Introduction to Neutron Scattering and ORNL Neutron Facilities

Student: Xiangshi Yin
(Email: xyin3@utk.edu)
Instructor: Elbio Dagotto
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Xiangshi Yin
Department of Physics and Astronomy, The University of Tennessee,
Knoxville, Tennessee 37996, USA

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Abstract

Neutron scattering has been a very powerful probe in the research to the lattice and magnetic structures of various kinds of materials now. In this paper, I will first give a brief introduction of the history of neutron scattering. Then, in the second part, I will talk about the properties of neutron scattering, present its basic mechanism and unique properties. After that, I will give some examples to illustrate the powerful function of neutron scattering in condensed matter physics research. And finally, I will talk a little about some general ways of getting neutrons and neutron facilities in Oak Ridge National Laboratory(ORNL).

1 Introduction

Neutron was first discovered by Chadwick [1] in 1932 when he observed a penetrating form of radiation emanating from beryllium metal when activated by alpha-particles from a radium source. After that, Elsasser [2] first suggested that the motion of the neutrons could be determined by wave mechanics and thus would be diffracted by crystalline materials. The experimental demonstration of this idea was done by Halban and Preiswerk [3] and also by Mitchell and Powers [4]. In 1936, Bloch[5] first pointed out that the scattering of neutron from the spin and orbital magnetic moment of atoms could be observable if the magnetic moment of neutron is at the same order as the measured value of proton. That means neutron could be a probe to the magnetic structure of solid. The real foundations of the neutron scattering technique were built in the 1940s and 1950s when researchers from U. S. and Canada got significant flux of neutron from the 1st generation reactors. Wollan and Shulls work in ORNL on the research into the ferimagnetic states of Fe_3O_4 between 1948 and 1955 laid the basis for the future neutron diffraction experiments. By means of neutron scattering, they determined the magnetic structures of various alloys. And almost in the mean time, Bertram Brockhouse developed the inelastic neutron scattering technique at Chalk Rivers NRX reactor in the mid 1950s. He designed the first triple-axis spectrometer to analyze the spectrum of neutrons after scattering. After his work, inelastic neutron scattering becomes a powerful tool to detect the elementary excitations in solid, such as phonons and spin waves. In 1994, both Shull and Brockhouse were awarded the Nobel Prize for their ”pioneering contributions to the development of neutron scattering techniques for the study of condensed matter”.

Table 1: Basic properties of neutron

Property	Value
Mass	$1.67492729(28) \times 10^{-27}$kg
Charge	0
Spin	0
Magnetic moment(in the unit of μ_N)	-1.9130427(5)

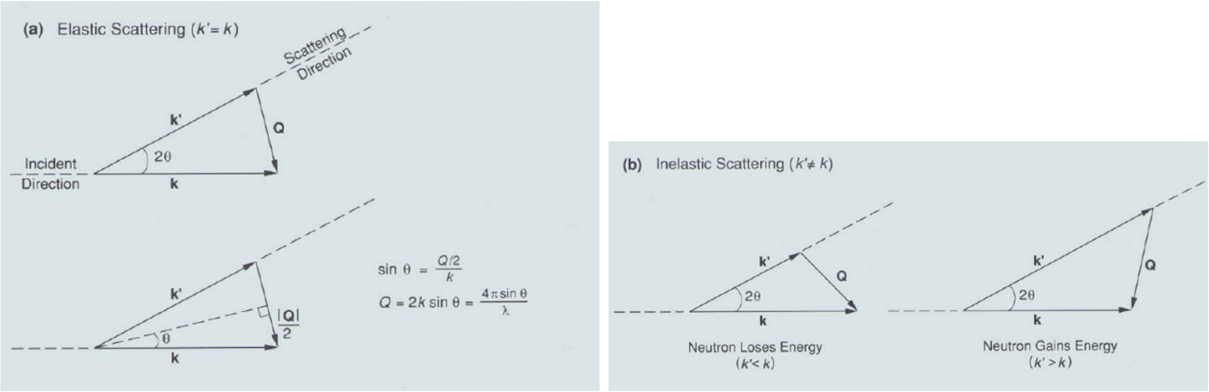


Figure 1: (a)Elastic scattering process (b)Inelastic scattering process[7]

2 Elementary properties and mechanism of neutron scattering

2.1 About neutron

2.1.1 Basic properties of neutron

When we start to look at neutron scattering technique, intuitively, everyone will ask such question—why do we use neutrons? Actually, there are a lot of probes we could use, STM, AFM, SEM, TEM, XRD and we could also use photo emission technique. Is neutron special to some extent? In order to answer this question, let's first have a quick look at some basic properties of neutron.

From table-1, it is obvious that neutrons have unique properties that make them a powerful probe of various materials. They have no charge, and unlike electromagnetic radiation or electrons, they only interact with atoms in the materials via short range nuclear-nuclear interactions. The interactions between incoming neutrons and the detected materials are so weak that generally neutrons can penetrate the materials far better than charged particles (like electrons) and X-rays(See Fig.2(Left)). So, it is a good probe to detect the bulk properties of materials. Neutrons also have spins and magnetic moments, so they can interact with the unpaired electrons of magnetic atoms. This property makes them ideally suitable to be used detecting the magnetic structures as well as magnetic excitations, such as spin waves.

2.1.2 Neutron temperatures

Neutrons are usually produced from a nuclear research reactor or a spallation source with energies usually millions of electron volts (MeV) [6]. Then, it will be slowed down by the so-calledmoderator to get proper wavelength comparable to the atomic

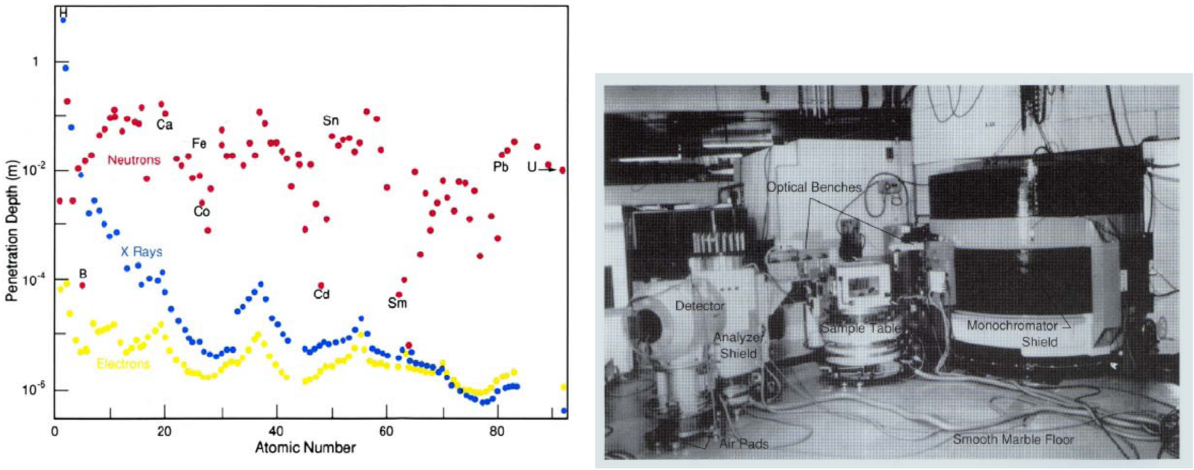


Figure 2: **(Left)**Comparison of penetration depth of different probes[7]. **(Right)**A triple axis spectrometer built at the institute Laue Langevin in Grenoble, France

spacing in solid or liquid and proper energy comparable to the dynamic processes in the materials. In the moderator, repeated collisions with atomic nuclei of small atomic weight (moderation) make the neutrons to a near-equilibrium distribution at a temperature at little above that of the moderator. A small tube through the reactor shielding admits a beam of the fast, intermediate and slow neutrons into the experimental hall. Depending on different moderating process, we could get neutrons with a wide range of energy as below: [8]

Fast neutrons have energy greater than 1 eV, 0.1 MeV or approximately 1 MeV, depending on the definition. **Slow neutrons** have an energy less than or equal 0.4 eV. **Epithermal neutrons** have energy from 0.025 to 1 eV. **Hot neutrons** have energy of about 0.2 eV. **Thermal neutrons** have energy of about 0.025 eV. **Cold neutrons** have energy from 5x10-5 eV to 0.025 eV. **Very cold neutrons** have energy from 3x10-7 eV to 5x10-5 eV. **Ultra cold neutrons** have energy less than 3x10-7 eV. **Continuum region neutrons** have energy from 0.01 MeV to 25 MeV. **Resonance region neutrons** have energy from 1 eV to 0.01 MeV. **Low energy region neutrons** have energy less than 1 eV.

The de Brogile wavelength λ could be expressed like:

$$\begin{cases} \lambda = \frac{h}{p} \\ p = \hbar k \\ E = \frac{p^2}{\sqrt{2mE}} \end{cases} \Longrightarrow \lambda = \frac{h}{\sqrt{2mE}}$$

So, if we consider the so-called thermal neutrons, it has energy of 25meV, the corresponding de Broglie wavelength should be 1.8 . This value is comparable with the interatomic distances in condensed matter, which makes the detecting of the special structure of materials possible. So thermal neutron scattering can yield structural information of condensed matter. Thermal and cold neutrons being in the range from 0.1meV to 100meV match very well with energies of elementary excitations of condensed matter. This enables one to investigate the dynamics of the elementary excitations of condensed matter. For excitations with energies higher than 100meV hot neutrons can also be used.

Although there are so many advantages for using neutron scattering as a powerful probe in condensed matter research, there are also some disadvantages in neutrons. The most significant one should be the weak flux we could get. For now, even use the most powerful neutron source, we could only get a flux in the order of $10^4 neutrons/mm^2 \cdot s$. However, in the X-ray instrument in the synchrotron source at the same energy scale, people could get much higher flux to the order of $10^{18} photons/mm^2 \cdot s$. Weak flux means relative weak intensity in the final scattering spectra, which is a not so good or people need to wait for much longer time to get a decent result. In other words, neutron scattering is a signal-limited technique.

Then, why people still use neutron scattering technique? Well, the answer maybe that people don't have other simpler and less expensive options to get such comprehensive information of materials as neutrons.

2.2 Rough picture of neutron scattering technique

2.2.1 Broad introduction[9]

As I said in the above section, there are basically two main interactions we need to consider in the neutron scattering process, one is the short range nuclear interaction, another is the magnetic interaction between the spin in the materials and the magnetic moments carried by neutrons. So, neutron scattering result is kind of superposition of both the nuclear scattering and magnetic scattering.

If we consider the energy loss in the scattering process, neutron scattering basically falls into two categories: elastic scattering and inelastic scattering.

Elastic scattering(Figure.1a) is a scattering process after which the emitting neutrons have more or less the same energy as the incident neutrons. It can be well applied to study the crystalline solids, gasses, liquids or amorphous materials and give complementary information containing both the lattice structure and the magnetic structure.

Inelastic scattering(Figure.1b) is a scattering process in which there is certain amount of energy transfer between the incoming neutrons and materials. So, the outgoing neutrons usually have different energy as the incoming ones. People usually use it to probe some elementary excitations in the materials, such as phonon excitations and spin waves.

Based on different research purposes, there are normally three categories of elastic scattering technique people usually use: neutron diffraction, small angle neutron scattering and neutron reflectory.

Neutron diffraction is basically the simplest elastic scattering process, like the X-ray diffraction technique, people could use it to investigate the lattice structure of condensed materials. And because of the deeper penetration, it is a good way to detect the bulk structure. Small angle neutron scattering is a good way to probe some larger scale structures. For example, in biophysics, people could use it to investigate some large molecules as DNA and protein. Neutron reflectory is a technique which could be used in the research to the surface structure and properties of materials. Although neutrons have deeper penetration and seems to be not a surface sensitive technique, it turns out that if the impinging angle between the incoming neutron wave vector and the surface is smaller than certain critical angle, the reflectivity of neutrons become significant. This could be used in surface science.

2.2.2 Sample preparation

Intuitively, for a complete scattering process, we should have a bullet and a target. The bullet here, obviously, is the neutron. As I mentioned above, we could get neutrons carrying various energies from neutron sources which could be used for different research purposes. For the sample, it could be either single crystal sample or polycrystalline sample. For single crystal sample, people usually interest in the properties of certain lattice plane. So, what people usually do before scattering experiment is to rotate the sample to find the correct orientation they want. For polycrystal sample, there is no such problem, because it is usually used to investigate the overall structure and properties containing informations from lattice planes with various orientations.

2.2.3 Scattering result

Normally, what people get from the neutron scattering experiment is the intensity of outgoing neutrons as a function of the momentum transfer Q and corresponding energy transfer E. There are a lot of ways to measure the momentum transfer depending on different neutron sources and different scattering process(elastic or inelastic).

In nuclear reactor source, people usually use monochromator to select single wavelength neutrons in elastic neutron scattering technique and triple-axis spectrometer in the inelastic scattering technique. The monochromator is usually an assembly of single crystals(often mad of either pyrolytic graphite, silicon or copper) each correctly oriented to diffract a mono-energetic beam of neutrons towards the scattering sample. The triple-axis spectrometer is also an assemblies of single crystals which are used to set the incident neutron wavevector and analyze the wavevector of the scattered neutrons. It records the scattered neutron intensity at a single wave vector transfer and single energy transfer.

In pulsed neutron sources, people usually use the so-called time-of-flight technique in which the detectors not only record the number of scattering neutrons at certain angular position but also the time when the neutrons arrive at the detector. Because the time when the neutrons come out from the source is known, the velocity of neutrons could be calculated and hence the momentum. So, basically the time-of-flight technique doesn't choose neutrons with certain wavelength, but only mark netrons with different energy.

2.3 Neutron scattering mechanism

2.3.1 Coherent and incoherent scattering

It turns out that[10]if we consider the interaction between neutron and materials to be the so-called pseudo potential as

$$V(\vec{r}) = \frac{2\pi\hbar^2}{m}\sum_i b_j 1 \delta(\vec{r} - \vec{r_i})$$

and if we apply Born approximation, we could get the intensity of outcoming neutrons as

$$I(\vec{Q}, E) = \frac{1}{h}\frac{k'}{k}\sum_{i,j} b_i b_j \int_{-\infty}^{\infty} \langle e^{-i\vec{Q}\cdot r_i\vec{(0)}} e^{i\vec{Q}\cdot r_j\vec{(t)}}\rangle e^{-iEt/\hbar} dt$$

Here, k and k' are wave vectors before and after scattering, b_i and b_j are the scattering length of different nucleus labeled by i and j. And the angluar bracket here means thermodynamic average value. Actually, the scattering length depends on specific spin states involved, and if we average the above equation over the nuclear spin states, the sumation part in the equation above becomes

$$\sum_{i,j}\langle b_i b_j\rangle A_{ij} = \sum_{i,j}\langle b\rangle^2 A_{ij} + \sum_i (\langle b^2\rangle - \langle b\rangle^2) A_{ii}$$

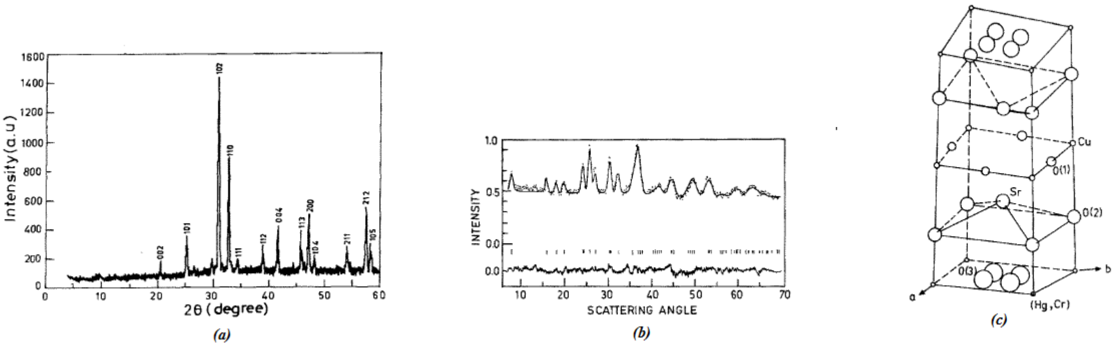
[11] The first term corresponding to the so-called coherent scattering, and the second part corresponding to the so-called incoherent scattering. Or, in the other words, the real scattering intensity we get is basically the superposition of the coherent scattering and incoherent scattering. The coherent scattering is actually the interference result of scattered neutrons from different neucleis, while the incoherent scattering is normally simple summation of neutron signals from different nuclei which means the phase of scattered neutrons are not correlated.

Of course, consider the energy loss, we have elastic coherent and inelastic coherent scattering, and also we have elastic incoherent and inelastic incoherent scattering. They normally carry different information of materials. Elastic coherent scattering tells us the equilibrium atomic distribution or the lattice structure of material, and inelastic coherent scattering carrys information of excitations of collective motions of atoms. Elastic incoherent scattering is normally the same in all directions, so it usually contributes as a background, while inelastic incoherent scattering usually contains information about the atomic diffusion in the material.

2.3.2 Neutron diffraction

Neutron diffraction is the simplest elastic neutron scattering. In this process, the Bragg's Law also works.

$$\begin{cases} 2dSin\theta = n\lambda \\ Q = \frac{4\pi Sin\theta}{\lambda} \\ d = \frac{2\pi}{Q} \end{cases}$$



(a) (b) (c)

Figure 3: (a)Powder X-ray diffraction pattern of $Hg_{0.7}Cr_{0.3}Sr_2CuO_{4.6}$ (b)Observed and profile-fitted diffraction patterns for $Hg_{0.7}Cr_{0.3}Sr_2CuO_{4.6}$, the difference pattern is shown at the bottom (c)A schematic of the refined structure[12]

Like the X-ray diffraction technique, what we usually get is a spectra of intensity of scattered neutrons as a function of the scattering angle 2θ as Fig.3(a). If we apply the Bragg's law, we could get a set of d values corresponding to various lattice planes with different orientations. Normally, it is not so easy to reproduce the real lattice structure from the diffraction sepctra, because it doesn't give us any information about the atomic distribution in the materials. So, people usually resort to some theoretical models,shift the atoms around until the predicted scattering spectra fits the real experimental result. Actually, Fig.3 is a really good example for this procedure, it is about a cuprate superconductor $Hg_{0.7}Cr_{0.3}Sr_2CuO_{4.6}$

2.3.3 Inelastic scattering

In reality, not all neutron scattering processes are energy conserved(neutron energy). In some cases, neutrons can experience some energy loss or even gain some energy. They can absorb or emit certain amount of quantized energy values which correspond to the phonon or spin wave energy. So, by measuring the energy change between outcoming and incident neutrons, we could get some idea of some elementary excitation process in condensed matters. And, the most frequently used method is the so-called "constant-Q method" developed by Brockhouse, in which people fixe the Q value and measure the corresponding energy loss and gain of neutrons. Fig.4(Left) is an example of phonon excitation peaks in the iron based superconductor $CaFe_2As_2$, and Fig.4(Right) is an example of the spin wave excitation peaks in the LCMO system with different dopings. In Fig.4(Right) you can see one peak splits into two peaks with positive and negative energy, which corresponds to the process of creat and annihilate a spin wave.

3 Neutron sources and neutron scattering facilities in ORNL

3.1 Neutron sources

There are two general ways to get neutrons, one is the research reactors and another is the spallation sources.

3.1.1 Research reactors

Research reactors are nuclear reactors that serve primarily as a neutron source. The intrinsic mechanism is the neutron-fission chain reactions (Fig.5(left)) where a neutron plus a fissionable atom causes fission resulting in a larger number of neutrons than was consumed in the initial reaction.

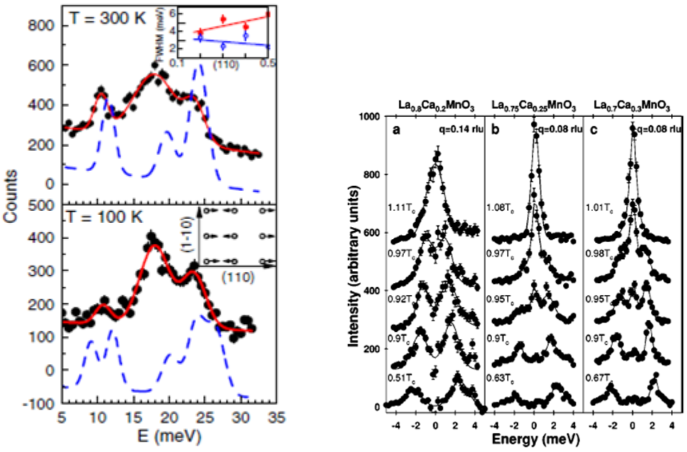


Figure 4: (**Left**) Energy scans taken at$Q = (2.5, 1.5, 0)$ of iron based superconductor $CaFe_2As_2$ at room temperature and at a temperature far below the structural phase transformation. The calculated phonon structure factors for nonmagnetic and spin-polarized are shown in the upper and lower panel, respectively. For better visibility the calculated profiles(dashed lines) in the lower and upper panels are shifted down by 200 counts. The insert in the upper panel shows the q dependence of the phonon linewidths of the brach around 18 meV(filled red circles) and around 24 meV(open blue circles). The lines were obtained by linear regression. The insert in the lower panel shows the displacement pattern of the zone boundary mode at $E = 18meV$. Only the Fe atoms are shown. All other atoms are at rest for this mode[13].(**Right**) Constant-q scans at[1+q,0,0] for LCMO20 at $q = 0.14rlu$ (a). LCMO25 at $q = 0.08rlu$ (b). and LCMO30 at $q = 0.08rlu$ (c). The solid lines are resolution-limited Gaussian fits to the data. The weak nonmagnetic contribution to the scattering at$\hbar\omega = 0$ has been subtracted from identical measurements at 10K[14]

This process will continue if the number of neutrons produced in a single reaction is capable of producing another fission process.

The so-called fissionable material here typically is the highly enriched uranium, typically 20% U-235. The elementary reactions involved are:

$$n +^{235}_{92} U \rightarrow^{236}_{92} U$$

$$^{236}_{92}U \rightarrow^{144}_{56} Ba +^{89}_{36} U + 3n + 177MeV$$

As I mentioned before, the neutrons coming from the reactor usually have high energy in the order of MeV. In order to slow them down, the reactors are usually surrounded by a neutron moderator such as regular water, heavy water, graphite, or zirconium hydride, and fitted with mechanisms such as control rods that control the rate of the reaction.

3.1.2 Spallation sources

Spallation is a process in which fragments of materials (spall) are ejected from a body due to impact or stress. Here, in the nuclear physics which we are interested in, the spallation is basically a process in which a heavy metal target (like mercury and tantalum) is hit by an incoming high energy species (like proton) and expel a large amount of neutrons(20 to 30 per impact). This concept of nuclear spallation was first coined by Nobelist Glenn T. Seaborg in this doctoral thesis on the inelastic scattering of neutrons in 1937[15]. Fig.5(Right) is a picture illustrating the basic process of nuclear spallation.

For the target, people tried the depleted uranium (DU, primarily composed of the isotope U-238) before because of its intense neutron production. However, its life time is too short. So generally, tantalum has been used.s

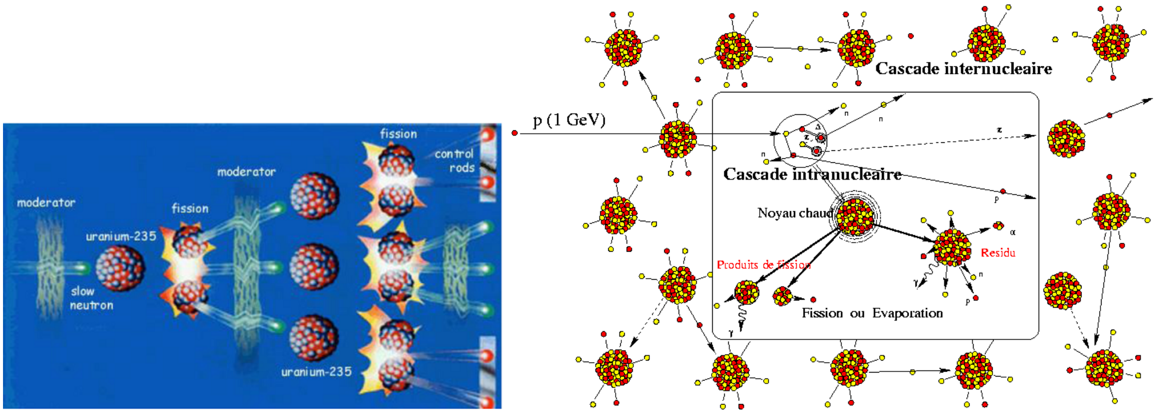


Figure 5: **(Left)**"Chain reaction process" **(Right)** Spallation process[16]

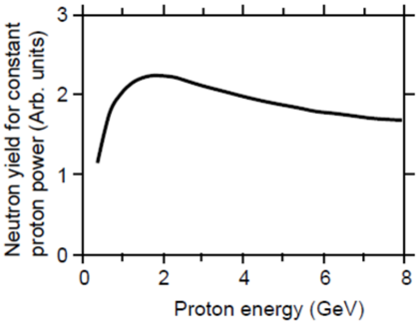


Figure 6: The relation between incident photon energy and the neutron yield[17]

Fig. 6 is a plot of the neutron yield as a function of the incoming proton energy. We can find that the proton energies in the range of 1 to 3 GeV prove optimal for neutron production via spallation reactions in heavy-metal targets, and production rate is directly related to the power deposited on the target.

As I mentioned before, the neutrons emerging fromthe spallation source usually carry energy in the order of MeV which is not suitable for the normal neutron scattering measurement. So, we also need a slow down process and corresponding moderator for this process. The velocity distribution is ultimately determined by the temperature of the moderator. Similarly, water at room temperature and liquid methane or liquid hydrogen at cryogenic temperature is most often used in it.

3.2 Neutron scattering facilities in ORNL

3.2.1 HFIR (High Flux Isotope Reactor)

The ORNL High Flux Isotope Reactor (HFIR) is the highest flux reactor-based source of neutrons for condensed matter research in the United States. HFIR is a beryllium-reflected, light-water-cooled and -moderated, flux-trap type reactor that uses highly enriched uranium-235 as the fuel. The preliminary conceptual design of the reactor was based on the "flux trap" principle, in which the reactor core consists of an annular region of fuel surrounding an unfueled moderating region or "island." This designation allows fast neutrons emerging from the reactor source to be moderated in the island and thus produces a region of very high thermal-neutron

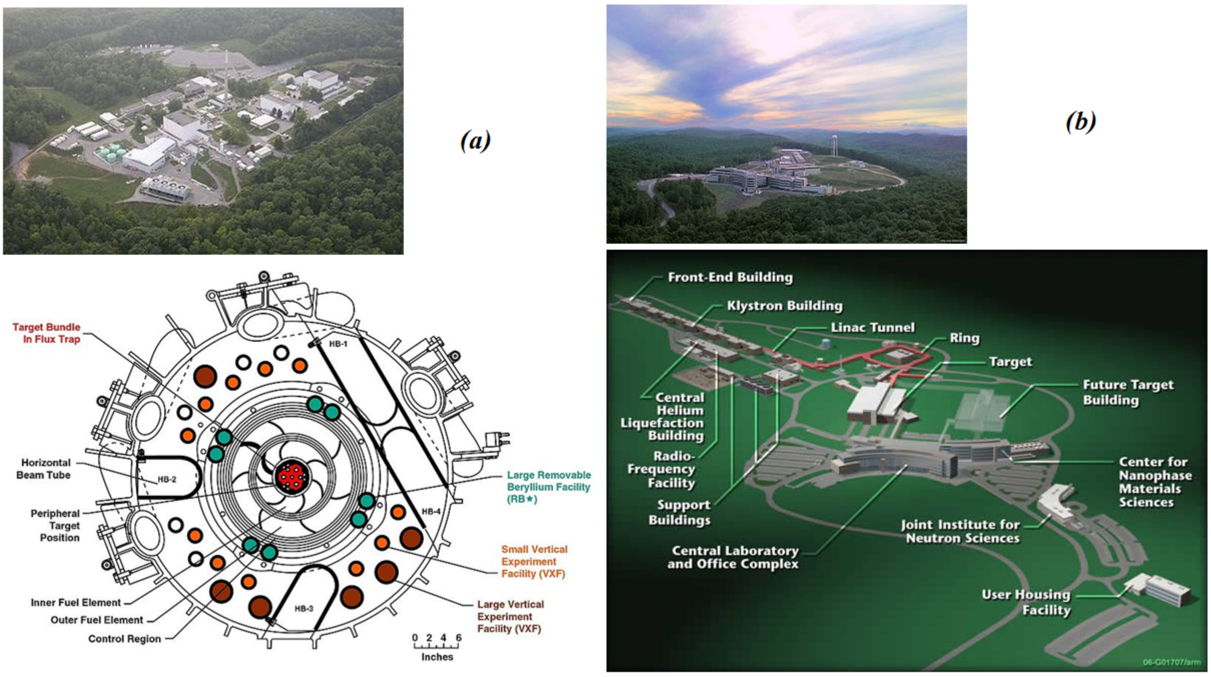


Figure 7: (a)Top view of HFIR and HFIR beam tubes and experiment locations (b)Top view of SNS and schematic picture of SNS components[18]

flux at the center of the island. This high flux neutrons may be tapped by extending empty "beam" tubes into the reflector, thus allowing neutrons to be beamed into experiments outside the reactor shielding. Finally, a variety of holes in the reflector may be provided in which to irradiate materials for experiments or isotope production.

3.2.2 SNS(Spallation Neutron Source)

SNS is an accelerator-based neutron source in Oak Ridge, Tennessee, USA. It provides the most intense pulsed neutron beams in the world for scientific research and industrial development.

The basic process for generating neutrons in SNS is like this:

Negatively charged hydrogen ions are produced by an ion source. Each ion consists of a proton orbited by two electrons. The ions are injected into a linear accelerator, which accelerates them to very high energies. The ions are passed through a foil, which strips off each ion's two electrons, converting it to a proton. The protons pass into a ring where they accumulate in bunches. Each bunch of protons is released from the ring as a pulse. The high-energy proton pulses strike a heavy-metal target, which is a container of liquid mercury. Corresponding pulses of neutrons freed by the spallation process will be slowed down in a moderator and guided through beam lines to areas containing special instruments such as neutron detectors. Once there, neutrons of different energies can be used in a wide variety of experiments.

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