Investigations of Neutron Collimators and Detector Systems Used in a Neutron-Capture Internal-Conversion Apparatus

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INVESTIGATIONS OF NEUTRON COLLIMATORS
AND DETECTOR SYSTEMS USED IN A NEUTRON-CAPTURE
INTERNAL-CONVERSION APPARATUS

by

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A thesis presented to the
Faculty of the School of Graduate Studies in partial fulfillment
of the
Degree of Master of Arts

Western Michigan University
Kalamazoo, Michigan
September 1964
ACKNOWLEDGMENTS

The author wishes to express his sincere appreciation for the invaluable help and guidance received during the course of this work from his advisors, Dr. S. B. Burson of the Argonne Physics Division and Dr. G. E. Bradley of the Physics Department of Western Michigan University.

The consultations with Dr. F. E. Throw regarding the composition of the manuscript are deeply appreciated. A word of acknowledgment is also due Dr. E. B. Shera and Teymoor Gedayloo of the Argonne Physics Division for their valuable participation in planning and executing the experiments.

Gratitude is expressed to Dr. C. C. Trail, Dr. S. Raboy, and Dr. H. H. Bolotin of the Argonne Physics Division for their cooperation in making some of the neutron beam experiments possible and, in addition, to Harry M. Mann of the Argonne Electronics Division, who provided the solid state detectors.

The author is indebted to the Argonne National Laboratory for providing the facilities used for this research and also to the Atomic Energy Commission under whose auspices the Argonne Graduate Thesis Program is conducted.
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PREFACE

During the past six decades considerable effort has been devoted to investigating and explaining the phenomena associated with the atomic nucleus. However, many important features of the nucleus still are not understood. Among these are such fundamental aspects as the structural composition of the nucleus, the nature of the forces between nucleons, and several of the basic processes that occur among the nuclear constituents. The goal of nuclear physics is to construct a unified theory which is able to explain all nuclear phenomena.

The experimentalist contributes toward this objective by gathering and classifying extensive amounts of experimental data in the conviction that some systematic patterns will develop and that these will suggest general trends. The theoretician uses these experimental data first as a guide in constructing new theories and then as a means to substantiate or refute his hypotheses. Thus, experimental data are prerequisite to the development of any acceptable theory.
CHAPTER I. INTRODUCTION

One method of determining the nuclear parameters that describe the states of a particular nuclide is studying the gamma radiation that results when the nucleus undergoes a transition from an excited state to a level of lower energy. From measurements of the energies, the multipolarities, and the relative transition probabilities of the emitted gamma rays, a decay scheme can be derived and spins and parities assigned to the various levels. A convenient experimental method for studying gamma transitions is provided by the internal-conversion process. In order to understand the processes of gamma radiation and internal conversion, consideration must be given to the charge distribution of the nucleus.

I. NUCLEAR MULTIPole FIELDS

As with any charge distribution, the electric field surrounding the nucleus can be expressed as a field produced by a combination of pure multipoles. If the distribution of the positive charges in the nucleus were spherically symmetric, the potential would simply be that of a single charge Ze located at the center of the nucleus, i.e., a monopole at the origin. However, the nuclear charge
distribution is not usually spherically symmetric and higher order multipoles must be included in the source.

In order for a nucleus to have a static dipole moment, the time-average charge distribution would require a greater concentration of protons on one side of a plane passing through the center of mass than on the other. That is, the center of charge would not correspond to the center of mass. The lack of any experimental observation of an average nuclear electric dipole moment indicates that it is unlikely that such a moment exists. Theoretical arguments point to the same conclusion.

A spheroidal charge distribution with its major axis along the nuclear spin axis is one more consistent with the symmetry requirements. Such a configuration can give rise to an electric quadrupole moment. Nuclear quadrupole moments of many nuclides have been measured. Nuclei with both negative and positive values have been found, the negative value corresponding to an oblate shape and the positive value to a prolate shape of the charge cloud. The largest values for the quadrupole moments occur for the nuclei in the regions of permanent deformation where \( A \approx 25 \), \( 150 < A < 190 \), and \( A > 222 \). These groups correspond to nuclei lying approximately midway between the magic numbers, i.e., closed shells of neutrons or protons.

Static magnetic dipole moments are also found in some nuclei. These are presumably due to proton motions which simulate current loops within the nucleus. The magnitude of higher order
static multipole moments is very small and the effects of higher order moments will not be considered here.

II. GAMMA RADIATION

The absence of static multipole moments in the nucleus does not rule out the possibility that there could exist an oscillating field whose time-average value is zero. A mode of vibration in the nucleus, most likely the result of some perturbation, could cause it to radiate in exactly the same manner as does an oscillating multipole. In fact, directional correlation experiments with gamma rays show that some nuclei do indeed emit electric-dipole radiation. Gamma rays corresponding to the radiations from higher order oscillating multipoles, both electric and magnetic, have also been observed.

Conventionally, the different multipoles are spoken of as \(2^L\) poles, where

\[
\begin{align*}
2^1 & = \text{dipole} \\
2^2 & = \text{quadrupole} \\
2^3 & = \text{octupole} \\
2^4 & = \text{hexadecapole (or simply a } 2^4 \text{ pole), etc.}
\end{align*}
\]

The notation used in classifying gamma rays specifies the type and multipole order of the oscillating field responsible for the radiation by giving the value of \(L\) prefixed by either \(E\) or \(M\). For example, a gamma ray due to electric-dipole radiation would be called \(E1\), that from a
magnetic quadrupole M2, and so on. The photon energy $h\nu$
will be equal to the difference between the energies of the two
states involved minus the recoil energy of the nucleus. The latter is
comparatively small and is usually neglected. Therefore,

$$h\nu = E_i - E_f,$$

where $E_i$ and $E_f$ are the energies of the initial and final states, respectively.

III. INTERNAL CONVERSION

Instead of emitting electromagnetic quanta, the nucleus
can interact through its multipole field with an orbital electron and transfer
energy to it. Thus, the nucleus can pass to a lower energy state by
ejecting an electron from the atom. This process, called internal
conversion, is a mode of de-excitation that competes with gamma emission.

Once outside of the Coulomb field of the nucleus, the
electron will have a kinetic energy $E_e$ equal to the energy of the transition
minus the electron binding energy $E_b$, i.e.,

$$E_e = h\nu - E_b,$$

Since the binding energies of the orbital electrons are tabulated for all the
elements, the transition energy can readily be determined from a
measurement of the kinetic energy of the emitted electron.
The similarity between Eq. (2) and Einstein's photoelectric equation explains the misleading name "internal conversion." It was first thought that the process involved the emission of a gamma ray from the nucleus; but instead of escaping from the atom, the gamma ray interacts with an orbital electron and ejects it from the atom by transferring all the gamma energy to it in a fashion analogous to external conversion. 

In order to explain the experimental fact that the probability for internal conversion depends upon the atomic number in a vastly different way than does external conversion, it was suggested that internal conversion was actually a primary quantum-mechanical process directly competing with gamma emission. This hypothesis was further supported by the fact that gamma radiation is strictly forbidden in a transition between two nuclear states of zero spin; whereas, internal conversion has been observed in 0-0 transitions. This is clear evidence that the interaction is directly between the atomic electrons and the nucleus, not through an intermediary gamma ray.

The ratio of the probability for internal conversion to that for gamma emission is called the total internal-conversion coefficient, usually designated by $\alpha$. Since internal conversion can occur in several of the electron subshells, $\alpha$ is the sum of the individual conversion coefficients for all the subshells, i.e.,

$$\alpha = \alpha_K + \alpha_{L_1} + \alpha_{L_2} + \cdots + \alpha_{M_1} + \cdots,$$
where $a_K$ is the internal conversion coefficient for the K shell, $a_{L_1}$ is the coefficient for the $L_1$ subshell, etc.

The theoretical values of the internal-conversion coefficients can be calculated. They are functions of (1) the energy of the transition $E$, (2) the atomic number of the nucleus $Z$, (3) the multipole order of the transition $L$, (4) the type of transition, electric or magnetic, and (5) the electronic wave function for the subshell in which conversion takes place. Tables of such calculated results can be found in the works of Rose and Sliv and Band. If the spin and parity of one of the states involved in a transition is known, the internal-conversion coefficients can be used in assigning the spin and parity to the other state. This is done by experimentally determining the conversion coefficients for the transition and comparing them with the theoretical values. From this the multipole order and the type of transition can be specified; and this, in turn, allows the spin and parity changes to be deduced.

IV. PROMPT INTERNAL CONVERSION

When a nucleus captures a thermal neutron, the result is a compound nucleus with excitation energy equal to the binding energy of the neutron, about 8 MeV. The compound nucleus has an extremely short lifetime ($<10^{-14}$ sec.). It decays to states of lower energy by means of gamma radiation or internal conversion. This two-step process of compound-nucleus formation and subsequent decay can be written symbolically

\[ \text{Neutron capture} \rightarrow \text{Compound nucleus} \rightarrow \text{Decay} \]
\[ Z^A_N + n \rightarrow Z^{A+1}_N \rightarrow Z^{A+1}_N + \gamma \text{ rays} \]
\[ \rightarrow \text{conversion electrons} \] (4)

where \( X \) represents the chemical symbol and the braces signify a compound nucleus. (Although particle emission is possible, the probability for this is relatively small and is not considered here.)

This short lifetime explains the common nomenclature, "prompt" radiation. However, during the decay process many intermediate states are populated, including longer lived isomeric states. This permits the properties of the isomeric states to be investigated also.

The phenomenon of prompt internal conversion was first reported in 1938 by Hoffman and Bacher. They found that when cadmium was irradiated with slow neutrons it would cause an x-ray film to become darkened. By using absorbers of different \( Z \) but the same number of atoms per square centimeter, they were able to show that the darkening was due to electrons. Since there was no long-lived activity when the neutron source was removed, it was deduced that the electrons were due to prompt internal conversion. Sala et al. were unable to measure the time interval between the capture of the neutron and the emission of the electron, but set an upper limit of \(<10^{-7}\) sec.

These findings were further supported by Amaldi and Rasetti, who detected prompt internal conversion electrons from Gd.
Using a completely different method, Wexler and Davies\textsuperscript{8} reported prompt internal conversion in Cd, Gd, and Sm. The generality of the phenomenon was further extended by Hibdon and Muehlhouse,\textsuperscript{9} who were able to detect prompt conversion electrons in Cd, Sm, Gd, Dy, and Hg. They also measured the energy of the electrons from these samples by use of a 180° focusing permanent-magnet beta-ray spectrograph. Similar studies have been reported by Church and Goldhaber,\textsuperscript{10} Groshev et al.,\textsuperscript{11} Motz,\textsuperscript{12} and Bieber.\textsuperscript{13}
CHAPTER II. DETECTION SYSTEMS

Since a silicon solid-state detector was basic to all the detection systems used in this work, the theory of operation of these counters is summarized here. A typical conversion-electron spectrum obtained with such a detector is shown in Fig. 1. The source used was Bi$^{207}$. The sharp peaks are due to the internal-conversion-electron groups from the 0.570- and 1.063-MeV transitions. The counts in regions A and B are mainly due to backscattered electrons which leave only a fraction of their energy in the detector. This "tailing" pulse distribution, which occurs with all monoenergetic lines when the detector is used in a conventional fashion, is undesirable since it obscures less intense lines of lower energy. Two different systems were tried in an effort to obviate this effect.

I. SOLID-STATE DETECTORS

Theory of Operation

A solid-state detector is a diode made by forming a junction of p-type and n-type semiconductors in a single crystal of high-purity material. When the junction is formed, electrons from the n-type semiconductor migrate, because of thermal agitation, to the
Fig. 1. A typical conversion-electron spectrum obtained with a lithium-drift solid-state detector. The source was Bi$^{207}$. The events in Regions A and B are primarily due to backscattered electrons.
p-type material and combine with holes. This process establishes a potential barrier which ultimately becomes sufficiently large to prevent any further migration of charge. The carrier exchange is a continuous process which finally reaches a temperature-dependent equilibrium condition. A narrow depletion region, relatively free of charge carriers, is thus formed at the junction.

When a reverse bias voltage is applied to the device, the width of the depletion region is increased since charge carriers are attracted away from the junction. If the small internal potential is neglected, the width $W$ of the depletion region is proportional to the square root of the product of the applied voltage $V$ and the resistivity $\rho$ of the semiconductor, i.e.,

$$W = k(V\rho)^{1/2}.$$  \hspace{1cm} (5)

The value of the constant $k$ depends upon the semiconductor material that is used. It is desirable to have the depletion region as large as possible since it is here that detection is achieved.

When radiation enters the depletion region, many free electron-hole pairs are produced by ionization. The amount of ionization depends upon the energy of the incident radiation and the nature of semiconductor used for the detector. In silicon, one electron-hole pair is liberated for approximately every 3.5 eV of energy lost by the radiation. The free electrons are collected at the positive terminal of
the detector and the holes at the negative electrode; the resulting current pulse can be measured electronically. The size of the output pulse is proportional to the energy lost in the depletion region. Therefore, this device provides a convenient means of measuring the energy of ionizing radiation.

The energy resolution of such a device depends upon several factors of which thermal noise is the most significant. Electron-hole pairs are continuously being freed by thermal agitation and are collected at the electrodes. This produces a reverse current that is statistical in nature. Thus a pulse due to incoming radiation is superimposed upon this noise and the statistical fluctuations are reflected by variations in the amplitude of the pulses. This effect can often be reduced by cooling the detector. However, the reverse current in good-quality detectors is inherently quite small.

The resolution also depends upon the efficiency with which the charge carriers are collected. Recombination sites, where electrons and holes can reunite, center around impurities and faults within the crystal. These result in incomplete charge collection and consequent attenuation of the output pulse. The effect can be minimized by the selection of extremely pure semiconductor material. Care must also be exercised in forming the n-p junction and providing for the electrical connections; otherwise impurities will be diffused into the semiconductor.
The existence of a dead layer (i.e., a nonsensitive region of the detector) also affects the resolution. If the incoming radiation has to penetrate the detector any distance before reaching the depletion region, its energy will be degraded by an amount dependent upon the number of collisions incurred. Thus, not all of the energy is deposited in the sensitive region and the output pulse corresponds to an energy less than the energy of the absorbed radiation. This results in a broadened line whose centroid is shifted downward an amount corresponding to the average energy loss in the dead layer.

The capabilities of a solid-state counter can be realized only if the auxiliary electronic equipment is compatible with the characteristics of the detector. This is particularly true with regard to the energy resolution. Since the signal from a semiconductor detector consists of a pulse of electrical charge, low-noise charge-sensitive amplifiers are usually employed. A more detailed discussion of solid-state radiation detectors and the associated electronics can be found in Ref. 14.

Surface-Barrier and Lithium-Drift Detectors

There are two basic classes of solid-state detectors—the surface-barrier and the ion-compensated type. Although they are similar in operation, they differ in method of production and in their suitability for particular applications. The junction of a surface-barrier counter is formed by etching the surface of a wafer of n-type semiconductor and
then exposing it to the atmosphere. Presumably, the exposed surface atoms chemically combine with oxygen which produces new quantum states for the electrons. These new states act as acceptor sites analogous to the holes in p-type semiconductors. Thus, an n-p junction of virtually zero thickness is formed at the surface. A very thin layer (≈ 150 Å) of gold is evaporated onto the surface to provide electrical contact. This thin film is the only dead layer through which particles must pass; its extreme thinness is the most advantageous property of surface-barrier detectors. However, the fact that the depletion region in surface-barrier detectors rarely exceeds 0.1 cm limits their usefulness for counting high-energy electrons.

The technique of ion compensation was originally introduced by Pell and was soon incorporated into the manufacture of radiation detectors. The lithium-drift radiation counter was first described by Mayer et al. The diode is formed by placing lithium metal on the surface of p-type semiconductor material and elevating the temperature to 260° to 600° C. At this temperature the lithium atoms thermally diffuse into the crystal. They act as donor impurities and produce a thin layer (≈ 200 microns) of n-type material which establishes the n-p junction. Unlike other donors, the lithium atoms do not displace the silicon from the lattice but take up interstitial sites within the crystal.

Gold is then evaporated onto the surfaces to provide electrical connections as in the surface-barrier detectors. A reverse bias voltage is
then applied while the temperature of the device is maintained at about 200°C. The electric field causes the Li\(^+\) ions to drift across the depletion region where they combine with acceptor atoms. The resulting compensation of the holes creates a region of very high resistivity. Depletion widths of up to 5 mm are not uncommon with detectors manufactured in this way. In contrast to the situation in surface-barrier detectors, the depth of the sensitive region of the lithium-drift detector is not a function of the bias voltage since the entire compensated region is readily depleted when the bias is applied.

This relatively large depletion region is the main advantage of lithium-drift detectors. However, it is achieved at the sacrifice of the extremely thin windows available with the surface-barrier detectors. When working with high-energy electrons, for which the thicker window is a less serious disadvantage than the narrow depletion width, lithium-drift detectors are often used.

Silicon solid-state detectors are quite insensitive to gamma rays because of the small photo cross section. Nevertheless, a pulse distribution that results from electrons due to Compton scattered gamma rays can usually be detected. Recently, germanium solid-state detectors have been developed for work with gamma rays. The probability of photoreactions in germanium is higher than in silicon because the atomic number is higher.
II. THE "THIN-THICK" SYSTEM

Description of Apparatus

The "thin-thick" detector arrangement is shown schematically in Figs. 2 and 3. This system was designed to suppress the tailing pulse distribution which results from backscattered electrons. A thin transmission-type surface-barrier detector was placed directly in front of a lithium-drift counter so that incident electrons had to pass through the thin detector before they could be detected in the thick counter. Since backscattered electrons from the thick detector must almost always re-enter the thin detector because of its proximity, this in principle offers a way of recognizing backscattered electrons. An incoming electron passes through the thin detector only once and deposits only a certain small amount of energy in it; whereas, a backscattered electron leaves additional energy when it re-enters the thin detector. A pulse-height analysis of the output pulses from the surface-barrier detector can therefore distinguish the events in which backscattering occurs.

Time marker circuits were used to shape the detector pulses from the preamplifiers. The window of a single-channel analyzer was adjusted so that it accepted only those pulses that corresponded to electrons that had made a single pass through the detector. The signal from the single-channel analyzer and the pulses from the time marker circuits were routed to a fast-slow coincidence system which controlled a linear gate on a multichannel
Fig. 2. "Thin-thick" detector arrangement. The lucite holder keeps the thin detector near the thick counter. The electrical leads are not shown.
Fig. 3. Block diagram of the electronics of the "thin-thick" detector system.
pulse-height analyzer. The gate was opened only when coincident events were detected. No events were counted unless the pulses from the lithium-drift detector were coincident with pulses from the surface-barrier detector which, in turn, had satisfied the conditions of the single-channel analyzer; i.e., it was intended that the multichannel analyzer count only those events that corresponded to a single pass through the thin detector.

A spectrum obtained from the thick detector showed peaks that corresponded to the energy of the incident electrons minus the energy that was lost in the thin detector. Since this energy loss was not constant, a linear summing circuit was used to add the coincident pulses. The use of this summed pulse for the pulse-height analysis automatically compensated for the energy lost in the thin detector.

The thin detector used in this work was a surface-barrier type with a sensitive width of about 92 µ. The front dead layer was less than 30 µg/cm² of gold—the back dead layer less than 40 µg/cm² of aluminum. The energy resolution (full width at half maximum) was 21 keV for the 5.5-MeV alpha particles from Am²⁴¹. The depletion region extended through the back of the detector when a reverse bias of 90 V was applied.

The lithium-drift silicon counter used for the thick detector has a sensitive width of about 2 mm. This device was operated with a back bias of 150 to 200 V. The surface density of the front dead layer was about 7 mg/cm².
A monoenergetic source of electrons was obtained by using a 180°-focusing magnetic β-ray spectrometer. A P$^{32}$ source was used and the spectrometer could be set to give a beam of electrons of any desired energy up to 1.71 MeV.

Results and Conclusions

It was expected that the spectrum from the thin detector would have two broad but resolved peaks—one from those electrons that passed through the thin detector and were then totally absorbed in the thick counter, the other from those that were backscattered. The spectrum obtained from a beam of 800-keV electrons is shown in Fig. 4. The large number of counts in the first few channels is due to detector noise. The spectrum shows little evidence of the expected structure. Apparently many incident electrons are severely degraded or stopped entirely with the result that pulses corresponding to the entire range up to the incident electron energy are produced.

Although this spectrum falls far short of meeting the idealized conditions, the system might still be feasible if the window of the single-channel analyzer were set to reject most of the pulses due to the backscattered electrons. To accomplish this the window would have to include only the very low-energy portion of the spectrum in order to assure the rejection of backscattered events. This would result in a decided reduction in the counting efficiency since many acceptable events would also be discarded.
Fig. 4. The pulse-height distribution due to 800-keV electrons incident upon the thin detector with the thick detector in place.
To determine the best setting of the discriminator, a two-parameter analysis was made by use of a 4096-channel analyzer. The output is in the form of a $64 \times 64$ matrix with the singles spectra from the thick and thin detectors stored in the first row and first column, respectively. Each remaining row of the matrix contains a 63-channel analysis of the thick-detector spectrum taken in coincidence with the thin-detector pulses that fall in the first cell of the row. This method simultaneously accumulates the equivalent of 64 different settings of the single-channel analyzer customarily used to gate the multichannel unit.

The spectrum from the thick detector used in a conventional manner has a peak-to-continuum ratio of about 12.5. These experiments showed that the "thin-thick" system could suppress the continuum by a factor of 3. However, this improvement was achieved at the expense of the counting efficiency which was reduced by a factor of more than 15. When the peak-to-continuum ratio was doubled, the efficiency dropped by a factor of 10 from the original value.

The serious loss of efficiency associated with the relatively small gain in the peak-to-continuum ratio makes this system only moderately successful at best. The situation is no better with a different monoenergetic source and is even worse when several monoenergetic electron groups are present.

The limited success of this system can be attributed to the fact that an idealized model was used as a guide in its design. However,
the deviations from the ideal often proved to be far greater than could be anticipated. The finite thickness and resolution of the thin detector were most significant.

If the detector is too thin, the average energy lost by incident electrons will not differ greatly from the total energy left by backscattered electrons. However, if the detector is too thick, degradation of the incident electrons becomes severe. After leaving a large fraction of its energy in the detector, a backscattered electron will have little energy left to contribute when it re-enters the detector. Thus the energy lost by the incident electron will again be nearly equal to the total energy lost by the backscattered electron. The ideal thickness obviously depends on the energy resolution of the detector. It should be made as thin as possible without losing the ability to distinguish between a single passage and a double passage of an energetic electron.

The thickness of the surface-barrier detector is important for another reason. Since the amount of energy an electron loses as it penetrates a thin slice of matter is statistical in nature, only the average energy lost can be specified. Therefore, the single-channel analyzer has to be adjusted so that a broad range of energies near the average value is accepted. It is possible that the amount of energy actually lost by a backscattered electron which makes a double passage will fall within the acceptance range and the event will not be rejected.

Furthermore, the average energy lost by a monoenergetic beam of electrons depends upon the electron energy; a slower electron
will lose considerably more energy than will a faster one. This prevents the window of the single-channel analyzer from being properly set for all electron energies. If the upper level of the window is set so that only those pulses resulting from an energetic electron making a single pass through the thin detector are accepted, many slower electrons will not be counted because of their larger energy loss. On the other hand, if the window is adjusted so that the larger pulses from the slower electrons are accepted, several pulses from energetic backscattered electrons will also be counted. A suitable compromise between the counting efficiency for slow electrons and the effectiveness for discarding events due to backscattering could not be achieved with the detectors used in these experiments.

III. THE PILOT B PLASTIC SYSTEM

Description of Apparatus

A second arrangement intended to improve the peak-to-continuum ratios for spectra obtained with solid-state detectors used a Pilot B scintillating plastic to detect the backscattered electrons. The system is shown in Fig. 5. A cylinder 1 1/2 in. in diameter and 1 in thick was made from the Pilot B plastic. In order to allow for the insertion of a solid-state detector, a hole 3/4 in. in diameter and 1 in. deep was drilled perpendicular to the axis and centered with respect to the ends of the cylinder. Concentric to this hole was a smaller one through which
Fig. 5. The Pilot-B plastic system.
electrons could reach the silicon detector. The same lithium-drift detector that was used in the "thin-thick" experiment was inserted into the larger hole and held in place by a small set screw. The plastic scintillator was then coupled to a 9536-B E. M. I. photomultiplier tube, and a light tight encasement was provided. The entrance hole was covered with a double thickness of aluminum-coated Mylar to provide an opaque window that was less than 2 mg/cm\(^2\) thick.

Since backscattered electrons leave the detector surface at all angles, many enter the Pilot B plastic where they produce scintillations that cause pulses in the photomultiplier tube. The output from the photomultiplier tube and the signals from the solid-state detector were routed to an anticoincidence circuit which controlled a linear gate on the multichannel analyzer. The signals from the solid-state detector were pulse-height analyzed except when the anticoincidence circuit recognized coincident events and closed the analyzer gate.

The effectiveness of this system depends upon what fraction of the backscattered electrons are detected by the Pilot B scintillator. If an electron is backscattered in a direction near the normal to the plane of the solid-state detector surface, it can leave the plastic through the entrance hole and will not be detected. Thus, these backscattered events will not be rejected.

The number of backscattered electrons that escape can be reduced by making the diameter of the entrance hole smaller and/or by
increasing its length. Either modification will decrease the solid angle through which they can escape; but the diameter of the entrance hole should be no smaller than the diameter of the sensitive region of the solid-state detector. If the hole is smaller than this, the Pilot B scintillator masks a portion of the counter and the efficiency is correspondingly impaired.

The length of the entrance hole is also limited. As this becomes longer, the source-to-detector length necessarily increases and consequently the solid angle for incoming electrons is decreased. If the strength of the source is limited, then the counting rate determines the largest acceptable length.

When very active sources are used, the dead time of the anticoincidence circuit becomes significant. This difficulty was overcome in part by shielding the plastic with a 1/4-in. lead mask. A 5/16-in. hole through the lead permits electrons to reach the window but shields the remainder of the plastic, thus reducing the superfluous pulses in the plastic.

Results and Conclusions

This system was, by far, the better of the two tried. The source used for these experiments was Bi$^{207}$. The continuum was definitely suppressed but not to the same degree throughout the spectrum, as seen in Fig. 6. The effectiveness of the system appears to be a function of the
Fig. 6. Spectra of Bi$^{207}$, taken with and without the anticoincidence circuit gating the analyzer. These spectra demonstrate the effectiveness of the Pilot-B plastic system.
energy of the backscattered electrons. The events in which most of the energy was deposited in the solid-state detector (so that the backscattered electron was left with little energy) were rejected less often than those events in which the backscattered electron was relatively energetic. This effect is minimized by using a low-noise high-gain photomultiplier tube.

The improvement in the peak-to-continuum ratio with this system over a detector used in a conventional fashion ranges from ≈2 to 5 depending on which portion of the spectrum is considered. The efficiency of the system was not noticeably less than that of the counter used in a conventional fashion. The fact that the solid angle for incoming electrons was reduced in order to achieve effective rejection of events due to backscattering seems to be the only serious disadvantage.
CHAPTER III. NEUTRON BEAM EXPERIMENTS

Several experiments were done at the CP-5 and Juggernaut reactors at Argonne National Laboratory for the purpose of developing a collimating system that would produce a neutron beam suitable for a neutron-capture internal-conversion apparatus. Preliminary experiments were done with neutron beams that were thought to have the necessary properties for the intended application. The results obtained from these preliminary experiments were used as a means of evaluating the results of further investigations at the west thermal column of the Juggernaut reactor.

Since these experiments began before those with the solid-state detectors were concluded, the detectors were used in a conventional fashion without taking special measures to discard the backscattered events. All the counters used were lithium-drift silicon devices.

PRELIMINARY EXPERIMENTS

The first set of preliminary experiments was conducted at the 6H3 beam hole of the Juggernaut reactor. A schematic diagram of the collimation system for this hole is shown in Fig. 7. A target of Sm$^{149}$ ($\sigma = 4.08 \times 10^4$) with a mean surface density of approximately 30 mg/cm$^2$ was prepared by dusting Sm$_2$O$_3$ powder on the adhesive side.
Fig. 7. Schematic diagram of the Juggernaut reactor 6H3 neutron-beam facility.
of cellophane tape. This source was then placed in the line of the neutron beam with a thermal flux of $5 \times 10^5$ neutrons cm$^{-2}$ sec$^{-1}$. The detector was situated $1\frac{1}{2}$ in. from the source and on a line perpendicular to the direction of the beam.

The resulting spectrum is shown in Fig. 8. The peaks are due to 340- and 440-keV transitions$^{9, 11, 13}$ in Sm$^{150}$. The detector was effectively shielded from the pile gamma rays and the room background by 4 in. of lead. The large number of counts composing the background continuum is primarily from the capture gamma rays emitted by the source. To verify this, further experiments were done at the 6H15 beam hole at the CP-5 reactor.

A neutron beam with a thermal flux of about $10^5$ neutrons cm$^{-2}$ sec$^{-1}$, relatively free of gamma rays, is available from this facility. The neutrons are reflected from a large magnetized cobalt mirror which separates them from the gamma ray beam. A large triple-walled tank houses the detector. The outer wall is 6 in. of paraffin; the middle is 1 in. of boron carbide; and the inner is 9 in. of lead.$^{18}$ The tank provides efficient shielding against both gamma rays and neutrons.

A 2 X 12-in. rectangular tube made of Boral (borated aluminum) provided a path through the shielding for the neutrons. The target was placed in the center of this tube near a port which allowed the electrons to reach the detector. A spectrum obtained with this apparatus is shown in Fig. 9. This spectrum displays characteristics very similar
Fig. 8. Prompt internal-conversion-electron spectrum of Sm$^{150}$ taken at the Juggernaut reactor. The peaks are due to the 340- and 440-keV transitions.
Fig. 9. Prompt internal-conversion-electron spectrum of Sm$^{150}$, taken at the CP-5 reactor.
to those of the spectrum in Fig. 8 obtained at 6H3 of the Juggernaut reactor. Again the major contribution to the spectral background is due to neutron-capture gamma rays emitted by the target.

The gamma-ray sensitivity of the detectors was experimentally determined by use of the radiations from Bi$^{207}$. The spectra accumulated with and without a beta shield over the detector are shown in Figs. 10 and 11, respectively. The total number of counts in the spectrum taken with the beta shield was assumed to be the total number of electromagnetic quanta detected. When this spectrum was stripped from the one taken without the beta shield, the remaining spectrum was assumed to be that of the internal-conversion electrons alone. When the conversion coefficient was considered, a calculation showed that the average sensitivity for the electromagnetic radiations from Bi$^{207}$ was 5%. The gamma sensitivity is a function of the photon energy—the higher the energy the lower the sensitivity. The spectrum in Fig. 10 clearly shows the photopeak from the bismuth x rays, but only the Compton distributions of the more energetic gamma rays can be seen.

Although the gamma sensitivity is far too low to make these detectors useful for gamma-ray spectroscopy, it is large enough to obscure the details of prompt internal-conversion-electron spectra. The reason is that whenever the conversion coefficient for a transition is less than 0.05, more gamma rays than electrons will be detected, and the resulting Compton distributions obscure low-intensity conversion lines.
Fig. 10. \( \text{Bi}^{207} \) spectrum taken with a lithium-drift solid-state detector which was shielded by \( \frac{1}{4} \) in. of aluminum. Only the Compton distributions are visible for the \( \gamma \) rays. The peak at the low-energy end is due to the bismuth x-ray.
Fig. 11. Bi$^{207}$ spectrum taken without any β shield.
A proposed method of increasing the solid angle for counting electrons and leaving the gamma rays unaffected will be discussed in the next chapter.

Subsequent experiments at the CP-5 reactor led to a number of valuable conclusions concerning the origin of the spectral background that was present when no target was in the beam. A sheet of 0.040-in. cadmium was placed over the entrance end of the Boral tube. This removed nearly all the thermal neutrons from the beam because of the high capture cross section of Cd$^{113}$ ($\sigma = 2.4 \times 10^4$ b). A spectrum taken with this configuration showed that the background decreased to 1% of the value observed when cadmium was not used. Thus, with no target, about 99% of the background is due to thermal neutrons whose capture in the Boral presumably gives rise to the unwanted background gamma rays.

The remaining 1% of the background is a result of the fast neutron flux in the beam. To verify this, cadmium was again used to block the thermal neutrons and a paraffin plug 8 in. long was placed inside the Boral tube. The background counting rate then was ten times that obtained when no paraffin was used. The paraffin moderates the neutrons to near thermal energies and thereby increases the neutron-capture processes in the surroundings.

Conclusions

Three important conclusions were formulated from the results of the preliminary experiments. (1) A method must be devised
for reducing the spectral background due to the high-energy capture gamma rays emitted from the source. (2) The fast-neutron flux in a beam suitable for the intended application should be small. (3) The thermal-neutron flux must be $10^6$ neutrons cm$^{-2}$ sec$^{-1}$ or larger.

One method that was used to eliminate the spectral background due to gamma rays was first to accumulate a spectrum and then to subtract the gamma background after inserting a thin beta absorber between the detector and the source. The beta absorber did not affect the gamma rays, but did prevent the counts due to electrons from being subtracted. The spectrum that resulted was the conversion-electron spectrum without the gamma-ray background.

The major disadvantage of this procedure is that the statistical uncertainties were increased by a factor of $\sqrt{2}$. Since both accumulation and subtraction must be done, the time needed to obtain the spectrum can also be a disadvantage.

The problems of reducing the fast-neutron flux and obtaining a maximum thermal flux are interrelated. To thermalize fast neutrons, a material such as graphite is placed in the beam. The amount of graphite used depends upon what degree of thermalization is needed to reduce the fast flux to a tolerable level. Initially, the addition of graphite causes an increase in the slow-neutron flux; but eventually the fast flux becomes so small that the net amount of thermalization is less than the attenuation of the slow-neutron flux because of the scattering and capture
processes in the graphite. Beyond a certain thickness, the addition of more graphite decreases the slow-neutron flux. Although this condition usually occurs before the fast flux has been sufficiently reduced, further additions of thermalizing material should be kept to a minimum.

INVESTIGATIONS AT THE WEST THERMAL COLUMN

The results and conclusions from the preliminary experiments were used as guides in further investigations directed toward the design of a collimation system for use at the west thermal column of the Juggernaut reactor. A drawing of the west thermal column is shown in Fig. 12. The inner plug was removed and a collimator was put into its place as shown in Fig. 13. This collimator was made by constructing a stainless steel shell of the desired shape and filling it with boron carbide. Because of its high capture cross section ($\sigma = 4.02 \times 10^3$ b), boron is a good thermal-neutron absorber. The collimator hole was tapered such that an extension of its walls to the face of the graphite around the reactor core would just outline the outer end of the 1-ft-square removable stringer section. An outward projection of the walls would outline a 1 x 3-cm rectangular target area at a distance of 6$\frac{1}{2}$ in. from the reactor face. This design takes advantage of the fact that the entire 1-ft-square graphite stringer section acts as a source of thermal neutrons.

To determine the effectiveness of the collimator, the flux was measured at various points along a line perpendicular to the axis of
Fig. 12. A schematic drawing of the west thermal column at the Juggernaut reactor.
Fig. 13. The collimator shown replacing the inner removable plug.
the beam at a distance of $6\frac{1}{2}$ in. from the face of the reactor. The results of these measurements are graphically shown in Fig. 14. The maximum thermal-neutron flux at the center of the target area was $2.7 \times 10^6$ neutrons cm$^{-2}$ sec$^{-1}$.

In order to reduce the large gamma-ray flux present in this beam, 4 in. of lead were placed in the void between the collimator and the gate. This lowered the gamma radiation to a tolerable level but also caused a serious attenuation of the thermal-neutron flux. To offset the drop in flux, a 2-ft length of graphite was removed from the center stringer sections. It was felt that the 2 ft of graphite caused unwarranted attenuation and that the remaining 4 ft would be sufficient to achieve the thermalization necessary to keep the fast flux at a reasonable level.

Removing the graphite did increase the thermal flux but not as much as had been anticipated. The gamma-ray flux was also increased; it was surmised that many of the gamma rays were due to thermal-neutron capture in the surroundings. This hypothesis was strengthened by the fact that 2 in. of lead arranged around the mouth of the collimator decreased the gamma background. To further decrease the gamma flux, more lead was added to that in the void, making a total of 10 in. between the graphite and collimator as shown in Fig. 15. These measures reduced the gamma radiation to an acceptable level.

Fig. 16 shows the configuration used in another attempt to increase the thermal flux. All but the last 12 in. of the center 4 X 4-in.
Fig. 14. A graphical analysis of the effectiveness of the collimator. It shows that there was a definite drop in flux as the perpendicular distance from the axis of the neutron beam increased.
Fig. 15. West thermal column with 2 ft of graphite removed and 10 in. of lead placed between graphite and collimator.
Fig. 16. West thermal column with all but the last 12 in. of the center stringer section removed. The 10-in. lead filter remains.
graphite stringer were removed. This configuration resulted in a satisfactory thermal flux ($\approx 10^7$ neutrons cm$^{-2}$ sec$^{-1}$), but the fast-neutron flux was so great that it created a personnel hazard. Therefore, the system was not acceptable for a permanent installation.

To determine whether the major contribution to the thermal flux came from the 4 x 4-in. end surface of the remaining 1 ft of center stringer, a cadmium tube and shield were placed in the beam. The tube, 4 in. square and 36 in. long, was inserted into the region vacated by the removed stringer. The shield measured 12 x 12 in. with a 4 x 4-in. square hole in the center and was situated adjacent to the stringer section so that its hole was aligned with the tube. The cadmium effectively absorbed all thermal neutrons except those originating from the 4 x 4-in. end surface of the center stringer.

This configuration caused a drop in thermal flux from $10^7$ to $1.2 \times 10^6$ neutrons cm$^{-2}$ sec$^{-1}$. Although this represents a drop by a factor of eight, it was still acceptable since the thermal flux was still greater than $10^6$ neutron cm$^{-2}$ sec$^{-1}$. In addition, the improved collimation was advantageous.

In the final design, a total of 24 in. of graphite is to be used in the center stringer section to reduce the number of fast neutrons in the beam. The collimator has been redesigned by making it of borated polyethylene instead of boron carbide. The advantage of this material is that the polyethylene acts as a good thermalizing agent and the boron
absorbs slow neutrons. Bismuth shielding will surround the mouth of the
collimator to absorb the capture gamma rays produced in the surroundings.

Experiments in which bismuth and lead were used in the
beam to reduce the gamma flux showed that less attenuation of the
thermal-neutron flux results with bismuth. The ratio of slow neutrons to
gamma rays with bismuth is four times as great as with the same amount
of lead. The bismuth had the added advantage that it helped to reduce the
fast-neutron flux because of its higher neutron-scattering cross section.
The disadvantage of bismuth is that it must be clad with another material
such as aluminum since polonium is formed when bismuth is irradiated
with neutrons. The polonium which flakes off the surface becomes a source
of radioactive contamination.
CHAPTER IV. SUMMARY OF CONCLUSIONS AND PROPOSED EXTENTION OF THE STUDY

The purpose of this research was to design an apparatus to be used for the investigation of internal-conversion-electron spectra associated with neutron capture process. The study mainly involved problems related to the design of neutron collimators and electron detection systems.

The experiments with solid-state counters showed that the pulse distribution due to backscattered electrons from the lithium-drift detectors can be reduced by as much as a factor of five by means of a system using a scintillating plastic. The plastic surrounds the lithium-drift device except for a small entrance hole and is used to detect the backscattered electrons so that the coincident events in the main detector can be discarded. It is probable that specially designed entrance holes through the plastic will result in an even greater improvement.

A second system which used a thin surface-barrier detector directly in front of a thick lithium-drift device was also tried as a method of suppressing the unwanted tailing pulse distribution. The operation of this system was marginal and the high loss in counting efficiency made the system impractical.

However, neither of the above systems was used since the Sm$^{150}$ spectra obtained at the nuclear reactors showed that the spectral
background arising from neutron-capture gamma rays emitted from the source was a more serious problem than that caused by backscattered electrons. A method for greatly enhancing the electron-counting efficiency has been proposed.

The proposed apparatus employs a superconducting solenoidal magnet, 10 in. long by 3 3/4 in. in diameter, which is capable of producing a magnetic field of 25 kG. A special Dewar has been designed which allows for a 1 1/2 in. diameter, room-temperature access through the 2 in. core of the magnet. A solid-state detector is located within the core region near one end of the magnet. The other end of the magnet is brought as close as possible to the target without permitting the neutron beam to irradiate the Dewar.

The strong magnetic field causes all the electrons which enter the core region to describe very tight helical trajectories about the magnetic field lines. These electrons are therefore guided to the detector. The solid angle for the electrons is roughly the solid angle of the 1 1/2-in. hole which is about 1 in. from the source. However, the magnetic field does not influence the gamma rays, and the solid angle for gamma rays is that of the detector 10 in. away. The ratio of the collection efficiency for electrons to that for gamma rays is thereby made 500—1000 times as great.

At the time of this writing, the magnet has been constructed and tested. A magnetic field of 28 kG was established at a current of
26.5 A. The quenching current was about 27.5 A. Additional tests on this magnet are still in progress.

Various neutron collimating configurations were tried in order to obtain a thermal-neutron beam relatively free of fast neutrons and gamma rays. Fig. 17 shows the present design which consists of 24 in. of graphite to thermalize the fast flux, 12 in. of bismuth to remove the gamma rays, and a borated polyethylene collimator. This apparatus is designed for the 6H1 beam hole of the Juggernaut reactor.

The Dewar containing the superconducting magnet is mounted on a vacuum chamber which houses the target as shown in Fig. 18. The neutron beam passes through the vacuum chamber and is stopped by a beam catcher. The final assembly of the apparatus should be completed as soon as the vacuum chamber and Dewar are constructed.
Fig. 17. Proposed configuration for the 6H1 neutron beam hole at the Juggernaut reactor.
Fig. 18. Schematic diagram of proposed Dewar and vacuum chamber.


