NUCLEAR DATA AND MEASUREMENTS SERIES

ANL/NDM-19

A Remark on the Prompt-Fission-Neutron Spectrum of ^{252}Cf

by

P. Guenther, D. Havel, R. Sjoblom, and A. Smith

March 1976

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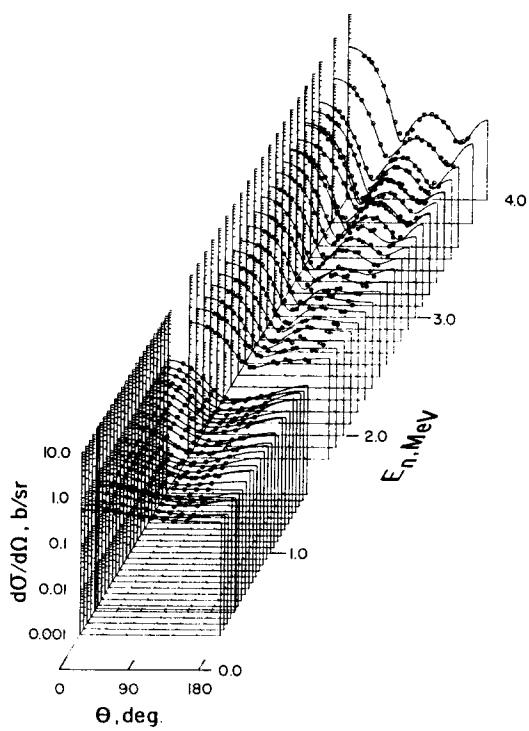

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In January 1975, the research and development functions of the former U.S. Atomic Energy Commission were incorporated into those of the U.S. Energy Research and Development Administration.

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A REMARK ON THE PROMPT FISSION NEUTRON SPECTRUM OF ^{252}Cf[a]

by

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ABSTRACT

Good resolution time-of-flight measurements show small structure in the prompt fission neutron spectrum of ^{252}Cf that is identified with air-scattering effects. The observed high-energy portion of the neutron spectrum is consistent with a Maxwellian distribution having a temperature of 1.42 MeV.

[a] This work supported by the U.S. Energy Research and Development Administration.

I. INTRODUCTION

Over several decades a number of detailed measurements have well defined the spectrum of prompt neutrons emitted at the spontaneous fission of ^{252}Cf over a wide energy range (1-13). The more recent of these results have been reviewed by Grundl and Eisenhauer and these authors conclude that the neutron spectral distribution is well represented by a Maxwellian distribution with a temperature of 1.42 ± 0.013 MeV (14). However, three areas of uncertainty remain: 1) the distribution at low energies ($\lesssim$ several 100 keV) where some results indicate a neutron intensity in excess of that predicted by a Maxwellian distribution, 2) the presence of fine structure in the distribution at energies of the order of an MeV, and 3) deviations from the Maxwellian shape at very high energies (e.g., $\gtrsim$ 10 MeV). This remark addresses the latter two of these areas.

II. EXPERIMENTAL METHOD

The experimental measurements used the fast time-of-flight technique where the neutron energy is determined from the time of transit over a known flight path. The timing sequence was initiated by the detection of a neutron and terminated with the detection of a fission event. The zero point of the time scale was established by the observation of the prompt gamma-rays emitted at fission. The velocity measurements were relativistically converted to neutron-energy distributions. The general experimental configuration is outlined schematically in Fig. 1.

The fission source-detector consisted of a gas scintillator containing a fractional microgram deposit of ^{252}Cf approximately 2 mm in diameter deposited upon a 0.025 cm thick platinum disc. This source-disc was mounted at the bottom of a brass can 5 cm deep and 5 cm in diameter with walls approximately 0.159 cm thick. A RCA 7264 photo-multiplier tube was sealed to the other end of the can with the cathode viewing the fission source. The

interior surfaces of the brass can were lined with 0.0075 cm thick aluminum foil which served as a light reflector. A continuous-flow gas system was arranged to keep the brass can filled with a mixture of 15 percent nitrogen and 85 percent argon gas at normal room temperature and pressure. The ^{252}Cf fission events were detected with very good efficiency with this device. A representative fission-fragment-pulse-height distribution, including a typical detection-bias point, is shown in Fig. 2. With this arrangement, the detected fission rate was approximately 3.0×10^5 events per second. Qualitative tests observing the prompt fission gamma-rays at a short flight path and with a fast coincidence apparatus demonstrated that the time response of the gas scintillator for the fission events was shorter than 2 nsec and thus sufficiently for good spectral measurements using the long flight paths employed in the neutron measurements.

The neutron flight path was defined by a massive collimator which shielded the neutron detector against the ambient room background. The detector shielding was very effective with low backgrounds observed even over very extended measurement periods (e.g., up to a week). The details of the collimator construction have been described elsewhere (15). The flight path was measured to be 507.2 ± 0.5 cm using a precision steel tape. The uncertainty was largely due to variations in the mean path of the neutron through the scintillator. The scintillator was "thin" to high energy (e.g., 5 MeV) neutrons and the mean path was approximately the geometric half thickness. At lower energies (i.e., several hundred keV) there was significant neutron attenuation in the scintillator resulting in mean paths somewhat less than the half thickness. Despite these uncertainties the flight path was known to a small fraction of a percent, and, as a consequence, the error did not appreciably effect the determination of neutron energies over the very large portion of the observed spectrum.

The neutron detector was an 11.4-cm-diameter by 2-cm-thick vial of NE-213 liquid scintillator coupled to a 58 AVP photo-multiplier tube (16). No special attention was given to the selection of the tube for particularly low noise response. In fact, the tube was not the best of a set of 10 employed in other measurements. Electronic circuitry was used to identify gamma-rays detected in the liquid scintillator using pulse-shape selection techniques. Events identified as due to gamma-rays were rejected by anti-coincidence logic.

The time-of-flight of the neutrons over the known flight path was measured using conventional electronic circuitry described elsewhere (15). The calibration of the timing system and the verification of its linearity were based upon a set of calibrated delay lines. These delay lines were a collection produced at various times by the Argonne National Laboratory Electronics Group using different material stocks and with independent calibrations. Calibrations of the timing system were made at delays of 0, 100, 180, 260, 340 and 420 nsec. The results were linear to within better than one timing channel of the detection system and typically gave a calibration of 2.727 ± 0.022 nsec/detection channel. The uncertainty was based upon a large-scale graphical estimate and was very conservative. Subsequently, the calibration was verified using a commercially available delay system which gave an entirely consistent result of 2.723 nsec/channel (17). The linearity of the timing system was further verified by moving the fission source-detector an appreciable distance away from the collimated neutron flight path and introducing a Pu-Be neutron source in a position so as to provide a modest response of the neutron detector. With this configuration, there should have been no time correlation between detected fission and neutron events. Time distributions were measured to approximately one percent statistical accuracy. They were essentially "flat" with no apparent slope over the range 0 to 500 nsec. An illustrative example is shown in Fig. 3.

The distribution of the counts in each time channel about the average was consistent with a standard deviation $\delta \equiv \sqrt{N}$ as shown in the figure. This distribution function was symmetric about the mean. Thus none of these tests showed significant time dependent structure or other experimental artifacts. Furthermore, the tests were repeated with an entirely different commercially available timing system with essentially the same results (17).

III. EXPERIMENTAL RESULTS AND CONCLUSIONS

A number of neutron-velocity spectra were obtained using the two different timing systems. The best neutron statistical accuracies were 1.5 to 2 percent. A typical spectrum is shown in Fig. 4. In all cases the backgrounds were small; $\lesssim$ 3 percent of the neutron spectrum maximum. The prompt gamma-ray peaks were very symmetrical with no evidence of delayed-fission gamma-ray emission observed in the range of a few tens of nsec. The long flight paths provided a clear separation between neutron and gamma-rays events and good neutron-velocity resolution.

All of the observed neutron spectra showed evidence of structure with a magnitude of several percent which persisted from measurement to measurement. This structure could not be attributed to a statistical artifact nor to instrument perturbations. Particularly persistent were fluctuations at the approximate neutron energies of 1030, 1130, 1200, 1350, 1590 and 1780 keV were clearly reproducible as is illustrated in Fig. 4. Much of this structure has the approximate energy and magnitude dependent of the results reported by Nefedov et al. (Ref. 9, noted by "N" in Fig. 4) and evident in Figure 6 of the work of Green et al. (Ref. 1, noted by "G" in Fig. 4). The consistency of this structure observed in the present with that from previous work is as good as can reasonably be expected from the resolutions and energy-scale calibrations of the various measurements.

Nefedov et al. (9) attribute this structure to delayed emission processes and their experimental procedures were unusual in greatly limiting fragment motion so as to stop the fragments in very short times. Thus delayed-neutron emission would be primarily from a static fragment and the delayed-neutron emission would be primarily from a static fragment and the delayed-neutron spectrum not distorted by the fragment motion. This was not the case for the present measurements and many, if not all, previous time-of-flight measurements of the ^{252}Cf spectrum.

The flight paths used in the present work were relatively long (> 5 m) with consequent good resolution. However, they did contain normal air which will attenuate neutrons passing from the source to neutron detector. This attenuation was calculated using oxygen and nitrogen cross sections as given in ENDF/B-IV (18) and a corresponding correction factor was deduced as illustrated in Fig. 4. The energy-average effect of this correction factor was to increase the apparent response of the neutron detector an average of 4 to 5 percent. In addition, this correction factor has prominent and often sharp structure corresponding to resonances in nitrogen and oxygen resulting in an additional few percent fluctuating correction to the detector response. Moreover, the structure in the correction factor is reasonably correlated with the observed fluctuations in the measured spectra in both magnitude and energy. There is also a good degree of correlation with the structure evident in the work of Nefedov et al. (9) and Green et al. (1).

In view of the above apparent correlation of structure and air attenuation a supporting measurement was made with approximately 10 cm of liquid nitrogen placed between neutron source and detector. The liquid nitrogen was contained in a thin-walled stainless steel vacuum dewar. Thus neutron transmission through the dewar was largely influenced by the liquid nitrogen contents. The resulting nitrogen-transmitted spectrum is shown in Fig. 4. Here the structure is very prominent and strongly correlated with the calculated air-attenuation correction and

with the fluctuations observed in the unfiltered fission spectrum. Moreover, the fluctuations observed in the nitrogen-filtered results are qualitatively energy-consistent with all but one of the fluctuations observed in the works of Nefedov et al. (9) and with the fluctuations evident in the measurements of Green et al. (1) between neutron energies of 1.0 and 4.0 MeV. The exception is in the vicinity of 3.3 MeV where the oxygen cross section changes rapidly by more than a factor of two (19). The nitrogen-transmission result is, of course, not sensitive to oxygen effects.

Attempts were made to correct the observed fission neutron spectra using the calculated air-transmission correction. The results were qualitatively successful in removing some of the fluctuations. However, the success of such corrections is very sensitive to the exact energy scales and experimental resolutions and there is no certainty that the ENDF/B cross-sections used in determining the correction factors are exact. These small discrepancies and/or uncertainties make quantitative corrections difficult and uncertain.

Generally, the small fission-spectrum fluctuations observed in the present work were qualitatively consistent in energy and magnitude with an air attenuation perturbation. It is suggested that the same qualitative conclusion is applicable to some extent to the work of Ref. 9 and Ref. 1. Both of the latter measurements were made at relatively long flight paths ($\sim$ 3.6 m) and the reports of both experiments do not describe air attenuation effects. However, it was noted above that only the work of Ref. 9 is arranged so as to limit fragment motion and thus enhance structure due to very short delayed neutron emission, if present. A number of other time-of-flight studies of the ^{252}Cf fission spectrum have generally employed shorter flight paths where the resolution was not comparable to that of the present experiments and where the attenuation due to air was much smaller. Thus it is reasonable that structure was not noted in these shorter flight-path studies.

Qualitatively, organic scintillators such as employed in present work should behave approximately as thin radiators at energies large compared to the bias point and as a "first approximation" have a relative energy-dependent response following the H(n,n) cross section. Detailed Monte-Carlo calculations such as those by Green et al. (1) and by Verbinski et al. (20) generally support this simple "first approximation" with relative efficiencies of scintillators, such as used in the present experiment, following the H(n,n) cross section to within 10 to 20 percent from 5 to 10 MeV. The deviation is toward larger efficiencies at higher energies due to secondary reaction processes particularly associated with the carbon of the scintillator. Assuming the ^{252}Cf fission spectrum follows a Maxwellian shape with a temperature of 1.42 MeV $\pm \sim 1\%$, as given in the evaluation of Grundl and Eisenhauer (14), the present spectra were used to deduce the relative efficiency of the detector. In doing so background effects were subtracted using a linear interpolation between background regions beyond either extrema of the fission spectrum. The background had a small slope qualitatively consistent with the effect of multiple events within the fission counter during the time interval of interest. The source intensity was limited to assure that this effect was small. A representative result is shown in Fig. 5. The shape of the sensitivity curve at energies below $\sim$ 5.0 MeV is very similar to that deduced from the observation of 5.0 MeV neutrons scattered from hydrogen at a number of angles in other calibration experiments (15). The relative efficiency from 5 to 12 MeV closely follows the H(n,n) cross section as illustrated in Fig. 5. Thus, if the above "first approximation" of relative detector response is valid, the present experimental results indicate no significant deviation of the spectrum from the Maxwellian of Grundl and Eisenhauer from $\sim$ 5 to 12 MeV. The more precise Monte-Carlo estimates of detector response could indicate a $\lesssim$ 20% deviation from a

Maxwellian at $\approx$ 10 MeV, toward lower neutron intensities. A trend of that nature was reported by Green et al. (1). Conversely, instrumental artifacts at very high neutron energies (e.g., overloads of linear circuit elements and malfunction of gamma-ray selection circuitry) probably would tend to reduce high-energy detector sensitivity from the simple "first approximation" estimate implying an enhancement of the spectrum relative to the Maxwellian at high energies. The present work does not preclude such a possibility. Generally, the present results from $\approx$ 5 to 12 MeV are consistent with Maxwellian distributions as deduced in the evaluation of Grundl and Eisenhauer (14).

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FIGURE CAPTIONS

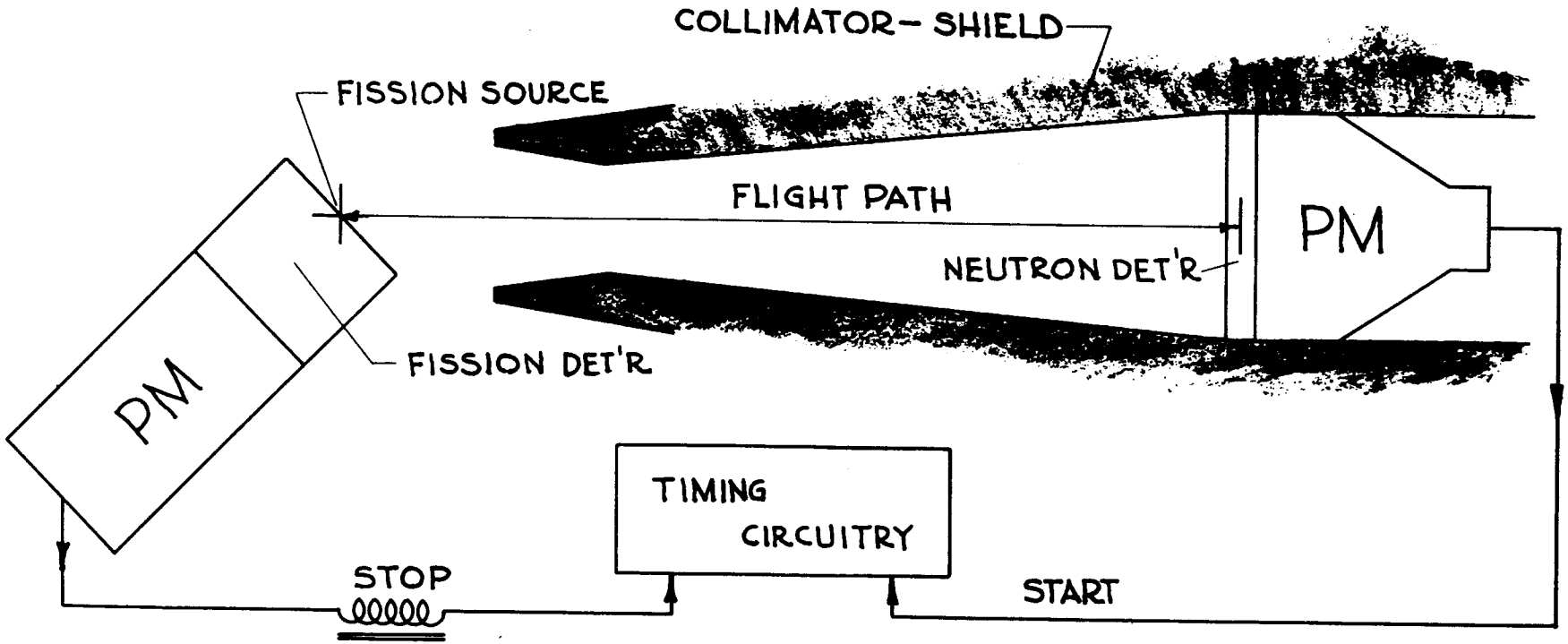
Fig. 1. Schematic Diagram of the experimental apparatus.
(ANL Negative No. 116-76-3)

Fig. 2. Typical fission-fragment distribution observed in the gas scintillator. The approximate bias point for determining the time of the fission is noted.
(ANL Negative No. 116-76-6)

Fig. 3. Illustrative tests of timing system linearity. A random time distribution is shown on the right. On the left the distribution of events about the average is shown where $\delta \equiv \sqrt{N}$.
(ANL Negative No. 116-76-6)

Fig. 4. Observed neutron-velocity spectra and the air-correction factor. The unfiltered spectrum and that filtered through $\sim$ 10 cm of liquid nitrogen are indicated by crosses the vertical magnitude of which denote statistical uncertainty. The solid curve indicates the air-attenuation correction factor on the same time scale. Dashed lines correlate structure in the spectra and correction factor at the indicated energies in keV. "N" and "G" respectively denote the position of similar structure reported in Refs. 9 and 1.
(ANL Negative No. 116-76-22)

Fig. 5. Relative neutron detector sensitivity assuming Maxwellian spectrum with temperature of 1.42 MeV. Measured values are noted by crosses with vertical magnitudes indicating uncertainties. The dashed curve indicates the H(n,n) cross section normalized at 5.0 MeV to the measured sensitivities.
(ANL Negative No. 116-76-2)



SCHEMATIC

Fig. 1.

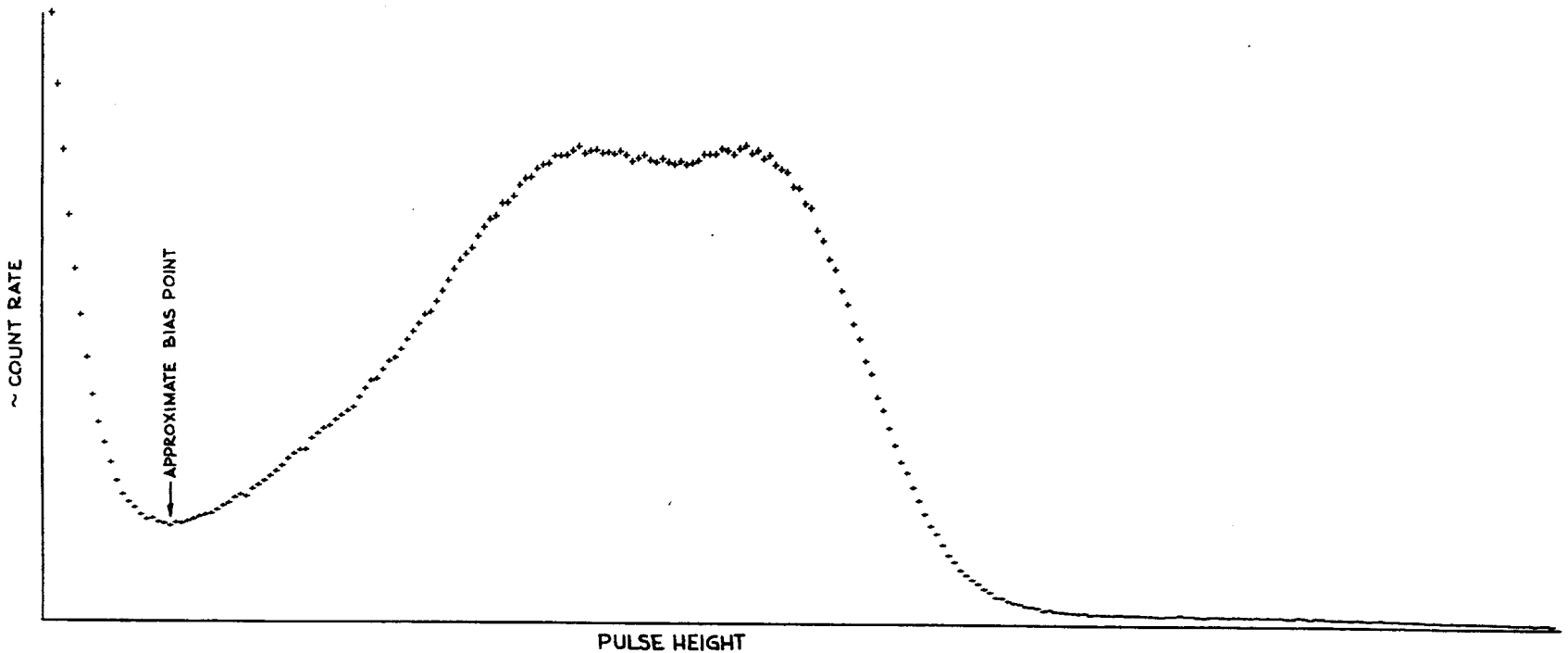


Fig. 2.

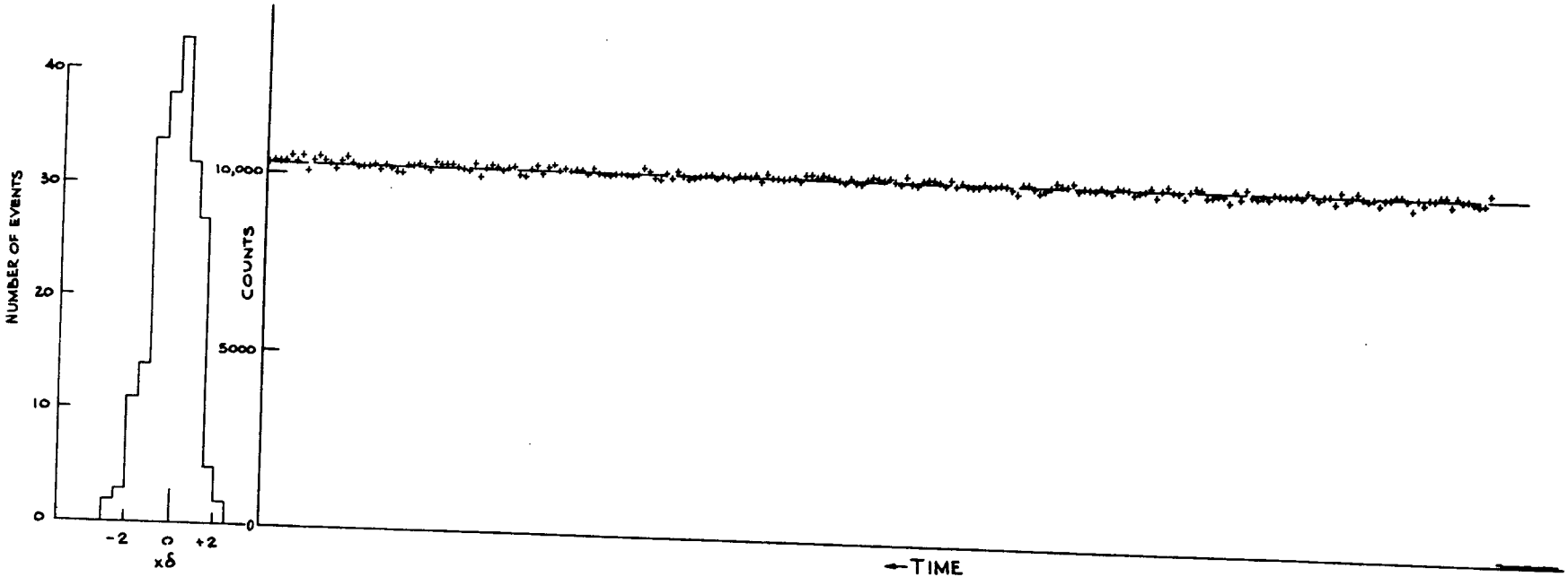


Fig. 3.

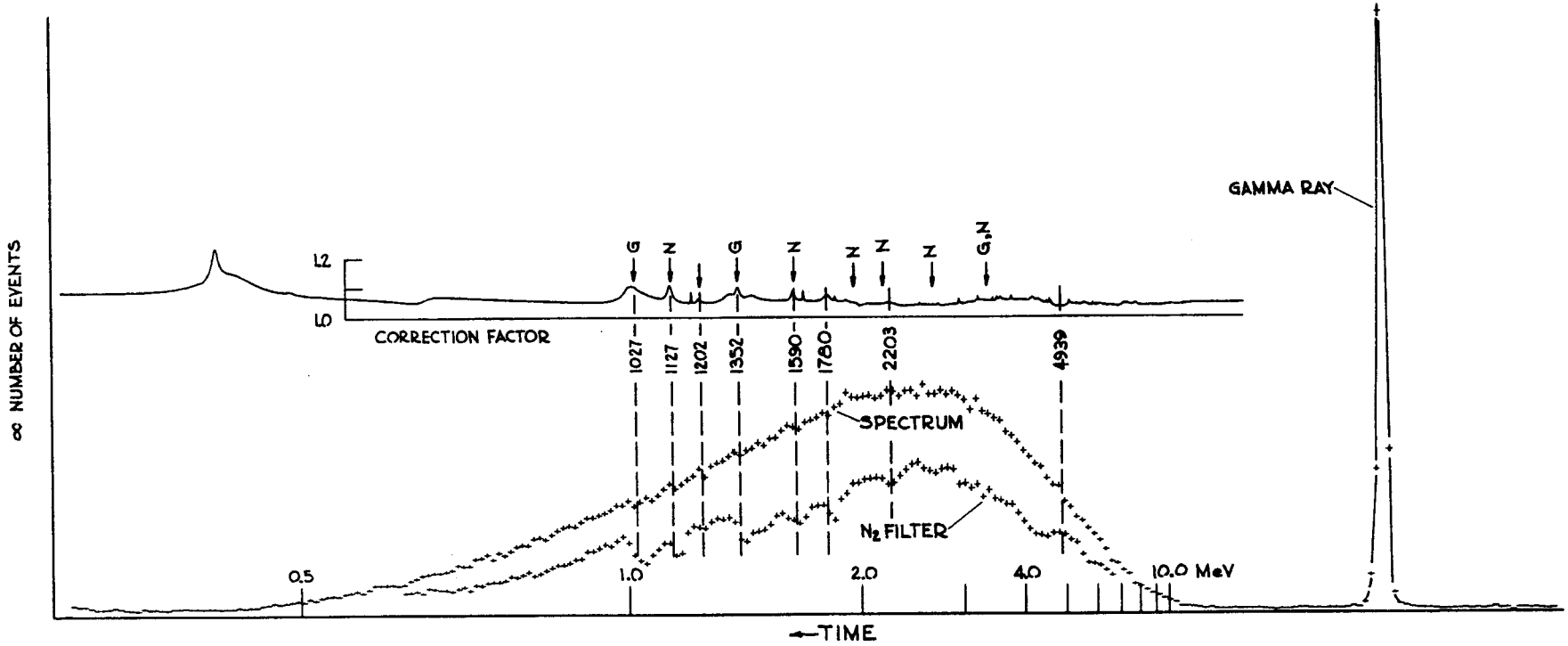

GAMMA RAY
G
N
G
N
N
N
N
G,N
1.2
1.0
CORRECTION FACTOR
1027
1127
1202
1352
1590
1780
2203
4939
SPECTRUM
N2 FILTER
0.5
1.0
2.0
4.0
10.0 MeV
TIME
∞ NUMBER OF EVENTS

Fig. 4.

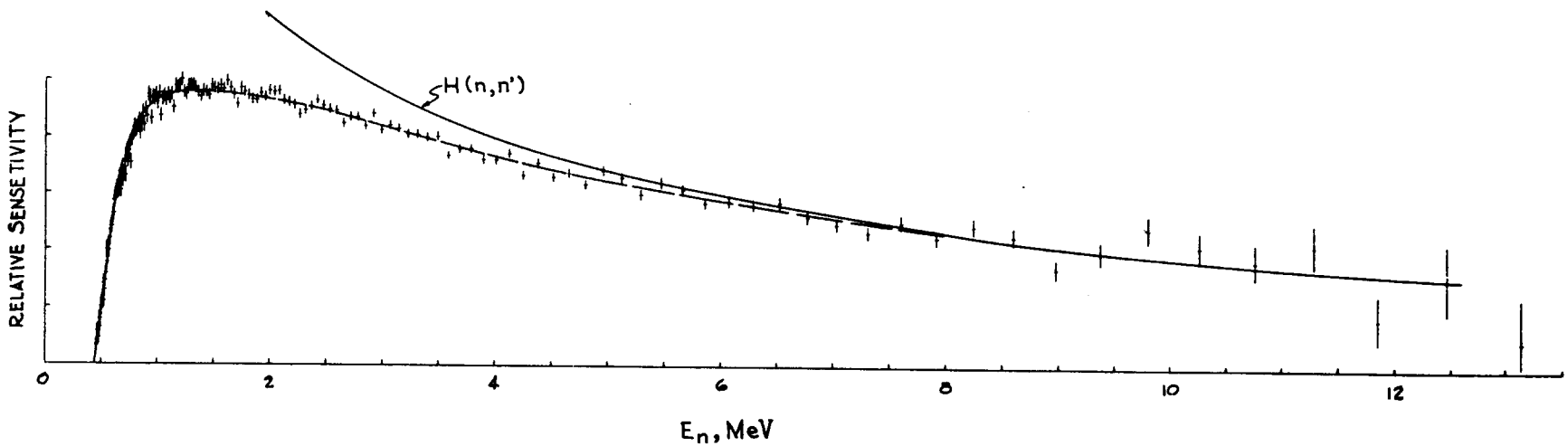


Fig. 5.