Report: DOE/ER/40246-7

NEUTRONS SCATTERING STUDIES IN THE ACTINIDE REGION

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Progress Report

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ABSTRACT

During the report period we have investigated the following areas:

(a) Prompt fission neutron energy spectra measurements.

(b) Neutron elastic and inelastic scattering from ^{239}Pu.

(c) Neutron scattering in ^{181}Ta and ^{197}Au.

(d) Response of a ^{235}U fission chamber near reaction thresholds.

(e) Two parameter data acquisition system.

(f) "Black" neutron detector.

(g) Investigation of neutron-induced defects in silicon dioxide.

(h) Multiple scattering corrections.

Four Ph.D. and one M.S. theses were completed during the report period. Publications consisted of three journal articles, four conference proceedings and eleven abstracts of presentations at scientific meetings. There are currently four Ph.D. and one M.S. candidates working on dissertations directly associated with the project. In addition three other Ph.D. candidates are working on dissertations involving other aspects of neutron physics in our laboratory.

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1. Prompt Fission Neutron Measurements

An accurate knowledge of the shape of the fission neutron spectrum is required to test the model of Madland and Nix[1]. To ensure that the relative shape of the spectrum is reported with sufficient accuracy, it is necessary that the time-of-flight (TOF) spectrometer be calibrated with great accuracy. The detector efficiency is being addressed by the use of the program "SCINFUL" from the Radiation and Shielding Information Center[2] (RSIC) and by the use of a ^{235}U fission chamber with a well known efficiency[3]. The energy calibration is being established by the use of the following reactions for the production of neutrons of known energies: $^{7}Li(p,n)^{7}Be$, $^{2}H(d,n)^{3}He$, $^{9}Be(d,n)^{10}B$ and $^{9}Be(^{3}He,n)^{11}C$. These reactions will enable us to calibrate the TOF spectrometer up to approximately 13 MeV.

The data analysis routines have been written and preliminary analysis of the spectra has been completed. Fig. 1 shows a sample fission neutron spectrum for ^{235}U at an incident neutron energy of 0.50 MeV. This energy spectrum has been corrected for time and energy resolution limits and detector efficiency. In addition bin width corrections have been made for the high energy region. Further results will be reported at the Fall Meeting of the Nuclear Physics Division of the American Physical Society at Santa Fe, NM. This work is part of the Ph.D. dissertation research of Parrish Staples.

2. Elastic and Inelastic Scattering From ^{239}Pu

In last year's progress report, we summarized some of our preliminary results of neutron scattering from ^{239}Pu at the incident energy of 570 keV. In order to improve the energy resolution in the past year we readjusted the operating conditions and the detector-target distance (flight path) from 97 cm to 175 cm. Under the new conditions, the differential cross sections at 72.5°, 107.5° and 135° were remeasured. In addition we have obtained cross sections for four more angles at this energy. The angular distributions for two two-level combinations, ground plus 7.9-keV states and 57.3-keV plus 75.7-keV states have been obtained. These angular distributions are shown in Figs. 2 and 3. This work is part of the Ph.D. dissertation of G.Yue.

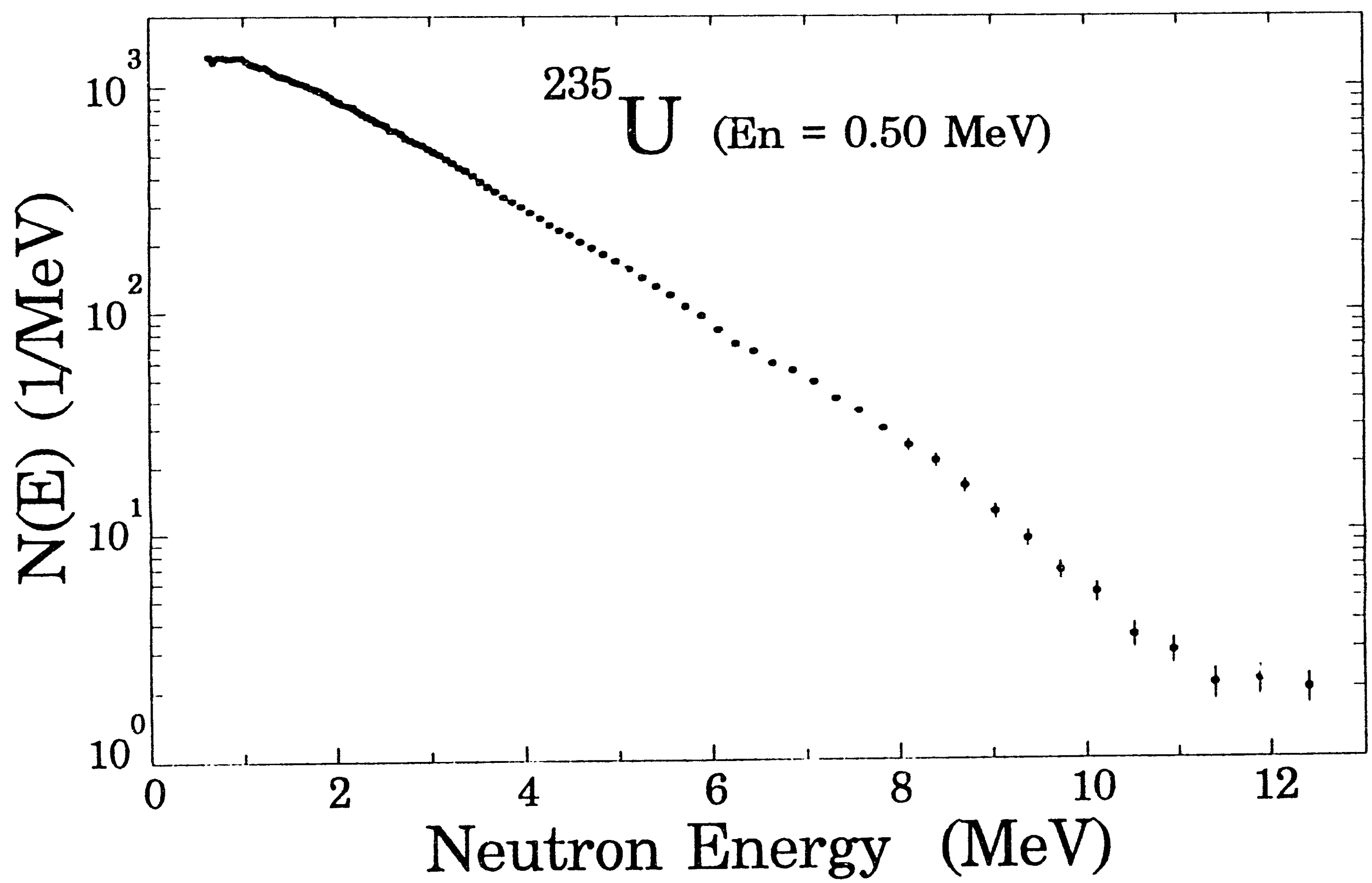

Fig. 1. ^{235}U fission neutron spectrum obtained with 0.5 MeV incident neutrons.

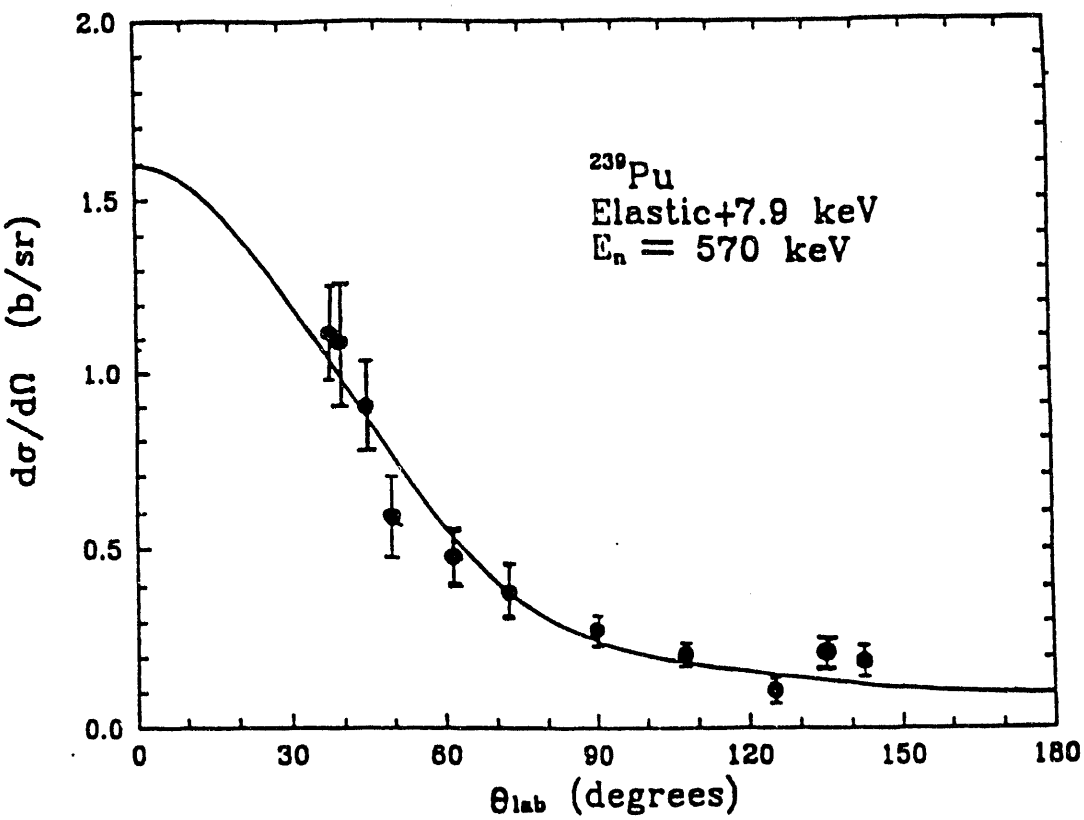

Fig. 2. Angular distribution for ground state and first excited (7.9 keV) states of ^{239}Pu for 570-keV incident neutrons. The solid line represents ENDF/B-VI.

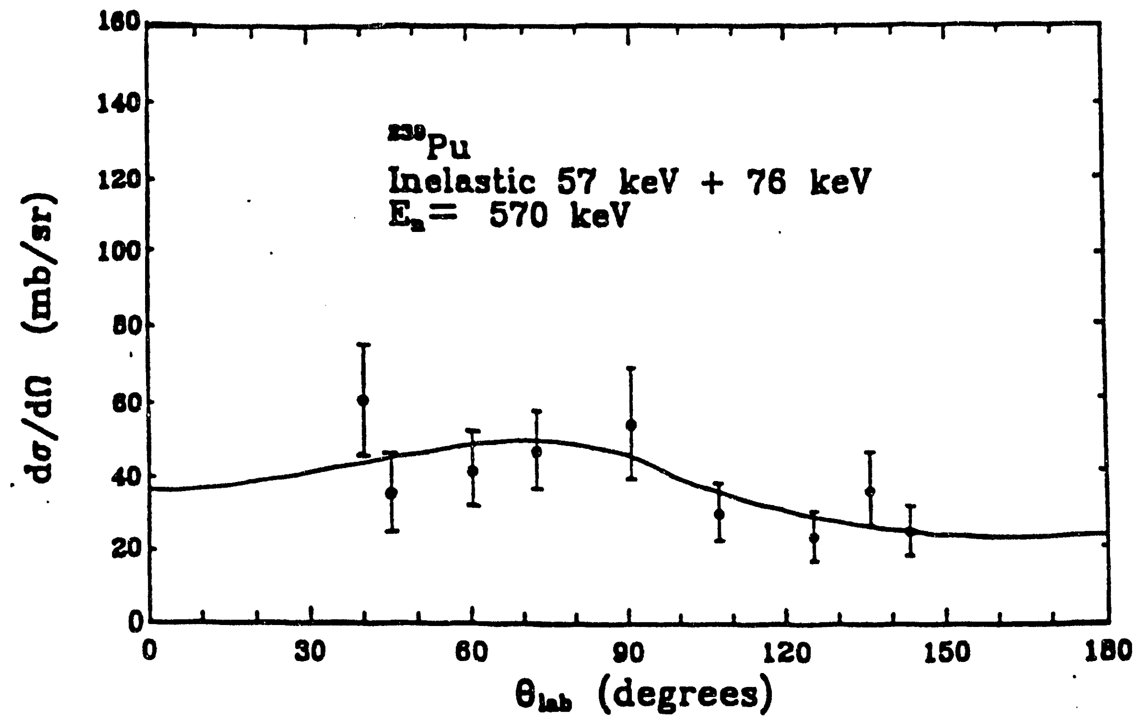

Fig 3. Angular distribution for second (57 keV) and third (76 keV) excited states of ^{239}Pu for 570-keV incident neutrons. The solid line represents ENDF/B-VI.

3. Neutron Scattering in ^{181}Ta and ^{197}Au

At the 1991 Fall Meeting of the Nuclear Physics Division of the American Physcial Society we presented preliminary results for 700-keV incident energy neutrons scattered from ^{181}Ta. The cross section for the ground-6.24 keV state doublet agreed favorably with ENDF/B-VI, but that of the 136.6-158.6 keV doublet exhibits more structure than ENDF/B-VI.

Continuing our work on odd-A nuclei, we did a comprehensive investigation as to what size ^{197}Au scatterer would produce the smallest neutron energy dispersion for purely geometrical effects. Within the constraint of maintaining a constant volume, we compared various sized disks and cylindrical scatterers for 1-MeV incident energy neutrons with the detector located at 125°. Using our computer code[4] IMBUI-G we determined that a disk sctatterer of radius 1.25 cm produces the minimum neutron energy dispersion (see Fig. 4).

We are also considering the merits of using a spectral peak fitting program which would be based on maximum entropy.[5] Currently we are using a least-squares procedure which works reasonably well, but we are always eager to examine a new approach. This work forms part of the Ph.D. dissertation of Michael O'Connor.

4. Response of a ^{235}U Fission Chamber Near Reaction Thresholds

A code has been written in FORTRAN (MS-FORTRAN v.4.0) to analyze efficiency measurements made with a ^{235}U fission chamber in the vicinity of the $^{7}Li(p,n)^{7}Be$ reaction threshold, where the neutron production at 0° becomes double-valued. The code compares the fission chamber yield to that of scintillation detector and computes the absolute efficiency of the latter assuming the efficiency of the former. The code takes into account the detailed kinematics (non-relativistic) of the reaction in three dimensions, the cross section of the reaction over the energy range of the incident protons, the fission cross section over the full range of neutron energies and angles of incidence on surfaces of the fission foils. A model of the expected fission chamber time-of-flight spectrum helps the experimenter to determine what weight to give to the contribution to the fission chamber response from the "second group", neutrons produced in the backward direction in the center-of-mass frame of reference. The

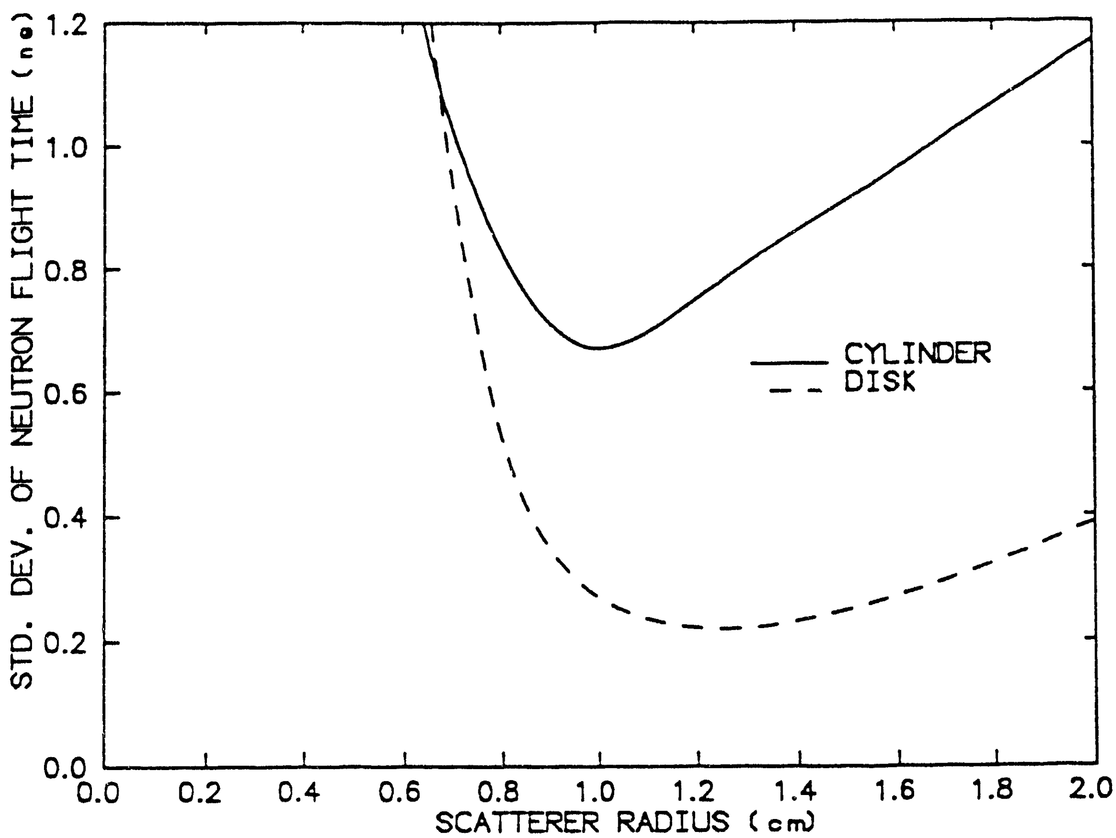

Fig. 4. The standard deviation of neutron flight time is compared for disk and cylindrical scatterers. Both scatterers have the same volume and the volume remained fixed as the radius was varied. The standard deviation of neutron flight time, which is plotted, arises purely due to geometrical effects. The plot illustrates that to obtain optimum time resolution disk scatterers should be used.

fission chamber and scintillator detector need not subtend the same solid angle, but must be normalized to the yield of a third detector.

The code was used to analyze the data from ^{232}Th, ^{238}U and ^{209}Bi scattering at 127 keV. This work is part of the Ph.D. dissertation of Christopher Horton.

5. Two Parameter Data Acquisition System

To facilitate the completion of the second phase of the fission neutron spectra measurements, the construction of a computer interface/buffer (2PIN) allowing two-parameter acquisition, data storage and analysis was undertaken. A diagram of the two-parameter interface in a data acquisition system is shown in Fig. 5. A diagram of the interface circuitry is shown in Fig. 6. Extensive software development to control 2PIN by the HP2100A computer and operate with the HP5416B analog-to-digital converter (ADC), and HP5410A multi-channel analyzer (MCA) was also completed. A simplified block diagram of the program execution is seen in Fig. 7.

The two-parameter interface is being tested in an experiment to measure lifetimes of nuclear states produced by Coulomb excitation. The experiment uses a barium fluoride (BaF_2) crystal coupled to a Hammamatsu R2083Q photomultiplier tube. Barium fluoride scintillators will be used in the second phase of the fission neutron spectra measurements for energies less than the incident energy to signal the occurence of a fission event by detecting the prompt fission gamma-rays.

The development and testing of the two parameter interface forms the basis of the M.S. thesis of Mitchell Woodring.

6. "Black" Neutron Detector

We have constructed a Black Neutron Detector of the Poenitz design.[6] It is a cylindrically shaped detector of length 15.75 inches and a diameter of 11 inches filled with 28 liters of BC501 liquid scintillator. This scintillator was chosen because of its pulse shape discrimination characteristics. Five photomultiplier tubes view the liquid scintillator. Figure 8 shows a picture of our black neutron detector. A detailed drawing is given in Fig. 9. Neutron sheilding consists of 144 five gallon tin cans filled with a mixture of water and boric acid. Polyethylene bags are used

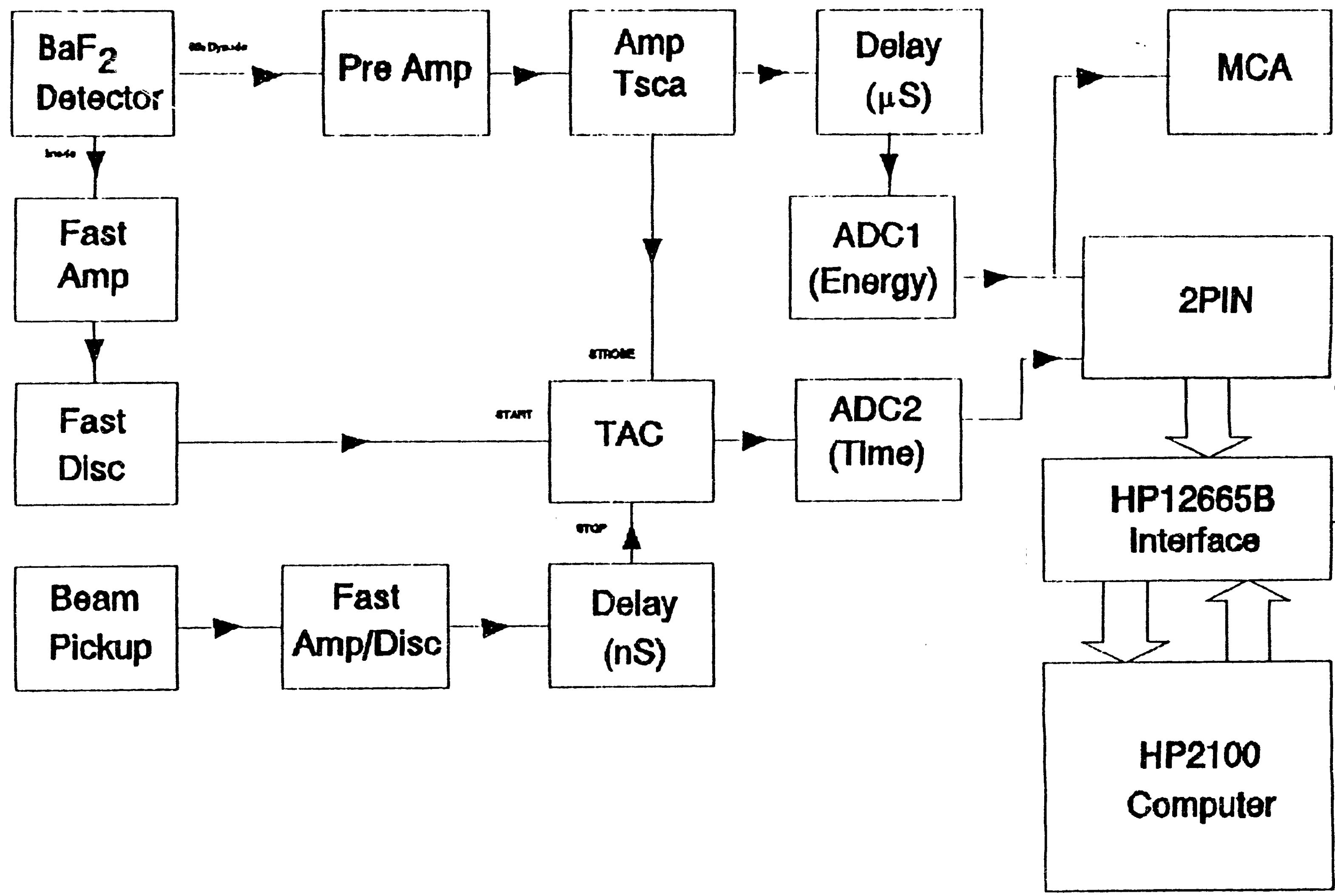

Fig. 5. Block diagram of of typical data acquisition electronics used in conjunction with the two-parameter interface.

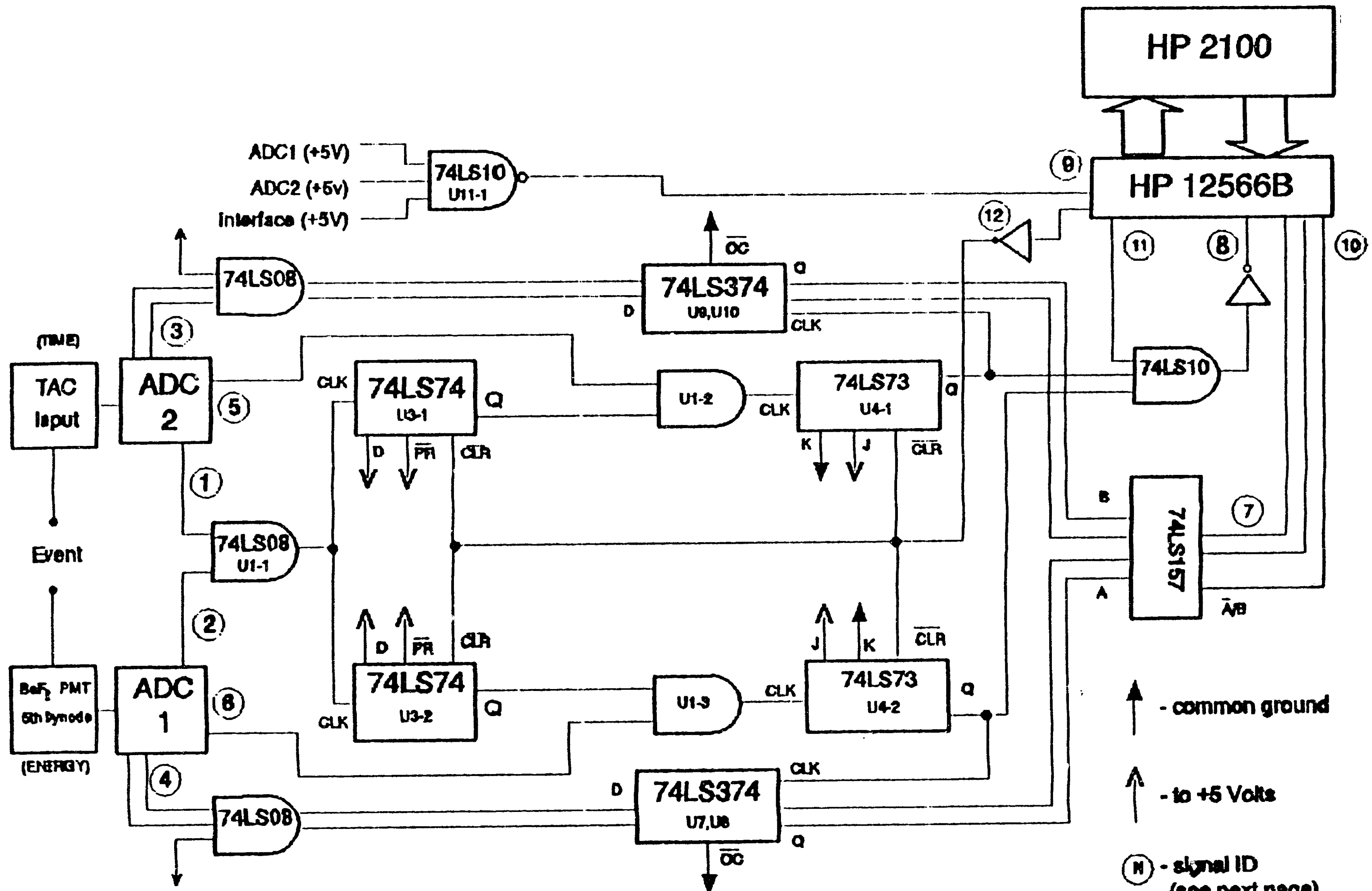

Fig. 6. Schematic of the two-parameter interface.

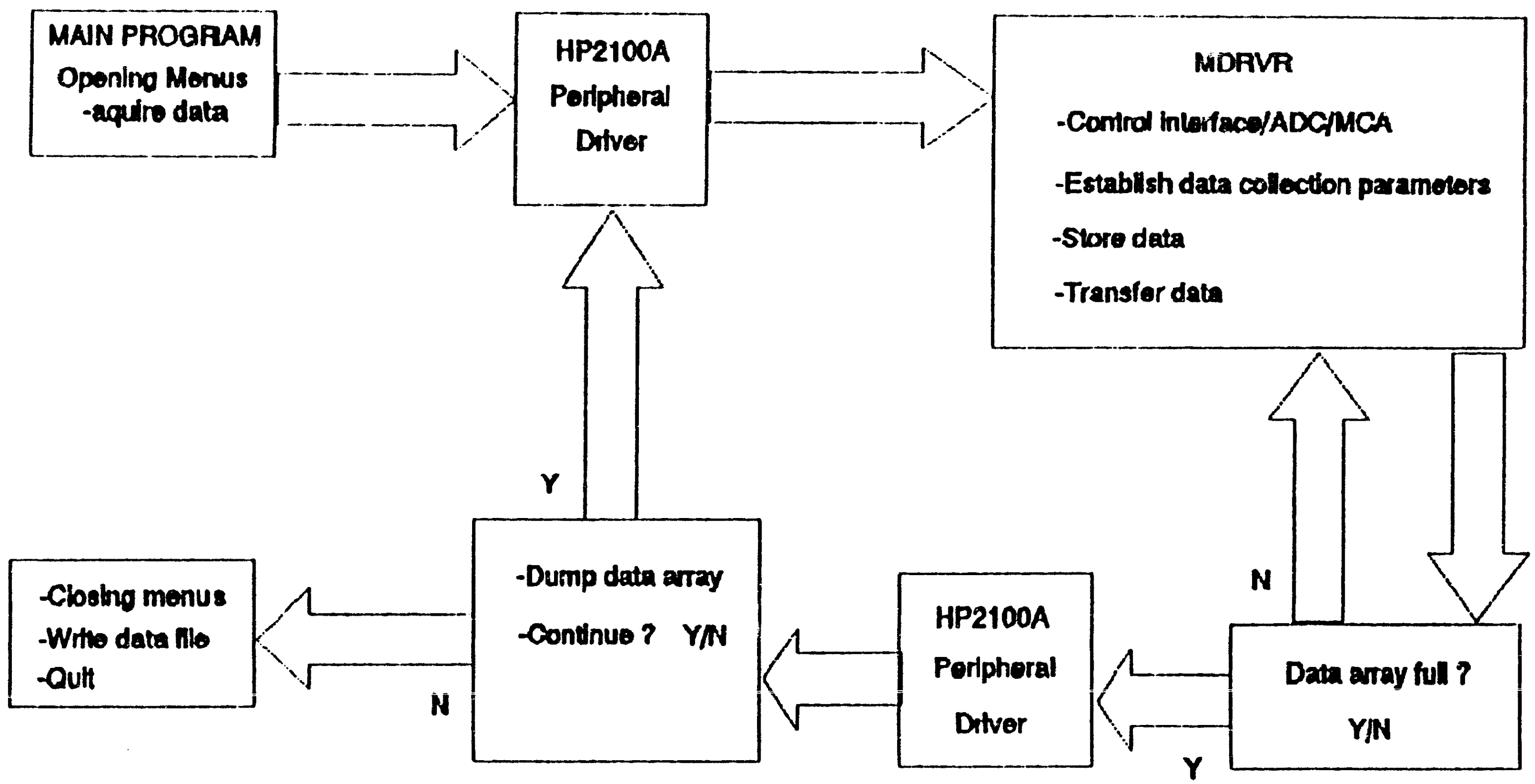

Fig. 7. Simplified block diagram and flow chart for the program controlling the two-parameter interface.

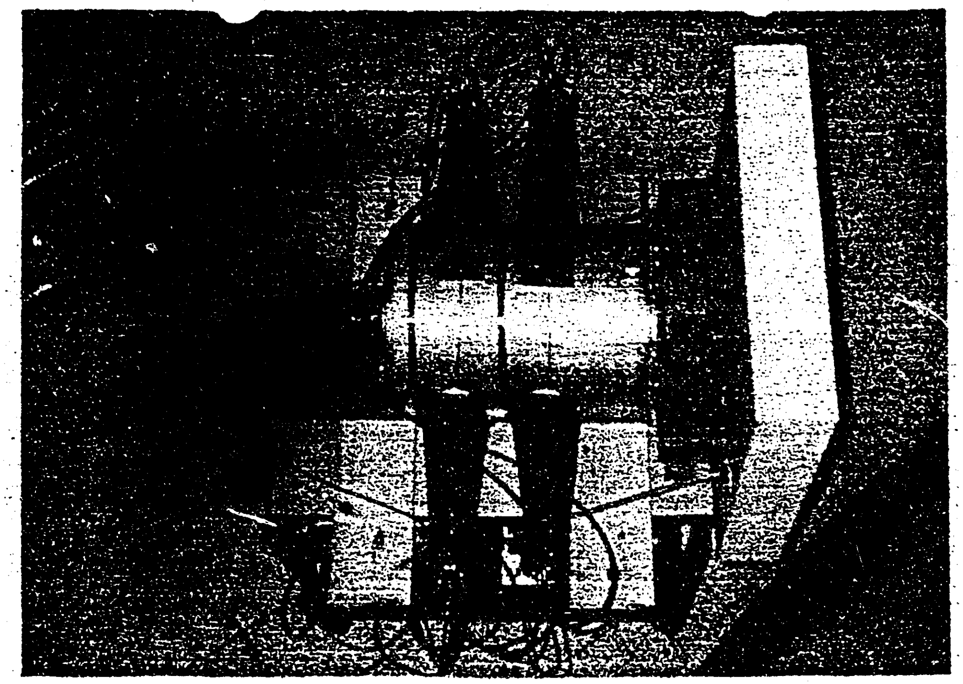

Fig. 8. A photograph of the black neutron detector with lead and borated polyetheylene shield at the front face (right side).

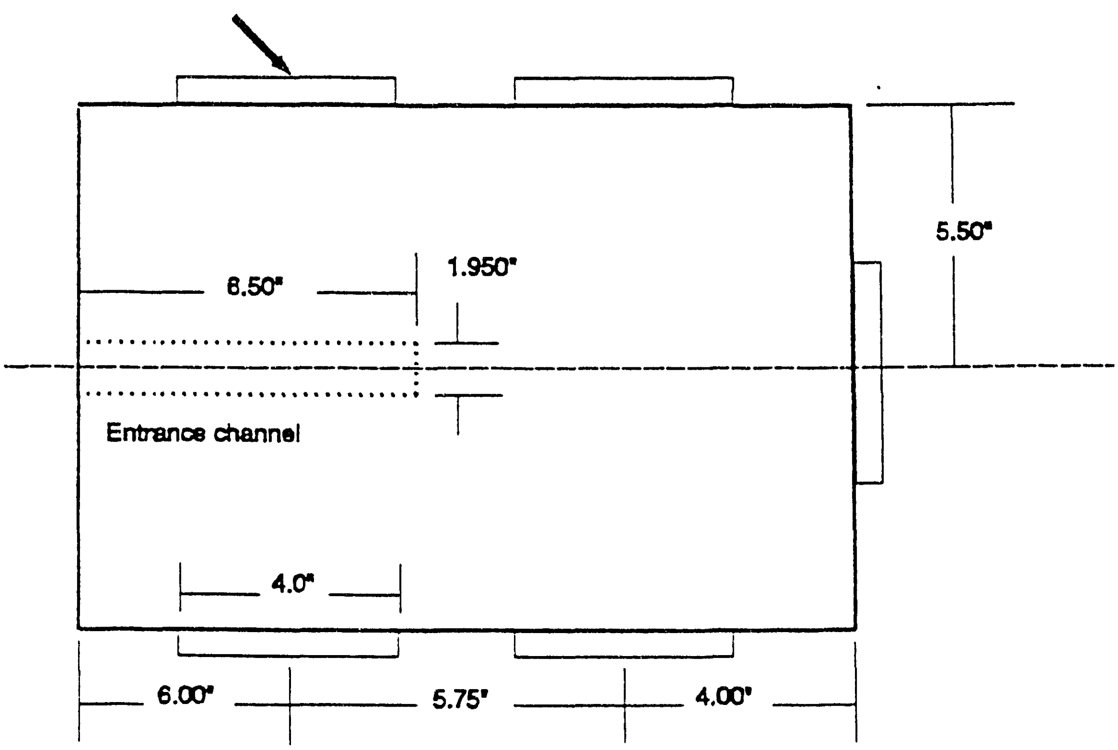

Fig. 9. Dimensions of the black neutron detector.

to line thė inside of the tin cans so that the boric acid solution does not erode the metal. Two inches of lead surrounds the detector for gamma-ray sheilding. Absorbed oxygen was removed from the BC501 by bubbling oxygen free nitrogen through it. We plan to incorporate preamplifiers into the bases of the photomultiplier tubes. This detector will be used as a flux monitor and in the investigation of the $^7Li(p,n)^7Be$ reaction as a neutron standard. This work is part of the Ph.D. research of David DeSimone.

7. Investigation of Neutron Induced Defects in Silicon Dioxide

A theoretical study of neutron damage in compound materials particularly SiO_2 was undertaken. Silicon dioxide is one of the primary constituents of electronic components, e.g. electronic devices used in nuclear instruments which have to function in radiation environments.

A paper titled "A framework for understanding fast-neutron induced defects in SiO_2 MOS structures." was published in the July 1992 Journal of Electronic Materials.[7] In this paper the pertinent theory, which is based to a large extent, on the work by Lindhard was reviewed.[8] Using the Lindhard partition function compute the total ionization energy deposited by neutrons was computed. In the same paper another approach is introduced viz. a computer simulation by using Monte Carlo methods. Because the Lindhard calculations are only applicable to single element materials, drastic approximations must be used in order to adapt the theory to multi-element cases. The problems encountered are cordially reduced by the computer simulation technique. The results from the two methods were compared in the publication. This work forms the Ph.D. dissertation research of William Chang.

8. Multiple Scattering Corrections

A detailed study of the multiple scattering of neutrons for disk scatterers was carried out using both Monte Carlo and analytical methods.

The Monte Carlo method involved simulation of our neutron scattering geometry. A program (NUSCAT) was developed in C using a powerful random number generator suggested by Marsglia et al [9,10], which passes all known randomness tests. NUSCAT calculates a finite size correction factor and a multiple scattering correction factor for various scattering orders. In order to maximize the efficiency of the generator a direction biasing method was developed. The direction biasing approach assigns a wieght to a scattered neutron according to its probability of reaching the detector based on the known angular distribution. This method increases the efficiency of the code and makes it practical to run it using a single-processor computer. The program generates a distribution for incident neutron energy and direction according to the $^{7}Li(p,n)^{7}Be$ reaction. The neutrons are scattered within the disk taking into consideration attenuation and the angular distribution. The program has been used to simulate both transmission and reflection geometries as illustrated in Fig. 10.

The program has been ported to DOS/PC, SunOs/Sun Sparc Station and is currently being ported to VAX/VMS.

NUSCAT was used to test a commonly used assumption, viz. that the ratio of the probability of (n+1)-order scattering to n-order scattering is a constant. Figure 11 shows a plot of this ratio for the various scattering orders for neutrons scattered from a ^{238}U disk. The ratio is not found to be a constant but varies significantly for small orders of scattering.

The corrections from NUSCAT have been applied to cross section data for ^{238}U for 185-keV and 128-keV incident neutrons.

In the analytical part of the study we used sparse matrix techniques[11,12,13] and eigenvalue methods to attempt to solve the multiple scattering integral. We were unable to obtain a simple analytic solution due to computing limitations with a single processor. However, we shall examine this problem in a more powerful computing environment. This work is the basis of R. Venugopal's Ph.D. dissertation.

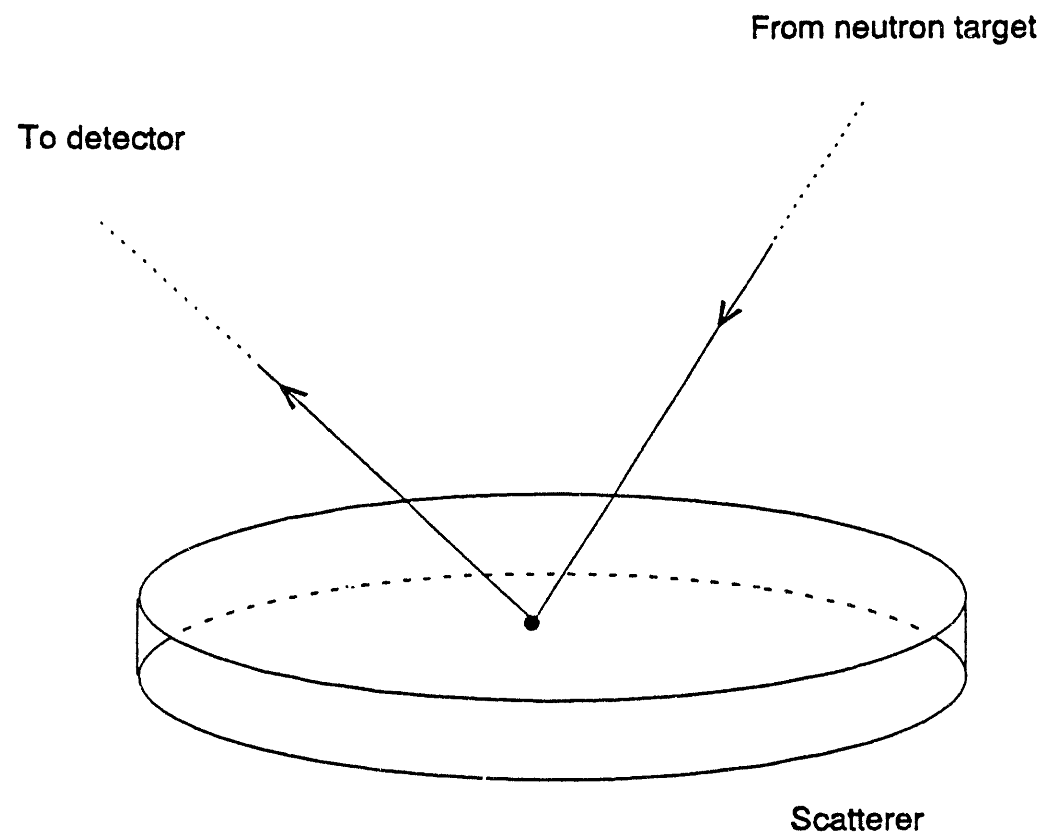

Fig. 10a Reflection geometry. Target and detector are located on the same side of the scatterer

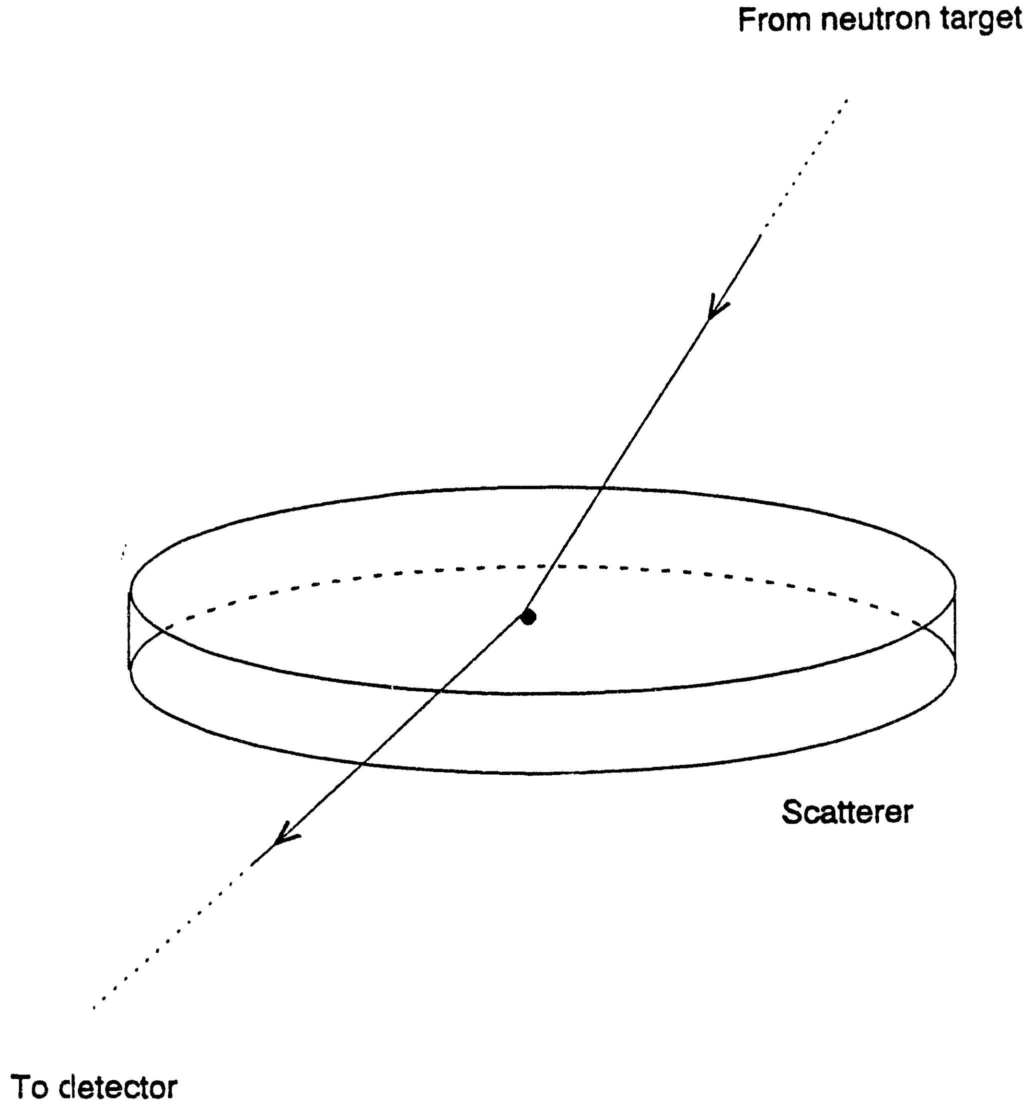

Fig. 10b Transmission geometry. Target and detector are located on opposite sides of the scatterer.

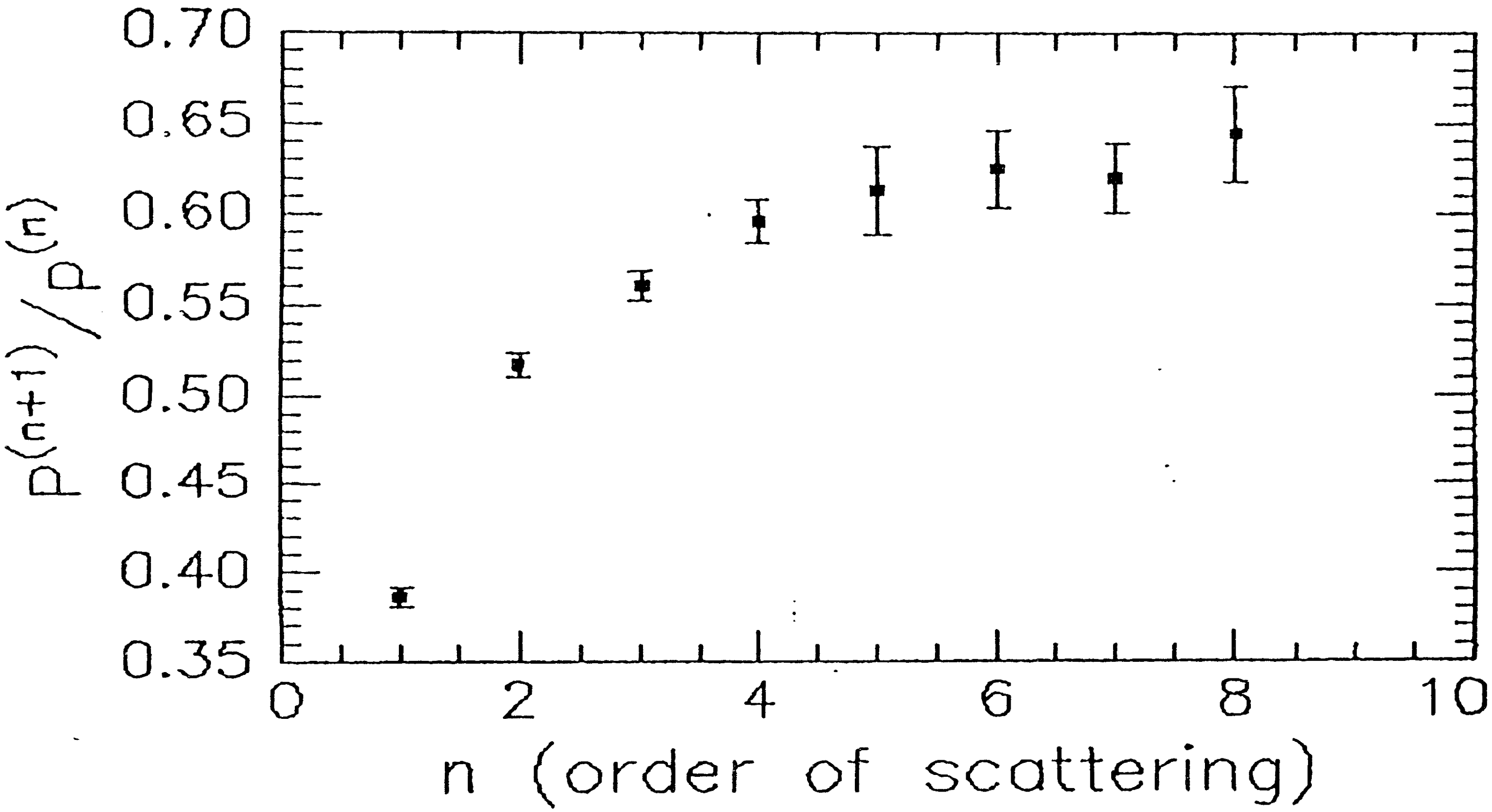

Fig. 11. Ratio of the probability of n+1 scatterings $p^{(n+1)}$ to n scatterings $p^{(n)}$. Simulation parameters: Distance from target to scatterer center = distance from scatterer center to detector = 100 cm. The scattering angle was 90 degrees. Only elastic scattering was considered. Each point in the graph represents an average of 11 runs, each run having 106 neutron histories.

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2. W. Chang "A Framework for Understanding Fast-Neutron Induced Defects in SiO_2 MOS Structures" J. Elect. Materials 21, 693 (1992).

3. Gunter H.R.. Kegel, Peter F. Bertone, Diane L. Case, David J. DeSimone, Causon K.C. Jen, Chandrika Narayan, Michael O'Connor and Parrish Staples, "Fluences and Spectra of Accelerator Generated Fast Neutrons", to be published in the Dec. 92 issue of the IEEE Transactions on Nuclear Science.

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1. J.J. Egan, G.H.R. Kegel, G. Yue, A. Mittler, P.A. Staples, D.J. DeSimone and M.L. Woodring, "Neutron Scattering in ^{239}Pu from 0.2 to 1.0 MeV", in Proceedings of the International Conference on Nuclear Data for Science and Technology, 13-17 May, 1991, Julich, Germany S.M. Qaim (Ed), Springer Verlag, Berlin, 1992, p.59.

2. G.H.R. Kegel, D.J. DeSimone, P.F. Dugan, J.J. Egan, G.K.C. Jen, A. Mittler and P.A. Staples, "Neutron Energy Spectra from Proton Irradiated Thick Li Targets", in Proceedings of the International Conference on Nuclear Data for Science and Technology, 13-17 May, 1991, Julich, Germany S.M. Qaim (Ed), Springer Verlag, Berlin, 1992, p.433.

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2. D.J. DeSimone, G.H.R. Kegel, C. Jen, J.J. Egan, D.L. Case, P. Bertone and A. Mittler, "Neutron Energy Spectra from Proton Irradiated Thick Li Targets", Bull. Am. Phys. Soc. 36, 2119 (1991).

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