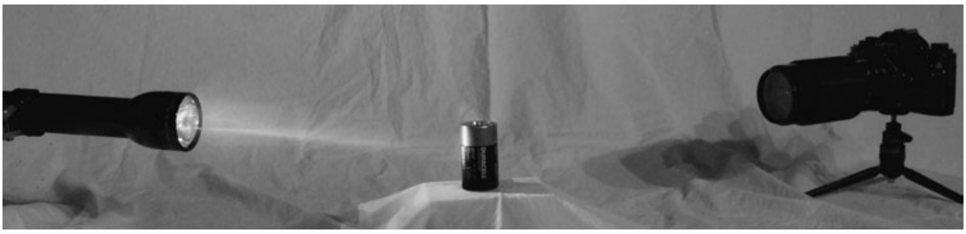
CAMBRIDGE

Cambridge University Press
978-0-521-85781-9 - Elements of Slow-Neutron Scattering: Basics, Techniques, and Applications
J . M. Carpenter and C.-k. Loong
Excerpt
More information

Introduction



A scattering setup: source, sample, and detector.

What is scattering?

When our neighbors and friends ask what we do, we often tell them that we shine beams of radiation onto an object and observe how they bounce off. From the details of the way they bounce, or *scatter*, we infer properties of the object. Studying the intensity variation of scattered light would reveal the optical transparency, the texture of the material, and its shape. We use neutron beams to study, on atomic scale, materials that we use in everyday life, as well as not-so-common substances that future technologies depend upon. From these studies, we reason how that fundamental information relates to the performance of materials and how to improve them. Other scientists, who study nuclei and subnuclear particles, sometimes say that their work is like hitting a watch with a sledgehammer and looking at the debris. We usually don't destroy our specimens in the process.

However, our methods differ fundamentally from what we are commonly accustomed to. With our eyes or a camera, we see a *direct-space* image of the structure of an object, that is, where each of the individual particles is. With scattering measurements, on the other hand, we record information on the correlations between positions of atoms or particles in space on the atomic and molecular scale. The measurements reveal the structure indirectly, through a mathematical relationship, a *Fourier transform*, which is a picture of the material in *reciprocal space*. Moreover, while direct-space images show each region of the object locally, scattering measurements are averages of the correlations over the entire volume of the sample material. Extending the analogy to particle motions, a camcorder records particle trajectories in a movie, whereas a scattering measurement infers dynamic information through color changes of the scattered light.

To record a clear picture, that is, one of high resolution, requires illuminating radiation whose wavelength is comparable to the size scale of interest in the object. To study excitations requires radiation whose energy is comparable to the excitation energies.

This book is about scattering – the radiations, the fundamental theory, the techniques, and the applications – and mainly about slow-neutron scattering as a probe of the atomic and magnetic structure and the motions in matter.

An overview of the field of neutron scattering

The very earliest slow-neutron scattering experiments were in-principle demonstrations using neutrons from radioisotope neutron sources and, later, low-energy cyclotron-driven sources. Available slow-neutron beam intensities grew immensely with the development of fission reactors after Enrico Fermi's demonstration in 1942. Pulsed electron linear accelerators giving bremsstrahlung photoneutrons entered the scene but were soon overshadowed by reactors that provided greater neutron fluxes. We briefly describe the evolution of reactors for slow-neutron scattering, then of accelerator-based pulsed spallation neutron sources and the advent of high-power proton synchrotrons.

Reactors

Early on, workers were mostly physicists investigating what kinds of measurements would be possible and quantifying fundamental aspects of neutron–matter interactions. In those years, the principal sources of neutron beams were the early nuclear reactors (see Mason *et al*. 2013), and the methods used were based on instruments developed for X-ray diffraction, a field then more than 40 years old. Soon, scientists began to apply neutron scattering methods to probe the properties of materials rather than just characterizing the interactions. The 1994 Nobel Prize in Physics acknowledged the early work of Clifford Shull and Bertram Brockhouse – Shull for his work on atomic and magnetic structure determination and Brockhouse for developing the triple-axis method for measuring the atomic and magnetic lattice excitations.

The pioneering works such as those of Enrico Fermi, Walter Zinn, Ernest Wollan, Shull and Brockhouse took place at reactors built in the 1940s and 1950s. The early reactors also contributed to important development of instrumentation,

Cambridge University Press
978-0-521-85781-9 - Elements of Slow-Neutron Scattering: Basics, Techniques, and Applications
J . M. Carpenter and C.-k. Loong
Excerpt
More information

in parallel with scientific discoveries. Ever more powerful research reactors evolved until the 1960s, when that technology reached engineering heat transfer limits and the available thermal neutron flux came to about 10^{15} n/cm^2/s. First at small reactors and later continued at larger sources, instruments and techniques were invented that enabled a greater variety and precision of measurements and a broader range of scientific investigations. Those developments continue to this day. The community of users expanded as the capabilities for measurements increased, the special properties of neutrons became more widely appreciated, and neutron scattering grew in importance as a probe for materials science investigations both independently and as a complement to other methods.

A milestone in these respects was the establishment of the Institut Laue-Langevin (ILL) in Grenoble and construction of the High Flux Reactor (HFR) in France, in 1972, with just over 10^{15} n/cm^2/s thermal neutron flux and 60 MW of power. The ILL HFR has not increased its neutron flux since its commissioning. But the scientific output of the ILL has steadily increased as special adaptations and new instruments came into play.

Cryogenic (liquid hydrogen) moderators that shift the neutron spectrum to greater-than-thermal-neutron wavelengths developed in the 1960s at the research reactors at Harwell, UK. The ILL reactor included two of these (20-K liquid D_2), and the extensive use of neutron guides made possible installing a large number of instruments. A hot source of 2000°C graphite extended the spectrum of useful neutrons to shorter-than-thermal wavelengths. Innovations and technical developments, along with a program of outreach to users outside the facility, fostered the growth of a very large and vigorous community of users. The same story played out elsewhere.

As this book is written, some of the older, less powerful reactors are being retired, while some highly optimized new reactors are coming online in response to demands for more and better facilities. The most recent of these are the FRM-2 reactor, in Garching, Germany, which grew out of the original AtomEi, FRM; OPAL at Lucas Heights, Australia; HANARO, in Taejon, Korea; and the China Advanced Research Reactor (CARR), in Beijing.

A pulsed reactor source is the main facility for neutron scattering in Russia, the 2-MW (average power) IBR-2 in Dubna. Research elsewhere along this line was quite active in the 1960s, but only the IBR-2 ever came into operation. Instantaneous neutron fluxes are very high, though pulse widths are somewhat too long for many applications, and with clever instrumentation, IBR-2 is very effective.

Accelerator-based neutron sources

Starting in the early 1970s with only a few tens of watts of proton beam power, accelerator-driven pulsed spallation sources have come to the fore. The pulsed time structure of the neutron beam is inherent in accelerator-based sources. Sorting neutrons according to the time they require to travel a known distance gives the neutron speed. The time-of-flight method, an efficient way to use more of the neutrons from the source, prevails at accelerator-based neutron sources. Precedents for time-of-flight methods, developed at cyclotrons and pulsed electron-linac-driven bremsstrahlung photoneutron sources, provided the basic ideas for neutron scattering applications. These sources had already reached the limits of power, about 50 kW, imposed by heat transfer engineering constraints on target design, and had limited neutron output because of the relative inefficiency of the neutron-producing process.

Spallation is the process induced by high-energy particles delivered by accelerators (usually giga-eV protons) striking nuclei in a massive target, which promptly emit neutrons to cool off. The spallation reaction produces several orders of magnitude less heat to be dissipated per useful neutron than the bremsstrahlung sources and about one-tenth as much heat as fission sources. *Pulsed* operation, the natural mode of most accelerator types, “on” and producing neutrons and heat during only a small fraction of the time and “off” the remainder of the time, enables heat removal at the time-average rate while the instantaneous power and neutron flux are very high. The pulses from accelerator-driven, *short-pulsed* spallation sources (SPSSs) are short enough to define the starting time for time-of-flight measurements. Pulses from *long-pulsed* spallation sources (LPSSs) are too long for most such purposes, but the duty-cycle heat transfer advantage remains. Compact low-energy particle accelerator-based sources are entering the scene.

Powerful accelerator-driven pulsed sources now coming online are poised to carry neutron scattering research to new levels of scientific effectiveness. And pulsed source instrument development, which depends heavily on the availability of computer resources, has made large strides as better and cheaper computers have continually allowed better capture and utilization of data (following Moore’s Law: computer capabilities double every one-and-a-half years). The number of detectors (and hence the efficiency) of instrumentation has grown remarkably. In addition, improvements in neutron optics (e.g., better guides and lenses) are opening up new opportunities.

Several laboratories have supported popular intermediate-level pulsed spallation sources: the Intense Pulsed Neutron Source (IPNS) at Argonne National Laboratory (ANL), 1981–2008; the Lujan Center at Los Alamos National Laboratory (LANL), 1985–2014; and KENS at the High Energy Laboratory (KEK), 1980–2008, in Tsukuba, Japan. Developments of pulsed spallation sources reached a high level in the United Kingdom at the ISIS facility, 1985, operating reliably with 160 kW of proton beam power, and now improved with 240 kW power. ISIS

Cambridge University Press
978-0-521-85781-9 - Elements of Slow-Neutron Scattering: Basics, Techniques, and Applications
J . M. Carpenter and C.-k. Loong
Excerpt
More information

showed the effectiveness of midlevel sources while continual improvements of instruments and techniques have supported a vigorous scientific program that is comparable to that of the ILL. And a new, second target station optimized for cold neutrons, came online in 2009, the ISIS TS-2. Also, megawatt-level pulsed spallation sources are coming online: the Spallation Neutron Source (SNS) at Oak Ridge National Laboratory (ORNL), 2006, for eventual 1.4-MW operation, and the Japan Spallation Neutron Source (JSNS), 2008, at the Japan Atomic Energy Agency (JAEA) complex in Tokai-mura, aiming for 1-MW operation. Recently authorities have chosen a site in Lund, Sweden, for the 5-megawatt European Spallation Source (ESS). A new project, the China Spallation Neutron Source (CSNS), has been launched in Guangdong, China.

Moderators in pulsed sources are more flexible than those in reactors – scientists can tailor their spectral and pulse-width properties to optimize the attached instruments. Workers realized early on that the pulsed sources readily adapt to installation of cryogenic moderators, *cold sources*. Relatively low nuclear heating power and radiation damage rates make possible the use of liquid hydrogen or deuterium, or even cryogenic hydrocarbons. Argonne's IPNS, for example, had three moderators – all cold and all methane (CH_4), two (solid) at 25 K and one (liquid) at 100 K – which produced copious fluxes of *cold* ($E < 0.005$ eV) *neutrons*, while also producing plenty of neutrons in the thermal and epithermal range, as well as pulse-width advantages. Still there is a great deal of room for further innovation of time-of-flight instruments, moderators, and target systems. IPNS, the Lujan Center, and KENS in Japan have been shut down, replaced by megawatt sources.

The steady spallation source SINQ at the Paul Scherer Institute in Switzerland, 1998, now operates with 1.3 MW of proton beam power. Because it employs the very efficient spallation reaction to produce neutrons, it generates neutron fluxes similar to reactors of $\sim$10 MW thermal power and, because it provides steady neutron beams, supports instruments with capabilities similar to those of midflux reactors.

The concept of the ESS, committed in 2009, is of a new type that has yet to be demonstrated, a Long-Pulse Spallation Source. In an LPSS, the moderators would be more efficient than in an SPSS, but most instruments would require pulse-width conditioning adaptations. An SPSS and an LPSS could share some of the same accelerator systems, or they could be separate facilities on different sites.

As existing steady and pulsed sources age and some are shut down, new, highly optimized facilities are replacing the old ones and extending their capabilities. Nevertheless, there is a projected shortfall in the number of available beams, as the community of users grows and demands more effective instruments (Comes 1994; Riste 1994).

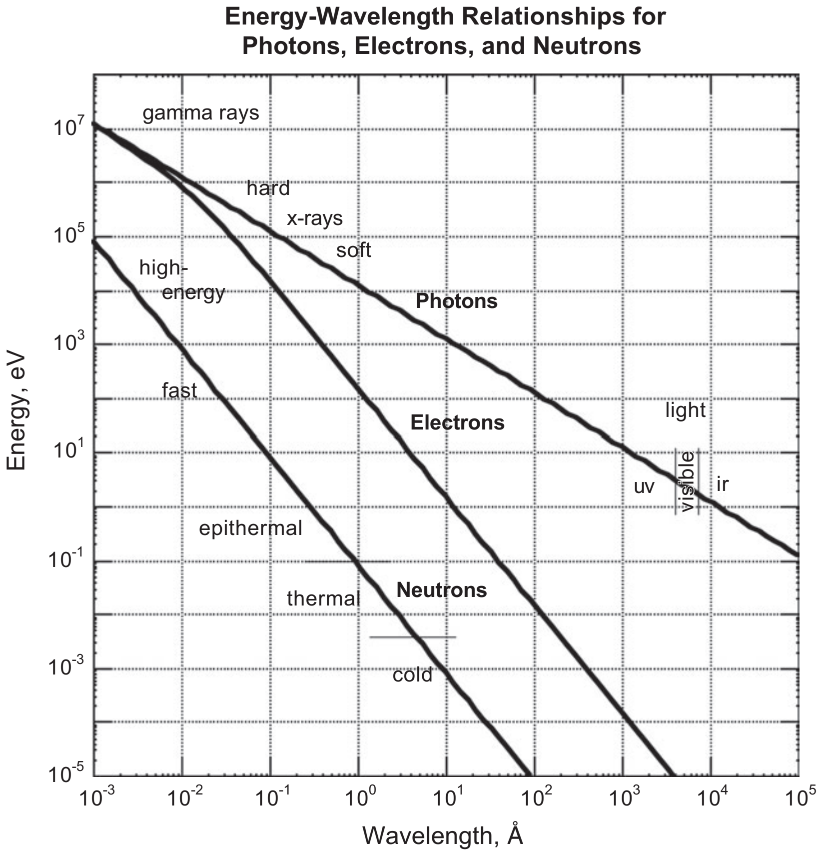


Figure I.1 Energy–wavelength (dispersion) relationships for neutrons, photons, and electrons.

Matters of general significance

Neutrons, photons, and electrons

The methods of slow-neutron, X-ray, and electron scattering are in many ways complementary, but each method presents its own advantages. A perceived difficulty with neutrons is the relative dimness of the sources, while X-rays from synchrotron radiation sources are very bright. Electron beams used in diffraction and microscopy are also relatively brighter than neutron beams by many orders of magnitude. In this book we devote most of our attention to neutrons and neutron scattering but offer some comparisons to other methods.

Here we examine the properties of neutrons, X-rays, and electrons as they relate to scattering studies. Figure I.1 shows the relationships between the energy and the wavelength of the various radiations used in materials science. Gamma rays are the most energetic photons in the electromagnetic spectrum. They are produced in violent cosmic events and, most commonly in our present context, in the decay

Cambridge University Press
978-0-521-85781-9 - Elements of Slow-Neutron Scattering: Basics, Techniques, and Applications
J . M. Carpenter and C.-k. Loong
Excerpt
More information

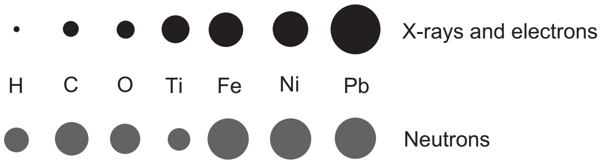


Figure I.2 The relative sizes (coherent scattering) of atoms as seen by (top) X-rays and electrons and (bottom) neutrons – different scales for each – for various elements.

of unstable nuclei. Strictly speaking, X-rays arise in the transitions among atomic electron states, but in materials science applications photons of similar energies come from acceleration and deceleration of electrons. Neutrons are unstable in free space (mean lifetime ~15 minutes, plenty of time for our uses.) Stable when bound in nuclei, they emerge at ~MeV energies from nuclear reactions and must be slowed down (*thermalized*) for our applications as described later. For scattering purposes the wavelengths are in the range 0.1 to 10 Å for X-rays and neutrons and 0.03 to 0.1 Å for electrons. Corresponding energies are 0.001 to 100 eV for neutrons, 10 to 100 keV for electrons, and 1 to 100 keV for photons.

For want of another place to do so, we note that, although Wilhelm Roentgen is widely said to have discovered X-rays in 1895, recent research[1] has revealed that Nicola Tesla had observed the same phenomena in 1891. Tesla never published his observations, however, but Roentgen published his.

Figure I.2 illustrates the relative scattering power of neutrons and X-rays for different elements.

Figure I.3, based on data from Rauch and Waschkowski (2003), illustrates the variation of X-ray and neutron scattering amplitudes as functions of atomic number Z. For X-rays, the scattering is proportional to Z and varies with the scattering parameter $s = \sin\theta/\lambda$ For neutrons, the nuclear scattering factors vary irregularly from isotope to isotope and are independent of wavelength because the interaction is with tiny atomic nuclei. There must be some theory governing the neutron-nuclear interaction, but it is so complicated that it is not possible to appeal to any underlying theory; consequently, the scattering amplitudes come from measurements.

Advantages of neutrons for scattering studies

Neutrons offer the following advantages as scientific probes:

- Neutrons are electrically neutral, they penetrate centimeters of most materials, enabling in-situ studies, and they are a "gentle probe" nondestructive of samples.

---

[1] James Mahaffey (2014), *Atomic Accidents*. New York: Pegasus Books. See page 5.

Cambridge University Press
978-0-521-85781-9 - Elements of Slow-Neutron Scattering: Basics, Techniques, and Applications
J . M. Carpenter and C.-k. Loong
Excerpt
More information

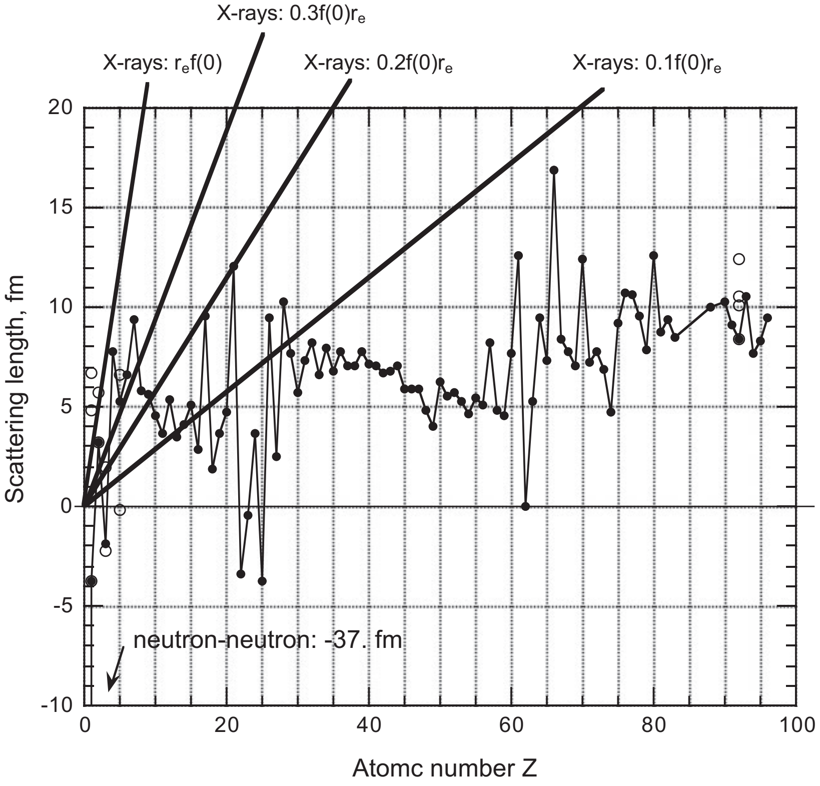


Figure I.3 Bound-atom neutron coherent scattering lengths and X-ray scattering amplitudes. In the figure, r_e is the classical electron radius, and (solid lines) the prefactors are representative ratios of $f(s)/f(0)$. (Closed symbols indicate neutrons.) The plot includes (open symbols) separated-isotope values for hydrogen, helium, lithium, boron, and uranium.

- Neutron cross sections exhibit no regular dependence on atomic number and are similar in magnitude across the periodic table, so they are sensitive to light elements, especially hydrogen, in the presence of heavy ones and capable of distinguishing similar-mass elements in the periodic table within a composite material.
- Certain large differences in isotopic scattering cross sections (e.g., H/D and ^{6}Li/^{7}Li) make neutrons especially useful for the study of light atoms in materials.
- The range of momentum transfers available allows researchers to examine structures on a broad range of length scales (0.1 to 10^5 Å).
- Slow neutrons cover a range of energies sufficient to probe a wide range of atomic and magnetic excitations (1.0 to 10^{-7} eV), as well as slow dynamic processes on time scales up to 10^{-7} s.
- Neutrons have magnetic moments and are sensitive probes of magnetic ordering and excitations.

- Neutrons can be polarized, allowing separation of the nuclear and magnetic cross sections.
- The simplicity of the magnetic and nuclear interactions facilitates straightforward interpretation of measurements.

Without requiring high resolving power relative to their energy, neutrons can be used to investigate, separately or simultaneously, energy levels or spatial arrangements of condensed matter. It is not true that various types of radiation can sense only the region around the dispersion curves of Figure I.1, because sensitive instrumental techniques sometimes make possible very high resolving power.

Neutrons suffer a major disadvantage in relation to synchrotron X-rays: the brightness (i.e., angular current density per unit fractional wavelength bandwidth) of the highest flux neutron sources is many orders of magnitude ($\sim 10^{18}$) smaller than the brightness of the brightest X-ray beams. This observation prompts a natural question "how can neutrons compete?" The answer is that the advantages listed previously overwhelm the intensity disadvantage in large fields of materials science, and for these applications neutrons predominate over X-rays.

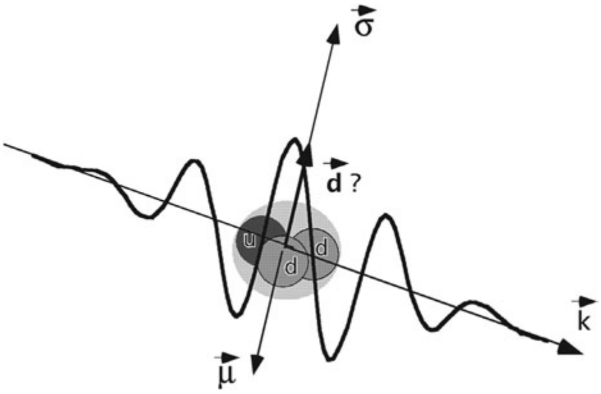
We should explain why we say *slow* rather than *thermal* neutrons, as it has been conventional to refer to the business of neutron scattering. It is because neutrons now used in scattering experiments range in energy far above and far below the 300-K, 25-meV nominally "thermal" range, as we explain further in Chapter 2. We collectively refer to all these energy categories as *slow neutrons*.

In addition to scattering studies of materials, sensitive experiments with slow neutrons are helping to unravel mysteries of quantum physics, cosmology, gravitation, and the standard model of particle physics. These applications are beyond the scope of this book, but see Snow (2013).

Our subject is, by parts, that of the source (flashlight), the beam (neutrons), the interactions with the sample (object), the recording device (camera), the image (picture), and its interpretations.

1

About neutrons



The neutron and its quark structure (udd), wave packet, wave vector, spin, and magnetic dipole and electric dipole moments.

1.1 The neutron as an elementary particle

As a fundamental particle, the neutron is one of the basic building blocks of material at the nuclear scale – the other is the proton, the nucleus of a hydrogen atom. James Chadwick, in 1932, first recognized the neutron as a product of irradiating beryllium with alpha particles in the reaction that we now denote $^9\mathrm{Be}(\alpha, n)^{12}\mathrm{C}$. Chadwick's discovery resolved numerous questions about the nature of atomic nuclei and for the first time allowed scientists to organize the atomic nuclei as consisting of neutrons and protons, which we now call the nucleons. As a member of the family of isotopes and elements, stable and radioactive, the neutron fills the "zero charge" slot, completing the list of nuclides on the low-mass side. Neutrons do not exist for long in free space – free neutrons have a mean lifetime of only about 881 seconds, ~15 minutes, (half-life 611 s, 10.2 min) and decay into a proton, an