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Neutron Scattering

Here we present a short introduction to the history, theory, and application of neutron scattering.

History

1932 - James Chadwick discovers neutron (Nobel Prize in 1935)

While experimenting with the bombardment of Beryllium with Alpha Particles, a resultant radiation was observed with the property that it could penetrate matter much further than ordinary radiation (gamma rays, x-rays). This radiation could penetrate 10-20 cm of Lead,[1] while protons with similar momenta penetrated only 1/4 mm. This led Chadwick to conclude that the radiation was formed of neutral particles with mass comparable to protons (he was able to observe atoms recoiling and by estimating the maximal momenta change of the alpha particles, conclude the particle nature of the neutral, as well as it's mass).

1938 - Noble Prize Fermi for atomic scattering and absorption cross-sections of thermal / slow neutrons.

Fermi began bombarding elements with neutrons and found the absorption cross-sections of Hydrogen rich compounds (paraffin wax, water) were sufficiently high to cause the neutrons to transfer all their momenta, resulting in neutrons with velocities corresponding to thermal agitation (e.g. thermal neutrons)[2].

1943 - Graphite reactor at Oak Ridge National Lab establish basic principles of neutron scattering.

Use of graphite pile to thermalize neutrons produced as one of the plutonium production reactions. (Fission/Reactor source). The addition of a neutron to U-235 leads to the particularly unstable U-236, which breaks down by the reaction shown, producing three more neutrons, which are capable of repeating the reaction with other U-235 unless they have their energy significantly reduced by collisions (thus the graphite pile, although other reactors since have used significant pools of water to cool excess neutrons).

$$n + {}^{235}U \longrightarrow {}^{236}U \longrightarrow {}^{92}Kr + {}^{141}Ba + 3n$$

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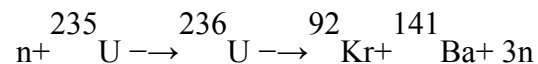
[1] Chadwick Nobel Lecture

[2] Fermi Nobel Lecture

1955 - First measurement of phonons by Brockhouse and Shull (Nobel Prize 1994)

Neutron scattering on Vanadium show an inelastic component that increases with temperature. Rules out Einstein model, and produces evidence for Debye model of specific heat.

1972 - Institut Laue-Langevin (ILL) in Grenoble, France comes online — one of the most intense thermal neutron sources in world.

1987 - Neutron Scattering helps identify the role of magnetism in high temperature super conductors.

Magnetic scattering due to the magnetic moment of the neutron allow for the inspection of magnetic characteristics in materials.

2010 - Lund, Sweden is chosen as the location for a new European Spallation Source. ESS to be completed in 2018-2019 with neutron beams up to 30 times brighter than today's.

Basic Principles

Neutron scattering is the result of (1) Bragg's scattering condition and (2) the neutrality of neutron.

Bragg's Condition

Bragg's condition is the result that for additive interference between scattered neutron (or x-ray) wave functions to occur, the incident wave vector, k_0, and scattered wave vector, k, must be related by $k_0 - k = Q$, where Q is a reciprocal lattice vector. This is equivalent to saying $Q \cdot R = 2 \pi l$ for any lattice vector R.[3]
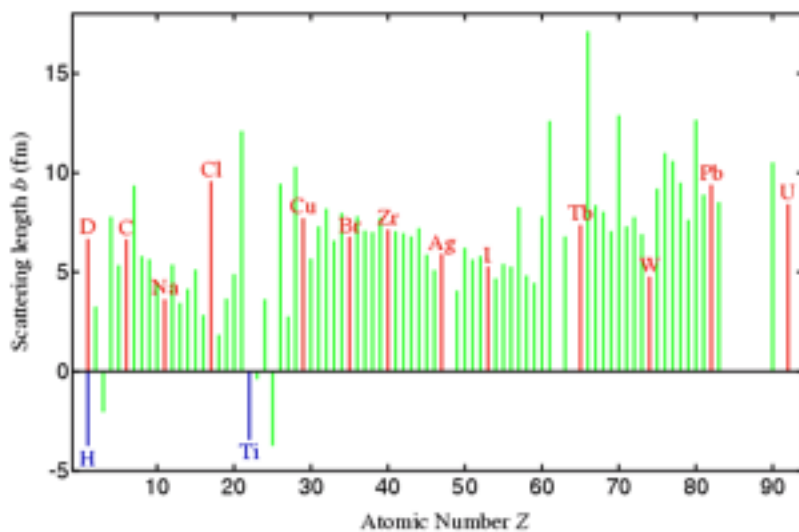
Plot of Scattering Length vs. Atomic Number from Birkbeck College, London

Neutrality of the neutron

The neutrality of the neutron allows for the neutron to penetrate much deeper into compounds than any charged or radiative source. This characteristic was what led to the discovery of neutrons by Chadwick in

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[3] Condensed Matter Physics, Marder, p.49

1932. As a consequence of non-interaction with the Coulomb potential, the neutron scatters off the nucleus due to the strong nuclear force (repulsive at ~1 fm). Compared to the x-ray interaction distance (on the order of 1 Å), this leads to the isotropic nature of neutron scattering off a single nucleus (neglecting magnetic interactions) because the nucleus is so small compared to the wavelength of the neutron.[4] Since the interaction between incident neutrons and nuclei is governed by the nuclear instead of Coulomb force, the amplitude of scattering does not increase with atomic number, rather it behaves erratically. This makes neutron scattering viable for small Z atoms such as oxygen or carbon.

However, the probing capabilities of neutron scattering come at a price: extremely small cross sections and the difficulty of production. The ability of neutron to penetrate into a material also gives the neutron the ability to fly right through without interacting, which means high flux beams are necessary to make experiments viable. Unfortunately, high flux beams are difficult to produce as there are only two primary techniques for the production of neutron beams: spallation sources and fission reactions.
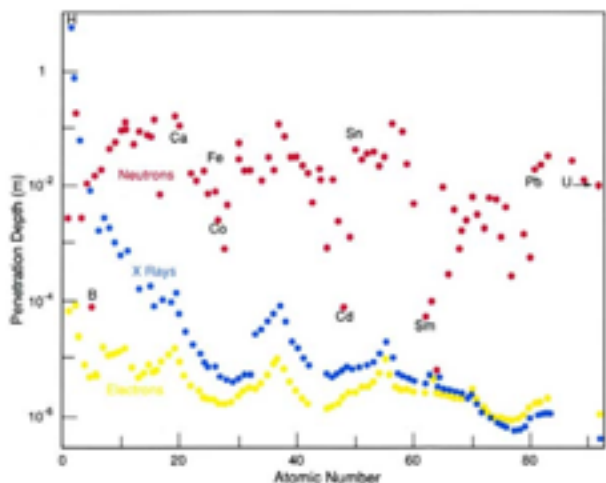
Spallation sources operate as follows; an ion source leads to a proton and two electron beam that is accelerated through a LINAC after which the electrons are stripped off by a foil, leaving the protons to enter the proton-accumulator ring, which bunches the protons (90% speed of light) before releasing them in pulses (60 Hz) to strike a liquid mercury sample, which generates neutrons (about 20-30 per proton).[5]

Fig. 2.1 The plot shows how deeply a beam of electrons, X-rays, or thermal neutrons penetrates a particular element in its solid or liquid form before the beam's intensity has been reduced by a factor 1/e, that is about 37% of its original intensity. The neutron data are for neutrons having a wavelength of 0.14 nm

Pynn Chapter 2 (The yellow plot is electron diffraction, it's difficult to see but electron is written in yellow).

A reactor source utilizes the breakdown of Uranium to generate neutrons and (typically) water to thermalize the neutrons prior to collection.

There are a limited number of neutron sources around the world, the larger ones are ILL at Grenoble, France, ISIS Rutherford Appleton Lab, UK, SNS Oakridge, J-PARC, Japan, and a new source is being planned for 2018-2019 in Lund, Sweden. These sources are respectively, reactor, spallation, spallation, spallation, and spallation. The reactor source at Oakridge was shut down in 1963, and the spallation source constructed in 2006.

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[4] Introduction to Neutron Scattering, Pynn, p. 4

[5] Spallation Neutron Source, Oak Ridge National Laboratory

<u>**Applications**</u>

The primary applications of neutron scattering are through powdered diffraction, and magnetic scattering. These techniques can be applied to metallic alloys, powdered minerals, ceramics, isotope-substituted materials, low atomic number compounds, antiferromagnetic, and ferromagnetic substances. Typically neutron scattering will be employed in addition to other techniques, such as x-ray diffraction.

Powdered Diffraction

Neutron beams can be used for powder diffraction, where the type of source dictates how the experiment is carried out. When a reactor source is used a filtering device called a monochromator is used to select neutrons with the same wavelength. After that, the neutron beam impinges upon the sample and the scattering patterns that emerge are concentric rings of varying amplitudes of intensity. A simple equation can be derived to relate the intensity of scattering and the radius of the ring; $K = 2k_0 \sin(\Theta/2)$, where K is the magnitude of a reciprocal lattice vector, k_0 is the magnitude of the incident wave, and theta is the scattering angle. This equation can be used to match the lengths of reciprocal lattice vectors with high intensity scattering angles.

If a pulsed spallation source is used instead of a reactor, the sample is hit by pulses of neutrons with wide ranges of energies. The resultant scattering is measured both as a function of the scattering angle, and as a function of when the neutron hits the detector (time of flight). The time of impact can be used to find the wavelength of the scattered neutron, and filter the superposition of rings corresponding to different wavelengths, resulting in a number of figures similar to those obtained from a single wavelength. The potential gain from this is a more efficient use of neutron beam flux as the beam doesn't have to be filtered to isolate one wavelength.

Magnetic Scattering

Neutrons are charge-less, but their quark composition leads to a (small) magnetic moment. This can be utilized in scattering experiments where the sample has ferromagnetic characteristics — the neutron can interact with the magnetic response of the sample instead of just the nuclear collisions in non magnetic materials. The magnetic moment of a neutron has a direction, and so contrary to the nonmagnetic scattering, the magnetic scattering is anisotropic. By aligning the moments, one can consider peaks where the reciprocal lattice vector is parallel to the magnetization, thereby eliminating the magnetic peaks and leaving only the nuclear ones.[6] This technique was applied by Bednorz and Müller to perovskite-type oxides to help elucidate high temperature superconductivity, for which they won the noble prize in 1987.[7]

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[6] Introduction to Neutron Scattering, Pynn, p. 10

[7] Nobel Lecture Bendorz and Müller

Inelastic versus Elastic Scattering

Elastic scattering occurs when the incident neutron energy is the same as the scattered neutron energy. This differs with inelastic scattering, which occurs when an incident neutron scatters off a vibrating nucleus allowing for either a gain or loss of energy. Specifically, the energy difference between k0 and k gives the energy imparted to the crystal (or taken from it), phonon energy = $\hbar\omega_{\vec{k}\nu}(n+\frac{1}{2})$; This gives a measurement of w_kv, which can be used to estimate the lattice constant.

Neutron Scattering applied to high-entropy alloys (HEAs)

High-entropy alloys are alloys produced with a nearly equi-molar ratio of 5 to 13 different elements. The large fraction of different elements produces distortion from typical crystal structures, which can strengthen the alloy. The experiment of Guo, Dmowski, and their colleagues was to analyze a ZrNbHf alloy with neutron and x-ray scattering. The former was preformed as powdered diffraction using a pulsed spallation source at Los Alamos National Laboratory, NM. The sample was tested for 14 hours, and the following plot obtained.
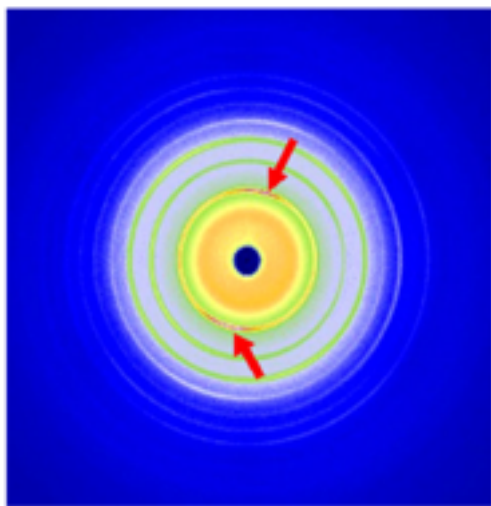

Fig. 1—Diffracted ring pattern of the ZrNbHf thin film. The continuous rings indicate fine grain microstructure and the high intensity regions arrowed in the first ring indicate preferred orientation.

Typically uniform rings are expected, however, Guo speculates that the sample being tested may have developed some texture on the thin film during production, which would result in the non-uniform bands seen.

The f.c.c. structure has 12 nearest neighbors while the b.c.c. structure has 8. So when the b.c.c. structure is distorted, some of the second nearest neighbors may be come nearest neighbors, blurring the distinction between the f.c.c. and b.c.c. phase. In fact, this is exactly what is proposed by Guo, which is partially verified by the radial distribution function plot shown below. Compared to the first, the second nearest neighbor shell is significantly broader, which corresponds to the distortion of the b.c.c. structure.
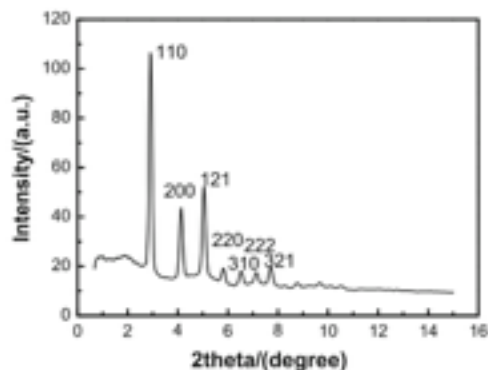
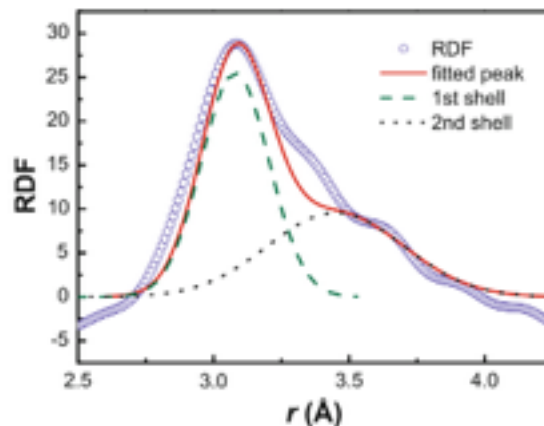Obtained by integrating the diffraction ring pattern seen above.

Fig. 6—The first and second peaks of the RDF for the $Zr_{1/3}Nb_{1/3}Hf_{1/3}$ alloy. A fitted peak originates from two Gaussian peaks, reflecting the two nearest neighbor shells in b.c.c. lattice.

References

1. Marder, Condensed Matter Physics Second Edition (2010).

2. Nobel Lecture Chadwick (1935).

3. Nobel Lecture Fermi (1938).

4. Nobel Lecture Bendorz and Müller (1987).

5. Pynn, Introduction to Neutron Scattering.

6. Pynn, Applications of Neutron Scattering.

7. W. Guo, W. Dmowski, J. Noh, P. Rack, P. Liaw, and T. Egami. Local Atomic Structure of a High-Entropy Alloy: An X-ray and Neutron Scattering Study (2012)