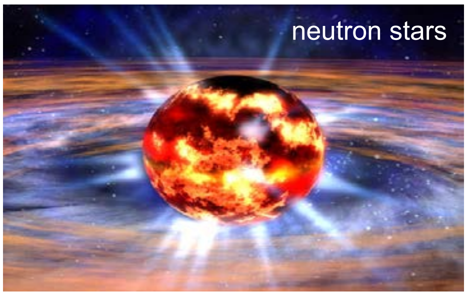
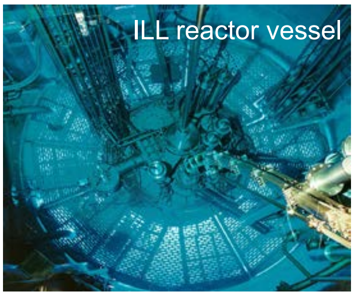
NEUTRONS + MATTER - A BRIEF GLIMPSE THROUGH THE LECTURE

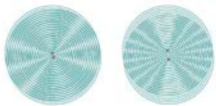




matter is made of neutrons (but not only!)

neutrons are both tools and objects for studies

thermal neuron beams: a gentle scattering probe



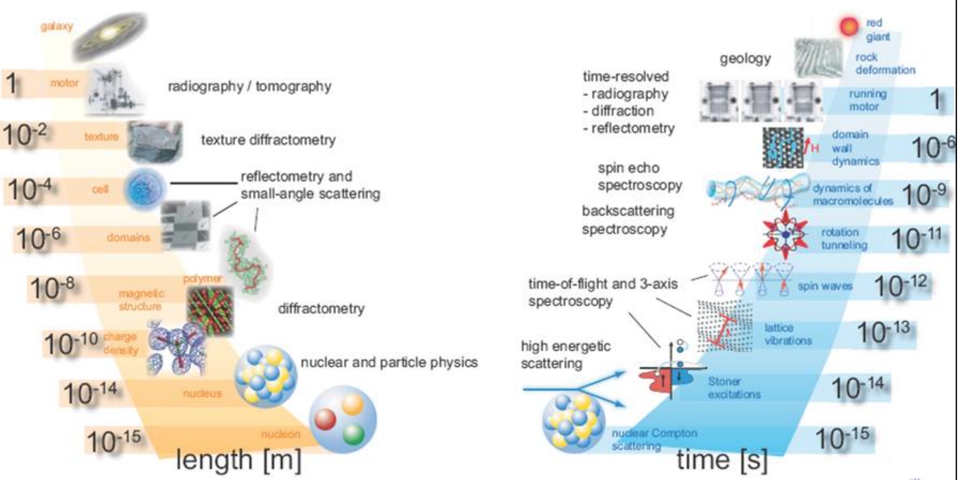
neutrons: no charge, mass close to proton – 1.675 10^{-23} kg

spin 1/2 – magnetic moment – 1.93 μ_N

NEUTRONS + MATTER - A BRIEF GLIMPSE THROUGH THE LECTURE

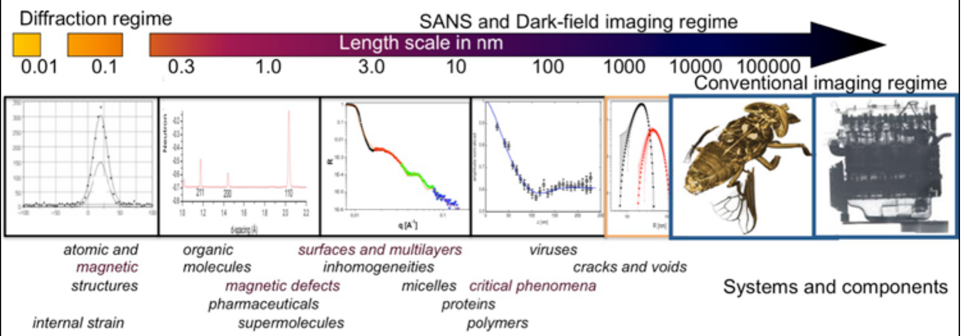
neutrons interact with nuclei and magnetic fields

they tell us where atoms & magnetic moments are and how they 'move'



NEUTRONS + MATTER - A BRIEF GLIMPSE THROUGH THE LECTURE

focusing on the thermal neutron domain – wavelength in the 0.1-1nm range

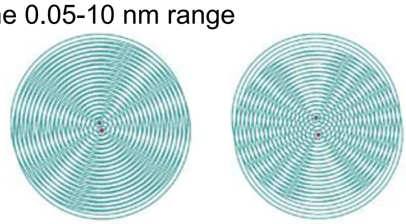


BEING A BIT MORE SPECIFIC

neutrons are massive particles - kinetic energy

useful range in the meV-eV region or below

wavelengths of associated waves in the 0.05-10 nm range



neutrons (neutral particles carrying a spin ½) interact with:

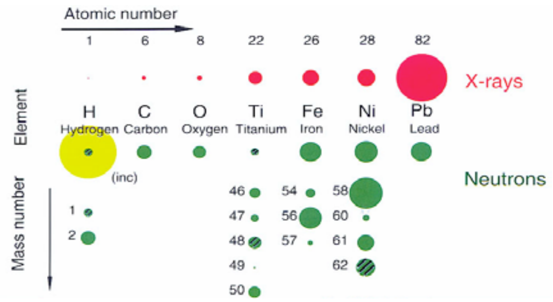
- nuclei
- magnetic densities 
- magnetic and nuclear interactions have similar strengths

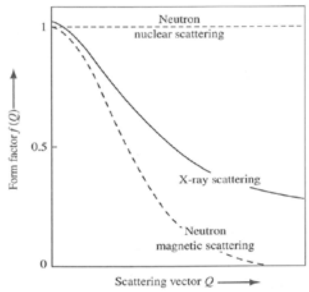
BEING A BIT MORE SPECIFIC

scattering form factors – they represent the 'shape' of scattering objects

angular dependence is different for 'nuclear' and 'magnetic' scattering

nuclear scattering is 'point-like'





random variation of nuclear 'b' across periodic table – isotopic effects

neutron and x-ray scattering amplitudes have 'similar' orders of magnitude

neutron sources are not as efficient as X-ray sources

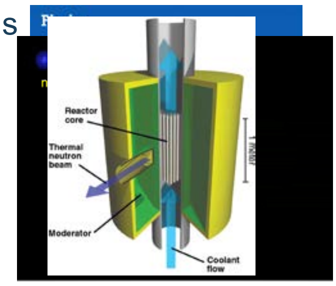
NEUTRON SOURCES - NUCLEAR REACTORS

a neutron source – we need 'free' neutrons

how does it work?

fission reactor

slow neutrons splitting nuclei



ILL reactor

D_2O moderator

hot source: graphite block 2400K

cold sources: liquid hydrogen 25K

thermal flux 1.5 10^{15} n/cm^2/sec

high power density (~1.2 MW/litre)/max achievable rate of cooling reactor core

NEUTRONS FROM ACCELERATORS

pulsed sources – to overcome cooling problems

pulsed reactors – accelerator driven sources

series of installations at Argonne Nat Lab, Rutherford Lab, KEK

ESS in construction

new projects for compact sources

photo-fission

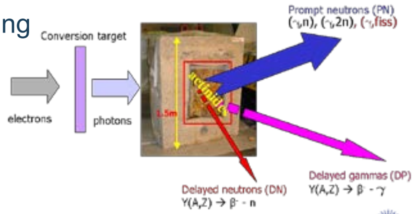
using high-energy electrons on heavy metal targets

deceleration leads to Bremsstrahlung

γ-rays produce fast neutrons
not efficient

3,000MeV per neutron

intense γ-ray beams



NEUTRON SOURCES - NUCLEAR REACTORS

pulsed reactors

pulsing reactivity to become supercritical

- mechanically (fissile materials or reflector)
- electron pulses in the core - photo-fission process
- combination of the two above.

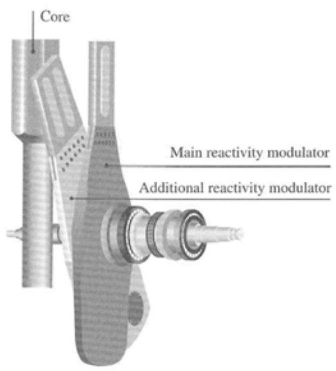
IBR-2 at Dubna, Russia

average power 2 MW

peak power 1,500 MW

rep rate 5 Hz, pulse width 215 μsec

peak flux 1.5 10^{16} n/cm^2/sec



NEUTRONS FROM ACCELERATORS

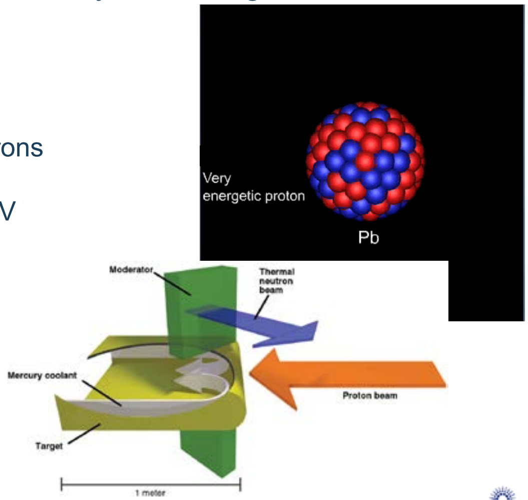
spallation sources

firing high-energy protons on heavy metal targets

'evaporation' of 30 fast neutrons

accelerator   proton 2.0 GeV

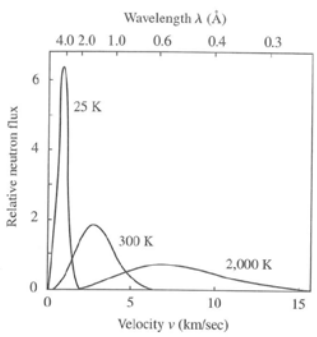
high Z metal target

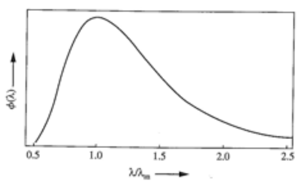


NEUTRON MODERATORS

fission/spallation neutrons are in the MeV range
moderation process
thermal equilibrium with moderator

Maxwellian spectrum if
thermal equilibrium





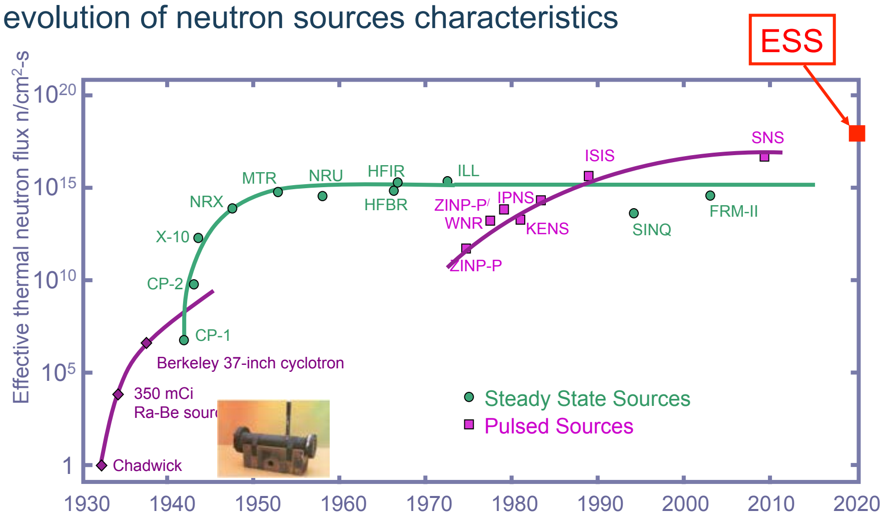
$$\lambda_m = \frac{h}{(5 m_n k_B T)^{1/2}} = \frac{19.483}{(T)^{1/2}} \ (\text{Å})$$

choice of moderator depends on applications

hot neutrons	<0.6 Å
thermal neutrons	1- 4 Å
cold neutrons	4-20 Å

NEUTRON SOURCES

evolution of neutron sources characteristics



(Updated from *Neutron Scattering*, K. Skold and D. L. Price, eds., Academic Press, 1986)

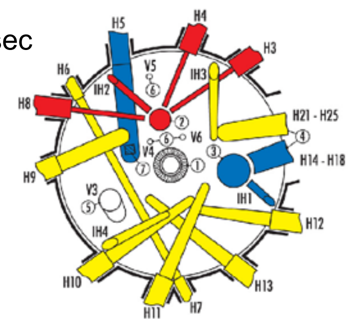
NEUTRON SOURCES

low brightness sources

$\sim 10^{14}\, n/cm^2/steradian/sec$

compared with a 60 W (900 lm) light bulb

$\sim 10^{19}\, ph/cm^2/steradian/sec$



a neutron reactor (ILL) produces $\sim 10^{19}$ n/s

usually neutron instruments receive a total of $\sim 10^{15}$ n/s

NEUTRON SOURCES

small compact sources – optimising brightness!

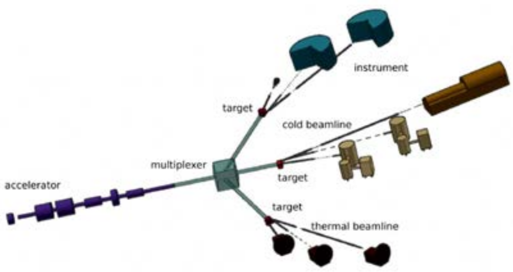
why small sources?

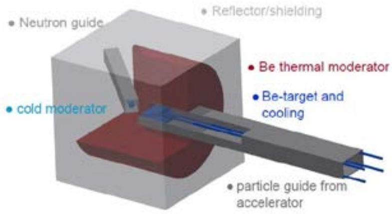
small is less expensive!

need for a network of regional sources

D^+ beams 100 kW – bombarding light element target

but 'directional' moderation





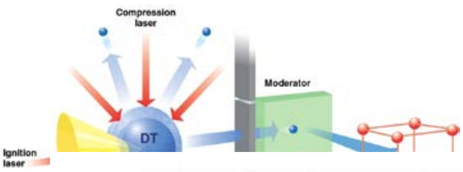
project being assessed

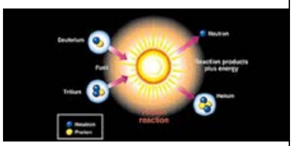
Jülich, LLB, Bilbao

http://www.fz-juelich.de/jcns/EN/Leistungen/High-Brilliance-Neutron-Source/_node.html

NEUTRON SOURCES

a 'dream' neutron source?



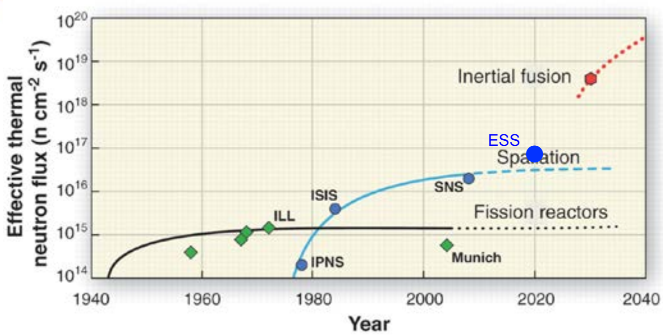


inertial fusion

D and T compressed by lasers

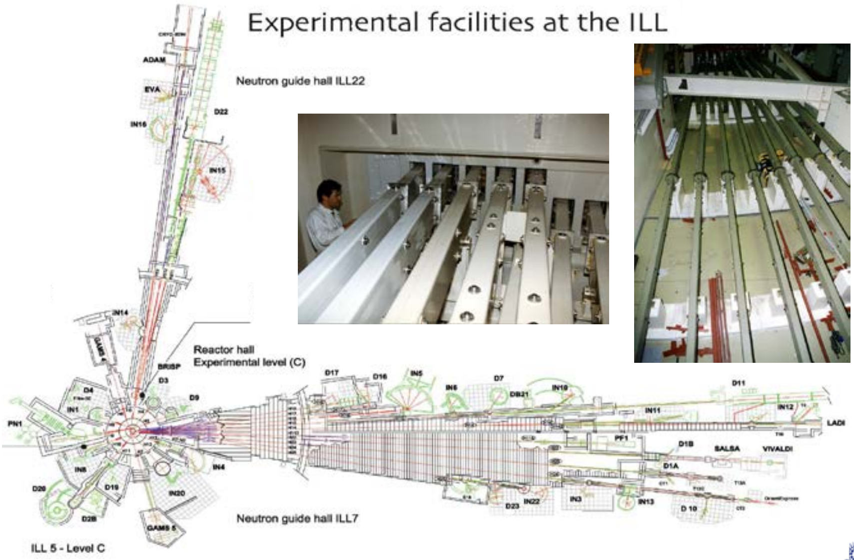
rons plus α

lear reaction



NEUTRON TRANSPORT

neutron guide halls



WHY DO WE USE NEUTRONS?

we use neutrons as probes to study matter at the atomic / molecular level

neutrons tell us about the positions and motions of atoms/magnetic moments in condensed matter:

- neutrons interact with nuclei and magnetic moments
- the two interactions have similar 'strengths'
- neutrons are penetrating: bulk materials can be studied

  sample can be placed in a special environment
- interactions with matter are gentle

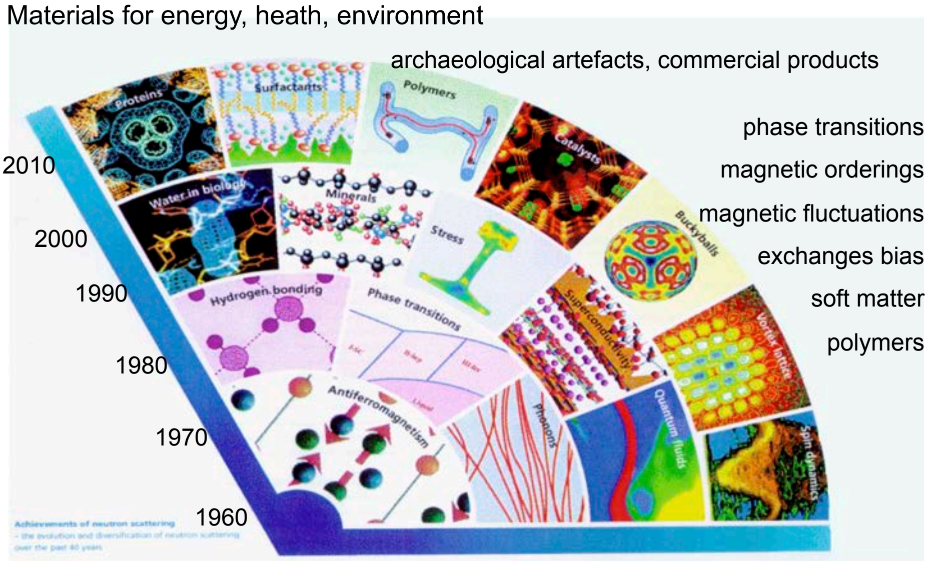
  scattering data are easy to interpret

- although neutron sources are not as efficient as X-ray sources

WHY DO WE USE NEUTRONS?

Materials for energy, heath, environment

archaeological artefacts, commercial products



phase transitions

magnetic orderings

magnetic fluctuations

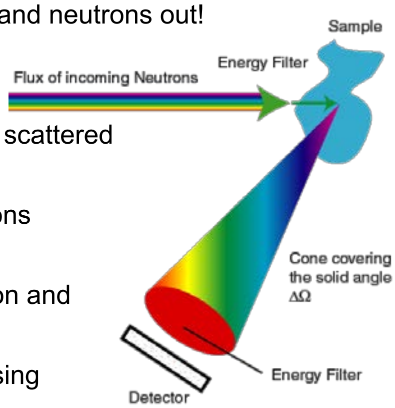
exchanges bias

soft matter

polymers

WHAT DO WE MEASURE?

Scattering experiments: neutrons in and neutrons out!



We measure the number of neutrons scattered by a sample

against the number of incident neutrons (neutron flux)

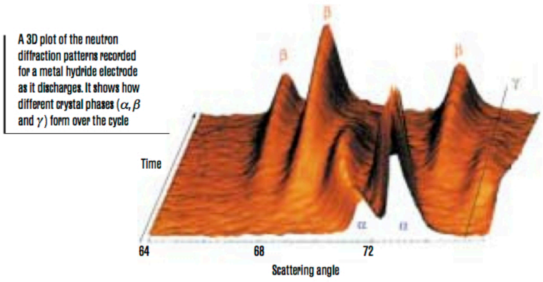
as a function of the change in direction and energy of the scattered neutrons

as a function of polarisation or polarising magnetic fields

- Scattered intensities involve positions and motions of scattering centres: atoms/magnetic moments
- Scattered intensities are proportional to Fourier transforms (in space and time) pair correlation functions

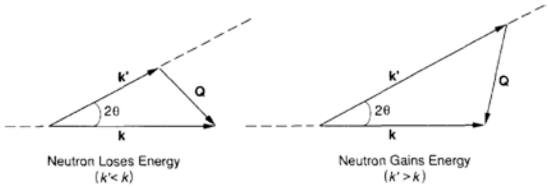
WHAT DO WE OBTAIN AS EXPERIMENTAL RESULTS?

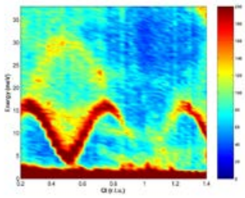
elastic scattering – diffraction experiment



time resolved powder diffraction

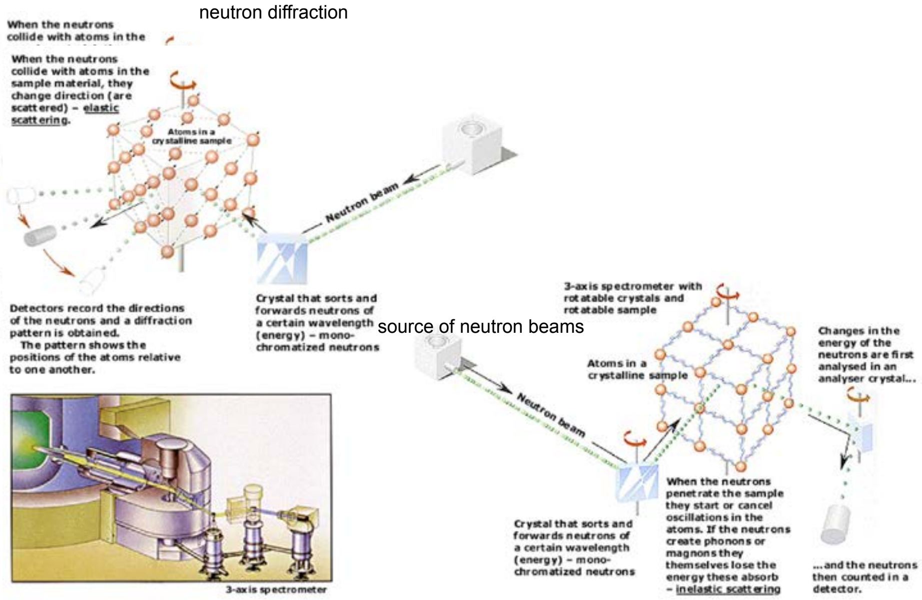
inelastic scattering: position in Q space and energy transfer





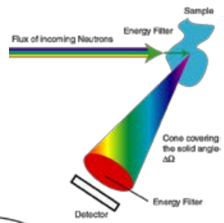
HOW DO WE MEASURE?

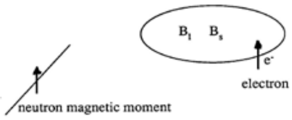
neutron diffraction

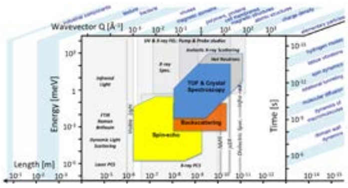


OVERVIEW OF THE LECTURE

- a bit history
- neutron properties
- interactions between neutrons and matter
- measured quantities
- scattering by atoms/nuclei
- scattering by magnetic moments
- key messages







NEUTRONS: INTRODUCTION

A bit of history:

W. Bothe & H. Decker -1930

discovered very penetrating radiation emitted when α particles hit light elements

I. Curie & F. Juliot -1932

observed creation of p^+ in paraffin sheets & thought new radiation was γ-rays

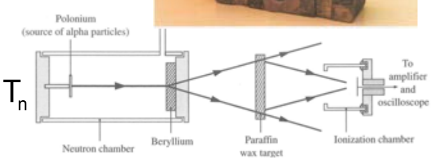
J. Chadwick -1932 a few months later

discovers the 'neutron', a neutral but massive particle

Nobel Prize in Physics $\quad {}_2^4\mathrm{He} + {}_4^9\mathrm{Be} \rightarrow {}_6^{12}\mathrm{C} + {}_0^1\mathrm{n}$

$${}_2^4\mathrm{He} + {}_5^{11}\mathrm{B} \rightarrow {}_7^{14}\mathrm{N} + {}_0^1\mathrm{n}$$

$$(m_{He} + m_B)c^2 + T_{He} = (m_N + m_n)c^2 + T_N + T_n$$



$$m_n = 1.0067 \pm 0.0012 \text{ a.m.u}$$

NEUTRONS: INTRODUCTION

A bit of history:

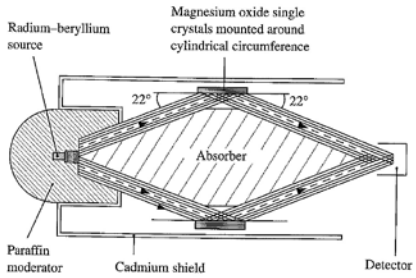
E. Fermi showed that neutrons moderated by paraffin could be captured by various elements, producing artificial radioactive nuclei

importance of neutron energy range

D.P. Mitchell & N. Powers / H. v. Halban & P. Preiswerk -1936

showed that thermal neutrons can be diffracted by crystalline matter

MgO crystals oriented (200) planes
22°corresponds to Bragg angle for peak of wavelength distribution of thermal neutrons
~0.16nm



NEUTRONS: INTRODUCTION

A bit of history:

• O. Hahn, F. Strassmann & L. Meitner -1938

discovered the fission of ^{235}U nuclei through thermal neutron capture

• H. v. Halban, F. Joliot & L. Kowarski -1939

showed that ^{235}U nuclei fission produced 2.4 n^0 on average – chain reaction

• E. Fermi & al. -1942

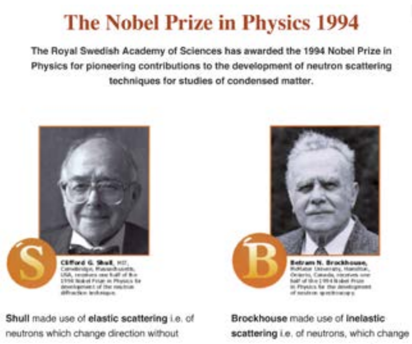
first self-sustained chain reaction reactor CF

• C.G. Shull -1949

Proof of antiferromagnetic order in M

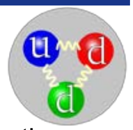
• C.G. Shull & B.N. Brockhouse -1994

Nobel Prize in Physics



NEUTRONS: NEUTRON PROPERTIES

free neutrons are unstable: β-decay proton, electron, anti-neutrino



life time: two values 888 ± 2.1 sec and 880 ± 0.6 sec !!!

wave-particle duality: neutrons have particle-like and wave-like properties

- mass: m_n = 1.675 x 10^{-27} kg = 1.00866 amu. (unified atomic mass unit)
- charge = 0
- spin =1/2 magnetic dipole moment: $\mu_n = -1.913\ \mu_N$
- velocity (v) kinetic energy (E) temperature (T) wavevector (k) wavelength (λ)

$$E = m_n v^2/2 = k_B T = (hk/2\pi)^2/2m_n \qquad k = 2\pi/\lambda = m_n v/(h/2\pi)$$

$$\lambda\,(\mathrm{nm}) = 395.6/v(\mathrm{m/s}) = 0.0286/(E)^{1/2}\ (E\ \mathrm{in\ eV}) \qquad 1\,(\mathring{A}) \approx 82\ \mathrm{meV} \approx 124\ \mathrm{THz} \approx 950\ \mathrm{K}$$

$$E\ (\mathrm{meV}) = 0.02072\ k^2\ (k\ \mathrm{in\ nm^{-1}})$$

$$T = \frac{L}{v} = 252.77\ \mu\mathrm{sec} \cdot \lambda\left[\mathring{A}\right] \cdot L[m]$$

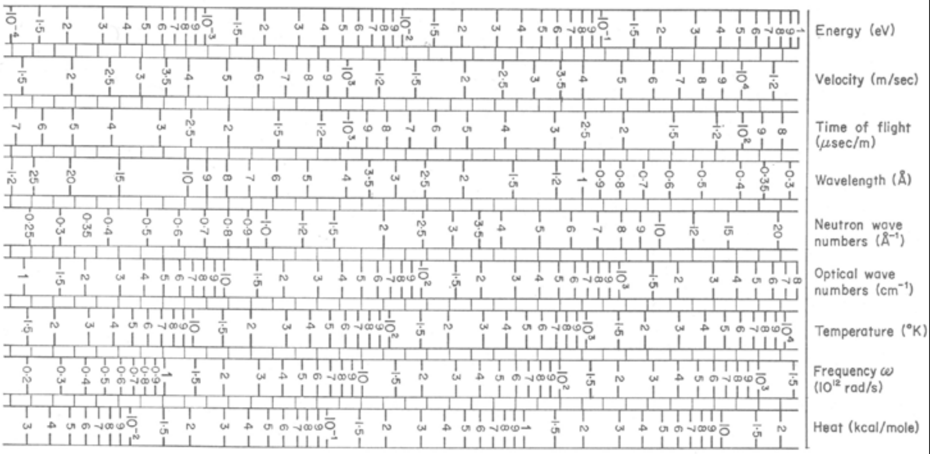
Example λ = 4 Å v=1000m/s
E= 5 meV

fortunately large value!

monochromatisation: diffraction or time of flight

NEUTRONS: NEUTRON PROPERTIES

Conversion chart

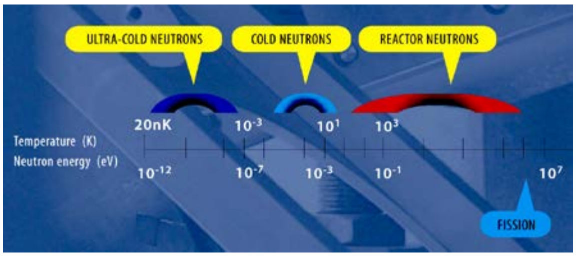


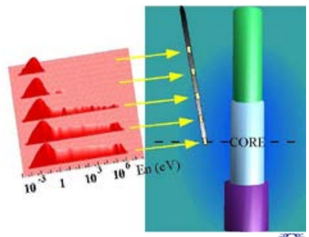
P. A. Egelstaff ed. - Thermal Neutron Scattering Academic Press 1965

NEUTRONS: NEUTRON PROPERTIES

Neutron energy ranges

	Energy	Temperature (K)	Wavelength (nm)	velocity (m/s)
Ultra cold neutrons	< 10 µeV	< 0.05	> 30	< 15
Cold neutrons	100 - 5000 µeV	1 - 60	0.4 - 3	150 - 1000
Thermal neutrons	5 - 50 meV	60 - 600	0.13 - 0.4	1000 - 4000
Hot neutrons	0.05 - 0.5 eV	600 - 6000	0.04 - 0.13	4000 - 10000
Epi-Cadmium neutrons	0.5 - 1 eV	> 6,000	< 0.005	> 13 km/s
"Slow" neutrons	1 - 10 eV			
Resonance neutrons	10 - 300 eV			
Intermediate neutrons	0.3 - 1 MeV			
Fast neutrons	1 - 20 MeV			
Relativistic neutrons	> 20 MeV			





ULTRA-COLD NEUTRONS

the very cold side

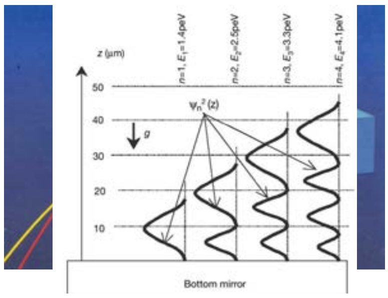
v ≈ 20m/s E_{kin} ≈ 2 μeV T= 0.023 K

λ = 200 Å

effect of gravity - neutrons are massive!

mirror ~ potential well for ultra-cold neutrons

neutrons are 'stacked' at distinct height levels (in the micrometer range!)

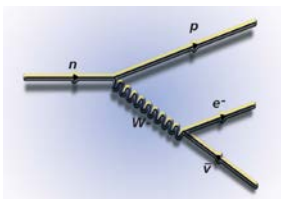


note: cold neutron beams are bent by gravity ~ 1.2 cm at 100 m for 20 Å neutrons

neutrons : objects to study fundamental interactions

neutron β-decay

free neutrons are not forever



KEY MESSSAGES

- neutrons are not elementary particles
- they are not for ever
- neutrons are not only a powerful probe, they can be studied as objects

NEUTRON SCATTERING: INTERACTIONS

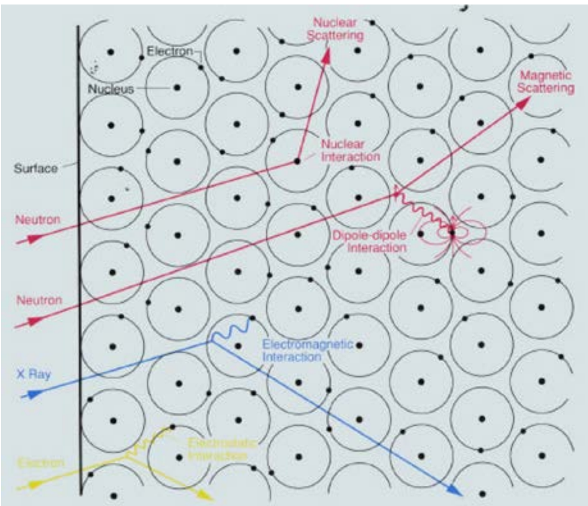
Neutron scattering exploits 'cool' neutrons: 0.05 meV< E_n < 500 meV

0.4 Å < λ < 40 Å

Neutrons

X-rays

Electrons



R. Pynn

Neutron scattering – a primer

NEUTRON SCATTERING: INTERACTIONS

Neutrons interact with nuclei:

capture: absorption, emission of particles

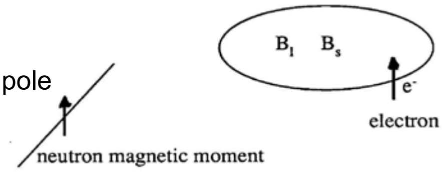
diffusion arising from very short range nuclear forces

neutron wavelength much longer than nucleus size

they can't solve nuclear structure

Neutrons interact with electrons:

magnetic interactions dipole-dipole



'Interactions' lead to 'scattering'

Strength of interactions? Measured through 'scattering cross sections'

Nuclear and magnetic neutron scattering terms have 'similar strength'

Neutrons have no charge, but they interact (extremely weakly) with charges/electrical fields spin-orbit/Schwinger scattering

see non-resonant magnetic X-ray scattering

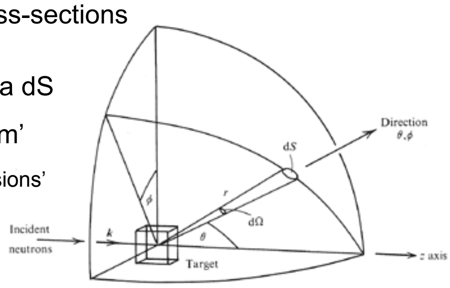
NEUTRON SCATTERING: WHAT DO WE MEASURE?

Scattering is measured in terms of cross-sections

neutron counter set up (θ, Φ) with area dS

at a distance *r* from 'scattering system'

large compared compared to sample 'dimensions'



Φ = number of incident neutrons per area per second

σ = number of neutrons scattered per second / Φ an area (barns 10^{-24} cm^2)

$$\frac{d\sigma}{d\Omega} = \frac{\text{number of neutrons scattered per second into } d\Omega}{\Phi \; d\Omega}$$

$$\frac{d^2\sigma}{d\Omega dE} = \frac{\text{number of neutrons scattered per second into } d\Omega \text{ \& } dE}{\Phi \; d\Omega \, dE}$$

applies to all types of scattering events

we have ignored the initial and final neutron spin states – to be seen later

HOW NEUTRONS INTERACT WITH MATTER – NUCLEAR SCATTERING

nuclear scattering from a single (fixed) nucleus

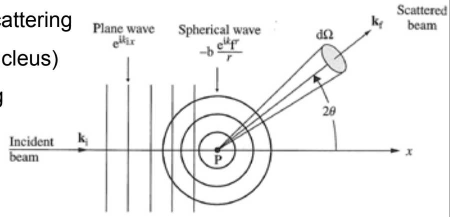
range of nuclear forces << neutron wavelength

point-like scattering so-called s-wave scattering

elastic scattering (fixed nuclei and no change in nucleus)

same velocity before and after scattering

no absorption (far away from resonance)



incident plane wave

$e^{i\mathbf{k}_i.x}$ incident flux = neutron density x velocity = v = $\frac{\hbar}{m_n}k$ neutron density = $\left|e^{i\mathbf{k}_i.x}\right|^2$

spherical scattered wave $-b\,\frac{e^{i\mathbf{k}_f.\mathbf{r}}}{r}$ b : scattering length $V(\mathbf{r}) = \frac{2\pi\hbar^2}{m_r}\,b\,\delta(\mathbf{r})$

of the order of nucleus's 'size'

number of neutrons per second into $d\Omega$ $\quad v\left|\Psi_{scatt}\right|^2 r^2\, d\Omega = v\,\frac{b^2}{r^2}\, r^2\, d\Omega = v\; b^2\, d\Omega$

cross sections $\frac{d\sigma}{d\Omega} = b^2 \qquad \sigma = 4\pi b^2$ b expressed in *fm* 10^{-15}m

HOW NEUTRONS INTERACT WITH MATTER – NUCLEAR SCATTERING

orders of magnitude:

nuclear scattering lengths, b's, depend on isotope, nuclear eigenstate, and nuclear spin orientation relative to neutron spin

Nuclide	Combined spin	b/fm	Nuclide	Combined spin	b/fm
^{1}H	1	10.85	^{23}Na	2	6.3
	0	−47.50		1	−0.9
^{2}H	$\frac{3}{2}$	9.53	^{59}Co	4	−2.78
	$\frac{1}{2}$	0.98		3	9.91

nuclear scattering from an assembly of nuclei:

atom at $\mathbf{R}_i$ incident wave: $e^{i\mathbf{k}_i.\mathbf{R}_i}$ scattered wave at $\mathbf{r}$: $\quad e^{i\mathbf{k}_i\cdot\mathbf{R}_i}\left[-b_i\,\frac{e^{i\mathbf{k}_f.(\mathbf{r}-\mathbf{R}_i)}}{\left|\mathbf{r}-\mathbf{R}_i\right|}\right]$

cross section $\quad \Psi_{scatt} = \sum_i e^{i\mathbf{k}_i\cdot\mathbf{R}_i}\left[-b_i\,\frac{e^{i\mathbf{k}_f.(\mathbf{r}-\mathbf{R}_i)}}{\left|\mathbf{r}-\mathbf{R}_i\right|}\right]$

$$\frac{d\sigma}{d\Omega} = \frac{vdS\left|\Psi_{scatt}\right|^2}{vd\Omega} = \frac{dS}{d\Omega}\left|e^{i\mathbf{k}_f.\mathbf{r}}\sum_j b_j\left[\frac{e^{i(\mathbf{k}_i-\mathbf{k}_f).\mathbf{R}_j}}{\left|\mathbf{r}-\mathbf{R}_j\right|}\right]\right|^2 = \sum_{i,j} b_i^* b_j e^{-i(\mathbf{k}_i-\mathbf{k}_f).(\mathbf{R}_i-\mathbf{R}_j)}$$

wavevector transfer $\boldsymbol{K}$ is defined by $\boldsymbol{K} = \mathbf{k}_i - \mathbf{k}_f$ $\quad \frac{d\sigma}{d\Omega} = \sum_{i,j} b_i^* b_j e^{-i\boldsymbol{K}.(\mathbf{R}_i-\mathbf{R}_j)}$

beware! X-ray boys use different sign convention!

Fourier transform

NUCLEAR SCATTERING – COHERENT/INCOHERENT

coherent and incoherent scattering

consider an assembly of similar atoms/ions – spins/isotopes are uncorrelated at different sites

$$\frac{d\sigma}{d\Omega} = \sum_{i,j \text{ averaged over all states}} b_i^* b_j e^{-iK\cdot(\mathbf{R}_i - \mathbf{R}_j)}$$

for a single nucleus $b_i = \langle b \rangle + \delta b_i$ where $\langle \delta b_i \rangle = 0$ taken as real number

$$b_i b_j = \langle b \rangle^2 + \langle b \rangle (\delta b_i + \delta b_j) + \delta b_i \delta b_j \quad \text{with } \langle \delta b_i \delta b_j \rangle = 0 \text{ unless i=j}$$

$$\langle \delta b_i^2 \rangle = \langle b_i - \langle b \rangle \rangle^2 = \langle b^2 \rangle - \langle b \rangle^2$$

$$\frac{d\sigma}{d\Omega} = \langle b \rangle^2 \sum_{i,j} e^{-iK\cdot(\mathbf{R}_i - \mathbf{R}_j)} + \left(\langle b^2 \rangle - \langle b \rangle^2\right) N \qquad \sigma_{coh} = 4\pi \langle b \rangle^2 \qquad \sigma_{incoh} = 4\pi\left(\langle b^2 \rangle - \langle b \rangle^2\right)$$

coherent scattering: correlations between different sites

incoherent scattering: correlations on the same site (at different times)

particular to neutron scattering

sources of incoherent scattering:

isotopic distribution and nuclear spin

NUCLEAR SCATTERING – COHERENT/INCOHERENT

If single isotope and zero nuclear spin, no incoherent scattering
If single isotope and non-zero nuclear spin I

nucleus+neutron spin: I+1/2 and I-1/2 scattering length b^+ and b^-

If neutrons and nuclei are un-polarised:

probability 'plus' $f^+ = \frac{I+1}{2I+1}$ probability 'minus' $f^+ = \frac{I}{2I+1}$

$$\langle b \rangle = \frac{1}{2I+1}\left[(I+1)b^+ + Ib^-\right] \qquad \langle b^2 \rangle - \langle b \rangle^2 = \frac{I(I+1)}{(2I+1)^2}(b^+ - b^-)^2$$

To reduce incoherent scattering (background):

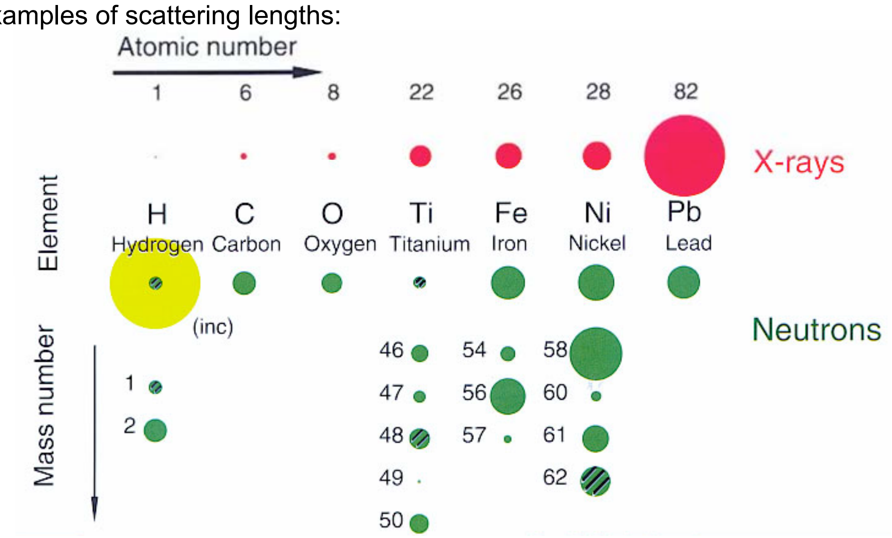
use isotope substitution

use zero nuclear spin isotopes

polarise nuclei and neutrons

NUCLEAR SCATTERING – COHERENT/INCOHERENT

examples of scattering lengths:



neighbouring elements can be easily identified

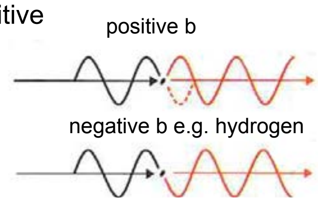
NUCLEAR SCATTERING – SCATTERING LENGTHS

most neutron scattering lengths are positive

(same for X-rays)

phase changes by after scattering

no change in phase at scattering point



ZSymbA	p or $T_{1/2}$	I	b_c	b_+	b_-	c	σ_{coh}	σ_{inc}	σ_{scatt}	σ_{abs}
0-N-1	**10.3 MIN**	**1/2**	**-37.0(6)**	**0**	**-37.0(6)**		**43.01(2)**		**43.01(2)**	**0**
1-H			**-3.7409(11)**				**1.7568(10)**	**80.26(6)**	**82.02(6)**	**0.3326(7)**
1-H-1	99.985	1/2	-3.7423(12)	10.817(5)	-47.420(14)	+/-	1.7583(10)	80.27(6)	82.03(6)	0.3326(7)
1-H-2	0.0149	1	6.674(6)	9.53(3)	0.975(60)		5.592(7)	2.05(3)	7.64(3)	0.000519(7)
1-H-3	12.26 Y	1/2	4.792(27)	4.18(15)	6.56(37)		2.89(3)	0.14(4)	3.03(5)	< 6.0E-6
2-He			**3.26(3)**				**1.34(2)**	**0**	**1.34(2)**	**0.00747(1)**
2-He-3	0.00013	1/2	5.74(7)	4.374(70)	9.835(77)	E	4.42(10)	1.532(20)	6.0(4)	5333.0(7.0)
2-He-4	0.99987	0	3.26(3)				1.34(2)	0	1.34(2)	0
3-Li			**-1.90(3)**				**0.454(10)**	**0.92(3)**	**1.37(3)**	**70.5(3)**
3-Li-6	7.5	1	2.0(1)	0.67(14)	4.67(17)	+/-	0.51(5)	0.46(5)	0.97(7)	940.0(4.0)
3-Li-7	92.5	3/2	-2.22(2)	-4.15(6)	1.00(8)	+/-	0.619(11)	0.78(3)	1.40(3)	0.0454(3)

NUCLEAR SCATTERING – CONTRAST VARIATION

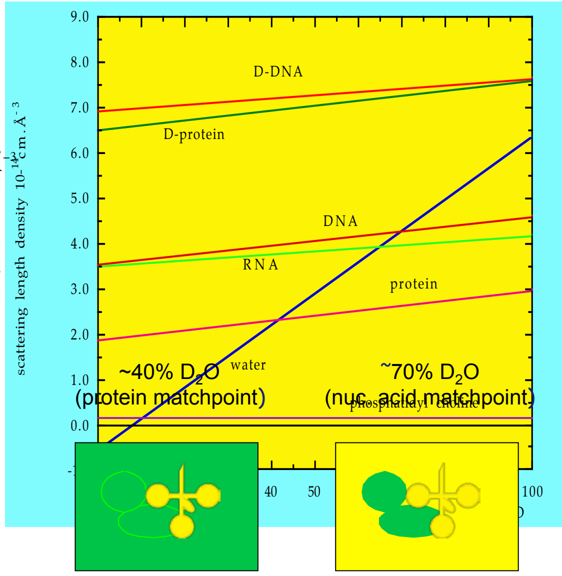
application to biology

water H_2O

$\rho = N(2b_H + 2b_O) = -0.53\,10^{-14}\,cm\,\mathring{A}^{-3}$

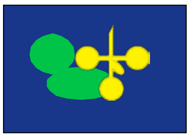
heavy water D_2O

$\rho = N(2b_D + 2b_O) = +6.3\,10^{-14}\,cm\,\mathring{A}^{-3}$



0% D_2O

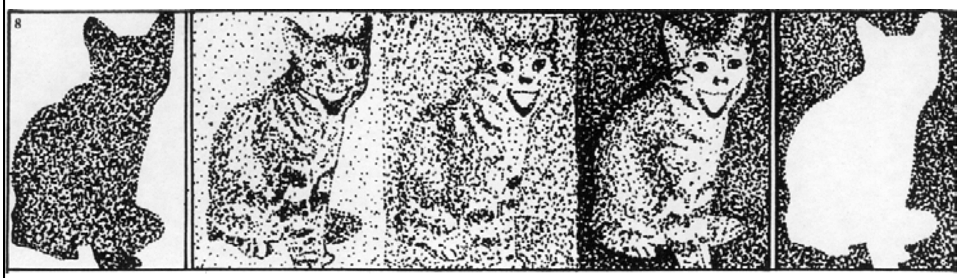
“natural” RNA/protein contrast



NUCLEAR SCATTERING – CONTRAST VARIATION

Mixing ^1H and ^2H (H and D) allows contrast variation

- full contrast : external shapes
- intermediate contrast : details



100 % H_2O → 100 % D_2O

useful for imaging and scattering (small angle neutron scattering, liquids)

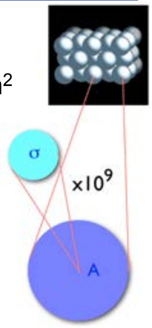
SCATTERING LENGTHS

Orders of magnitude - numbers for neutron scattering

cross sections are in ~ barns   1 barn = 10^{-24} cm^2

area per atom ~ 10 Å^2 = 10 10^8 barns

1 atom gives 10^{-9} probability scattering when beam hits it!



To obtain 1% scattering (over 4π) requires 10^7 atoms

~ 0.1 cm of sample!

Take a single atom of C: b = 6.65 pm = 6.65 10^{-15}m = 6.65$10^{-13}$cm

σ = 5.56 barns

The probability to observe a single scattered neutron per second requires a huge flux: $I_0 = \Phi\sigma \quad \Phi = I_0/\sigma = 1/\left(5.56\,10^{-24}\right) \approx 1.8\,10^{23}$ particules/cm^2/sec

actual neutron flux ≈ 10^7 n/cm^2/sec, we must either wait for

$1.8\,10^{16}$ sec $\approx$ 570 million years   or use ~2 10^{16} atoms ~ 0.4 μg only

SCATTERING LENGTHS

Numbers for neutron scattering

typical neutron flux ~10^7 n/cm^2/sec

sample volumes in the fraction of mm^3 to cm^3 range

counting time for 'incoherent scattering' from Vanadium (σ_{incoh} ~ 5 barns)

sample volume 1x1x0.1 cm^3 i.e. ~ 8.7 10^{21} atoms

count rate ~ 4 10^5 n/sec over 4π

detector angular aperture ~ 1% leads to ~ 4 10^3 n/sec

Questions about statistics:

- experimental data are 'counts in the detector', independent events but with a fixed probability (scattering cross sections!): Poisson's like
- usual goal is to achieve 1% error per information unit:
  - requires ~10,000 counts per bin
  - i.e. ~ 0.5 -10 minutes for typical elastic peak ($\frac{d\sigma}{d\Omega}$)
  - i.e. at least 10 times longer for inelastic studies ($\frac{d^2\sigma}{d\Omega dE}$)

SCATTERING LENGTHS – APPLICATIONS

Absorption

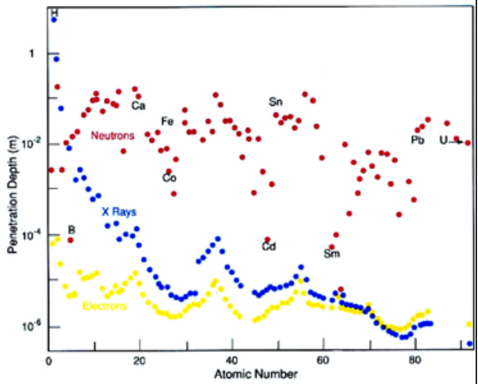
essentially neutron capture

large penetration depth (a few cm’s)

random variation with

atomic number and isotopes

energy-dependent



Applications for detection

neutrons are captured by nuclei

capture creates charged particles

recoiling particles ionise gaseous materials

$${}^{3}_{2}He + {}^{1}_{0}n \rightarrow {}^{3}_{1}H + p + 0.764\ MeV$$

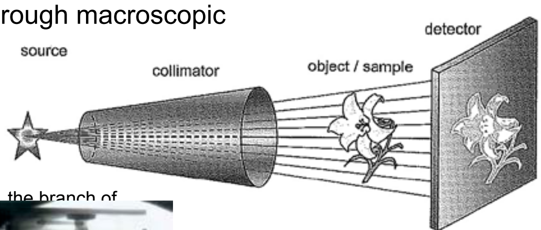
$${}^{10}_{5}B + {}^{1}_{0}n \rightarrow {}^{7}_{3}Li + {}^{4}_{2}He + 2.3\ MeV$$

SCATTERING LENGTHS – APPLICATIONS

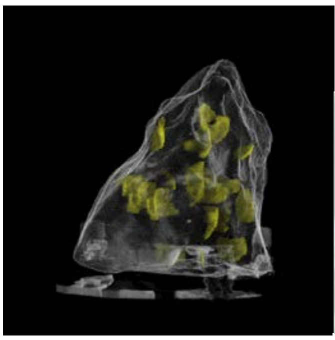
Imaging with neutrons

selectivity of neutrons – selective imaging through absorption

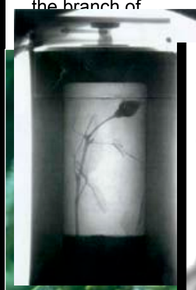
direct transmission through macroscopic

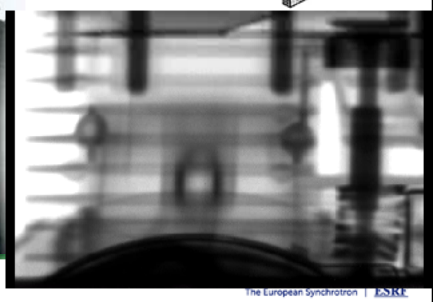


a piece of rock from the Antartic



the branch of





SCATTERING LENGTHS – APPLICATIONS

Refractive index for neutrons

For a single nucleus, Fermi pseudo-potential $V(\mathbf{r}) = \frac{2\pi\hbar^2}{m_r} b\,\delta(\mathbf{r})$

Inside matter, $\overline{V} = \frac{2\pi\hbar^2}{m}\rho$ scattering length density $\rho = \frac{1}{\text{volume}}\sum_i b_i$

neutrons obey Schrödinger's equation $\left[\nabla^2 + \frac{2m}{\hbar^2}(E - \overline{V})\right]\Psi(\mathbf{r}) = 0$

In vacuo, $\overline{V} = 0$ and $E = E_{cin}$ $k_i^2 = 2mE/\hbar^2$

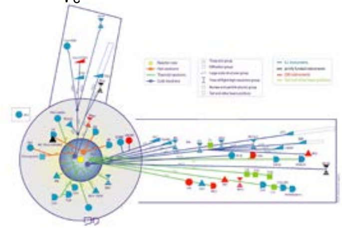
Inside the medium, $k_f^2 = 2m(E - \overline{V})/\hbar^2 = k_i^2 - 4\pi\rho$ $n = k_f/k_i \approx 1 - \lambda^2\rho/2\pi$

With b>0, n<1 and neutrons are externally reflected by most materials

SCATTERING LENGTHS – APPLICATIONS

Applications

neutron guides: critical angle $\gamma_c \approx \lambda\sqrt{\rho/\pi}$ Ni (Ni^{58}) $\gamma_c \approx 0.1^\circ Å^{-1}$

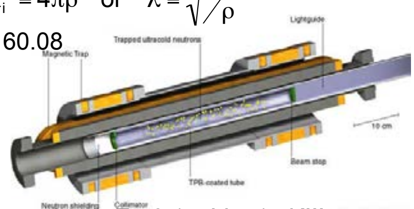


Neutron bottles: a bottle imposes n = o, $k_i^2 = 4\pi\rho$ or $\lambda = \sqrt{\pi/\rho}$

example SiO_2 density: 2.66 molecular weight: 60.08

$N = 10^{-24}(2.66/60.08)N_{Avogadro} = 0.0267\,Å^{-3}$

$\rho = N(b_{Si} + 2b_O) = 4.21 10^{-6}\,Å^{-2}$



This works with very cold neutrons: λ > 864 Å v~ 4.6 m/s E~ 0.1μeV ~ 1mK!!! Nature 403, 62 (2000)

KEY MESSAGES

- neutrons interact with nuclei – very short range interaction
    - isotropic scattering amplitude
    - random variation of b's with atomic number
    - contrast and isotopic substitution
    - coherent and incoherent scattering
    - low absorption

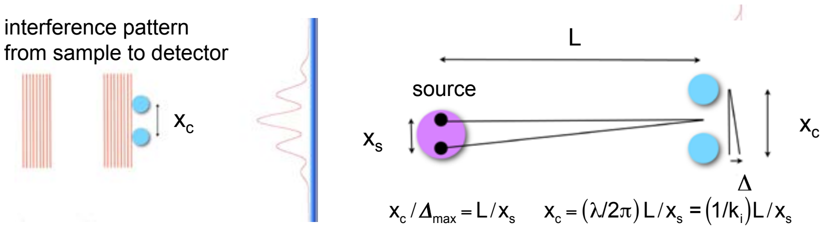
HOW TO 'ACCUMULATE' INTENSITIES?

So far, we have added individual scattering intensities. How to combine them?

neutron sources are chaotic:

emission over 4π, at ill-defined times with wide distribution of energies, neutrons are moderated

Do we have to take the neutron source into account? Coherence?



$x_c / \Delta_{max} = L / x_s \quad x_c = (\lambda/2\pi) L / x_s = (1/k_i) L / x_s$

typical values: L ~ 50-100 m x_s ~ 10cm x_c ~ y_c ~10 nm with neutrons

(X-ray tomography with pin-hole source x_c~500μm)

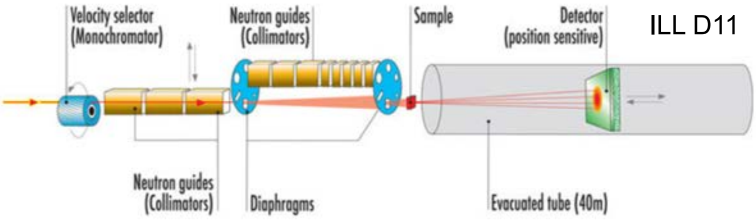
$$z_c = \lambda^2 / \Delta\lambda \approx 50 \times \lambda \approx 10 \text{ nm}$$

HOW TO 'ACCUMULATE' INTENSITIES?

another question for small angle neutron scattering:

what is the largest object that can be measured? (within the sample)

ILL D11



size given by the lateral coherence length $x_c = (\lambda/2\pi)\, L / x_s$

L~40 m, λ = 12Å, x_s~10^{-2} m (adjustable with slits) x_c ~ 1μm

Typically, objects smaller than 1μm are studied by scattering methods

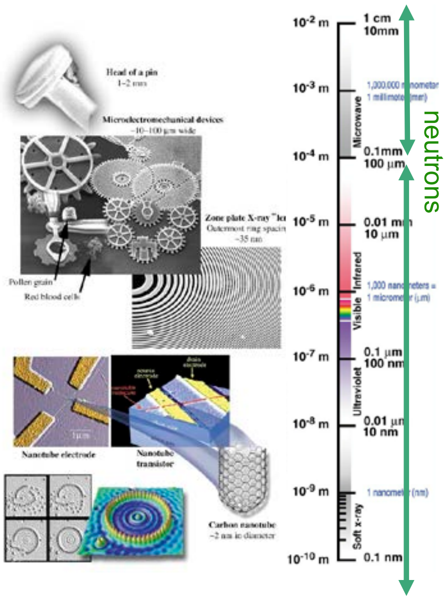
objects larger than 1μm are 'imaged'

In general, ignore coherence of neutron sources and focus on constructive interference of scattered waves

KEY MESSAGE

neutrons cover a wide range of length scales

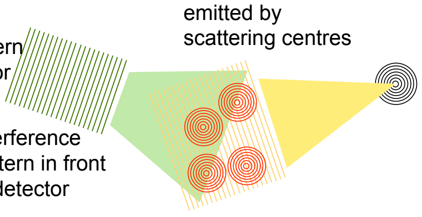
imaging/scattering



HOW TO 'ACCUMULATE' INTENSITIES?

Assume plane waves for incident neutrons

'far away' from source



How to combine scattered waves (amplitudes and phases)?

HOW TO 'ACCUMULATE' INTENSITIES?

Different ways to combine scattered waves

Born approximation – kinematic theory

neutron wavefunction un-perturbed inside sample

in general OK, away from Bragg reflections and total reflection

Dynamical theory of scattering

takes into account of the change in the neutron wave in the system

see refractive index – but generalised for all scattering vectors

important near total reflection

may be needed near Bragg reflections from perfect crystals with highly collimated beams

In most cases, kinematic theory applies for neutrons

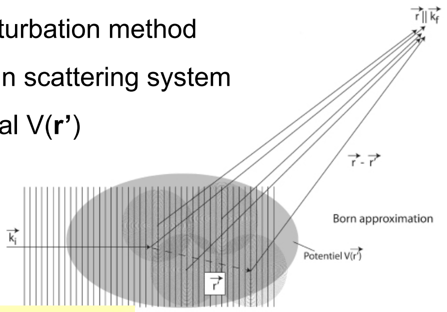
HOW TO 'ACCUMULATE' INTENSITIES?

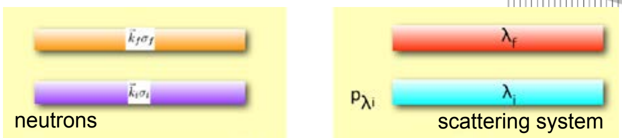
Born approximation, i.e. first order perturbation method

The incident wave travel unperturbed in scattering system

scattering centres with potential V(**r'**)

From the definition, $\frac{d\sigma}{d\Omega} = \frac{\sum_{k_f\, in\, d\Omega} W_{k_i,\lambda_i \to k_f,\lambda_f}}{\Phi\ d\Omega}$





Use Fermi's Golden rule to calculate transition probabilities $\sum_{k_f\, in\, d\Omega} W_{k_i,\lambda_i \to k_f,\lambda_f}$

$$\sum_{k_f\, in\, d\Omega} W_{k_i,\lambda_i \to k_f,\lambda_f} = \frac{2\pi}{\hbar}\rho_{\mathbf{k}_f}\left|\langle \mathbf{k}_f\lambda_f|V|\mathbf{k}_i\lambda_i\rangle\right|^2$$

where $\rho_{\mathbf{k}_f}$ is the density of **k**-states in dΩ per unit energy range

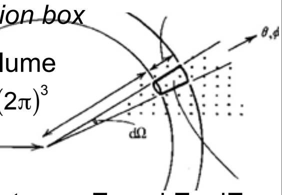
HOW TO 'ACCUMULATE' INTENSITIES?

some algebra and manipulations

$e^{i k_i . x}$ as incident wavelength - plane wave in *normalisation box*

neutron states are periodic in **k**-space, unit cell volume

neutron flux $\Phi = \frac{\hbar}{m_n} k$ $\qquad V_k = (2\pi)^3$



$\rho_{\mathbf{k}_f} dE_f = \frac{1}{V_k} k_f^2 dk_f d\Omega$ is the number of n-states in dΩ between E_f and E_f+dE_f

kinetic energy: $dE_f = \frac{\hbar^2}{m_n} k_f dk_f$ therefore: $\rho_{\mathbf{k}_f} = \frac{1}{(2\pi)^3} k_f \frac{m_n}{\hbar^2} d\Omega$

scattering cross section with $\mathbf{k}_i$, λ_i and λ_f fixed $\left(\frac{d\sigma}{d\Omega}\right)_{\lambda_i \to \lambda_f} = \frac{k_f}{k_i}\left(\frac{m_n}{2\pi\hbar^2}\right)^2 \left|\langle \mathbf{k}_f\lambda_f|V|\mathbf{k}_i\lambda_i\rangle\right|^2$

energy of neutrons+ scattering system must be conserved $E_i + E_{\lambda_i} = E_f + E_{\lambda_f}$

$$\left(\frac{d^2\sigma}{dE_f\, d\Omega}\right)_{\lambda_i \to \lambda_f} = \frac{k_f}{k_i}\left(\frac{m_n}{2\pi\hbar^2}\right)^2 \left|\langle \mathbf{k}_f\lambda_f|V|\mathbf{k}_i\lambda_i\rangle\right|^2 \delta\left(E_i - E_f + E_{\lambda_i} - E_{\lambda_f}\right)$$

partial differential cross section for all scattering potentials

DIFFERENTIAL SCATTERING CROSS SECTION – NUCLEAR SCATTERING

more algebra and manipulations

insert a potential V – Fermi pseudo-potential - short range, scalar and central

V has the form $V=\sum_j V_j(\mathbf{r}-\mathbf{R}_j)=\sum_j V_j(\mathbf{x}_j)$ with $\mathbf{x}_j=\mathbf{r}-\mathbf{R}_j$

$$\langle \mathbf{k}_f\lambda_f|V|\mathbf{k}_i\lambda_i\rangle=\sum_j\int \chi^*_{\lambda_f}\exp(-i\mathbf{k}_f\cdot\mathbf{r})V(\mathbf{x}_j)\chi_{\lambda_i}\exp(i\mathbf{k}_i\cdot\mathbf{r})\,d\mathbf{R}_1 d\mathbf{R}_2 d\mathbf{R}_3\ldots d\mathbf{R}_j\ldots d\mathbf{r}$$

$$\langle \mathbf{k}_f\lambda_f|V|\mathbf{k}_i\lambda_i\rangle=\sum_j V_j(\boldsymbol{K})\langle\lambda_f|\exp(i\boldsymbol{K}\cdot\mathbf{R}_j)|\lambda_i\rangle \quad \text{where} \quad \boldsymbol{K}=\mathbf{k}_i-\mathbf{k}_f$$

$$V_j(\boldsymbol{K})=\int V_j(\mathbf{x}_j)\exp(i\boldsymbol{K}\cdot\mathbf{x}_j)\,d\mathbf{x}_j$$

$$\langle\lambda_f|\exp(i\boldsymbol{K}\cdot\mathbf{R}_j)|\lambda_i\rangle=\int\chi^*_f\exp(i\boldsymbol{K}\cdot\mathbf{R}_j)\chi^*_f\,d\mathbf{R}_1 d\mathbf{R}_2 d\mathbf{R}_3\ldots d\mathbf{R}_j\ldots$$

Fermi pseudo-potential $V_j(\mathbf{x}_j)=\frac{2\pi\hbar^2}{m}b_j\delta(\mathbf{x}_j)$ $\quad V_j(\boldsymbol{K})=\frac{2\pi\hbar^2}{m}b_j$

$$\left(\frac{d^2\sigma}{dE_f\,d\Omega}\right)_{\lambda_i\to\lambda_f}=\frac{k_f}{k_i}\left|\sum_j b_j\langle\lambda_f|\exp(i\boldsymbol{K}\cdot\mathbf{R}_j)|\lambda_i\rangle\right|^2\delta(E_i-E_f+E_{\lambda_i}-E_{\lambda_f})$$

DIFFERENTIAL SCATTERING CROSS SECTION – NUCLEAR SCATTERING

even more algebra and manipulations

introduce time and energy to reach thermodynamics

$$\delta(E_i-E_f+E_{\lambda_i}-E_{\lambda_f})=\frac{1}{2\pi\hbar}\int_{-\infty}^{+\infty}\exp\{i(E_{\lambda_f}-E_{\lambda_i})t/\hbar\}\exp(-i\omega t)\,dt \quad \hbar\omega=E_i-E_f$$

$$\left(\frac{d^2\sigma}{dE_f\,d\Omega}\right)_{\lambda_i\to\lambda_f}$$

$$=\frac{k_f}{k_i}\sum_{jj'}b_j b_{j'}\langle\lambda_i|\exp(-i\boldsymbol{K}\cdot\mathbf{R}_{j'})|\lambda_f\rangle\langle\lambda_f|\exp(i\boldsymbol{K}\cdot\mathbf{R}_j)|\lambda_i\rangle\times\frac{1}{2\pi\hbar}\int_{-\infty}^{+\infty}\exp\{i(E_{\lambda_f}-E_{\lambda_i})t/\hbar\}\exp(-i\omega t)\,dt$$

$$\left(\frac{d^2\sigma}{dE_f\,d\Omega}\right)_{\lambda_i\to\lambda_f}=\frac{k_f}{k_i}\frac{1}{2\pi\hbar}\sum_{jj'}b_j b_{j'}$$

$$\times\int_{-\infty}^{+\infty}\langle\lambda_i|\exp(-i\boldsymbol{K}\cdot\mathbf{R}_{j'})|\lambda_f\rangle\langle\lambda_f|\exp(iHt/\hbar)\exp(i\boldsymbol{K}\cdot\mathbf{R}_j)\exp(-iHt/\hbar)|\lambda_i\rangle\exp(-i\omega t)\,dt$$

where H is the Hamiltonian of the scattering system

Introduce time-dependent operators $\mathbf{R}_j(t)=\exp(iHt/\hbar)\exp(i\boldsymbol{K}\cdot\mathbf{R}_j)\exp(-iHt/\hbar)$

DIFFERENTIAL SCATTERING CROSS SECTION – NUCLEAR SCATTERING

To get measured cross section, sum over all final λ_f (with fixed λ_{i}) and average over all λ_i.

$$\delta\left(E_i - E_f + E_{\lambda_i} - E_{\lambda_f}\right) = \frac{1}{2\pi\hbar}\int_{-\infty}^{+\infty} \exp\left\{i\left(E_{\lambda_f} - E_{\lambda_i}\right)t/\hbar\right\}\exp(-i\omega t)\,dt \quad \hbar\omega = E_i - E_f$$

Introduce partition function $Z = \sum_\lambda \exp\left(-E_\lambda / k_B T\right)$, probability $p_\lambda = \frac{1}{Z}\exp\left(-E_\lambda / k_B T\right)$

$$\left(\frac{d^2\sigma}{dE_f\, d\Omega}\right) = \sum_{\lambda_i \lambda_f} p_{\lambda_i}\left(\frac{d^2\sigma}{dE_f\, d\Omega}\right)_{\lambda_i \to \lambda_f}$$

$$= \frac{k_f}{k_i}\frac{1}{2\pi\hbar}\sum_{jj'} b_j b_{j'} \int_{-\infty}^{+\infty}\left\langle \exp\left\{-i\boldsymbol{K}\cdot \mathbf{R}_{j'}(0)\right\}\exp\left\{i\boldsymbol{K}\cdot \mathbf{R}_j(t)\right\}\right\rangle \exp(-i\omega t)\,dt$$

A compact form, but not easy to calculate:

measure of 'pair correlation functions'

information on scattering system contained in time-dependent ops. and wavefunctions

DIFFERENTIAL SCATTERING CROSS SECTION – NUCLEAR SCATTERING

Consider a simple system with a single element but different b's

$$\frac{d^2\sigma}{d\Omega dE_f} = \frac{k_f}{k_i}\frac{1}{2\pi\hbar}\sum_{jj'}\left\langle b_{j'} b_j\right\rangle \int_{-\infty}^{+\infty}\left\langle j', j\right\rangle \exp(-i\omega t)\,dt \quad \langle b\rangle = \sum_i f_i b_i \quad \left\langle b^2\right\rangle = \sum_i f_i b_i^2$$

no correlation between b's on different sites $\left\langle b_{j'} b_j\right\rangle = \langle b\rangle^2, j' \neq j \quad \left\langle b_{j'} b_j\right\rangle = \left\langle b^2\right\rangle, j' = j$

$$\left.\frac{d^2\sigma}{d\Omega dE_f}\right)_{coh} = \frac{\sigma_{coh}}{4\pi}\frac{k_f}{k_i}\frac{1}{2\pi\hbar}\sum_{jj'}\int_{-\infty}^{+\infty}\left\langle \exp\left\{-i\boldsymbol{K}\cdot \mathbf{R}_{j'}(0)\right\}\exp\left\{i\boldsymbol{K}\cdot \mathbf{R}_j(t)\right\}\right\rangle \exp(-i\omega t)\,dt$$

$$\left.\frac{d^2\sigma}{d\Omega dE_f}\right)_{incoh} = \frac{\sigma_{incoh}}{4\pi}\frac{k_f}{k_i}\frac{1}{2\pi\hbar}\sum_{j}\int_{-\infty}^{+\infty}\left\langle \exp\left\{-i\boldsymbol{K}\cdot \mathbf{R}_{j}(0)\right\}\exp\left\{i\boldsymbol{K}\cdot \mathbf{R}_j(t)\right\}\right\rangle \exp(-i\omega t)\,dt$$

Coherent scattering: pair correlations – probability of finding a particle at **R**+**r**(t) when there is a particle at **R**(0) interference effects

Incoherent scattering: self correlation – probability of finding a particle at **R**+**r**(t) when the same particle was at **R**(0) no interference effects

KEY MESSAGE

• in general, ignore coherence effects in neutron scattering

• neutron scattered intensities are proportional to space and time Fourier transforms of site correlation functions

CRYSTALLINE MATERIALS

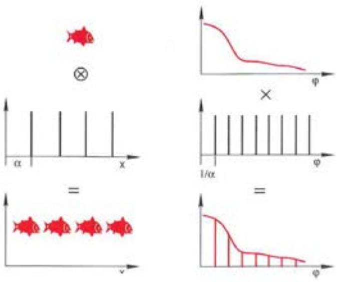
Examples of objects on a lattice - crystalline/ordered materials

Real space

1-d system:

convolution of objects and lattice of Dirac functions

N objects, N large

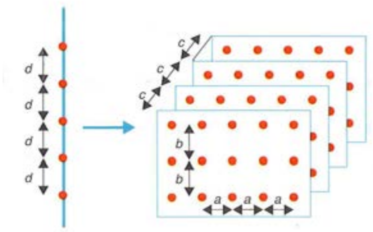


Reciprocal space

1-d system:

Fourier transforms

For N large



Similarly we define associated reciprocal spaces that reflect the symmetry and periodicities of real space lattices

NUCLEAR SCATTERING FROM CRYSTALLINE MATERIALS

Crystalline materials:

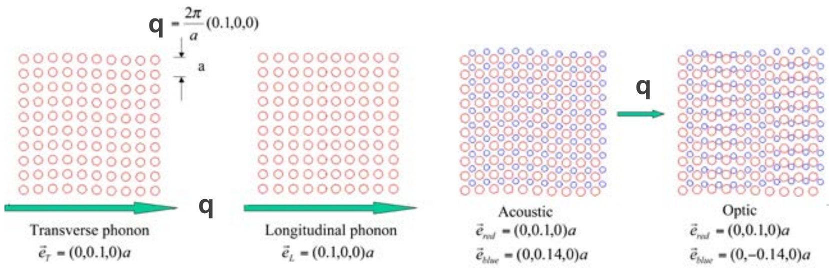
all atoms (nuclei) have an equilibrium position and they move about it

atom in cell *j*: $\mathbf{R}_j(t) = \mathbf{j} + \mathbf{u}_j(t)$ can be generalised to non-Bravais lattices

$$\sum_{jj'} \left\langle \exp\{-\mathrm{i}\boldsymbol{K}\cdot\mathbf{R}_{j'}(0)\} \exp\{\mathrm{i}\boldsymbol{K}\cdot\mathbf{R}_j(t)\} \right\rangle = \mathrm{N}\sum_j \exp(\mathrm{i}\boldsymbol{K}\cdot\mathbf{j}) \left\langle \exp\{-\mathrm{i}\boldsymbol{K}\cdot\mathbf{u}_0(0)\} \exp\{\mathrm{i}\boldsymbol{K}\cdot\mathbf{u}_j(t)\} \right\rangle$$

$$\sum_{j} \left\langle \exp\{-\mathrm{i}\boldsymbol{K}\cdot\mathbf{R}_{j}(0)\} \exp\{\mathrm{i}\boldsymbol{K}\cdot\mathbf{R}_j(t)\} \right\rangle = \mathrm{N} \left\langle \exp\{-\mathrm{i}\boldsymbol{K}\cdot\mathbf{u}_0(0)\} \exp\{\mathrm{i}\boldsymbol{K}\cdot\mathbf{u}_0(t)\} \right\rangle$$

displacements **u**(*t*) can be expressed in terms of normal modes or phonons



NUCLEAR SCATTERING FROM CRYSTALLINE MATERIALS

Coherent part

exp U exp V

$$\sum_{jj'} \left\langle \exp\{-\mathrm{i}\boldsymbol{K}\cdot\mathbf{R}_{j'}(0)\} \exp\{\mathrm{i}\boldsymbol{K}\cdot\mathbf{R}_j(t)\} \right\rangle = \mathrm{N}\sum_j \exp(\mathrm{i}\boldsymbol{K}\cdot\mathbf{j}) \left\langle \exp\{-\mathrm{i}\boldsymbol{K}\cdot\mathbf{u}_0(0)\} \exp\{\mathrm{i}\boldsymbol{K}\cdot\mathbf{u}_j(t)\} \right\rangle$$

it can be shown (Squires) $\langle \exp U \exp V \rangle = \exp\langle U^2 \rangle \exp\langle UV \rangle$

$$\left.\frac{d^2\sigma}{d\Omega dE_f}\right)_{coh} = \frac{\sigma_{coh}}{4\pi}\frac{k_f}{k_i}\frac{\mathrm{N}}{2\pi\hbar}\exp\langle U^2\rangle \sum_j \exp(\mathrm{i}\boldsymbol{K}\cdot\mathbf{j}) \int_{-\infty}^{+\infty} \exp\langle UV\rangle \exp(-\mathrm{i}\omega t)\,dt$$

Debye-Waller factor $2W = -\langle U^2 \rangle = \left\langle \{K\cdot u\}^2 \right\rangle$

$$\left.\frac{d^2\sigma}{d\Omega dE_f}\right)_{coh} = \frac{\sigma_{coh}}{4\pi}\frac{k_f}{k_i}\frac{\mathrm{N}}{2\pi\hbar}\exp(-2W)\sum_j \exp(\mathrm{i}\boldsymbol{K}\cdot\mathbf{j}) \int_{-\infty}^{+\infty} \left(1+\langle UV\rangle+\frac{1}{2!}\langle UV\rangle^2+\ldots..\right) \exp(-\mathrm{i}\omega t)\,dt$$

zero-th order: coherent elastic scattering - Bragg scattering

1st order: coherent one-phonon scattering

……

NUCLEAR SCATTERING FROM CRYSTALLINE MATERIALS

Coherent elastic scattering diffraction

$$\left.\frac{d^2\sigma}{d\Omega dE_f}\right)_{coh} = \frac{\sigma_{coh}}{4\pi}\frac{k_f}{k_i}\frac{N}{2\pi\hbar}\exp(-2W)\sum_j \exp(i\boldsymbol{K}\cdot\mathbf{j})\int_{-\infty}^{+\infty}\exp(-i\omega t)\,dt$$

$$\int_{-\infty}^{+\infty}\exp(-i\omega t)\,dt = 2\pi\hbar\,\delta(\hbar\omega)$$ purely elastic scattering $|\mathbf{k}_i| = |\mathbf{k}_f|$

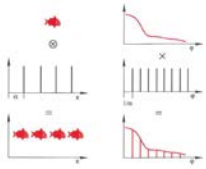
$$\left.\frac{d^2\sigma}{d\Omega dE_f}\right)_{coh\,el} = \frac{\sigma_{coh}}{4\pi}N\,\exp(-2W)\sum_j \exp(i\boldsymbol{K}\cdot\mathbf{j})\,\delta(\hbar\omega)$$

$$\left.\frac{d\sigma}{d\Omega}\right)_{coh\,el} = \int_0^\infty \left.\frac{d^2\sigma}{d\Omega dE_f}\right)_{coh\,el} dE_f = \frac{\sigma_{coh}}{4\pi}N\,\exp(-2W)\sum_j \exp(i\boldsymbol{K}\cdot\mathbf{j})$$

$$\left.\frac{d\sigma}{d\Omega}\right)_{coh\,el} = \frac{\sigma_{coh}}{4\pi}N\frac{(2\pi)^3}{v_0}\exp(-2W)\sum_\tau \delta(\boldsymbol{K}-\tau)$$

N: number of unit cells

v_0: volume of the unit cell



non-Bravais lattice with different atoms *d* in the unit cell

$$\left.\frac{d\sigma}{d\Omega}\right)_{coh\,el} = N\frac{(2\pi)^3}{v_0}\sum_\tau \delta(\boldsymbol{K}-\tau)|F_N(\boldsymbol{K})|^2 \qquad |F_N(\boldsymbol{K})| = \sum_d \langle b_d\rangle \exp(i\boldsymbol{K}\cdot\mathbf{d})\exp(-W_d)$$

structure factor

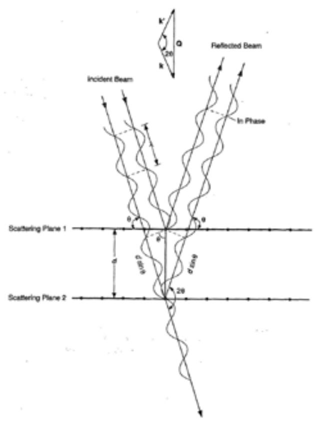
NUCLEAR SCATTERING FROM CRYSTALLINE MATERIALS

Bragg's law

$\lambda = 2d\sin\theta$

Practical application

monochromators!



Collecting intensities at Bragg peaks gives access to 'squared' values of Fourier components of the structure

Collect as many as possible to overcome the phase problem

NUCLEAR SCATTERING FROM CRYSTALLINE MATERIALS

Coherent elastic scattering (diffraction) provides:

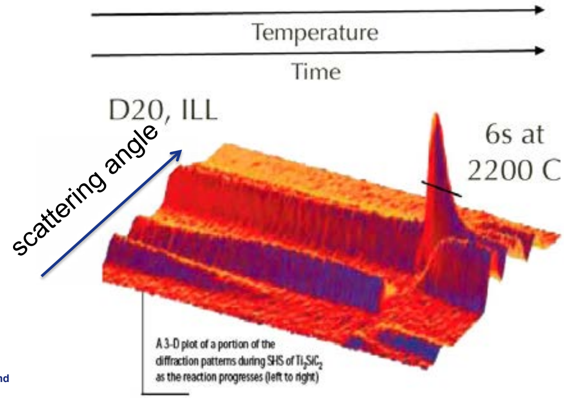
- periodicity in space, lattice symmetry and lattice constants
- positions of atoms in cells from $|F_N(\boldsymbol{K})|^2$
- powder and single crystals methods

But requires inversion of intensities into phases/amplitudes

Self propagating synthesis

High temperatures

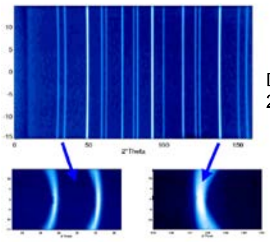
Phase transitions visible



A 3-D plot of a portion of the diffraction patterns during SHS of Ti_3SiC_2 as the reaction progresses (left to right)

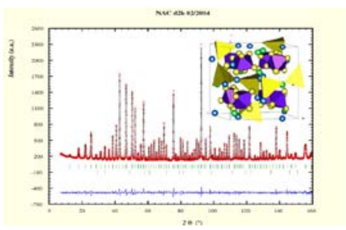
CRYSTALLINE MATERIALS : DIFFRACTION INSTRUMENTS

How does it work?



Detector
2D

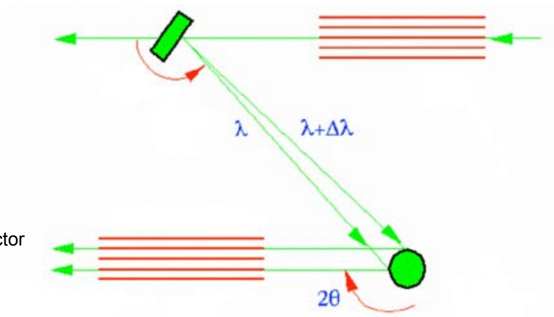
detector



Detector
1D

monochromatisation
crystal – diffraction
or time of flight

neutron
source



CRYSTALLINE MATERIALS : BEAM FILTERS

Maximum wavelength for which no Bragg scattering can occur

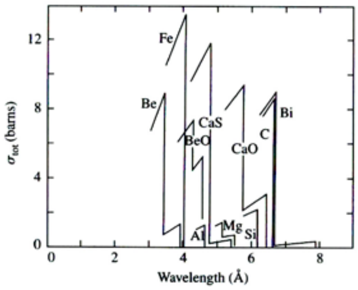
$$\lambda_{max} = 2d_{max}(\sin\theta)_{max} = 2d_{max}$$

Bragg cut-off

d_{max} is the maximum plane spacing

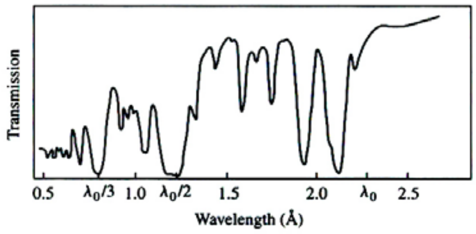
Beyond λ_{max} σ_{tot} drops

$\sigma_{tot} = \sigma_{scatt} + \sigma_{absor}$



Cut-off wavelengths for polycrystalline materials

Be: 3.9 Å graphite: 6.7 Å



Transmission of pyrolytic graphite

KEY MESSAGES

- neutron diffraction is an essential tool for structures determination
- it complements other diffraction methods
- neutrons probe bulk samples

NUCLEAR SCATTERING FROM CRYSTALLINE MATERIALS

Inelastic coherent scattering (Bravais lattice)

$$\left.\frac{d^2\sigma}{d\Omega dE_f}\right)_{coh} = \frac{\sigma_{coh}}{4\pi}\frac{k_f}{k_i}\frac{N}{2\pi\hbar}\exp(-2W)\sum_j \exp(i\boldsymbol{K}\cdot\mathbf{j})\int_{-\infty}^{+\infty}\left(1+\langle UV\rangle+\frac{1}{2!}\langle UV\rangle^2+\ldots\right)\exp(-i\omega t)\,dt$$

one-phonon coherent scattering (Bravais lattice)

<UV> involves creation and annihilation of phonon modes

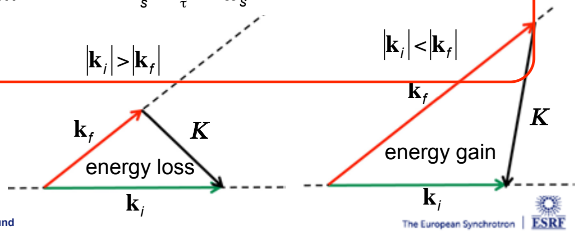
$$\langle UV\rangle = \frac{\hbar}{2MN}\sum_s \frac{(\boldsymbol{K}\cdot\mathbf{e}_s)}{\omega_s}\left[\exp\{-i(\mathbf{q}\cdot\mathbf{j}-\omega_s t)\}\langle n_s+1\rangle+\exp\{i(\mathbf{q}\cdot\mathbf{j}-\omega_s t)\}\langle n_s\rangle\right]$$

$$\left.\frac{d^2\sigma}{d\Omega dE_f}\right)_{coh\pm1} = \frac{\sigma_{coh}}{4\pi}\frac{k_f}{k_i}\frac{(2\pi)^3}{v_0}\frac{1}{2M}\exp(-2W)\sum_s\sum_\tau\frac{(\boldsymbol{K}\cdot\mathbf{e}_s)}{\omega_s}\langle n_s+1/2\pm1/2\rangle$$
$$\times\,\delta(\omega\mp\omega_s)\,\delta(\boldsymbol{K}\mp\mathbf{q}-\tau)$$

conservation laws!

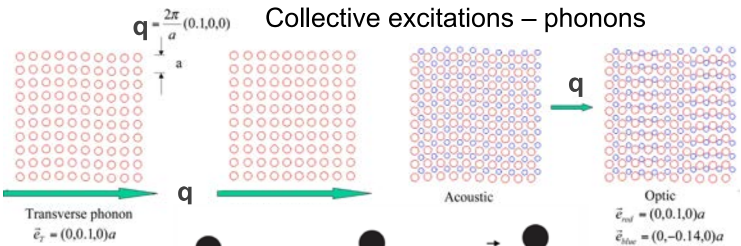
$\boldsymbol{K} = \tau \pm \mathbf{q}$

$E_i - E_f = \hbar\omega = \pm\hbar\omega_s$

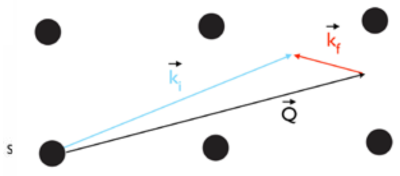


NUCLEAR INELASTIC SCATTERING

Collective excitations – phonons



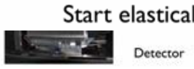
How to measure?



Go to a given $\boldsymbol{K} = \tau \pm \mathbf{q}$ and energy transfer $\hbar\omega$

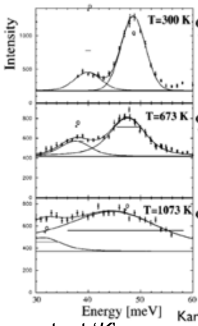
Orient sample: sample frame in coincidence with instrument frame

Start elastically, i.e. $k_i = k_f$



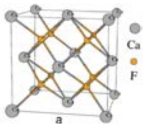
PHONON MODES

classical instruments: triple-axis machines



Karin Schmalzl

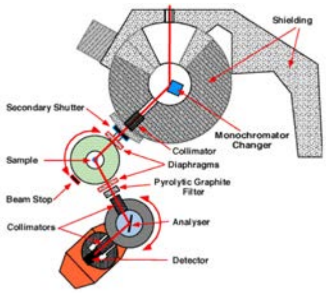
constant ‘*K*’



scan point by point

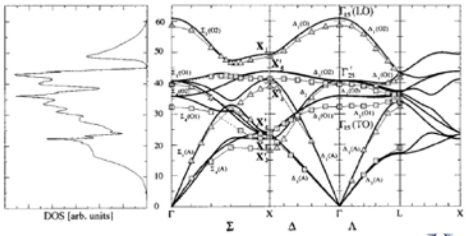
either constant ‘*K*’

or constant ‘energy’



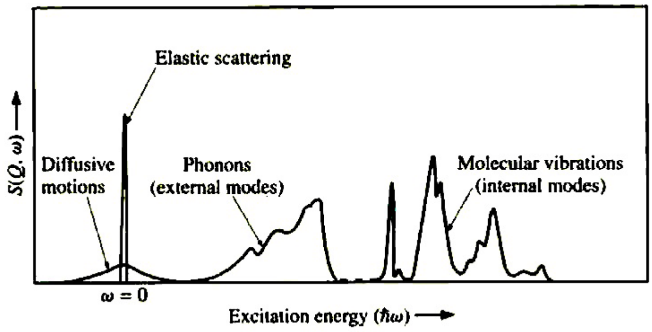
applications:

- lattice interactions, ‘soft mode’
- phase transitions
- superconductivity, …



SPECTROSCOPY

Spectroscopy – internal modes – little dispersion of modes



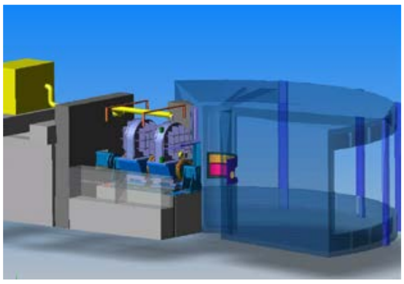
Excitations in hypothetical molecular crystal J.Eckert SpectroChim. Acta 48A, 271 (1992)

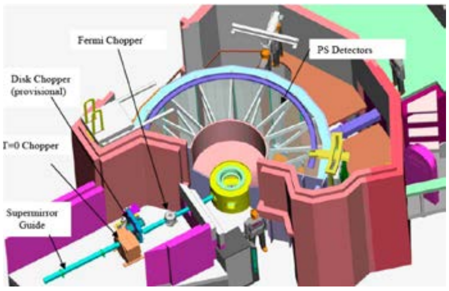
Use incoherent scattering

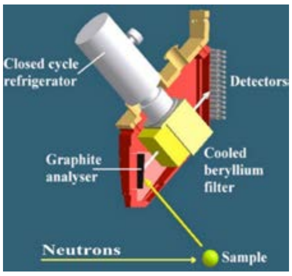
$$\left.\frac{d^2\sigma}{d\Omega dE_f}\right)_{incoh\pm1} = \frac{k_f}{k_i}\sum_s \delta(\omega\mp\omega_s)\frac{\langle n_s+1/2\pm1/2\rangle}{2\omega_s}\sum_r \frac{(\sigma_{incoh})_r}{4\pi}\frac{1}{M_r}|\mathbf{K}\cdot\mathbf{e}_r|^2\exp(-2W_r)$$

SPECTROSCOPY

more global picture: time of flight and large detector coverage







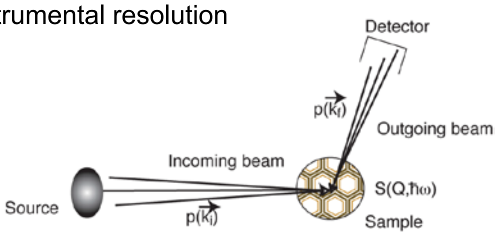
TIME DOMAIN

Time domain is reached through inelastic scattering

the range of energy transfer that can be covered determines the 'accessible' time domain (Fourier transform!)

Depends on 'incident' neutron energy

and instrumental resolution

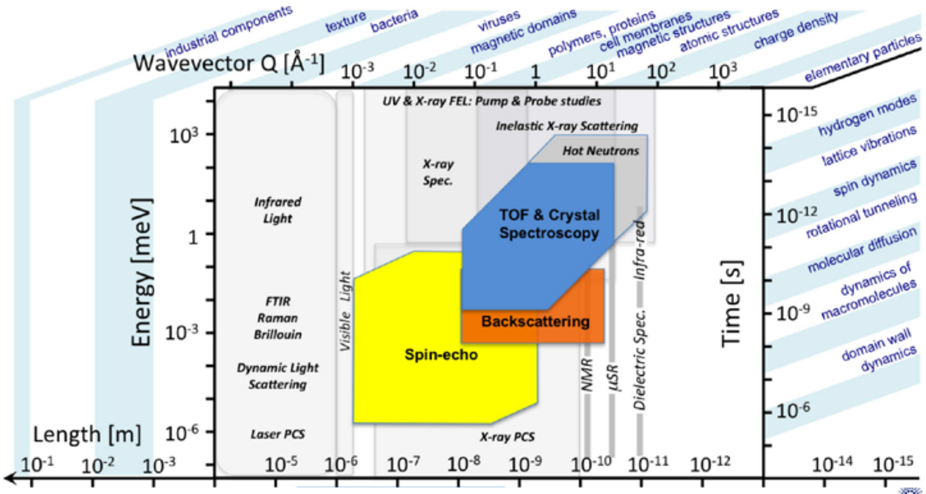


Typical resolution in wavelength (and energy) are ~ a few percents

except special techniques (backscattering, NSE)

TIME DOMAIN AND COMPLEMENTARITY

complementarity with other methods



KEY MESSAGES

- accessible time and space domains cover a wide range of applications
- no single probe can cover the whole (K,ω) space that would allow an ideal Fourier transformation
- use complementary probes

DISORDERED MATERIALS

liquids, glasses, ..no equilibrium positions for atoms

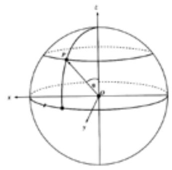
local order but no long range order

consider mono-atomic system – $\dfrac{d\sigma}{d\Omega} = \sum_{i,j} b_i^* b_j e^{-i\boldsymbol{K}\cdot(\mathbf{R}_i - \mathbf{R}_j)}$

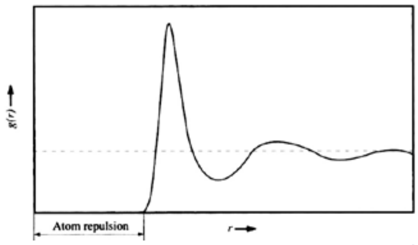
for most liquids, glasses, ... scattering depends on magnitude of

averaging over polar angles

$$\left\langle \exp\left\{-i\boldsymbol{K}\cdot\left(\mathbf{R}_i - \mathbf{R}_j\right)\right\}\right\rangle = \frac{\sin(Kr_{ij})}{Kr_{ij}}$$



replace sum over atoms by radial distribution function



g(r) for monoatomic liquid

DISORDERED MATERIALS - LIQUIDS

mono-atomic liquid

$$\frac{d\sigma}{d\Omega} = N\langle b^2\rangle + \sum_{i\neq j}^{N} \langle b_i\rangle\langle b_j\rangle e^{-i\boldsymbol{K}\cdot(\mathbf{R}_i - \mathbf{R}_j)} = N\langle b^2\rangle + 4\pi\rho\langle b^2\rangle \int_0^\infty r^2 g(r)\frac{\sin(Kr)}{(Kr)}dr$$

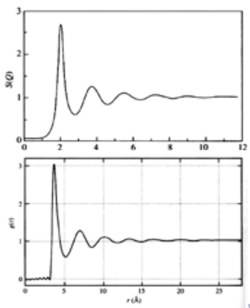
note that $\displaystyle\int_0^\infty r^2 \frac{\sin(Kr)}{(Kr)}dr = 0$ unless $K = 0$

$$\frac{d\sigma(K\neq 0)}{d\Omega} = N\langle b^2\rangle S(K) \quad with = S(K) = 1 + \frac{4\pi\rho}{K}\int_0^\infty r\left[g(r)-1\right]\sin(Kr)\,dr$$

S(K): structure factor

very different from $F_N(\boldsymbol{K})$ in crystalline materials

- relates to intensity (not amplitude)
- not well structured, only a few peaks



Liquid Argon

T= 85K

Experimental data

Inverted $\rho(r)$

(Fourier transform)

J.L. Yarnell et al.

PRA 7, 2130 (1973)

DISORDERED MATERIALS - LIQUIDS

In n-component systems, there are n(n+1)/2 site-site radial distributions

To be measured, using isotopic substitution.

In liquids, strictly speaking there is no elastic neutron scattering:

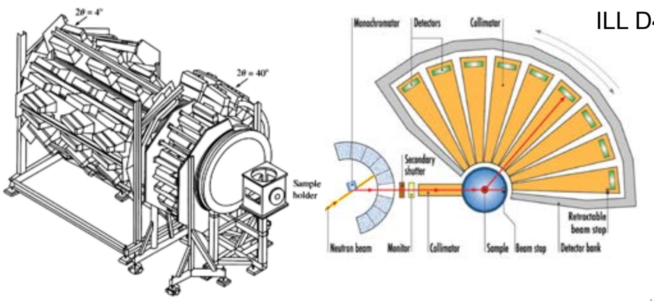
nuclei recoil under neutron impact

need to span all ($\boldsymbol{K}$,ω) space

or apply inelasticity corrections   Placzek PRB 86, 377 (1956)

Typical instruments

Sandals ISIS



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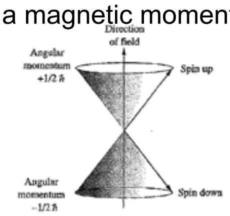
MAGNETISM!

Neutrons have a spin ½ and therefore carry a magnetic moment

neutron beams can be polarised



$$\boldsymbol{\mu}_n = -\gamma \mu_N \boldsymbol{\sigma} \quad \text{where } \mu_N = \frac{e\hbar}{2m_p} \quad \gamma = 1.913$$

For comparison, electrons have $\boldsymbol{\mu}_e = -2\mu_B \mathbf{s}$ where $\mu_B = \frac{e\hbar}{2m_e}$

Neutron magnetic moments feel magnetic fields created in materials:

- electrons: dipole moments and currents

- nuclei: dipole moments (neglected here)

The potential V in the scattering cross section should include these effects

$$\left(\frac{d^2\sigma}{dE_f\, d\Omega}\right)_{\lambda_i \to \lambda_f} = \frac{k_f}{k_i}\left(\frac{m_n}{2\pi\hbar^2}\right)^2 \left|\langle \mathbf{k}_f \lambda_f | V | \mathbf{k}_i \lambda_i \rangle\right|^2 \delta\left(E_i - E_f + E_{\lambda_i} - E_{\lambda_f}\right)$$

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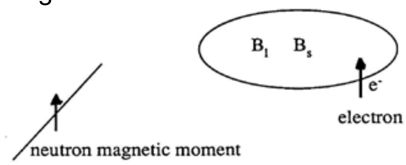
MAGNETISM!

Magnetic field created at distance ***R*** electron with momentum ***p***



$$\boldsymbol{B} = \boldsymbol{B}_S + \boldsymbol{B}_L = \frac{\mu_0}{4\pi}\left\{\mathrm{curl}\left(\frac{\boldsymbol{\mu}_e \times \mathbf{R}}{R^2}\right) - \frac{2\mu_B}{\hbar}\frac{\mathbf{p}\times\mathbf{R}}{R^2}\right\}$$

Potential of a neutron in B $\quad \boldsymbol{V}_m = -\boldsymbol{\mu}_n \cdot \boldsymbol{B} = -\frac{\mu_0}{4\pi}\gamma\,\mu_N\, 2\,\mu_B\left\{\mathrm{curl}\left(\frac{\boldsymbol{s}\times\mathbf{R}}{R^2}\right) + \frac{1}{\hbar}\frac{\mathbf{p}\times\mathbf{R}}{R^2}\right\}$

During scattering, neutron changes from state $\boldsymbol{k}_i,\sigma_i$ to $\boldsymbol{k}_f,\sigma_f$

$$\left(\frac{d^2\sigma}{dE_f\, d\Omega}\right)_{\sigma_i\lambda_i\to\sigma_f\lambda_f} = \frac{\boldsymbol{k}_f}{\boldsymbol{k}_i}\left(\frac{m_n}{2\pi\hbar^2}\right)^2 \left|\langle \mathbf{k}_f\sigma_f\lambda_f|\boldsymbol{V}_m|\mathbf{k}_i\sigma_i\lambda_i\rangle\right|^2 \delta\left(E_i - E_f + E_{\lambda_i} - E_{\lambda_f}\right)$$

Complex evaluation:

- magnetic interaction is long range
- scattering entities are not point-like – form factors
- magnetic forces are not central

MAGNETISM!

After long calculations

$$\left(\frac{d^2\sigma}{dE_f\, d\Omega}\right)_{\sigma_i\lambda_i\to\sigma_f\lambda_f} = \frac{k_f}{k_i}(\gamma r_0)^2 \left|\langle\sigma_f\lambda_f|\sigma\cdot \mathbf{Q}_\perp|\sigma_i\lambda_i\rangle\right|^2 \delta\left(E_i - E_f + E_{\lambda_i} - E_{\lambda_f}\right)$$

r_0: classical radius of electron 2.8$10^{-13}$ cm

'strength' of scattering ~ $(\gamma r_0)^2$ of the order of 0.3 barn see nuclear scatt.

$$\boldsymbol{Q}_\perp = \sum_{\text{electrons } x}\exp\left(i\boldsymbol{K}\cdot\mathbf{r}_x\right)\left\{\hat{\boldsymbol{K}}\times\left(\boldsymbol{s}_x\times\hat{\boldsymbol{K}}\right) + \frac{i}{\hbar K}\left(\boldsymbol{p}_x\times\hat{\boldsymbol{K}}\right)\right\}$$

whereas for nuclear scattering $\sum_j b_j \exp\left(i\boldsymbol{K}\cdot\mathbf{R}_j\right)$

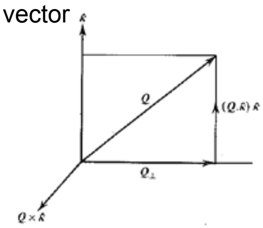
The $\boldsymbol{Q}_\perp$ operator is related to the magnetisation of the scattering system

separating spin and orbital contributions

$\mathbf{Q}_\perp(\boldsymbol{K}) = \hat{\mathbf{K}}\times\left(\mathbf{Q}(\boldsymbol{K})\times\hat{\mathbf{K}}\right)$ where $\mathbf{Q}(\boldsymbol{K}) = -\frac{1}{2\mu_B}\mathbf{M}(\boldsymbol{K})$ $\quad\hat{\mathbf{K}}$ is a unit vector

M(**K**) is the Fourier transform of ***M***(***r***)

$\boldsymbol{Q}_\perp$ is the vector projection of **Q** on to the plane perpendicular to ***K***



KEY MESSAGES

• 'nuclear' and 'magnetic' interactions have similar strengths

$\gamma r_0 \approx 5.4$ fm similar to many b's

• interactions with electrons connect to magnetisation densities

• 'magnetic' scattered intensities are proportional to space and time Fourier transforms of site correlation functions for magnetic moments

• magnetic form factors

• geometrical dependence of the scattering

MAGNETISM!

Similarly to nuclear scattering, magnetic neutron scattering probes 'correlations'

$$\left(\frac{d^2\sigma}{dE_f\, d\Omega}\right) = \frac{k_f}{k_i}\frac{(\gamma r_0)^2}{2\pi\hbar}\sum_{\alpha\beta}\left(\delta_{\alpha\beta} - \hat{K}_\alpha \hat{K}_\beta\right)\int\left\langle Q_\alpha(-\boldsymbol{K},0)Q_\beta(\boldsymbol{K},t)\right\rangle \exp(-i\omega t)\,dt$$

geometrical factor

$$Q_\beta(\boldsymbol{K},t) = \exp(iHt/\hbar)Q_\beta(\boldsymbol{K})\exp(-iHt/\hbar)$$

equivalent to nuclear scattering

Elastic scattering – thermal average at infinite time

$$\left(\frac{d\sigma}{d\Omega}\right)_{el} = (\gamma r_0)^2\sum_{\alpha\beta}\left(\delta_{\alpha\beta} - \hat{K}_\alpha \hat{K}_\beta\right)\left\langle Q_\alpha(-\boldsymbol{K})Q_\beta(\boldsymbol{K})\right\rangle$$

or $\left(\frac{d\sigma}{d\Omega}\right)_{el} = \left(\frac{\gamma r_0}{2\mu_B}\right)^2\left|\hat{\boldsymbol{K}}\times\left\{\left\langle \mathbf{M}(\boldsymbol{K})\right\rangle\times\hat{\boldsymbol{K}}\right\}\right|^2$ with $\mathbf{Q}(\boldsymbol{K}) = -\frac{1}{2\mu_B}\mathbf{M}(\boldsymbol{K})$

$\mathbf{M}(\boldsymbol{K})$ contains all information on magnetic arrangements

symmetry, periodicity, moments, …..

MAGNETIC DIFFRACTION

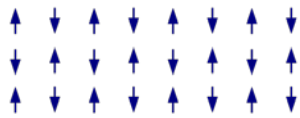
Ferromagnets

localised magnetic system has the same periodicity as lattice

$$\left(\frac{d\sigma}{d\Omega}\right)_{el} = (\gamma r_0)^2 N \frac{(2\pi)^3}{v_0} \langle S^\eta \rangle^2 \sum_\tau \left\{\frac{1}{2} g F(\tau)\right\}^2 \exp(-2W) \times \left\{1 - (\hat{\tau} \cdot \hat{\eta})_{Aver}{}^2\right\} \delta(\boldsymbol{K} - \tau)$$

- magnetic intensity on top of nuclear intensity
- ‘magnetic form factor’ not constant as b –

  spatial distribution of ‘magnetic’ electrons
- Measurements of intensities give $F(\tau)$ which allow M(r) to be calculated



Non-ferromagnets

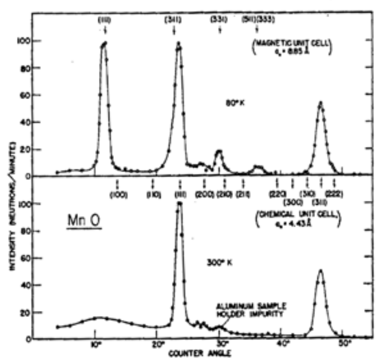
new periodicity in space leads to new Bragg peaks

MAGNETIC DIFFRACTION

Non-ferromagnets

neutrons allow to probe local magnetic order

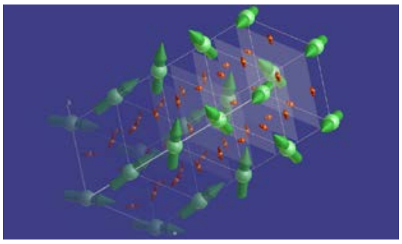
C. Shull et al. 1949



Powder samples or single crystals

‘easy’ and routine experiments!

One of the very strong points for neutrons



More complex materials

Important for new devices

KEY MESSAGE

- neutron diffraction (powder) is the the method of choice to determine magnetic structures (if not the only one …)
- a lot of software has been developed including symmetry arguments
- beware of progress made by other techniques …..

INELASTIC MAGNETIC SCATTERING OF NEUTRONS

Inelastic magnetic neutron scattering probes magnetic 'correlations'

$$\left(\frac{d^2\sigma}{dE_f\, d\Omega}\right)_{\sigma_i\lambda_i\to\sigma_f\lambda_f} = \frac{k_f}{k_i}(\gamma r_0)^2 \left|\langle\sigma_f\lambda_f|\sigma\cdot \mathbf{Q}_\perp|\sigma_i\lambda_i\rangle\right|^2 \delta\left(E_i - E_f + E_{\lambda_i} - E_{\lambda_f}\right)$$

neutron spin states can change by ΔS= -1, 0, +1

- simple localised 'magnetic' excitations - crystal field levels

  rare-earth systems magnetism carried by 4*f* electrons

  Russel-Saunders coupling J good marker of states

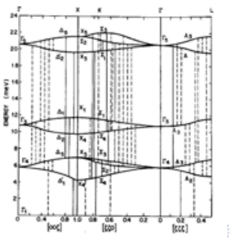
  spherical symmetry is broken by local crystal symmetry

  origin of magnetic anisotropy

  Pr ions in PrSb: Pr^{3+}: 2 4*f* electrons L=5 S=1 J=4

  4 states in cubic symmetry – singlet ground state

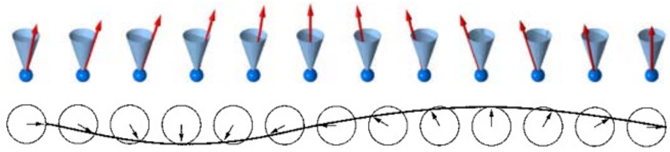
  dispersion – presence of exchange couplings



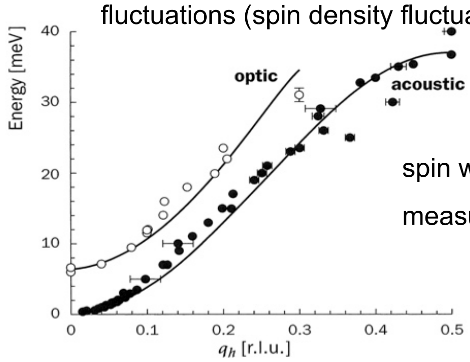
INELASTIC MAGNETIC SCATTERING OF NEUTRONS

Collectives magnetic excitations

spin waves



fluctuations (spin density fluctuations)



spin waves in $La_{1.2}Sr_{1.8}Mn_2O_7$

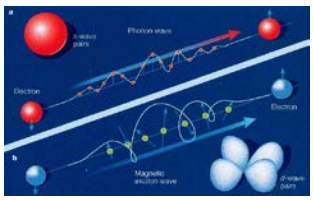
measured on TAS instruments

INELASTIC MAGNETIC SCATTERING OF NEUTRONS

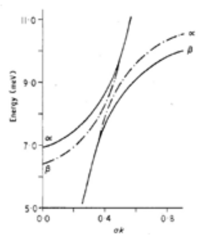
collectives magnetic excitations

spin density fluctuations

investigating pairing mechanisms



interactions between lattice and magnetism



how to separate lattice and magnetic modes?

Q-dependence: Q^2 versus form factor

polarisation analysis

KEY MESSAGES

- neutron inelastic magnetic scattering: a unique probe to study magnetic interactions
- it covers the 'interesting' energy range
- it is enhanced by neutron polarisation methods

NEUTRON POLARISATION

scattering cross section involves neutron spin states

another neutron degree of freedom

use of polarised neutrons

use of polarisation

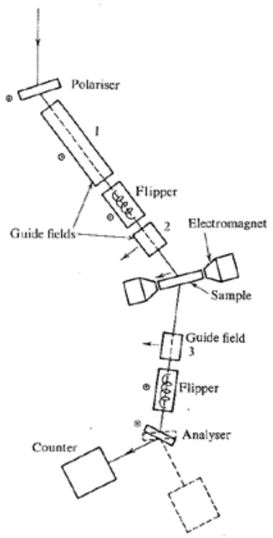
use of neutron spin precession in fields

Larmor precession

spin manipulation

spin filters

NSE methods



NEUTRON POLARISATION

polarised neutron beams, *up (u)* and *down (v)* states $P = \frac{n_+ - n_-}{n_+ + n_-}$

previous cross-sections gives rise to 4 cross-sections $u \to u \quad v \to v \quad u \to v \quad v \to u$

coherent nuclear scattering

$$\left.\begin{matrix} u \to u \\ v \to v \end{matrix}\right\} \quad \bar{b} = \left\langle \frac{(I+1)b^+ + Ib^-}{2I+1} \right\rangle_{isotopes}$$

$$\left.\begin{matrix} u \to v \\ v \to u \end{matrix}\right\} \quad \bar{b} = 0$$

incoherent nuclear scattering

$$\left.\begin{matrix} u \to u \\ v \to v \end{matrix}\right\} \quad \langle b^2 \rangle - \langle b \rangle^2 = \left\langle \left(\frac{(I+1)b^+ + Ib^-}{2I+1} \right)^2 \right\rangle_{isotopes} - \left\langle \frac{(I+1)b^+ + Ib^-}{2I+1} \right\rangle^2_{isotopes} + \frac{1}{3} \left\langle \left(\frac{b^+ - b^-}{2I+1} \right)^2 I(I+1) \right\rangle_{isotopes}$$

$$\left.\begin{matrix} u \to v \\ v \to u \end{matrix}\right\} \quad \langle b^2 \rangle - \langle b \rangle^2 = \frac{2}{3} \left\langle \left(\frac{b^+ - b^-}{2I+1} \right)^2 I(I+1) \right\rangle_{isotopes}$$

particular cases: unpolarised neutrons

Ni: all isotopes with I=0

Vanadium: only one isotope

NEUTRON POLARISATION

Polarisation 'induces' interference between nuclear and magnetic scattering

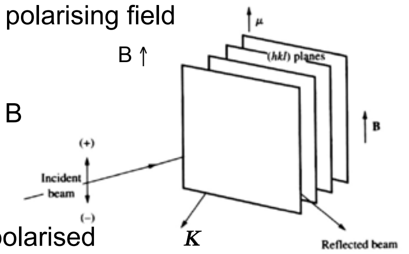
$$\frac{d\sigma}{d\Omega} = N \frac{(2\pi)^3}{v_0} \left\{ |F_N(\mathbf{K})|^2 + 2(\hat{P} \cdot \hat{\mu}) |F_N(\mathbf{K})| |F_M(\mathbf{K})| + |F_M(\mathbf{K})|^2 \right\}$$

In ferromagnets, $|F_N(\mathbf{K})|$ and $|F_M(\mathbf{K})|$ are non-zero for the same *K* vectors

polarising field

application: polarisation devices

B ↑



$(\hat{P} \cdot \hat{\mu}) = \pm 1$ for neutrons (anti-)parallel to B

$$\frac{d\sigma}{d\Omega} = N \frac{(2\pi)^3}{v_0} |F_N(\mathbf{K}) \pm F_M(\mathbf{K})|^2$$

if matching F_N and F_M reflected beam is polarised

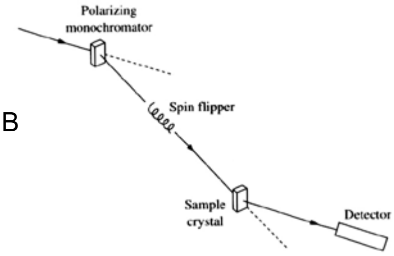
guide fields

Similar effects/applications in reflectometry – 'magnetic' optical index

polarising neutron guides

NEUTRON POLARISATION

application: precise measurement of weak magnetic signals



apply B perpendicular to ***K***

moments are aligned parallel to B

$$\frac{d\sigma}{d\Omega} = N\frac{(2\pi)^3}{v_0}\left|F_N(\boldsymbol{K}) \pm F_M(\boldsymbol{K})\right|^2$$

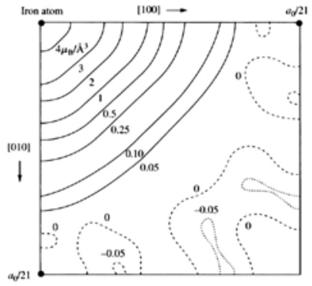
measure flipping ratio R

$$R = \left.\frac{d\sigma}{d\Omega}\right)^{+} \Big/ \left.\frac{d\sigma}{d\Omega}\right)^{-} = \left(\frac{1-\gamma}{1+\gamma}\right)^2 \text{ with } \gamma = F_M(\boldsymbol{K})/F_N(\boldsymbol{K})$$

if γ is small, R~1-4γ

allows to measure spin densities

Iron C.G Shull et al. J.Phys.Soc.Japan 17,1 (1962)

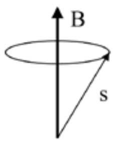


MANY OTHER FIELDS IN NEUTRON METHODS

neutron spin-echo: use of Larmor precession of neutron's spin

time evolution of s=1/2 in magnetic field B

$$\frac{d\vec{s}}{dt} = \gamma\,\vec{s} \times \vec{B} \quad \omega_L = |\gamma| B \quad \text{with } \gamma = -2913 * 2\pi \text{ Gauss}^{-1} \cdot \text{s}^{-1}$$

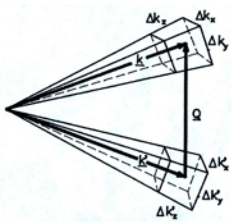


total precession angle $\phi = \omega_L t = \gamma \text{ B d/v}$ depends neutron's velocity

with B=10 Gauss ~29 turns/m for 4Å neutrons

neutron spin-echo encodes neutron velocity – quite high resolution

without loss in intensity



NSE breaks the awkward relationship between intensity and resolution: the better the resolution, the smaller the resolution volume and the lower the count rate!

neutron reflectometry, SANS, …

SUMMARY OF KEY MESSAGES

- neutrons have no charge – low absorption
- 'nuclear' and 'magnetic' interactions have similar strengths
- interaction with nuclei very short range

  isotropy, isotope variation and contrast
- interactions with electrons allow studies of magnetisation densities

  neutron diffraction the method of choice to determine magnetic structures
- scattered intensities are proportional to space and time Fourier transforms of site correlation functions (positions and magnetic moments)
- accessible time and space domains cover a wide range of applications
- caveat: neutron sources are not very efficient …..

FURTHER READING

Introduction to the Theory of Thermal Neutron Scattering

G.L. Squires Reprint edition (1997) Dover publications ISBN 04869447

Experimental Neutron Scattering

B.T.M. Willis & C.J. Carlile (2009) Oxford University Press ISBN 978-0-19-851970-6

Neutron Applications in Earth, Energy and Environmental Sciences

L. Liang, R. Rinaldi & H. Schober Eds Springer (2009) ISBN 978-0-387-09416-8

Methods in Molecular Biophysiscs

I.N. Serdyuk, N. R. Zaccai & J. Zaccai Cambridge University Press (2007) ISBN 978-0-521-81524-6

Thermal Neutron Scattering

P.A. Egelstaff ed. Academic Press (1965)