  - Toxic and disliked by physicists
  - Now rarely used
- Gas pressurised (5-10 bar)

  ➜ improves efficiency

Neutron Detectors

- Solid State Detectors
  - ➢ Use (n,γ) reaction, but γ-ray converted to visible photon
    - – Measure with photomultiplier tube
    - – Early versions very sensitive to γ background
  - ➢ Use lithium salts or rare-earth glasses
    - – e.g. $Gd_2O_3.Gd_2S_3$
  - ➢ Advantage is high efficiency
    - – Very thin
      - ➜ important for Time-of-Flight measurements

Neutron Monitors

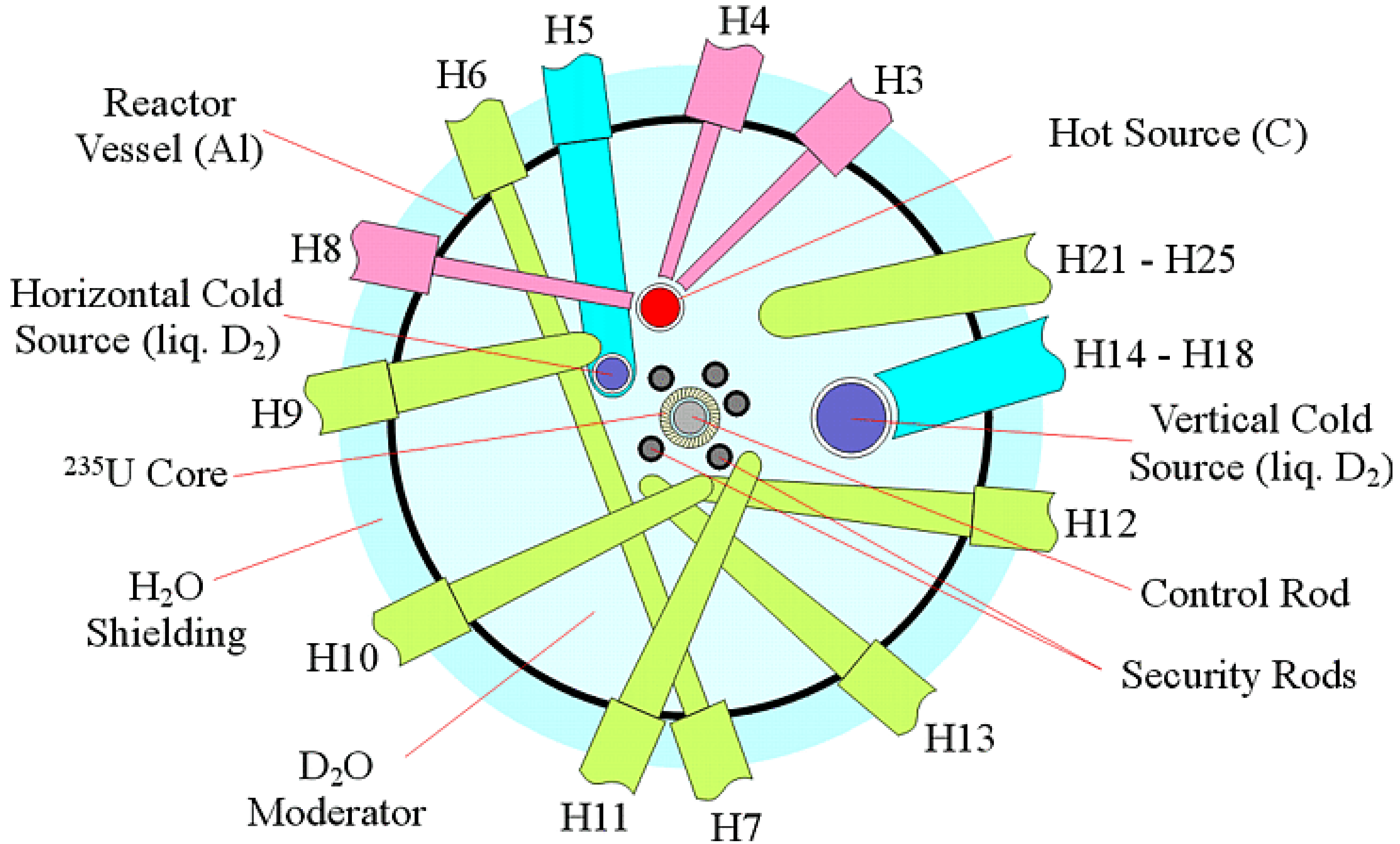
- Required to Monitor Incident Flux
  - No X-ray laboratory equivalent
    - X-ray tube assumed to produce constant flux
- Inefficient Detectors
  - e.g. 99.9% ^{4}He with 0.1% ^{3}He
    - Dilution optimised for particular wavelength & flux

Reactor Neutron Sources

- ILL, Grenoble
  - ➢Still World's Highest Flux Neutron Source
  - ➢Other European sources are much lower flux

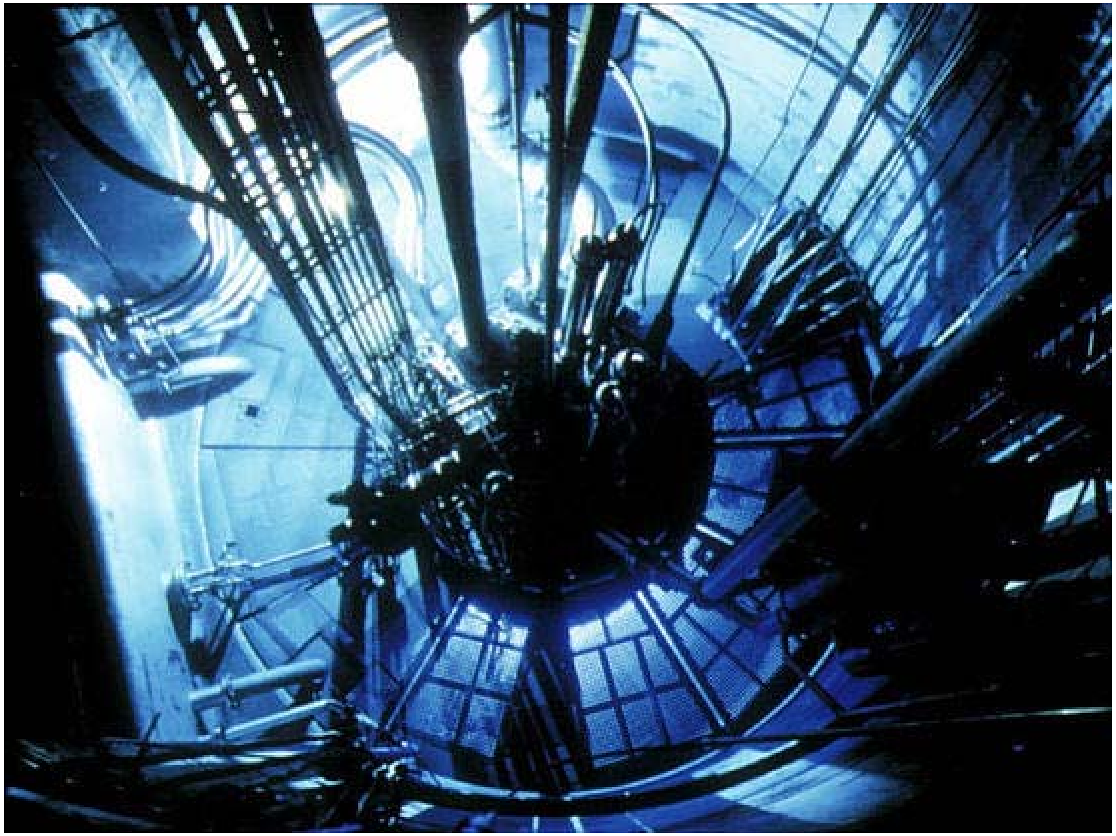
Centre of the Reactor

- 10 kg of ^{235}U heavy water cooled at 35°C



Light Water Biological Shield

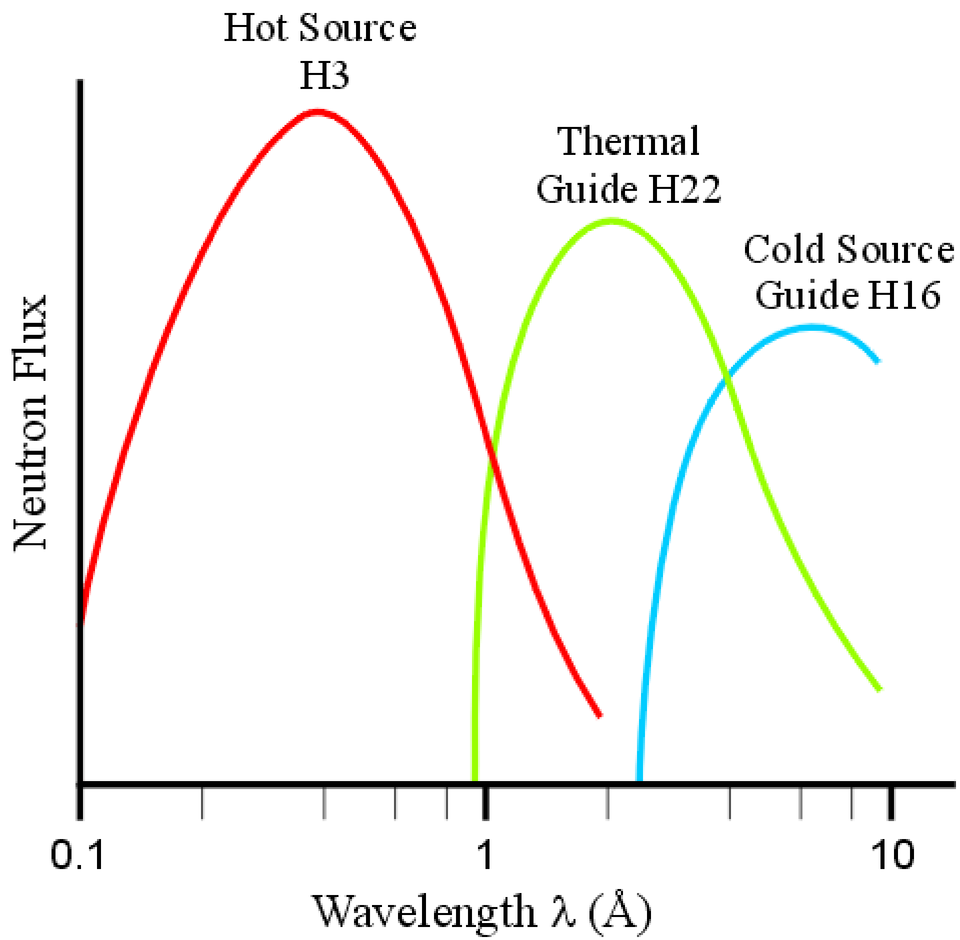
- Cerenkov Radiation



(*Photo supplied by ILL Publicity Office*)

Hot and Cold Neutrons

- Cold Source
  - ➢ Liquid H_2 fridge
  - ➢ Long λ neutrons
    - – SAS
- Hot Source
  - ➢ Solid graphite
    - – Self-heated
  - ➢ Short λ neutrons
    - – Small d spacings



Neutron Guide Halls

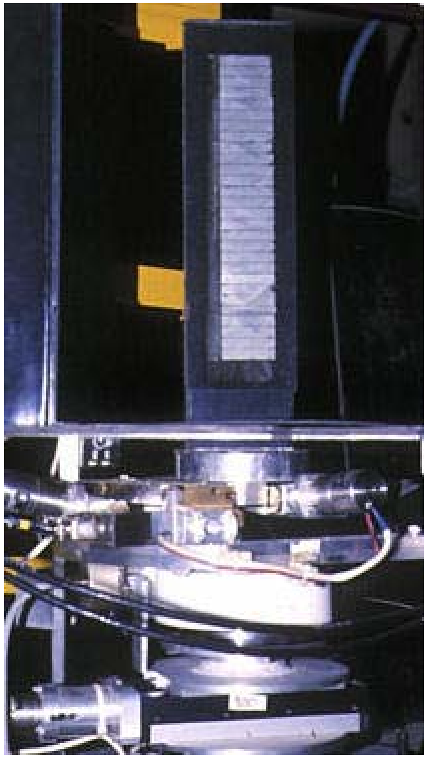
- Limited Space around Reactor Core
- Transmit Neutrons along curved Guides
  - Total internal reflection (especially long λ)
  - Removes γ-radiation (straight lines)

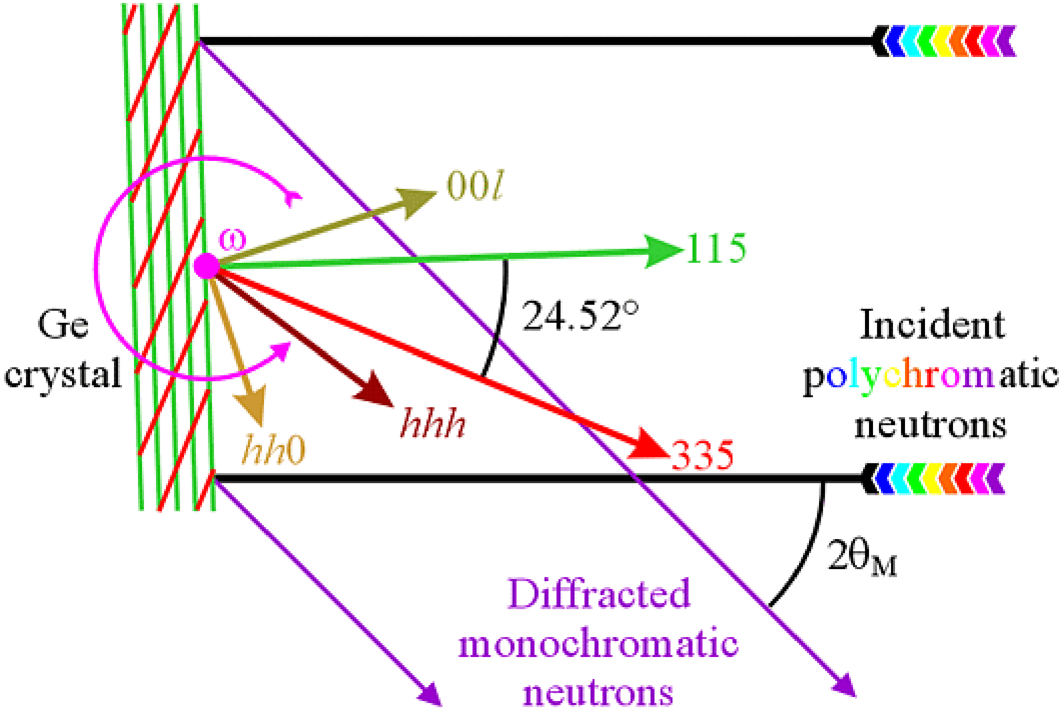
Neutron Guides

- Nickel coated glass
  - Isotopic ^{58}Ni (b = 14.4 fm) improves reflectivity
  - OK for thermal neutrons
  - Excellent for cold neutrons
  - Unsuitable for hot neutrons

Monochromators

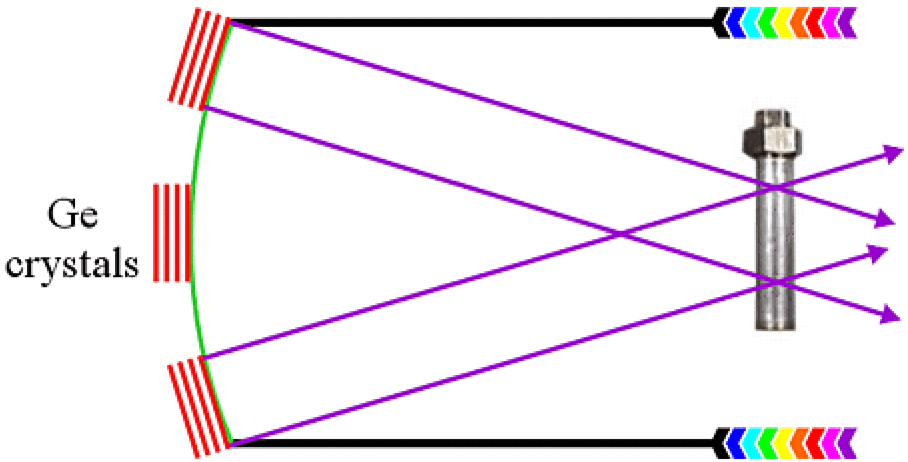
- White Beam with no Time Structure
  - Require monochromators (Ge, C, Cu) for single λ
  - Single bounce – Rotate to obtain different *d* spacings





Focussing

- Relatively Low Flux at Sample
  - ➢ Use vertically-focussing monochromators
  - ➢ Increase angular divergence
    - – Lower resolution



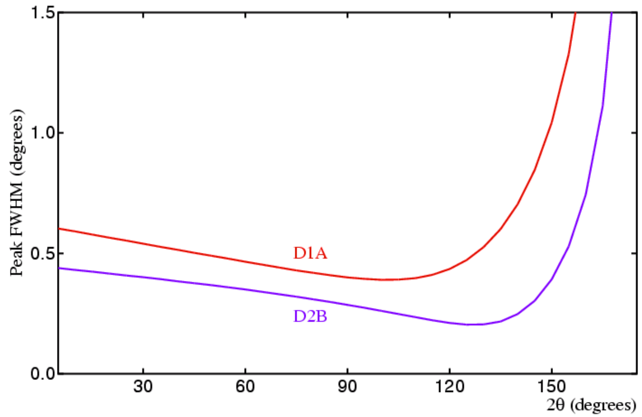
Instrumentation

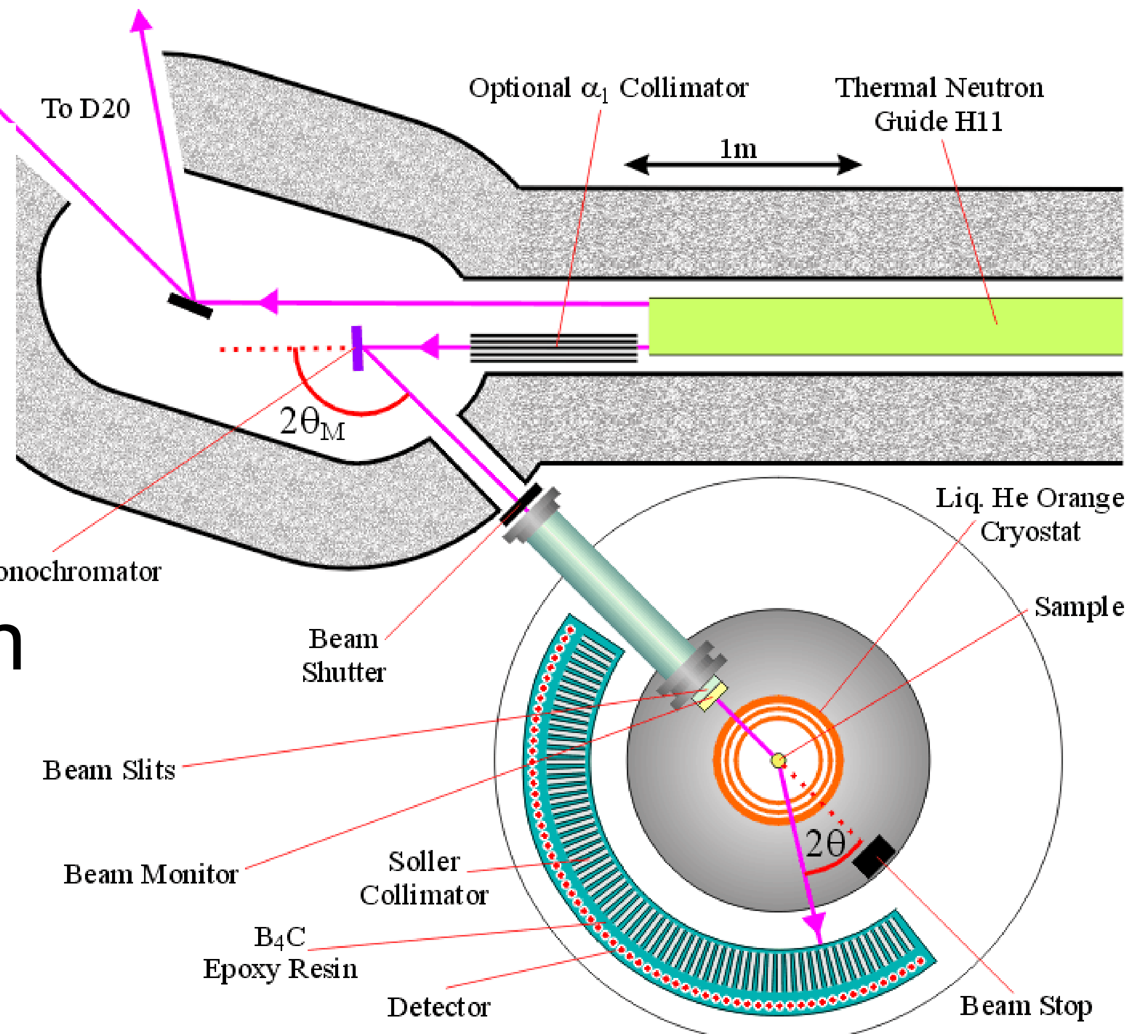
- Single Crystal Diffraction at ILL
  - D9 (Hot)
  - D19 & Vivaldi (Thermal)
  - D10 & D15 (Thermal, plus additional features)
  - D16 (Cold, plus powder diffraction)
- Powder Diffraction at ILL
  - D2B, D1A (Thermal, high resolution)
  - D20, D1B (Thermal, high flux)

Single-Crystal Neutron

- Comparison with X-rays
  - Relative large crystals (10s mm^3 – 1 mm^3)
  - Use χ-circles
  - Modernised instruments have big detectors
    - PSD or image plate
    - Older ones have point detectors
  - Some experiments require manual setting crystal orientation
    - Understanding UB orientation matrices helps
    - Some data sets still collected one reflection at a time
  - Data acquisition times are long

Powder Diffraction



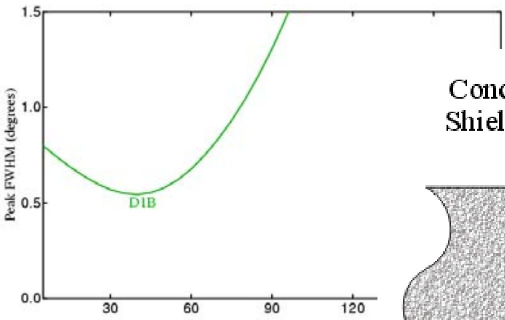


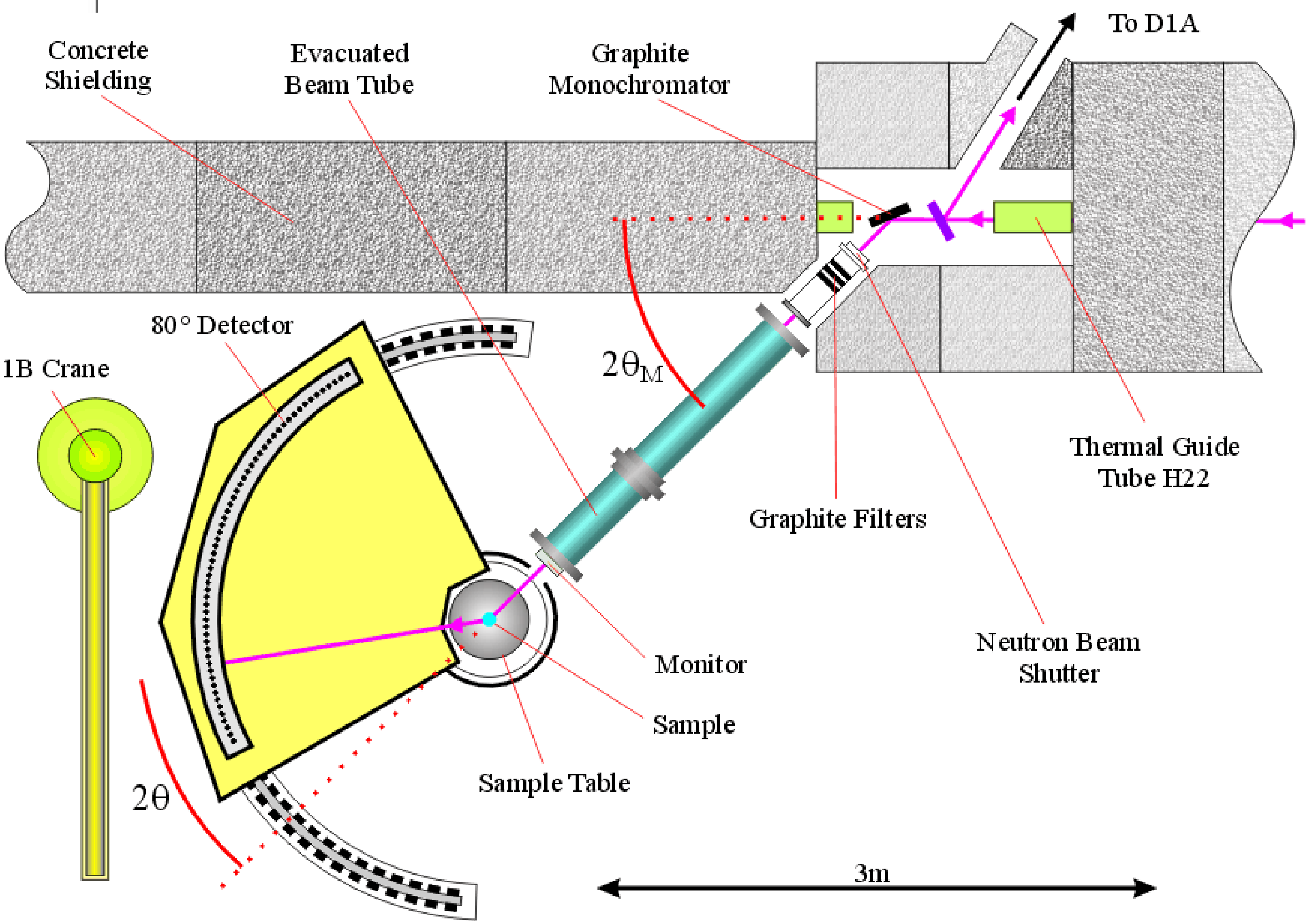
High Resolution

Improving Statistics

- 1st Generation
  - (1960s to early 1970s)
  - ➢Single Point Detectors
- 2nd Generation
  - (Late 1970s to early 1990s)
  - ➢Multiple Detectors
- 3rd Generation
  - (Late 1990s to present day)
  - ➢Sophisticated PSDs
    - Delay line ^{3}He tubes

Powder Diffraction

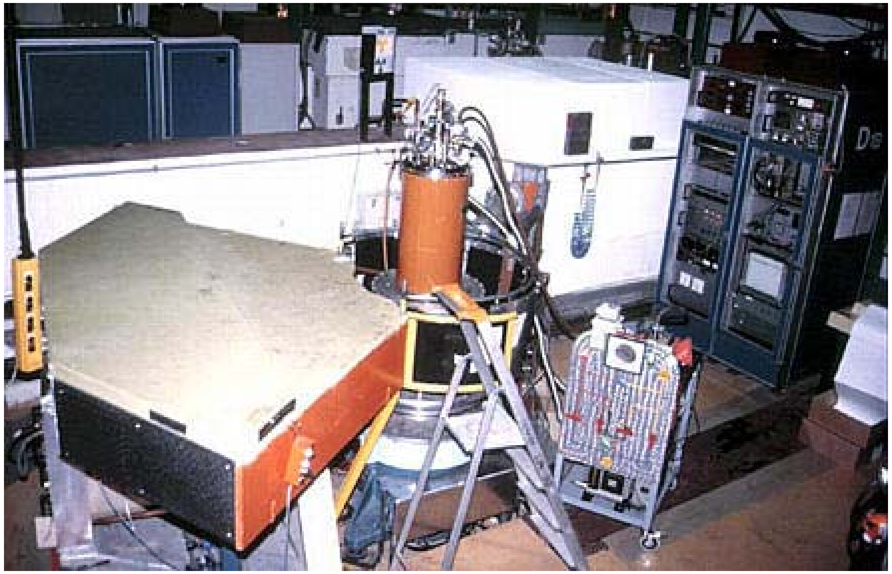




High Flux

View of Old High Flux P.D.

- Instrument with the ILL's best publication record these last 5 years!
  - Now a CRG instrument



Sample Holders

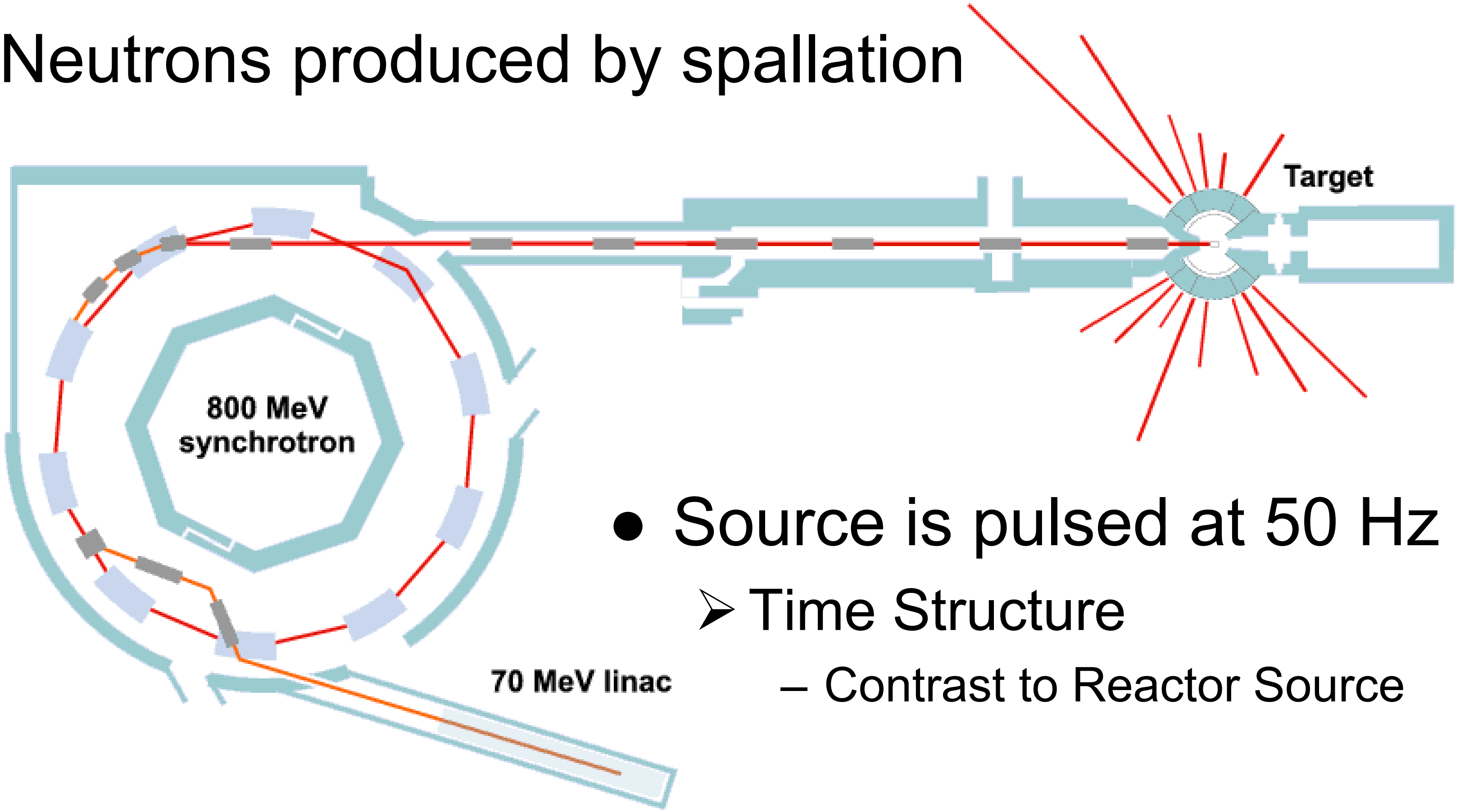
- Vandium Cans or Quartz (not Glass) Tubes
- Relatively Large Quantities of Powder

Pulsed Neutron Sources

- Pulsed Neutron Sources share some similarities to Synchrotron Sources
  - Both accelerate particles in a ring
- Use Charged Heavy Particles e.g. H^- ions
  - Accelerate in a linear accelerator (linac)
- Strip Electrons to produce H^+ beam
- Accelerate to High Energy in Storage Ring
- Collide with Heavy Metal Target (water cooled)
  - Ta, U, W
  - Produce smaller nuclei plus neutrons

Pulsed Neutron Sources

- ISIS World's Most Intense PNS
  - ➢Neutrons produced by spallation



- Source is pulsed at 50 Hz
  - ➢Time Structure
    - – Contrast to Reactor Source

TOF Concepts

- Essential to Use Time Structure of Pulsed Neutrons
  - ➢ Time-averaged flux is relatively low
- de Broglie equation $\lambda = h / mv$
  - – Planck's constant $h = 6.626\times10^{-34}$ Js
  - – Neutron mass $m = 1.675\times10^{-27}$ kg
  - ➢ Velocity, v, is inversely proportional to λ
  - ➢ In 1 ms a neutron will travel a few metres
    - – Time over a fixed path length, L, to obtain λ

TOF Concepts

- Combine de Broglie and Bragg Equations:

$$\lambda = ht \,/\, mL = 2d \sin\theta$$

- d spacing is proportional to time, t
- Timing is between start of the pulse and arrival time on the detector
  - "Time-of-Flight" (TOF)
  - Need thin detectors to reduce uncertainty in path length

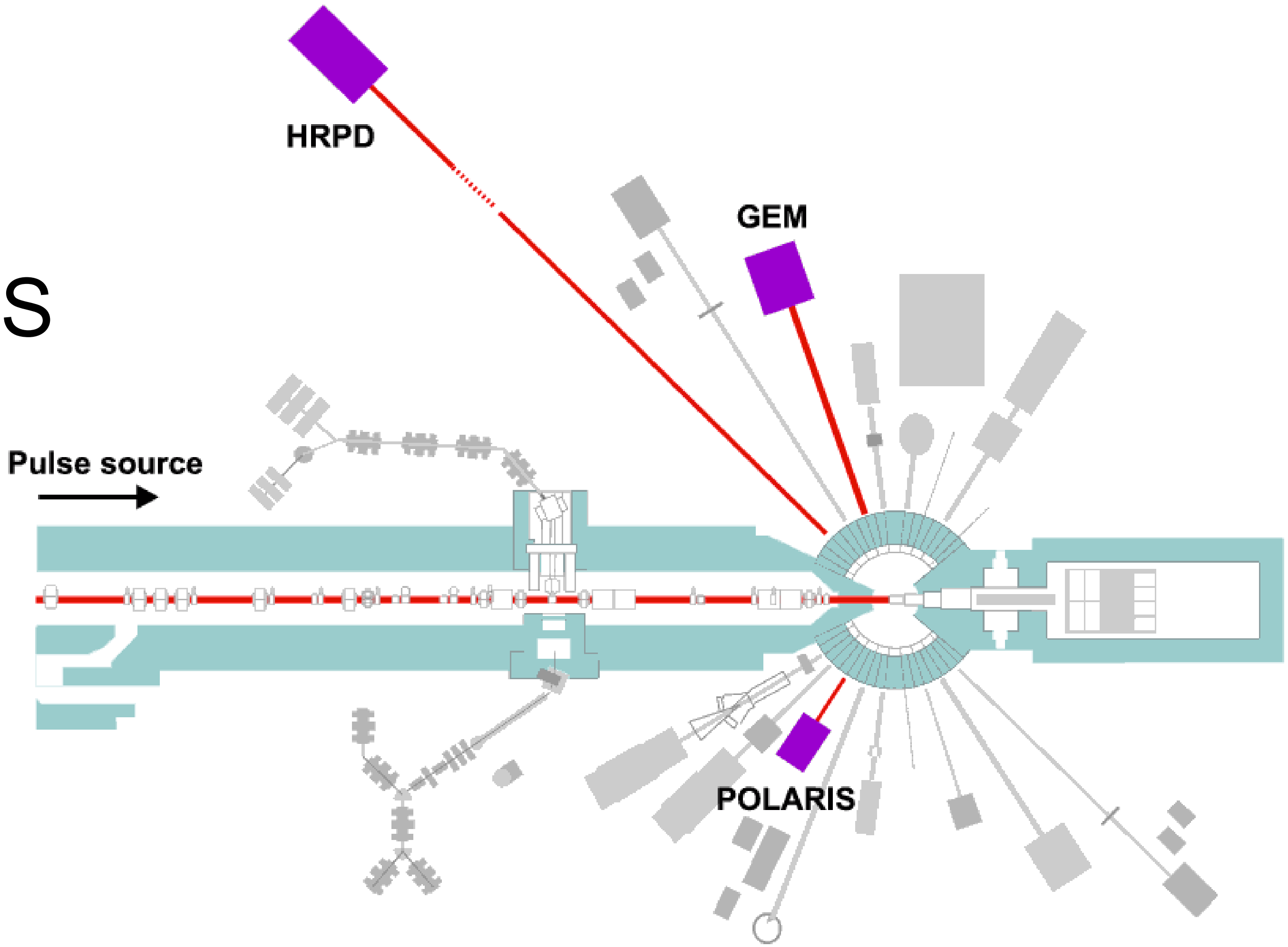
TOF Single Crystal

- SXD
  - ➢ Measure all of reciprocal space
  - ➢ Bragg peaks *plus* space between peaks
    - – Useful for some experiments

      e.g. incommensurate phase transitions

TOF Powder Instruments

- HRPD
- GEM
- POLARIS

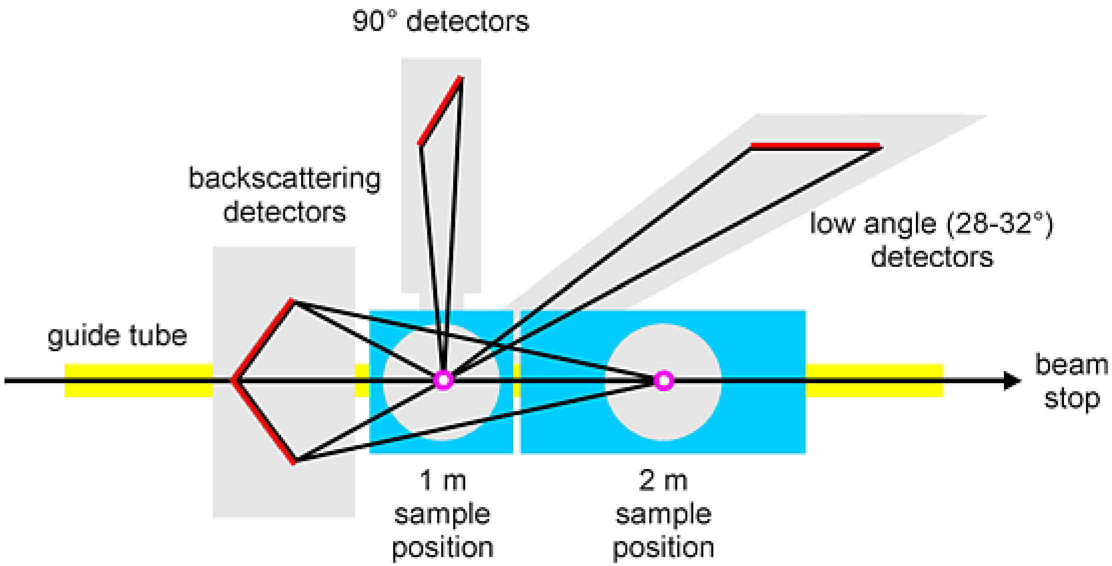


Inside ISIS Instrument Hall

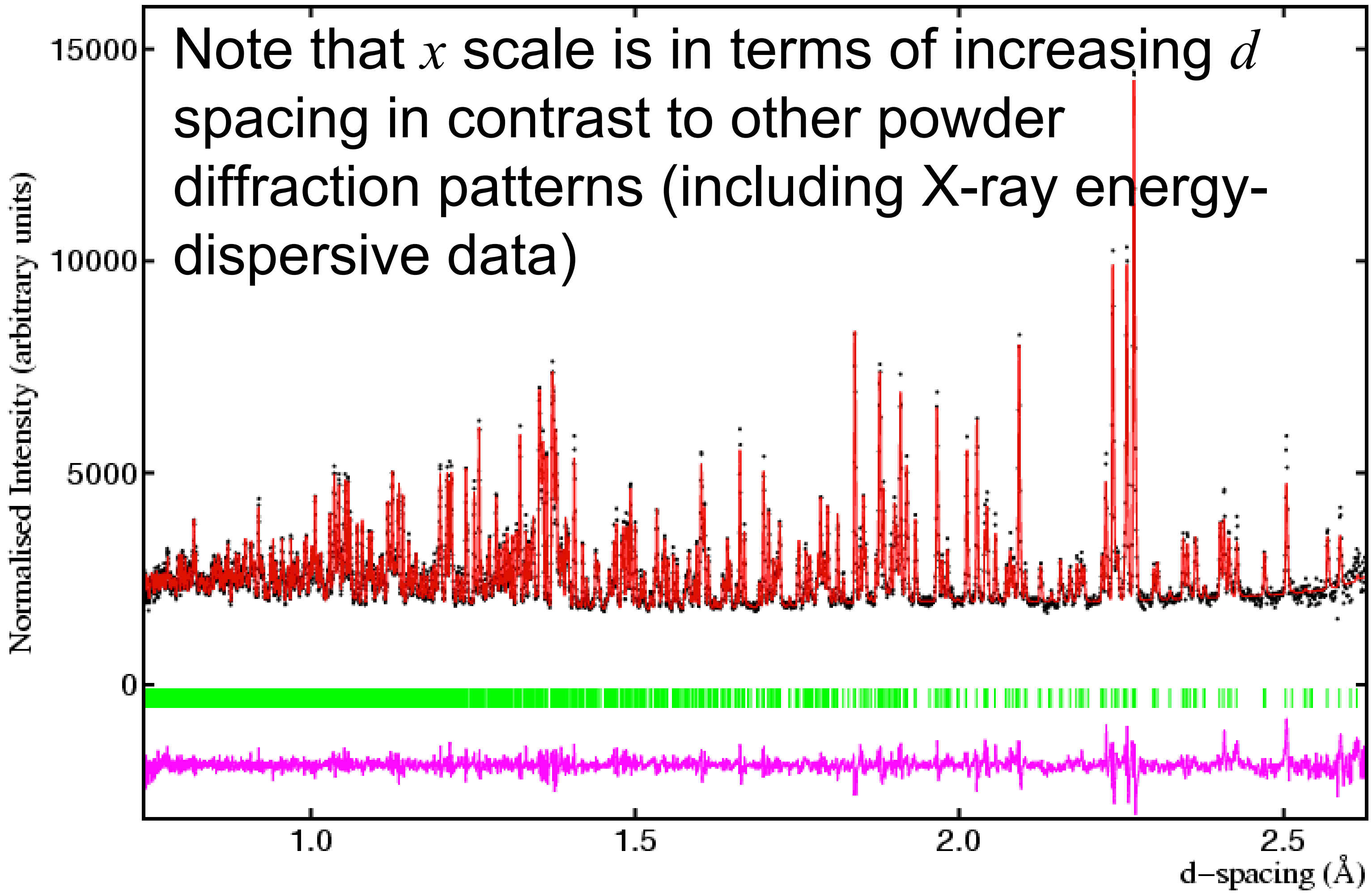
- Require Heavy Shielding – Less to See

Instrument Design

- Resolution versus Flux
  - Flight Path & Detector Angle
  - Massive Solid Area Coverage – GEM
- Resolution determined by errors in t, L, 2θ



Typical TOF Powder Data



Typical Experiments

- Single-crystal
  - Precise location of H
    - Especially in organometallics
    - Hydrogen-bonding studies
  - Precise location of nucleus
    - Needed for X − N electron density difference maps
  - High-accuracy thermal motion studies
    - No form factors
  - High-accuracy disorder studies
    - No form factors
  - Magnetic Crystal Structures

Typical Experiments

- Powders
  - Precise location of atoms including anisotropic B-values by Rietveld refinement
    - Metal oxides, e.g. ceramics, high-temperature superconductors
    - Hydrogen bonding (but with deuterated samples)
  - Solid-state phase transition studies
    - Non-magnetic and magnetic
  - Solid-state reactions
  - Texture measurements (bulk)

Typical Experiments

- Historically
  - Many experiments requiring non-ambient conditions
    - Expensive
    - Often developed by physicists
- Non-ambient now available in many chemistry laboratories
- Better done with hard synchrotron X-rays?

Question Time / Dinner

?