International Tables for Crystallography (2018). Vol. H, Section 2.3.3.2, pp. 72–73.

the Intense Pulsed Neutron Source (IPNS) at the Argonne National Laboratory (operational from 1981 to 2008, 15 µA of 450 MeV protons, 30 Hz, target depleted U, moderator solid/liquid methane) are both worthy of mention for their work on techniques and applications at pulsed neutron sources; notable are contributions from IPNS on the subjects of high-temperature superconductors (Jorgensen *et al.*, 1987) and colossal magnetoresistance (Radaelli *et al.*, 1997).

The specifications and performance of modern currently operating spallation neutron sources will be presented in Section 2.3.3.3.

2.3.3.2. *Fission reactors for neutron-beam research*

Reactors used for neutron-beam research all rely on the fissile uranium isotope[9] ${}^{235}_{92}U$. This constitutes only about 0.7% of natural uranium; however, enrichment in this isotope is possible. A representative fission event would be

$$ {}^{1}_{0}n + {}^{235}_{92}U \rightarrow {}^{236}_{92}U \rightarrow {}^{141}_{56}Ba + {}^{92}_{36}Kr + 3{}^{1}_{0}n + 170\ \text{MeV}. $$

This equation indicates that a neutron of thermal energy is captured by ${}^{235}U$ to form ${}^{236}U$ in an unstable state, and in the majority of cases (88%) this breaks up almost instantly to yield fission products of intermediate mass, fast neutrons and energy. The unstable ${}^{236}U$ can break up in many different ways – there are usually products of intermediate but unequal masses, with masses distributed around 95 and 135 (Burcham, 1979), with the release of usually 2 or 3 neutrons (average 2.5; one of these neutrons is needed to initiate the next fission event), and of different amounts of energy (average around 200 MeV). As explained in Section 2.3.3.1, a chain reaction becomes possible if the fast neutrons released in the fission process are moderated to thermal energies so that they can be captured by another ${}^{235}U$ nucleus. Neutrons will lose energy most rapidly through collisions with nuclei of mass equal to the neutron mass, namely nuclei of hydrogen atoms, but collisions with other light nuclei are also quite effective. Hydrogenous substances are evidently useful, and water would seem ideal; however, there is some absorption of neutrons in water, so in some reactors, heavy water (D_2O, where D is ${}^{2}_{1}H$) is used since, as can be seen from the absorption cross sections (Table 2.3.2), thermal neutron capture in D is orders of magnitude less than for H. It has not been possible to achieve a self-sustaining chain reaction using natural uranium and light water as a moderator – for this reason uranium fuel enriched in ${}^{235}U$ and/or heavy-water moderators are in use. Adjacent to the reactor core is a so-called reflector, which is simply in place to moderate neutrons and prevent their premature escape. The energy released in the fission process ends up as heat, which must be dissipated (or used), so cooling is required – where light or heavy water is used as the moderator it can also serve as the coolant. Control rods are also essential – these are rods containing highly neutron absorbing materials, such as boron, cadmium or hafnium, which can be inserted into or withdrawn from the reactor to increase, maintain or reduce the thermal neutron flux as required. These control rods provide the means for reactor shutdown.

The neutrons in a reactor core range from the fast neutrons (∼1 MeV) released in the fission process, through epithermal neutrons (in the range eV to keV), which are neutrons in the process of slowing down, to thermal neutrons (∼25 meV), which are neutrons in equilibrium with the moderator (see Carlile, 2003). Evidently, for sustaining the chain reaction and for providing neutrons for diffraction instruments, the thermal neutrons are of the greatest interest. Neutrons in thermal equilibrium with the moderator have a Maxwellian distribution of energies, such that the number of neutrons with energies between E and $E + \mathrm{d}E$ is given by $N(E)\,\mathrm{d}E$, where

$$ N(E) = \frac{2\pi N_0}{(\pi k_B T)^{3/2}} (E)^{1/2} \exp(-E/k_B T). \quad (2.3.9) $$

Here N_0 is the total number of neutrons, T is the temperature (in kelvin) of the moderator, and k_B is Boltzmann's constant. The neutron flux is the product of the neutron density with the neutron speed, so the energy dependence of the flux distribution takes the form

$$ \varphi(E) = \varphi_0 \frac{E}{(k_B T)^{3/2}} \exp(-E/k_B T). \quad (2.3.10) $$

This distribution takes its peak value at $E = k_B T$; for a temperature of 293 K, this leads to a peak in the flux distribution at 25.2 meV (*cf.* Section 2.3.1). In the diffraction context the wavelength dependence of the flux is of more interest. Making use of the relationships $E = h^2/2m\lambda^2$ and $\mathrm{d}E/\mathrm{d}\lambda = -h^2/m\lambda^3$, we find that the variation of flux with wavelength can be described by $\varphi(\lambda)\,\mathrm{d}\lambda$, where

$$ \varphi(\lambda) \propto \lambda^{-5} \exp(-h^2/2m\lambda^2 k_B T). \quad (2.3.11) $$

This distribution peaks at $\lambda = h/(5mk_B T)^{1/2}$; at 293 K the peak in this wavelength distribution is at 1.15 Å. For some applications of neutron diffraction it may be desirable to have a greater neutron flux at shorter or longer wavelengths; as indicated in Fig. 2.3.5 this can be achieved by cooling or heating strategically placed special moderators.

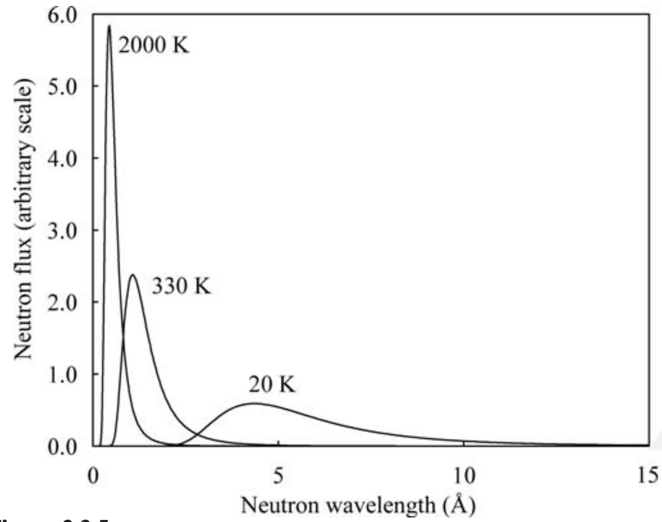

Figure 2.3.5
The Maxwellian distribution of neutron wavelengths produced within moderators at different temperatures. Reproduced from Kisi & Howard (2008) by permission of Oxford University Press.

As one specific example of a research reactor, we consider the NBSR located at the National Institute of Standards and Technology, Gaithersburg, USA. This reactor uses highly enriched (93% ${}^{235}U$) uranium in U_3O_8-Al as fuel, and heavy water as moderator and coolant. The thermal neutron flux in this reactor is 4×10^{14} n cm^{-2} s^{1}. It uses four cadmium control blades. An early plan view of this reactor and a cutaway view of the core assembly

[9] Bombardment with >1 MeV neutrons can cause the fission of the predominant uranium isotope ${}^{238}U$; however, there are too few neutrons at these energies to support a chain reaction based on this isotope.

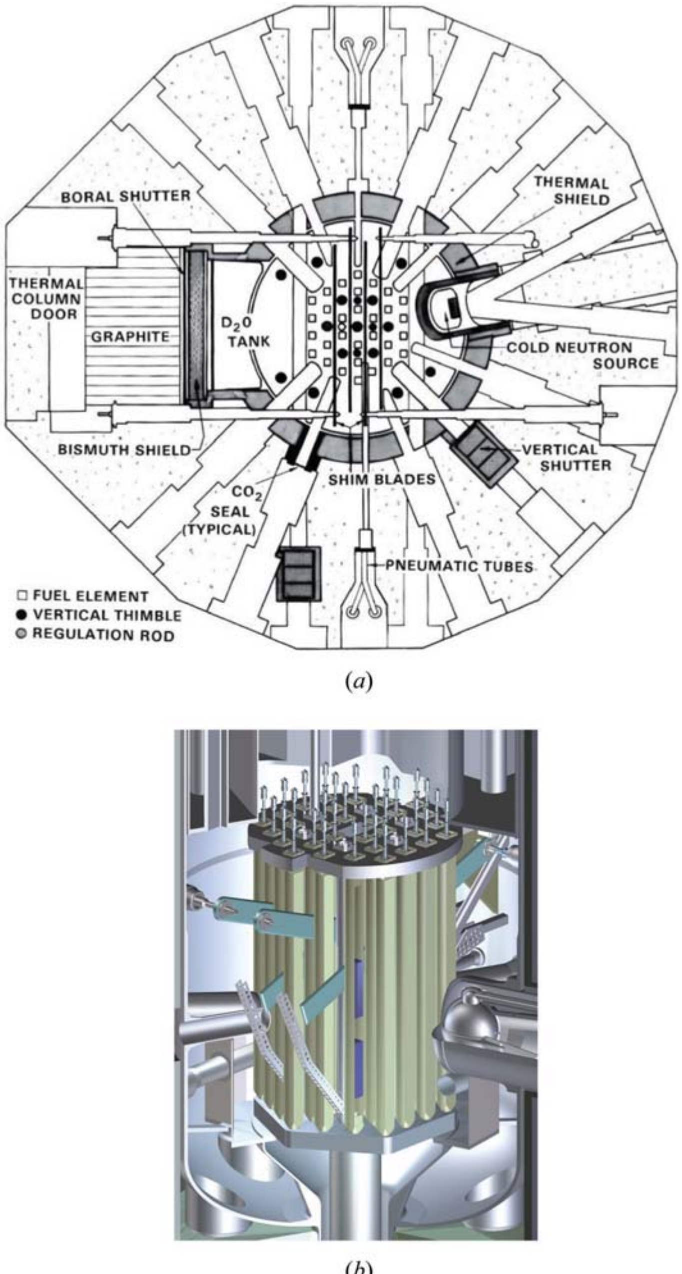

Figure 2.3.6
The NBSR at the National Institute of Standards and Technology Center for Neutron Research. Part (*a*) is a plan view (reproduced from Rush & Cappelletti, 2011) while (*b*) is a recent cutaway view of the reactor core showing the liquid-hydrogen cold source on the right-hand side.

are shown in Fig. 2.3.6. Note the presence of numerous beam tubes that allow neutrons to be taken out from the vicinity of the reactor core. This view of the NBSR (Fig. 2.3.6*a*) shows provision for a cold neutron source, and for beam tubes to transport cold neutrons to experiments, but it was years before any cold neutron source was installed. The first cold source, installed in 1987, was frozen heavy water; this was replaced in 1995 by a liquid-hydrogen cold source, and that was upgraded in turn in 2003. The NBSR first went critical in December 1967; the history of its subsequent development and use in neutron-beam research has been recounted by Rush & Cappelletti (2011).

The HFR at the Institut Laue–Langevin (ILL), considered to be the premier source for reactor-based neutron-beam research, serves as our second example. It too uses highly enriched uranium, here in a single centrally located U_3Al_x-Al fuel element, and it relies on heavy water for moderator and coolant. It operates at 58 MW and the thermal neutron flux is 1.5 × 10^{15} n cm^{-2} s^{-1}. The reactor incorporates two liquid-deuterium cold sources, operating at 20 K, and a graphite hot source operating at 2000 K. In the HFR, being of modern design and purpose-built for neutron-beam research, the beam tubes do not view the core directly, but are 'tangential' to it (Fig. 2.3.7); this reduces the unwelcome fast-neutron component of the emerging beams. The HFR achieved criticality in July 1971. More details on this reactor can be found in the 'Yellow Book' which is maintained on the ILL web site, https://www.ill.eu.

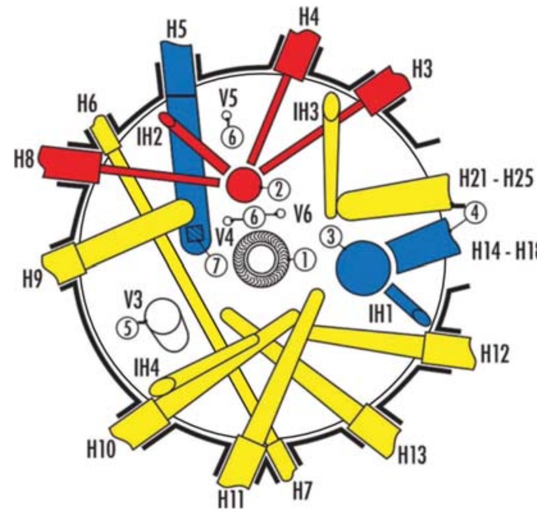

Figure 2.3.7
Schematic diagram of the HFR operated by the Institut Laue–Langevin in Grenoble, France. It has a compact core – the beam tubes avoid viewing the central core in favour of the surrounding moderator. This reactor also features hot (red) and cold (blue) sources. (Diagram reproduced with permission from the ILL from The Yellow Book 2008, https://www.ill.eu/fileadmin/users_files/Other_Sites/Yellow-Book2008CDRom/index.htm.)

From the opening paragraph of this section, it might be concluded that the more heavy water deployed, and the more highly is the uranium enriched in the fissile isotope ^{235}U, the greater the neutron fluxes that can be obtained. This conclusion would be correct, but concerns about nuclear proliferation have brought a shift to the use of low-enrichment uranium (LEU) in which the ^{235}U is enriched to less than 20%; however, in some reactors highly enriched uranium (HEU) with enrichment levels greater than 90% remains in use. Table 2.3.3 gives pertinent details on a number of research reactors important for neutron diffraction. Additional reactors are listed by Kisi & Howard (2008) in their Table 3.1, and a complete listing is available from the International Atomic Energy Agency Research Reactor Database (IAEA RRDB, https://nucleus.iaea.org/RRDB/RR/ReactorSearch.aspx).

2.3.3.3. *Spallation neutron sources*

The bombardment of heavy-element nuclei by high-energy protons, *i.e.* protons in the energy range 100 MeV to GeV, causes the nuclei to break up with the release of large numbers of neutrons. The word 'spallation' might suggest that neutrons are simply being chipped off the target nucleus, and indeed neutrons can be ejected by protons in a direct collision process with transfer of the full proton energy, but such simple events are relatively rare. In most cases there is a sequence involving incorporation of the bombarding proton into the nucleus, intra- and internuclear cascades accompanied by the ejection of assorted high-energy particles, including neutrons, and then an 'evaporation' process releasing neutrons from excited nuclei with