Neutrons.

Sources and theory basics.

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View of J-PARC in May 2007

Site; Tokai, Ibaraki 150km to the north from Tokyo The first neutrons from liquid mercury target 30 May 2008!

Linac (400MeV -> 200MeV) Materials & Life Experimental Hall (MLF: JSNS+muon) 1MW, 25Hz ->0.6MW)

> 50 GeV synchrotron (MR)

Hadron Experimental Hall

Neutrino to

Super KAMIOKANDE (300km away)



600year old historic shrine

nchrotron

The European Spallation Source





Brightness comparison of different radiation sources



ILL, Grenoble, France



Neutron as a particle

According to quark theory, neutron consists of two down (d) and one up (u) quarks.

Quarks	Charge / e	Mass / m _e
'down' d	- 1/3	600
ʻup'u	+ 2/3	600

Neutron



mass m_n= 1.175×10^{-27} кг el. charge = 0; спин = ½ magnetic dipole moment μ_n = -1.913 μ_N , where nuclear magneton μ_N = eh/4 π m_p = 5.051×10⁻²⁷ JT⁻¹

Neutron as a particle



 $n \rightarrow p^+ + e^- + \overline{\nu}_e + 782 \text{ keV}$

$$\tau_n = 885.7 \pm 1.0 \text{ sec}$$

Neutron as a particle and a wave

Neutron possesses properties of both particle and a wave:

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



Ultra-cold cold thermal not	Ultra-cold	cold	thermal	hot
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Energy (meV)	<0.0001	0.001 - 10	5 - 100	100 - 500
Wavelength (Å)	~ 900	4 - 300	1 - 4	0.4 - 1
Temperature (K)	~ 0.001	0.01 - 120	60 - 1000	1000 - 6000



Neutrons for condensed ma	atter research
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Property	Applicability
No charge, small cross sections	 can investigate bulk material can treat scattering in first Born approximation complex sample environment no damage to biological samples
Wavelength of thermal neutrons in the range of interatomic distances	 can determine crystal structures and atomic positions
Magnetic moment	 can examine magnetic properties on microscopic scale
Kinetic energy in the range of elementary excitations	 can investigate dynamical properties and excitation energies
Scattering by nuclei	• can "see" hydrogen, can distinguish isotopes \rightarrow contrast variation
Coherent and incoherent scattering	collective phenomena as well as single atom effects



Neutrons for condensed matter



The diameters of the circles shown scale with the scattering amplitude f_x (sin θ =0) for X-rays, and $b_{coh} \times 10$ for neutrons. Hatching indicates negative scattering amplitudes



Penetration depth of thermal neutrons, low energy electrons and 8 keV X-rays into different elements

For neutrons: no regular wavelength dependence of scattering length (which is just a number, not the function of the scattering angle as in the case of X-rays!) and absorption coefficient from atomic number; enormous difference in scattering legths and absorption coefficients for H and D; huge absorption for Cd, B, Sm, Gd

Disadvantages

Relatively low intensity of neutron sources \Rightarrow weak signal, large sample volumes required etc. Strong absorption of some elements, but isotope substitution can cure the problem. Kinematical restrictions limit the achievable energy-momentum range..



Neutrons for condensed matter

racaarah

Neutrons bounce

against atomic nuclei

They also react to the

magnetism of the

atoms.

Research reactor

Neutrons behave as

articles and as

Neutrons reveal

The Nobel Prize in Physics 1994

Clifford G. Shull, MIT, Cambridge, Massachusetts, USA, receives one half of the 1994 Nobel Prize in Physics for development of the neutron diffraction technique.



Shull made use of elastic scattering i.e. of neutrons which change direction without losing energy when they collide with atoms.

Because of the wave nature of neutrons, a diffraction pattern can be recorded which indicates where in the sample the atoms are situated. Even the placing of light elements such as hydrogen in metallic hydrides, or hydrogen, carbon and oxygen in organic substances can be determined. The pattern also shows how atomic dipoles are oriented in magnetic materials since neutrons are affected by magnetic forces. Shull also made use of this phenomenon in his neutron diffraction technique.



atoms that he

Lay a neutron diffraction map (showing the positions of the nuclei) over an Xray diffraction map (gring the distribution of the electrons), it is then clear that the electron density is shifted in relation to the posi-

tions of the atomic nuclei. Since a chemical bond invol

direct picture of the chen

D.J. Hughes The Nuclear Reactor as a Research Instrument, SCIENTIFIC AMERICAN, VOL. 189, AUGUST 1953, P. 23. Lengeler and J.L. Finney The European Spallation Source EUROPHYSICS NEWS, VOL. 25, P.37, 1904-mation about the Nobel Prize in Dhysits 1994 (presselence), THE ROYAL SWEDTSH ACADEMY OF SCIE-

Neutrons see more than X-rays

Hydrogen, for example, which has only one electron, is no so easy to see. With neutrons, all kinds of atoms are visible,

X-rays are scattered by elect With X-rays it is easiest to s

Further reading:

Detectors record the directions of the neutrons and a diffraction pattern is obtained. The pattern shows the positions of the atoms relative

Atoms in a

0

crystalline sample

where atoms are

When the neutrons collide with atoms in the sample material, they

change direction (are

scattered) - elastic scattering.

to one anothe

forwards neutrons of a certain wavelength (energy) - mono-chromatized neutrons

Crystal that sorts and

Neutrons reveal inner stresses Neutrons show what atoms remember of their earlier positions when they move randomly in relation to each other in liquids and melts. Even here there is in fact some local other. The atoms cannot move infinitely close to each other. Some distances are more in an important metal aircraft part, Does the part match up? Neutron diffraction can show how much the dis-tance between the atoms has changed and hence th internal forces remaining round the hole after it has en punched 00 The curves show local expansion forces (positive) and compression forces (negative) in different directions (red, green and blue) in a direroft part (Saab39 Gripen). (1 ps = one mil lionth of a mi onth of a se ring. Corresponding "memory f ets e.g. near the Curie tempera



Crystal that sorts and forwards neutrons of a certain wavelength

(energy) - mono-chromatized neutrons

How it started

. how it continues

elastic properties of polymers

became available for peacetime research.

Thousands of researchers are now working at the many

super-installations the researchers are studying the structure o

new ceramic superconductors, molecular movements on surfaces of interest for catalytic exhaust cleaning, virus structures and the connection between the structure and the

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.

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



of inelastic scattering i.e. of neutrons, which change both direction and energy when they collide with atoms. They then start or cancel atomic oscillations in crystals and record movements in liquids and melts. Neutrons can also interact with spin waves in magnets.

With his 3-axis spectrometer Brockhouse measured energies of phonons (atomic vibrations) and magnons (magnetic waves). He also studied how atomic structures in liquids change with time.



and the neutrons then counted in a detector.





Neutrons for condensed matter Interdisciplinary character of research with neutrons







Each fission act produces in average ~2.5 neutrons with energy release about 200 MeV. In the case of chain (self sustained) reaction 1 neutron is necessary to ensure the next fission, 0.5 is lost because of absorption inside the neutron source and 1 neutron can be extracted for use in the experiment



Steady state nuclear reactor





Spallation reaction gives up to 30 neutrons per 1 proton. There exists *empirical* expression for the estimation of neutron yield from the target when proton energy exceeds 120 MeV

$$N_{n}(E,A) = \begin{cases} 0.1 \times E_{GeV} \times (A+20) - \text{for non-fissile target} \\ 50 \times E_{GeV} - \text{for uranium-238} \end{cases}$$



Principal scheme of spallation neutron source







Components of neutron scattering instruments

What do we need to perform good neutron experiment?

- Intense neutron source with efficient moderator
- neutron shaping, guiding and velocity selection system
- interesting sample for the study
- system to analyse parameters of scattered neutron beam
- advanced neutron detector



Typical scheme of neutron scattering experiment



Examples of registered patterns on a detector

From the analysis of the signal on a detector, using advanced mathematical models one can extract information about characteristics of a sample under investigation



Joint Institute for Nuclear Research

Cross-sections definitions

Let the sample is irradiated by flux Φ_{in} of neutrons per unit area per unit time. We define I_s and I_a as a number of neutrons scattered and absorbed by the sample per unit time, respectively. Then the total cross sections of scattering and absorption of neutrons by the sample are defined as:

 $\mathbf{I}_{s} = \Phi_{in}\sigma_{s}$

 $\mathbf{I}_a = \Phi_{in} \sigma_a$

Dimensionality of the total cross sections is [barn]:

 $1barn = 10^{-24} cm^2$.

By definition the flux $\Phi_{in} = 1/S \cdot t = v_0 / V$, where S – is the area irradiated by neutrons, t – time of irradiation, v_0 – velocity of neutrons, V – volume of the irradiated sample.



The interaction of neutron with nuclei is described by Fermi pseudopotential:

$$H = \frac{2\pi\hbar^2}{m_n} b\delta(\vec{r} - \vec{R})$$

Where m_n – neutron mass, b – scattering length, <u>R</u> – vector defining the position of a nucleus in space.

Let us first consider the scattering of a neutron on isolated nucleus under the following assumptions:

>neutron wavelength is much larger than the scattering length which is the radius of action of nuclear force (this is the limitation of applicability of Fermi pseudopotential)

≻the scattering is purely elastic, e.g. the energy of neutron is conserved before and after the scattering

> the absorption of neutrons by the nucleus can be neglected





$$\Phi_{\text{out}} = v_0 \Psi_{\text{out}}^* \Psi_{\text{out}} = \left| \mathbf{b} \right|^2 \frac{1}{r^2} \frac{v_0}{V}$$



The number of neutrons passing through the element of spherical surface $S = r^2 d\Omega$ is equal

$$dI_{out} = \Phi_{out}(\vec{r})r^2 d\Omega = \left|b\right|^2 \frac{v_0}{V} d\Omega$$

And differential scattering cross section (i.e. the probability that neutron is scattered in a direction defined by a vector \underline{r} into an element of solid angle $d\Omega$) is equal

$$\frac{d\sigma}{d\Omega} = \frac{dI_{out}}{\Phi_{in}d\Omega} = \left|b\right|^2$$

And the total scattering cross section will be

 $\sigma = \int (d\sigma/d\Omega) d\Omega = 4\pi |b|^2$ ISOTROPIC!!!





Let us now consider the scattering by a nucleus which position is defined by a vector \vec{R}_i . The wave function of incident neutrons will again be a plane wave:

$$\Psi_{in} = \frac{1}{\sqrt{V}} e^{i\vec{k}_0\vec{r}}$$

And for the scattered neutrons

$$\Psi_{out} = \frac{1}{\sqrt{V}} e^{i\vec{k}_0\vec{R}_i} \left| \frac{-b_i}{\left|\vec{r} - \vec{R}_i\right|} e^{ik_0\left|\vec{r} - \vec{R}_i\right|} \right|$$

 \sqrt{V}

$$\left| \vec{r} - \vec{R}_i \right| = r - \frac{\vec{r}}{r} \vec{R}_i + O\left(\frac{1}{r}\right)$$

For $r \rightarrow \infty$ we have:

$$\mathbf{e}^{i\mathbf{k}_{0}|\vec{r}-\vec{R}_{i}|} = \mathbf{e}^{i\mathbf{k}_{0}r}\mathbf{e}^{-i\mathbf{k}_{0}\frac{\vec{r}}{r}\vec{R}_{i}} = \mathbf{e}^{i\mathbf{k}_{0}r}\mathbf{e}^{-i\vec{k}_{1}\vec{R}_{i}}$$
$$\Psi_{out} = \frac{1}{\sqrt{n}}\mathbf{e}^{-i\vec{k}_{1}\vec{R}_{i}}\left(\frac{-\mathbf{b}_{i}}{-\mathbf{b}_{i}}\mathbf{e}^{i\mathbf{k}_{0}r}\right)$$

r



The resulting neutron flux after scattering on the nuclei $\underline{R}_{\underline{i}}$ and $\underline{R}_{\underline{i}}$ will be equal

$$\Phi_{out}^{i,j} = \frac{v_0}{V} \Psi_{out}^{i} \Psi_{out}^{j} = \frac{v_0}{V} b_i b_j e^{i\vec{k}_1 \vec{R}_i} e^{-ik_1 R_j}$$

Therefore, differential cross section for N nuclei:

$$\frac{d\sigma}{d\Omega} = \sum_{i,j}^{N} b_i b_j e^{i\vec{Q}(\vec{R}_i - \vec{R}_j)}, \text{ where } \vec{Q} = \vec{k}_1 - \vec{k}_0$$





Now, consider more common case, when the velocity of neutron is changing during the scattering process due the fact that atoms perform thermal vibrations and their coordinates in space become time dependent. This means that $v_1 \neq v_0$ and $R_i = R_i(t)$. Then the above formula can be written in a more common form giving the double differential cross section. This cross section defines the probability of neutron being scattered in a direction <u>r</u> into an element of solid angle $d\Omega$ with the neutron energy change $E = E_1 - E_0$ in an interval dE.



$$\frac{d^2\sigma}{d\Omega d\omega} = \frac{k_1}{k_0} \sum_{i} \sum_{j} \int_{-\infty}^{+\infty} \left\langle b_i b_j e^{i\vec{Q}\vec{R}_i(t)} e^{-i\vec{Q}\vec{R}_j(0)} \right\rangle e^{-i\omega t} dt$$

Angular brackets mean averaging over all values of nuclei coordinates in a space $R_i(t)$, isotope content and all possible spin states of nuclei. It is known, that the scattering length depends on a relative orientation of nucleus and neutron spins. For parallel spins denote the scattering length as b^+ , and for anti parallel as b^- . Scattering length is also different for different isotopes of the chosen chemical element. Then one obtains:

$$\left\langle b_{i}b_{j}\right\rangle = \begin{cases} \left\langle b_{i}\right\rangle \left\langle b_{j}\right\rangle & \text{for } i \neq j \\ \left\langle \left|b_{i}\right|^{2}\right\rangle & \text{for } i = j \end{cases}$$







Consequently, neutron cross sections will consist of two terms – coherent and incoherent:

$$\sigma = \sigma_{coh} + \sigma_{inc}; \qquad \sigma_{coh} = 4\pi b_{coh}^{2}; \qquad \sigma_{inc} = 4\pi b_{inc}^{2}$$

$$\frac{d\sigma}{d\Omega} = \left(\frac{d\sigma}{d\Omega}\right)_{coh} + \left(\frac{d\sigma}{d\Omega}\right)_{inc} = \left|\underbrace{\sum_{i}^{N} \langle b_{i} \rangle e^{i\vec{Q}\vec{R}_{i}}}_{i}\right|^{2} + \underbrace{\left(\langle \left|b_{i}\right|^{2} \rangle - \left|\langle b_{i} \rangle\right|^{2}\right)}_{inc}$$

$$\begin{split} &\frac{d^2\sigma}{d\Omega d\omega} = \left(\frac{d^2\sigma}{d\Omega d\omega}\right)_{coh} + \left(\frac{d^2\sigma}{d\Omega d\omega}\right)_{inc} \\ &\left(\frac{d^2\sigma}{d\Omega d\omega}\right)_{coh} = \frac{k_1}{k_0}\sum_i\sum_j b_i^{coh}b_j^{coh}S(\vec{Q},\omega), \ rgamma rga$$

Which is the probability, that if the arbitrarily chosen nucleus j at a time t=0 had a coordinate $\underline{\mathbf{R}}_{i}(0)$, then at a moment t≠0 another atom i will have a coordinate $\underline{\mathbf{R}}_{i}(t)$.



$$\begin{split} &\left(\frac{d^2\sigma}{d\Omega d\omega}\right)_{inc} = \frac{k_1}{k_0} \sum_i \left(b_i^{inc}\right)^2 S^{inc}(\vec{Q},\omega) \\ &S^{inc}(\vec{Q},\omega) = \frac{1}{2\pi} \iint d\vec{r} dt \ e^{i(\vec{Q}\vec{r}-\omega t)} G_a(\vec{r},t) \\ &G_a(\vec{r},t) = \sum_i \iint \left\langle \delta\left[\vec{r}-\vec{r}'+\vec{R}_i(0)\right] \delta\left[\vec{r}'-\vec{R}_i(t)\right] \right\rangle \end{split}$$

 $G_a(\vec{r},t)$ - Is called an AUTOCORRELATION FUNCTION,

Which is the probability, that if at a time t=0 the arbitrary chosen atom i had a coordinate $\underline{\mathbf{R}}_{\underline{i}}(0)$, then at a time t≠0 its coordinate will be $\underline{\mathbf{R}}_{\underline{i}}(t)$.



Possible types of neutron scattering experiments and corresponding scientific applications

Type of scattering	Q	ω	r	t	Scattering law	Scientific applications
elastic	Q	0	ř	∫dt	S(Q ,0)	Atomic and magnetic structures
	$\vec{Q} \rightarrow 0$	0	$\vec{r} \rightarrow \infty$	∫dt	S(Q,0)	Submolecular structures
total	Q	∫dω	r	0	S(Q)	Disordered structures
	Q	ω	r	t	S (Q ,ω)	Collective excitation
inelastic	Q	ω	ŕ	t	S(Q ,ω)	Atomic and molecular spectroscopy
	∫Q	ω	r = 0	t	S(ω)	Density of vibrational states
quasi- elastic	Q	ω ↓ 0	ŕ	t ↓ ∞	S(Q,ω)	Diffusion of atoms and molecules

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Magnetic neutron scattering

The detailed evaluation of the expressions for cross sections of elastic and inelastic neutron scattering on different magnetic structures is rather complicated. Therefore, let's see only the final result for several most common cases.

General expression for double differential cross section (unpolarised neutrons)



Magnetic neutron scattering

Magnetic form factor F(Q) is the Fourier transform of the magnetisation density in a crystal. It is important to note, that neutron magnetic form factor is defined by distribution of electrons with uncompensated spins only.

 $r_0 = e^2/m_e c^2$ – electromagnetic radius of the electron

 γ = -1.913 – neutron magnetic moment in Bohr magnetons

 $\delta_{ij} = \begin{cases} 0, \text{if } i \neq j \\ 1, \text{ if } i = j \end{cases} - \text{Krone ker symbol} \\ \text{Polarisation factor} \qquad (\delta_{\alpha\beta} - \frac{Q_{\alpha}Q_{\beta}}{Q^2}) & \text{indicates that neutrons interact} \end{cases}$ only with the components of atomic spin which are perpendicular to the scattering vector \mathbf{Q} . This fact allows uniquely measure in an experiment the directions of atomic spins and polarisation of spin waves. \mathbf{r}_{0} value indicates that the magnetic neutron scattering cross section is of the order 10⁻²⁴ cm², therefore comparable with nuclear scattering cross section.



Uses of neutron scattering in magnetism

Static properties (elastic scattering)

Spin arrangements in ordered magnetic structures Static spin correlations in disordered or frustrated magnets Magnetisation density (magnetic form factor) Flux distributions in superconductors Magnetism of surfaces (reflectometry)

Magnetism of surfaces (reflec

etc

Dynamic properties (inelastic scattering)

Crystal field excitations

Inter-multiplet atomic excitations

Spin waves in ordered magnetic structures

Spin fluctuations in strongly correlated or frustrated magnets

Magneto-phonon coupling

etc





Magnetic scattering

$$\mathbf{S}^{\alpha\beta}(\mathbf{Q},\omega) = \frac{1}{2\pi\hbar} \int dt \ e^{-i\omega t} \frac{1}{N} \sum_{\mathbf{R}\mathbf{R}'} e^{i\mathbf{Q}\cdot(\mathbf{R}-\mathbf{R}')} < S^{\alpha}_{\mathbf{R}}(0) S^{\beta}_{\mathbf{R}'}(t) >$$



Modern challenges - novel fields of neutron scattering application "There are no such things as applied sciences, only applications of science." "A bottle of wine contains more philosophy than all the books in the world."









What neutron autoradiography tells us about Old Masters: The genesis of Jan Steen's "Wie die Alten sungen, so zwitschern die Jungen"

By Dr. K.Kleinert (HZB, Berlin) and M.Reimelt (Gemaldegalerie, Berlin)



Jan Steen, "Wie die Alten sungen, so zwitschern dte Jungen", 1665/66, сапvas, 84.8x 100.4 cm, Gemaldegalerie Berlin







The X-ray image already shows that large areas of the painting, like the drapery on the upper part of the archway on the right have been changed during the painting process.





Neutrons allow the visualisation of structures and layers beneath the surface and, in addition, enable the detailed identification of the elements contained in the pigments. Neutrons immediately clearly reveal the drapery and the landscape in the archway.





Today's version of the painting



Contour drawing of the reconstructed original version



(IBR-2, FSD, October 2011)

Top: 3D plot and map of the neutron diffraction pattern near (200) reflection during scan along zcoordinate in the "parasitic" grain region. Bottom: Total neutron diffraction pattern from single crystal turbine blade. The inset demonstrates (200) reflection shape evolution during z-scan.









Date 8 January 1989 Summary Engine fan blade fracture (design flaw), <u>Pilot error</u> Site Kegworth, Leicestershire, England <u>52°49'55"N 1°17'57.5"WCoordinates</u>: <u>52°49'55"N 1°17'57.5"W</u> Passengers 118 Crew 8 Injuries (non-fatal) 79 Fatalities 47 Survivors 79 Aircraft type Boeing 737–4Y0 Operator British Midland Flight origin London Heathrow Airport Destination Belfast International Airport



Neutron Reflectivity Reveals Suspected Air Layer under Water Drops on Lily Pads

• Hydrophobic forces govern protein folding, lipid aggregation, and hence life itself

 Dew drops roll off lily pads because their surfaces are hydrophobic ("water fearing")

An air layer has been long-suspected under such a drop

Removal of dissolved gases reduced the layer thickness; aeration increased it

Dhaval A. Doshi, Erik B. Watkins, Jaroslaw Majewski, Jacob Israelachvilli, PNAS Water

Air (10 Å)







Hydrophobic Polymer





Clean transport based on hydrogen technology requires synthesis of advanced proton conducting materials





Neutrons are unique to study light atoms positions in structurally disordered phases

CsDSO,

































































Tomography of padlock









