PILE NEUTRON RESEARCH

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by

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PREFACE

The neutrons now available in high intensity from chain-reacting piles are being extensively used as tools for basic and applied research in many fields, either immediately at the piles or by means of radioisotopes produced by neutrons within the piles. The first controlled nuclear chain reaction was attained in December of 1942 and it was then possible to perform with ease experiments that had been almost impossible the preceding year. More difficult experiments were performed as time passed, and the complexity of the techniques involved increased correspondingly.

"Pile neutron research" is a distinct yet broad field of endeavor in which physicists, chemists, biologists, and engineers take advantage of the specific facilities that have been developed to make use of pile neutrons. It is the purpose of the present book to describe the techniques of this new field as completely as possible, with special emphasis on those methods that are peculiar to it. It is written primarily for those doing research utilizing the pile, including not only physicists studying the interactions of neutrons with matter as an end in itself, but chemists, biologists, and engineers who are using the neutrons as tools for studies in their own fields. Because of the special nature of the techniques involved, the book is not a text in the sense of one that deals with a basic subject such as mechanics, but it is designed to be useful in any basic course in science or engineering that involves the use of chain-reacting piles or radioisotopes. It should also be useful as a sourcebook in nuclear physics courses that cover the interactions of neutrons with nuclei, for many of the important results have been obtained with pile neutrons and we shall consider their measurement in our treatment of research techniques.

The general level of the book is that of a first-year graduate course in physics. However, the explanations for much of the physical phenomena involved are placed at a simpler level, in view of the needs of scientists in other fields, and in some cases, such as in the discussion of the various types of incoherent scattering in connection with neutron diffraction and reflection, the discussion is necessarily at a more advanced level. An effort has been made to include references that give thorough coverage of the basic physical principles whenever they cannot be treated fully. Coverage of every published pile neutron research finding in the manner of a review article is certainly not a goal; detailed descriptions and literature references are included only if they add to the understanding of neutron research. Reference to Atomic Energy Commission reports not generally available is avoided whenever possible, and they have been used only when space does not permit adequate treatment in the text. The existence of classified material in pile neutron research has not presented a serious problem because such a large fraction of the material is now unclassified. The recent release of constants of the fissionable materials (Chapter 1, reference 13) has aided greatly in the writing of Chapter 2, and has made it possible to include cross sections for fission neutrons hitherto unpublished (Section 4-2). Some pre-pile work has been included because it aids in the understanding of the later techniques, but descriptions of the old methods, many of which are of historical interest only, in general are not given.

The fundamental interactions of neutrons with matter, an understanding of which is essential for the discussions of the entire book, are first discussed (Chapter 1). The object here is not to present a mathematical development of these interactions, but to give a clear physical picture of neutron cross sections and such neutron phenomena as diffusion and moderation, that are based on cross sections. The mathematics is limited to that necessary in the explanation of the physical principles involved and for the later description of experimental pile research. The physical principles of the chain-reacting pile are treated in Chapter 2, again using only the necessary mathematics. The principal object of the discussion is to present the physical basis for the distribution of the neutrons in energy and position in the pile. The common types of piles are described, as well as the general facilities for research that are available with them. In all research with piles, a quantitative knowledge of the intensities of available neutrons is essential. The methods of determining neutron source strengths and fluxes, although somewhat complex, should be available to and understood by research workers, so Chapter 3 is devoted to the methods of "neutron standardization."

The actual research techniques that have been developed are considered in the order of the neutron energy, beginning with "fast" neutrons, of energy 1 to 14 million electron volts, in Chapter 4. The intermediate or "resonance" energy region, extending down to one electron volt, is treated in Chapters 5 and 6, while the broad field of slow (below one electron volt) neutron research occupies Chapters 7 through 11. In many of the methods of utilizing pile neutrons, such well-known instruments as Geiger-Mueller counters or ion chambers are used. No detailed discussion of these instruments is included because they are adequately treated elsewhere, and the techniques of pile measurements are concerned with the special use of these instruments, rather than with the instruments themselves. The new instruments unique to piles, however ("choppers," crystal spectrometers, and neutron mirrors, for example), are covered in detail, for their descriptions are generally unavailable.

Although research piles are of low power, the radiations emerging from them, and from the radioisotopes manufactured in them, are extremely dangerous if adequate precautions are not taken. Chapter 12 is devoted to a short discussion of the measurement of radiation intensity in terms of the effect on living tissue, and to the methods for avoiding radiation damage. Experience with chain-reacting piles has shown that a reasonable amount of care ("health physics") ensures protection from radiation accidents.

A complete table of slow neutron (thermal) cross sections, based on a critical evaluation of all experimental data on coherent and incoherent scattering, absorption, and activation cross sections, is included in the Appendix. These cross sections are in constant need for research at the pile as well as for preparation of radioisotopes, but are not usually available in one authoritative list. Those constants, conversion factors, formulas, and mass values that are frequently needed in connection with neutron research are also collected in the Appendix for convenient reference.

Preparation of this volume would have been impossible without the generous help of many workers in supplying data and in critical reading of the manuscript. I hope that they will in part be repaid by the usefulness of the book in their future research. The work of John Garfield and George Cox of the Brookhaven Graphic Arts Department has been of great value in the preparation of the illustrations. I wish to extend particular thanks to Miss Adele Kentoffio, without whose untiring efforts in typing and retyping, the task would have required twice the time.

D. J. H.

September 1952

CONTENTS

CHAPTER 1

FUNDAMENTAL CONCEPTS OF NEUTRON PHYSICS

1–1 The neutron. In 1930, physicists were seeking to understand a radiation, more penetrating than gamma-rays, that was produced when beryllium was bombarded with alpha-particles. It was clear that it did not consist of charged particles because ions were not produced in gas by the passage of the radiation. In addition, it was known that it could expel high-energy hydrogen and nitrogen atoms from materials, presumably by elastic collision. In 1932 Chadwick showed that this penetrating non-ionizing radiation could produce recoil protons of the energies observed only if its mass were approximately equal to that of the proton. Chadwick called this new particle the *neutron*, a term that had been used as early as 1921 in speculation concerning the possible existence of a neutral particle of protonic mass. The removal of a neutron from the beryllium by bombardment with alpha-particles is an example of a common type of nuclear reaction,*

$$Be^9 + He^4 \rightarrow C^{12} + n,$$

which is usually written more simply as

$$Be^9(\alpha,n)C^{12}.$$

The discovery of the neutron furnished the solution to a fundamental problem of nuclear physics, the nature of the particles in the nucleus. In the early 1930's, atomic nuclei were thought to be composed of protons and electrons surrounded by orbital electrons. The presence of electrons in nuclei was extremely unlikely because an exceedingly high energy would be necessary in order for the electron wavelength† to be as small as the size of nuclei (about 10^{-12} cm). However, no other method could be devised for explaining the fact that nuclear masses are about twice (in units of the proton mass) the nuclear charge (in units of the proton charge). The discovery of the neutron gave the answer, for it was necessary only to assume that nuclei were composed of approximately equal numbers of neutrons and protons. The neutrons thus add to the nuclear mass without increasing the nuclear charge and hence give rise to the observed charge-to-mass

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* An elementary discussion of the basic principles involved in the writing of an equation for a nuclear reaction is given in Chapter 5 of *Applied Nuclear Physics*, E. Pollard and W. L. Davidson, Jr. (Wiley, 1951) and in D. Halliday, *Introductory Nuclear Physics* (Wiley, 1950), pp. 369–379.

† This wavelength is the de Broglie wavelength, associated with every particle, which can be taken as a measure of the size of the particle; it is evaluated in Eq (1–4).

ratio. The large mass of the neutron relative to the electron, and hence the small wavelength, allows for the presence of neutrons of moderate energy (about 20 Mev) within nuclear dimensions.

For some time the only method of producing neutrons was by means of the (α,n) reactions. The most intense (α,n) sources were prepared[1] by mixing finely divided radium salt with powdered beryllium, to obtain intimate contact and thus prevent absorption of the alpha-particles in the radium itself. One curie of radium well mixed with several grams of powdered beryllium produces about 10^7 neutrons per second. These (α,n) neutron sources, although very weak compared with modern sources, proved to be extremely valuable for the study of neutron-induced nuclear reactions. The efficacy of neutrons in producing nuclear disintegrations arises from the fact that no electrostatic potential barrier exists for a neutron entering a nucleus. The nonexistence of the potential barrier was shown in a striking way when it was found that for most substances the probability of disintegration *increased* as the neutron velocity decreased. Such behavior would never be observed for charged particles, of course, because at low velocities they would not be able to approach the nuclei through the potential barrier produced by electrostatic repulsion. As the interaction of neutrons with nuclei was further investigated, it was found that the variations with neutron velocity and from element to element were indeed complex. It soon became clear that the nuclear interactions of neutrons could not be studied in the necessary detail with the sources utilizing natural radioactivity.

The available neutron intensities were increased greatly about 1937 when artificially accelerated particles were substituted for the natural alpha emitters. Deuterons and protons produced in cyclotrons, Van de Graaffs, and cascade transformers could be used to create neutron intensities that would have required tens of curies of natural alpha emitters. By modulating the sources in cyclotrons,[2] it was also possible to produce neutrons in sharp bursts so that those having a particular velocity could be selected by measurement of their time of flight to a detector. With the attainment of the nuclear chain reaction in 1942, neutron intensities of an entirely new order of magnitude became available, and the many techniques described in this book were developed to take advantage of the copious neutron fluxes.

1-2 Properties of the neutron. We have seen that the mass of the neutron is one of the properties that led to its discovery. The first measurements of the neutron's mass were made by an application of the laws of conservation of energy and momentum to the collision of neutrons with nuclei. Such measurements were only approximate but did show that the neutron had a mass very close to that of the proton. At the present time the neutron mass is accurately determined from mass spectrometer results combined with the measured energy changes in certain nuclear reactions. The mass difference between the deuteron and a neutron plus a proton (its

[1] Reference lists are at the ends of the chapters.

constituents) is known from the energy required, in the form of a gamma-ray, to disintegrate the deuteron:*

$$n + H^1 - H^2 = (\Delta M)_1. \tag{1-1}$$

The mass difference $(\Delta M)_1$ is obtained from the gamma energy E by use of the mass-energy conversion $E = (\Delta M)c^2$, or 1 atomic mass unit (physical scale) = 931 Mev. Another mass difference, that between the hydrogen molecule and the deuteron, has been carefully measured with the mass spectrometer:

$$2H^1 - H^2 = (\Delta M)_2. \tag{1-2}$$

These two mass differences can be combined to give the mass of the neutron in terms of the accurately known mass of the proton:

$$n = H^1 + (\Delta M)_1 - (\Delta M)_2. \tag{1-3}$$

The present value of the neutron mass obtained in this way, in atomic mass units, is†

$$M = 1.008989.$$

The last digit in the mass value represents millionths of a mass unit, or approximately kilovolts of energy. By this method, the neutron mass is established with an accuracy of several kilovolts. The neutron-proton mass difference can also be determined directly from the energy required in a (p,n) reaction, such as H^3(p,n)He^3 together with the beta energy of the target nucleus, in this case H^3. The mass of the neutron has been investigated by means of several (p,n) reactions recently[3] and the result, $M = 1.008982 \pm 0.000003$, which is somewhat more accurate than the value given above, is in excellent agreement with that value.

The neutrons emitted in fission are of sufficiently high energy so that they have a de Broglie wavelength‡ smaller than nuclear dimensions. The de Broglie wavelength, λ, is given by

$$\lambda = h/mv, \tag{1-4}$$

where h is Planck's constant and mv is the momentum of the neutron. The wavelength is expressed also in terms of the neutron energy, E:

$$\begin{aligned} \lambda &= h/\sqrt{2mE} \\ &= 2.87 \times 10^{-9}/\sqrt{E \text{ (in ev)}} \text{ cm} \\ &= 0.287/\sqrt{E \text{ (in ev)}} \text{ Ångstrom units (A).} \end{aligned} \tag{1-5}$$

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* Equation (1–1) states that the combined mass of the neutron and proton (more accurately, hydrogen atom) exceeds that of the deuteron by an amount $(\Delta M)_1$. In order to convert the deuteron to a neutron plus a proton, this amount of energy must be supplied, for instance by a gamma-ray.

† The actual values used to obtain the neutron mass are $(\Delta M)_1 = 0.002391$ mass units, $(\Delta M)_2 = 0.001552$, $M_H = 1.008150$.

‡ An interesting discussion of the wave properties of matter may be found in F. K. Richtmeyer and E. H. Kennard, *Introduction to Modern Physics* (McGraw-Hill, 1942), pp. 259–294, and W. Heitler, *Elementary Wave Mechanics* (Oxford, 1944), Chapters I and II.

The equations for the neutron wavelength are given in the simple nonrelativistic form because pile neutrons are of sufficiently low energy so that the relativistic corrections are negligible. As the neutrons lose energy by collisions in the pile, their wavelength increases rapidly. The wavelength of these slow neutrons becomes larger than nuclear, and even atomic, dimensions. The wave characteristics of the neutrons then predominate, and such phenomena as diffraction and interference become more and more evident. The most probable velocity for a neutron distribution in equilibrium with matter at room temperature corresponds to a wavelength of 1.8 A (Section 2–8) and neutrons have been isolated in beams with wavelengths as great as 25 A. Useful relations and values for neutron wavelength, energy, velocity, and temperature are given in Appendix 1.

In common with all elementary particles, the neutron has an intrinsic angular momentum, or spin. The spin is expressed in units of $\hbar = h/2\pi$ (h, Planck's constant $= 6.62 \times 10^{-27}$ erg-sec) and it is known that nuclei containing odd numbers of particles have spins that are equal to odd integral multiples of $\hbar/2$, that is, $\hbar/2$, $3\hbar/2$, $5\hbar/2$, etc. From the general behavior of neutrons it is quite clear that the correct spin must be $\hbar/2$, but the correctness of this spin can be shown very directly by means of the reflection of neutrons from mirrors (Section 11–4). The neutron has a magnetic moment associated with its spin and this magnetic moment has been accurately measured by utilization of "polarized" pile neutrons (Section 11–8). The present value for the magnetic moment is

$$\mu = -1.913 \text{ nuclear magnetons.}$$

The unit of the magnetic moment is the *nuclear magneton,* which is defined as $eh/4\pi Mc$, in analogy with the Bohr magneton, and differs from the latter only in the replacement of the electron mass by the proton mass M. A negative moment is one oppositely directed to the spin and, if resulting from a circulating charge, necessitates a negative charge. The actual methods involved in the neutron spin and magnetic moment work will be described in some detail in Chapter 11.

Inasmuch as the neutron is heavier than the proton, it is to be expected that it will disintegrate into the latter plus a beta-particle with an end-point energy given by the neutron-proton mass difference. The radioactivity of the free neutron has recently been demonstrated at the Oak Ridge Laboratory[4] and the Chalk River Laboratory[5] in Canada, using high intensity beams of thermal neutrons as sources of decaying neutrons. The half-life of the neutron has been shown to be in the range 5–15 minutes, and the energy of the beta-particle emitted is just that predicted from the neutron-proton mass difference, which is 0.78 Mev (Appendix 2).

The neutron is one of the fundamental particles that make up the structure of matter, and the question naturally arises concerning the structure of this fundamental particle. The neutron has no charge, yet possesses angular momentum and a magnetic moment, so it seems likely that the neutron itself has a reasonably complex structure. Modern *meson* theory seeks to explain the forces that bind nuclear particles by means of mesons of mass about 300 times the electron mass, which are exchanged between the nuclear particles. According to this picture, even a free neutron will emit negative mesons and quickly reabsorb them. It is thought that the meson "charge cloud" circulating around the proton, which is left after negative meson emission from the neutron, produces the magnetic moment of the latter. There should be a small electrostatic force between a neutron and an electron when the former is dissociated into a proton and meson; we shall see in Sections 10–5 and 11–7 how neutron interference and mirror experiments are used to measure this "neutron-electron interaction."

1–3 Cross sections as nuclear area. The many possible interactions of neutrons with matter are expressed quantitatively in terms of the *cross sections* for the various interactions. The concept of a nuclear cross section can be most easily visualized as the cross-sectional area, or "target area" presented by a nucleus to an incident neutron. If we consider nuclei as spheres of radius R cm and the neutrons as point projectiles, then the target area, or cross section σ, of each nucleus will be given by

$$\sigma = \pi R^2 \text{ cm}^2. \tag{1-6}$$

A neutron which passes normally through a thin sheet of material of area A containing N_T nuclei (Fig. 1–1) will have a probability $N_T\sigma/A$ of collision, provided that $N_T\sigma/A$ is small (i.e., that "overlapping" of the nuclei is negligible). The quantity N_T/A, which is the number of nuclei per cm² or the surface density of nuclei, is equal to Nt, where N is the number of nuclei per cm³, and t the thickness of the sheet. For an incident beam containing n neutrons per cm³, moving with velocity v, the number of neutrons passing through the sheet will be nv per cm² per second, and the collision rate can be expressed as

$$\text{collisions per cm}^2 \text{ per second} = nv \times N_T\sigma/A = nv\sigma Nt.$$

This result can be rewritten to give a simple evaluation of collision cross section:

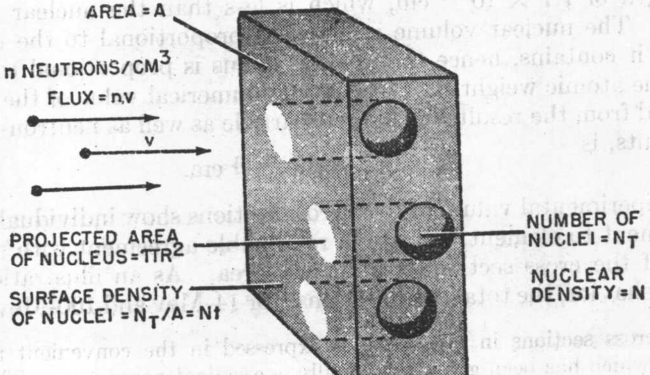

FIG. 1–1. Neutron cross sections as geometrical area of nuclei.

$$\sigma = \frac{\text{collisions per cm}^3\text{ per second}}{nvN}. \tag{1-7}$$

The quantity nv, which is an important item in most neutron experiments, would be called *flux density* in strict analogy with electrodynamics but the shorter term *flux* has come to be generally used in nuclear physics. Equation (1–7) shows that the cross section for collision is just the number of collisions per unit volume per second for unit incident flux and unit nuclear density.

The simple mechanical picture used to derive Eq. (1–7) involved only the collision cross section, without reference to the course of events ensuing after the collision. There are many possible nuclear interactions, but the cross-section expression of Eq. (1–7) applies to all these interactions as well as to the collision itself. The cross section for the particular process considered, whether neutron scattering, capture, or fission, is always proportional to the number of such processes occurring per cm³ per unit incident flux. The scattering cross section can be further divided into various *differential* scattering cross sections which give the probability for scattering per unit solid angle in particular directions. The number of neutrons removed from the beam, of course, is given by the sum of all the processes which can take place, and the collision cross section, which corresponds to the sum of all the processes, is usually called the *total* cross section. The partial cross sections for scattering, capture, etc., can be thought of as representing partial areas of the total cross-sectional area, σ_T.

Equation (1–7) shows directly that if the interaction rate per cm³ depends on the neutron density n and the density of nuclei N, but not on v, then σ must be proportional to $1/v$. Thus, the well-known "$1/v$ law," which applies in general to slow neutron capture, follows in a plausible way from Eq. (1–7), although the rigorous treatment of slow neutron cross sections is necessarily quantum mechanical and somewhat involved. The visualization of nuclei as spheres, of size larger than the neutrons, leads to reasonably correct values of total cross sections for neutrons of several Mev energy. According to Eq. (1–5) a 14-Mev neutron, for instance, has a wavelength of 7.7×10^{-13} cm, which is less than the nuclear radius for large A. The nuclear volume is taken as proportional to the number of particles it contains, hence the nuclear radius is proportional to the cube root of the atomic weight A. The actual numerical value of the radius, as calculated from the results of charged-particle as well as neutron-scattering experiments, is

$$R = 1.5 \times 10^{-13} A^{\frac{1}{3}} \text{ cm}. \tag{1-8}$$

The experimental values of total cross sections show individual variation from element to element, but are in reasonable agreement with the simple picture of the cross section as a nuclear area. As an illustration of this general behavior, the total cross sections* for 14-Mev and 100-Mev neutrons

* The cross sections in Fig. 1–2 are expressed in the convenient magnitude, 10^{-24} cm², which has been given the equally convenient name *barn*. There are, of course, the obvious secondary units, milli-, micro-, and mega-, barns. The cross sections plotted are from the 1952 compilation of the AEC Neutron Cross Sections Advisory Group.[6]

are given in Fig. 1–2 for a number of elements, as well as the cross section that follows from Eqs. (1–6) and (1–8):

$$\sigma = \pi(1.5 \times 10^{-13})^2 A^{\frac{2}{3}} \text{ cm}^2. \tag{1-9}$$

The actual cross section is expected to be larger than πR^2 because the neutron has a finite size and because the neutron waves are deflected somewhat by diffraction at the nucleus. This diffraction or "shadow scattering" is exactly analogous to optical diffraction and hence has an angular spread of the order of λ/R, where λ is the neutron wavelength. If the detector is so small that the diffracted neutrons are not detected then the cross section will be increased by πR^2 to a total of $2\pi R^2$. The cross section becomes $4\pi R^2$ at low energy, as will be seen from Eq. (1–13); the variation of cross section in the transition region is discussed by Feshbach and Weisskopf.[7] At 14 Mev, λ/R is of the order unity, so the total cross sections are expected to be $2\pi R^2$. The results of Fig. 1–2 agree well with the model of nuclei as impenetrable spheres, and indicate that at high energy the cross section is directly related to the geometrical size of the nucleus. The 14-Mev points lie slightly above the curve because of the appreciable size of λ; the 100-Mev points slightly below because of nuclear "transparency," which will be mentioned in Section 1–6.

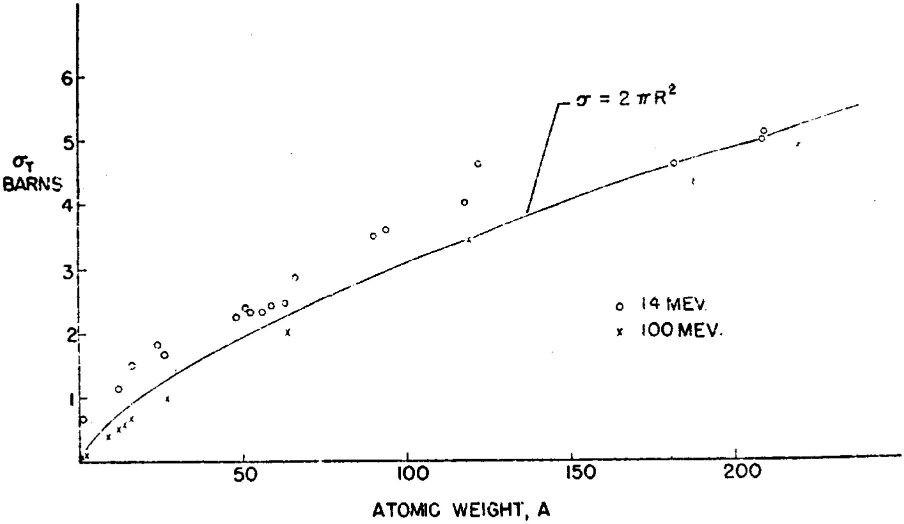

FIG. 1–2. Total cross sections[6] for neutrons at 14 Mev and 100 Mev, compared with $2\pi R^2$, the value for a nuclear area of πR^2 plus diffraction scattering equal to πR^2.

1–4 Quantum mechanical analysis of neutron cross sections. When interactions other than total collision are considered, or when the neutron wavelength becomes larger than the nuclear size, the simple mechanical picture of cross sections breaks down and it is necessary to use the correct quantum mechanical formulation of nuclear interactions. The quantum mechanical treatment is correct for all processes and for all neutron energies, but usually reduces to simple forms for limiting cases, and these forms lend themselves to physical models. Several of the quantum mechanical results that are of general applicability will now be discussed briefly. Most of these results will be taken up in greater detail later in connection with specific parts of pile neutron research. In fact, it can be said that practically all of neutron research deals with the measurement of neutron cross sections, because most of the nuclear interactions involving neutrons can be expressed in terms of cross sections.

In the quantum mechanical treatment* of cross sections the incident neutrons are considered as a plane wave that interacts with the potential field of the nucleus, producing a secondary scattered wave. The differential scattering cross section is then proportional to the square of the amplitude of the scattered wave. If the nuclear potential can be considered as spherically symmetrical, the amplitude of the scattered wave can be expressed as the sum of several *partial waves*. Mathematically, the amplitude is expanded in terms of Legendre polynomials and each partial wave corresponds to a particular polynomial. The differential cross section is the square of the amplitude,

$$\sigma(\theta) = \left|\frac{\lambda\!\!\!^{-}}{2i}\sum_{l=0}^{\infty}(2l+1)(e^{2i\delta_l}-1)P_l(\cos\theta)\right|^2,$$

$$\sigma(\theta) = \lambda\!\!\!^{-\,2}\left|\sum_{l=0}^{\infty}(2l+1)e^{i\delta_l}\sin\delta_l\,P_l(\cos\theta)\right|^2, \tag{1-10}$$

where $\sigma(\theta)$ is the scattering cross section per unit solid angle at the scattering angle θ, $\lambda\!\!\!^{-}$ is the neutron wavelength divided by 2π, and P_l is the Legendre polynomial of order l. Thus the P_l's give the angular distribution of the scattered neutrons. These distributions are more complicated for higher l's. For example, $P_0 = 1$, $P_1 = \cos\theta$, so $l = 0$ scattering is isotropic and $l = 1$ is a simple $\cos^2\theta$. The quantity δ_l, which is real for the case of scattering we are considering (no absorption), is the *phase shift* of the lth partial wave resulting from the nuclear potential, and its value determines the magnitude of the lth partial wave. All the symbols in Eq. (1–10) refer to the center of mass system (Section 1–10), which is the same as the laboratory system for scattering from heavy nuclei.

The total scattering cross section, valid in both the center of mass and laboratory system, is obtained by integration of $\sigma(\theta)$:

$$\sigma = 2\pi\int_0^{\pi}\sigma(\theta)\sin\theta\,d\theta = 4\pi\lambda\!\!\!^{-\,2}\sum_{l=0}^{\infty}(2l+1)\sin^2\delta_l, \tag{1-11}$$

and it is easily seen that the contribution to the cross section of any partial

* The present, necessarily brief, treatment of the partial wave analysis can be supplemented by reference to N. F. Mott and H. S. W. Massey, *The Theory of Atomic Collisions* (Oxford, 1949), pages 19–40, and L. L. Schiff, *Quantum Mechanics* (McGraw-Hill, 1949), pages 96–120.