

NEUTRONS AND THEIR INTERACTION WITH MATTER



INSTITUT MAX VON LAUE - PAUL LANGEVIN

Overview

- History neutrons and nuclear reactions
- Production reactors and spallation sources
 - •Properties as a particle and a probe
- •Instruments exploiting the probe to do science



A BIT OF HISTORY

The neutron

 1932: J. Chadwick, after work by others, discovers the 'neutron', a neutral but massive particle



 ${}^{4}_{2}\text{He} + {}^{9}_{4}\text{Be} \rightarrow {}^{12}_{6}\text{C} + {}^{1}_{0}\text{n}$ $(m_{\text{He}} + m_{\text{B}})c^{2} + T_{\text{He}} = (m_{\text{C}} + m_{\text{n}})c^{2} + T_{\text{C}} + T_{\text{n}}$

 $m_n = 1.0067 \pm 0.0012 a.m.u$





A BIT OF HISTORY

The nuclear reaction

- 1938: O. Hahn, F. Strassmann & L. Meitner discovered the fission of ²³⁵U nuclei through thermal neutron capture
- 1939: H. v. Halban, F. Joliot & L. Kowarski showed that ²³⁵U nuclei fission produced 2.4 n⁰ on average – chain reaction
- 1942: E. Fermi & al. demonstrated first self-sustained chain reaction reactor

Chicago pile: 360T of graphite 50T of U and UO 0.5W power





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NOBEL PRIZES, NEUTRONS AND THE ILL

Chadwick, Shull & Brockhouse

The Nobel Prize in Physics 1994

The Royal Swedish Academy of Sciences has awarded the 1994 Nobel Prize in Physics for pioneering contributions to the development of neutron scattering techniques for studies of condensed matter.



James Chadwick (1891 - 1974)



Shull made use of **elastic scattering** i.e. of neutrons which change direction without



Betram N. Brockhouse, MdMater University, Hamilton, Ontario, Canada, receives one half of the 1994 Nobel Prize in Physics for the development of neutron spectroscopy.

Brockhouse made use of inelastic scattering i.e. of neutrons, which change



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Haldane (1977 – 1981), Kosterlitz and Thouless for topological phase transitions and phases of matter (Electronic structure and excitation of 1D quantum liquids and spin chains)





NEUTRON SOURCES

Fission reactors

- Nuclear fission \rightarrow chain reaction with excess neutrons (1n \rightarrow 2.5n)
- Slow neutrons split U-235 nuclei
- Fission neutrons have MeV energies and need to be moderated (thermalized) to meV energies by scattering from water
- Thermalisation @ RT → thermal neutrons,
 @ 25K → cold neutrons and @ 2400 K → hot neutrons
- ILL flux 1.5 x 10¹⁵ n/cm²/s





NEUTRON SOURCES

Spallation sources

- Neutrons can be produced by bombarding heavy metal targets
- 2 GeV protons (90% speed-oflight) produce spallation – evaporation of ~30 neutrons





NEUTRON SOURCES



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



CONTINUOUS OR PULSED BEAMS

Integrated vs peak flux – ESS will have a time-integrated flux comparable to ILL







CONTINUOUS OR PULSED BEAMS

Integrated vs peak flux – ESS will have a time-integrated flux comparable to ILL





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N vs X

ESRF (hard X-rays)





N & X



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As a particle

- free neutrons are unstable: β -decay \rightarrow proton, electron, anti-neutrino life time: 888 ± 1 sec or 880 ± 1 sec
- wave-particle duality: neutrons have particle-like and wave-like properties
- mass: $m_n = 1.675 \times 10^{-27} \text{ kg} = 1.00866 \text{ amu}$. (unified atomic mass unit)
- charge = 0
- spin =1/2
- magnetic dipole moment: $\mu_n = -1.9 \ \mu_{N_i} \ \mu_p = 2.8 \ \mu_{N_i} \ \mu_e \sim 10^3 \ \mu_{n_i}$
- velocity (v), kinetic energy (E), temperature (T), wavevector (k), wavelength (λ)



As a particle

• velocity (v), kinetic energy (E), temperature (T), wavevector (k), wavelength (λ)

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

Neutron energy determines velocity and therefore time-of-flight (*tof*) over a given distance i.e. *tof* → energy determination

$$tof = \frac{L}{v} = 253\mu \sec{\lambda} \begin{bmatrix} o \\ A \end{bmatrix} \cdot L[m]$$



As a probe

	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







As a probe

Wavelengths on the scale of inter-atomic distances:
 Å - nm wavelengths to measure Å - μm
 distances/sizes

$$n\lambda = 2dsin\Theta$$

- Energies comparable to structural and magnetic excitations: *meV* neutrons to measure *neV – meV* energies
- Neutral particle gentle probe, highly penetrating (e.g. 30 cm of Al), no radiation damage
- Magnetic moment (nuclear spin) probes magnetism of unpaired electrons (N.B. $\mu_e \sim 1000 \text{ x} \mu_N$)



As a probe – interacting with matter – scattering from atoms

- Neutron flux at reactor core
- 1.5 x 10¹⁵ n/cm²/s
- Flux at an instrument sample position
- 10⁸ n/cm²/s
- \rightarrow 10⁻⁶ n/ μ m²/ μ s
- → 10⁻¹⁵ n/nm²/ns
- On these time and length scales, neutrons are being scattered one at a time
- Need wave-particle duality of neutrons





As a probe – interacting with matter – (elastic) scattering from a single fixed nucleus

- Nuclear size << neutron wavelength → point-like s-wave scattering
- b is the scattering length ('power') in fm
- #neutrons scattered per second per unit solid angle Ω : $\Psi^2 r^2 d\Omega$

 $d\sigma/d\Omega = b^2$

• σ is the cross-section: $4\pi b^2$ (in barns)





As a probe – interacting with matter – scattering from a set of nuclei

$$\frac{d\sigma}{d\Omega} = \sum_{j,k} b_j b_k e^{i\vec{Q}\cdot\left(\vec{R}_j - \vec{R}_k\right)}$$
$$\vec{Q} = \vec{k}_f - \vec{k}_i$$

- *Q* is called momentum transfer
- *Q*-dependence (eg angle) gives info about atomic positions





As a probe – interacting with matter – scattering from a set of identical nuclei – coherent and incoherent scattering

- Set of N similar atoms/ions spins/isotopes are uncorrelated at different sites
- *b* depends on spin/isotope
- Average is
- Incoherent scattering gives a *Q* independent background
- But it can be useful to probe the dynamics of single particles (later)

$$\frac{d\sigma}{d\Omega} = \langle b \rangle^2 \sum_{j,k} e^{iQ \cdot (R_j - R_k)} + \left(\langle b^2 \rangle - \langle b \rangle^2 \right) N$$

$$\sigma_{coh} = 4\pi \langle b \rangle^{2} \qquad \sigma_{coh} = 4\pi b_{coh}^{2}$$

$$\sigma_{incoh} = 4\pi (\langle b^{2} \rangle - \langle b \rangle^{2}) \qquad \sigma_{incoh} = 4\pi b_{inc}^{2}$$



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As a probe – interacting with matter – scattering from a set of identical nuclei – coherent and incoherent scattering

- 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^-
- To reduce incoherent scattering (background):
 - polarise nuclei and neutrons
 - use isotope substitution e.g. $H \rightarrow D$

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

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



Scattering lengths Light atoms Contrast





Scattering lengths can be positive or negative (nuclear physics)

- Positive *b* (most nuclei): phase change
- Negative *b*: no phase change at scattering point





ZSymbA	p or T _{1/2}	1	b _c	b+	b_	c	σcoh	σinc	σscatt	σabs
0-N-1	10.3 MIN	1/2	-37.0(6)	0	-37.0(6)		43.01(2)		43.01(2)	0
1-Н			-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)	Е	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)



Scattering lengths can be positive or negative → Contrast matching





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As a probe – interacting with matter - absorption

- Absorption neutron capture
- Several strong absorbers: He, Li, B, Cd, Gd,...
- Isotope dependent choose to your advantage





As a probe – interacting with matter - absorption - Neutron detection

- How to detect a weakly interacting, neutral particle?
- With a neutron absorber and measure the resulting signal

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





Scattering and absorption cause attenuation of a neutron beam \rightarrow imaging



NEUTRONS

X-RAYS



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Scattering and absorption cause attenuation of a neutron beam → imaging



As a probe – interacting with matter - summary

- Interaction with nuclei:
 - short range interaction \rightarrow angle independent scattering (no form factor)
 - scattering length can be positive or negative (\rightarrow contrast variation)
 - depends on isotope (\rightarrow selectivity) and nuclear spin
 - Coherent and incoherent scattering strength and weakness
 - Scattering contrast different from X-rays, favours light atoms
- A gentle probe meV neutron beam does not cause radiation damage like a ~10 keV photon beam (what about XFEL!)
- Magnetic moment probes magnetism of unpaired electrons





THE ILL'S INSTRUMENT SUITE





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GENERAL EXPRESSION FOR SCATTERING FROM A COMPLEX SYSTEM

Deriving the general scattering function

Based on

- Born approximation kinematic theory: neutron wavefunction un-perturbed inside sample
- Fermi's Golden Rule to calculate transitions of neutron (k) and system (λ) from initial and final state
- Hamiltonian to describe the system states (λ)

$$\left(\frac{d^{2}\sigma}{dE_{f}d\boldsymbol{\varOmega}}\right)_{\lambda_{i}\rightarrow\lambda_{f}} = \frac{k_{f}}{k_{i}}\left(\frac{m_{n}}{2\pi\hbar^{2}}\right)^{2}\left|\left\langle\mathbf{k}_{f}\lambda_{f}|\boldsymbol{V}|\mathbf{k}_{i}\lambda_{i}\right\rangle\right|^{2}\delta\left(E_{i}-E_{f}+E_{\lambda_{i}}-E_{\lambda_{f}}\right)$$

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

GENERAL EXPRESSIONS FOR SCATTERING FROM A SET OF MOVING ATOMS

Deriving the scattering function – end up with (after much algebra and manipulations!)

$$\left(\frac{d^{2}\sigma}{dEd\Omega}\right) = \frac{k_{f}}{k_{i}} \frac{1}{2\pi\hbar} \sum_{jk} b_{j} b_{k} \int_{-\infty}^{+\infty} \left\langle \exp\left\{-i\vec{Q}\cdot\vec{R}_{j}\left(0\right)\right\} \exp\left\{i\vec{Q}\cdot\vec{R}_{k}\left(t\right)\right\}\right\rangle \exp\left(i\omega t\right) dt$$

$$\left(\frac{d^{2}\sigma}{dEd\Omega}\right) = \frac{k_{f}}{k_{i}} \frac{1}{2\pi\hbar} S(\vec{Q},\omega)$$

- Experiment measures double differential crosssection which is simply related to $S(Q, \omega)$ (or I(Q, t))
- *S*(*Q*, *ω*) is the double Fourier transform of the timedependent pair-correlation function





GENERAL EXPRESSIONS FOR SCATTERING FROM A SET OF MOVING ATOMS

Deriving the scattering function – end up with – coherent & incoherent contributions

• For a simple system with a single element but different b's

$$\frac{d^{2}\sigma}{d\Omega dE_{f}}\right)_{coh} = \frac{\sigma_{coh}k_{f}}{4\pi k_{i}} \frac{1}{2\pi\hbar} \sum_{jk} \int_{-\infty}^{+\infty} \left\langle \exp\left\{-i\vec{Q}\cdot\vec{R}_{j}(0)\right\} \exp\left\{i\vec{Q}\cdot\vec{R}_{k}(t)\right\} \right\rangle \exp\left(-i\omega t\right) dt$$
$$\frac{d^{2}\sigma}{d\Omega dE_{f}}\right)_{incoh} = \frac{\sigma_{incoh}k_{f}}{4\pi k_{i}} \frac{1}{2\pi\hbar} \sum_{j} \int_{-\infty}^{+\infty} \left\langle \exp\left\{-i\vec{Q}\cdot\vec{R}_{j}(0)\right\} \exp\left\{i\vec{Q}\cdot\vec{R}_{j}(t)\right\} \right\rangle \exp\left(-i\omega t\right) dt$$

- Scattering function determined by positions *R* of different atoms at different times *t*
- Incoherent scattering can be useful: it measures the correlation between the same atom at different times → single particle dynamics
 diffusion



GENERAL SCATTERING EXPERIMENT

Scattering triangle – handling Q and ω

•
$$Q = k_f - k_i, \ \hbar \omega = E_f - E_i (E \sim k^2, \ k = 2\pi/\lambda)$$

- Elastic scattering: vary *Q* without changing ω
 E_i = *E_f* vary 2Θ (monochromatic) vary |*E*| fix 2Θ (t.o.f.)
- Quasi/in-elastic scattering: vary ω , normally Q will also change vary E_i or E_f and/or 2Θ





GENERIC INSTRUMENT

Energy selection

- How to measure the energy of a neutron beam?
- Or, how to monochromate a beam?
- Measure λ with Bragg reflection

 $n\lambda = 2dsin\Theta$

- d = distance between scattering planes
- Use neutron *t.o.f.* (or precession of neutron magnetic moments in a magnetic field)

$$tof = \frac{L}{v} = 253\mu \sec{\lambda} \begin{bmatrix} o \\ A \end{bmatrix} \cdot L[m]$$





Instruments (don't measure the final energy!) – D2b & LADI







Example – Formation and properties of ice XVI obtained by emptying a type sII clathrate hydrate



Instruments (don't measure the final energy!) – D2b & LADI







Example – Improving drug design: HIV-1 Protease in complex with clinical inhibitors (sample ~50 µg)







Simplified expressions for the scattering function - coherent scattering

In previous scattering expressions

 $R = R_0 + \delta R(t)$

For normal modes: $\delta R(t) \rightarrow$ displacement vectors **e** & frequencies ω Coherent scattering - Phonons:

- Short range coupling gives long range correlations
- Dispersion as a function of \boldsymbol{q} (or wavelength) guitar string!

$$\frac{d^{2}\sigma}{d\Omega dE_{f}}\right)_{coh\pm1} = \frac{\sigma_{coh} k_{f} (2\pi)^{3} 1}{4\pi k_{i} v_{0} 2M} \exp(-2W) \sum_{s} \sum_{\tau} \frac{\left(\overrightarrow{Q} \cdot \overrightarrow{e}_{s}\right)}{\omega_{s}} \langle n_{s} + 1/2 \pm 1/2 \rangle$$
$$\times \delta(\omega \mp \omega_{s}) \delta\left(\overrightarrow{Q} \mp \overrightarrow{q} - \overrightarrow{\tau}\right)$$



Instruments – varying $k_i \& k_f - TAS$, TOF







Example – phonon lifetimes in thermoelectrics - Complex Metallic Alloy - Al₁₃Co₄ Quasicrystal approximant











1,600

b

500

e

FOR SOCIETY

Simplified expressions for the scattering function - incoherent scattering

In previous scattering expressions

 $R = R_0 + \delta R(t)$

For normal modes: $\delta R(t) \rightarrow$ displacement vectors e & frequencies ω Incoherent scattering - Internal (molecular) modes:

• No long range correlations due to weak coupling

• No dispersion

$$\frac{d^{2}\sigma}{d\Omega dE_{f}}\right)_{incoh\pm1} = \frac{k_{f}}{k_{i}} \sum_{s} \delta(\omega \mp \omega_{s}) \frac{\langle n_{s} + 1/2 \pm 1/2 \rangle}{2\omega_{s}} \sum_{r} \frac{(\sigma_{incoh})_{r}}{4\pi} \frac{1}{M_{r}} \left| \overrightarrow{Q} \cdot \overrightarrow{e}_{r} \right|^{2} \exp(-2W_{r})$$



Instruments – TOF, Lagrange







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Example – endofullerenes





Nanolaboratories: physics and chemistry of small-molecule endofullerenes

Papers of a Theo Murphy Meeting Issue organized and edited by Malcolm H. Levitt and Anthony J. Horsewill





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Example – oxide ion conductors



Anode Electrolyte Cathode





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NEUTRONS FOR SOCIETY

Example – oxide ion conductors







SPECTROSCOPY – SPIN ECHO

Energy selection - precession of neutron magnetic moments in a magnetic field (depends on t.o.f. in B)





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SPECTROSCOPY – SPIN ECHO



Structure and dynamics – double differential cross-section

As for interactions with nuclei but

- Neutron spin probes local magnetic fields due to electron spin and orbital contribution
- Atomic form factor scattering from an atom is angular dependent due to electron cloud
- No incoherence effects

$$V_m = -\mu_n \cdot B = -\frac{\mu_0}{4\pi} \gamma \mu_N 2\mu_B \left\{ curl\left(\frac{s \times R}{R^2}\right) + \frac{1 \ p \times R}{\hbar \ R^2} \right\}$$

• N.B. σ and V in these equations

$$\left(\frac{d^2\sigma}{dE_f d\Omega}\right)_{\sigma_i \lambda_i \to \sigma_f \lambda_f} = \frac{k_f}{k_i} \left(\frac{m_n}{2\pi\hbar^2}\right)^2 \left|\left\langle k_f \sigma_f \lambda_f \left| V_m \left| k_i \sigma_i \lambda_i \right\rangle \right|^2 \delta\left(E_i - E_f + E_{\lambda_i} - E_{\lambda_f}\right)\right\rangle \right|$$



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Polarised neutrons – separate nuclear and magnetic signals & more precise information on magnetic structures





 Measure 2, 6 or 10 polarised scattering channels: u→u (non spin flip) and d→u (spin flip) in the simplest case





Polarised neutrons – separate nuclear and magnetic signals & more precise information on magnetic structures

- Polarised (optically pumped) 3He selectively absorbs one neutron spin state
 more versatile polariser
- Cryopad allows full control of incident and scattered neutron polarisation – spherical polarimetry







Example – Ground state selection under pressure in the quantum pyrochlore magnet $Yb_2Ti_2O_7$





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Example – How do electrons/spins organise in a triangular lattice? Spins pair into quantum-mechanical bonds and fluctuate...

	1
triangular-lattice quantum-spin-liquid	-
What is a Quantum What is a Quantum What is a Quantum where the second secon	



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The neutron

- Is Highly penetrating
- Interacts with nuclei favourable for light atoms (H, Li, O,...)
- Incoherent scattering is ideal for proton dynamics
- Isotopes provide selectivity contrast matching
- Interacts with unpaired electrons magnetism
- Probes 15 orders of magnitude in length & 10 in time

Neutron sources have relatively low intensity and are only available in large scale facilities – ILL, ISIS, PSI, FRM2 in Europe, SNS & NIST in US



ADDITIONAL READING

Search the web! Plus...

- 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)



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