event were of high energy. A subsequent investigation employing apparatus primarily designed to measure electron velocity spectra is contemplated. Such spectra are of interest because according to the Weizel-Beeck theory of ionization by ions, a quasi molecule is formed during an ionizing collision, and it seems likely that the fine structure indicated in the spectra of Fig. 6 is related to the energy levels of the quasi molecule.

The author acknowledges with thanks the very considerable help of Professor R. N. Varney under whose direction this work was done. The assistance of Research Corporation is also gratefully acknowledged.

PHYSICAL REVIEW VOLUME 104, NUMBER 3 NOVEMBER 1, 1956

Prompt Gamma Rays Accompanying the Spontaneous Fission of Cf^{252}†

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(Received July 23, 1956)

Single- and multiple-crystal scintillation techniques are utilized to measure the energy spectrum of prompt gamma rays emitted in the spontaneous fission of Cf^{252}. It is found that each fission yields an average of 10.3 photons having a total energy of 8.2 Mev. These photons are heterogeneous in energy. An upper limit is placed on the intensity of neutrons and quanta emitted in the order of 10^{-9} second after fission.

INTRODUCTION

A KNOWLEDGE of the prompt gamma rays accompanying fission is fundamental to a complete understanding of the fission process. Furthermore, since prompt emission constitutes a significant portion of the radiation emanating from a critical assembly, it is necessary that the number and energy of the quanta be known in order to design the optimum shielding for such facilities.

Early studies[1,2] of the neutron-induced fission of U^{235} indicated that two prompt quanta were emitted with a total energy of 4–5 Mev. Several recent studies at the Oak Ridge National Laboratory[3,4] show that within 0.3 microsecond after fission more than 7 Mev of energy are emitted as photons. However, the spectral distributions and absolute normalizations of these measurements are not in complete agreement. These discrepancies may well be attributable to the high background associated with the neutron atmosphere required for the experiments.

Recently, intense Cf^{252} spontaneous fission sources have become available. The spontaneous fission of Cf^{252} has been shown theoretically and experimentally[5–7] to be similar to the neutron-induced fission of U^{235}. This offers an opportunity to study the fission phenomenon without complicating backgrounds. By utilizing these clean experimental conditions, the prompt photons and neutrons emitted in the fission of Cf^{252} were measured. The results may be extrapolated to apply to the general fission act.

EXPERIMENTAL TECHNIQUES

All of the measurements carried out in this experiment are of a coincidence type requiring simultaneous response by the fission and neutron-gamma detectors. To obtain the maximum time resolution and minimum background, it is fundamental that the detection system be as fast as possible. The recently developed gas scintillator serves as an excellent fast-fission detector,[8] combining good energy resolution with high speed and an insensitivity to gamma radiation.

Because the prompt gamma-ray spectrum is expected to be of a heterogeneous nature, it is desirable that the technique employed respond to an incident photon in a unique manner. Two methods are utilized. In the first the pulse-height distribution from a single $1\times1\frac{1}{2}$ inch NaI(Tl) crystal is measured in coincidence ($\tau=0.3$ microsecond) with the pulses from the fission dectector. The response of this system to gamma rays is not unique but it is the most sensitive method available. In the second approach, a double-crystal Compton spectrometer is used[9–11] in coincidence ($\tau\geq2.5$ millimicroseconds) with the fission detector. This system does give a unique response to the incident quanta. In most applications of the Compton effect to

† This work is supported by the U. S. Atomic Energy Commission.
[1] M. Deutsch and J. Rotblat, Atomic Energy Commission Report AECD-3179 (unpublished).
[2] Kinsey, Hanna, and Van Patter, Can. J. Research **26**, 79 (1948).
[3] J. Francis and R. Gamble, Oak Ridge National Laboratory Report ORNL-1879 (unpublished).
[4] F. Maienschein *et al.*, Oak Ridge National Laboratory Report ORNL-1879 (unpublished).
[5] Smith, Friedman, and Fields, Phys. Rev. **102**, 813 (1956).
[6] L. Glendenin and E. Steinberg, J. Inorg. and Nuclear Chem. **1**, 45 (1955).
[7] R. B. Leachman, Phys. Rev. **101**, 1006 (1956).
[8] C. Eggler and C. Huddleston, Nucleonics **14**, 4 (1956).
[9] R. Hofstadter and J. McIntyre, Phys. Rev. **78**, 134 (1950).
[10] F. Maienschein *et al.*, Oak Ridge National Laboratory Report-1142 (unpublished).
[11] P. R. Howland *et al.*, U. S. Naval Radiological Defense Laboratory Report USNRDL-TR-65 (unpublished).

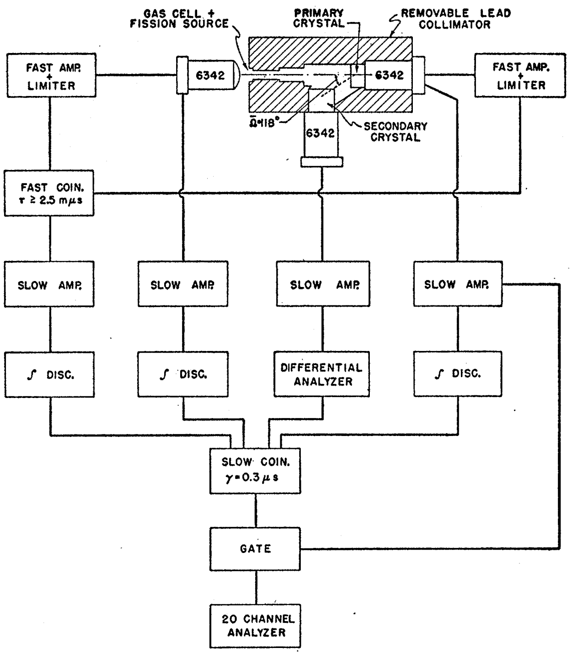

FIG. 1. Block diagram of the experimental apparatus.

spectrometry, energy resolution is the primary concern. However, in experiments of this nature sensitivity is also of importance, so experimental parameters are adjusted to give an optimum compromise between these two characteristics. The response of the secondary crystal is differentially selected in such a manner that the efficiency of this detector for Compton backscattered quanta is much greater than for annihilation radiation. This extends the usefulness of the technique to higher energies. A composite diagram of the apparatus is shown in Fig. 1.

EXPERIMENTAL MEASUREMENTS

The gas cell is coated with carrier-free Cf^{252} yielding about 5000 fissions/minute.

The pulse-height distribution resulting from prompt-fission gamma rays incident on the single NaI crystal is shown in Fig. 2. Toward higher energies the distribution is seen to decrease monotonically. At very low energies it falls off sharply, with evidence of a small photopeak at ~60 kev. The source is mounted on a piece of aluminum 0.010 inch thick and no heavy shield material is used, so it appears that this 60-kev line must be associated with the fission event rather than the secondary x-ray effects as has been proposed.[12] In the following, this line will be considered only in its integrated contribution to the over-all spectrum.

The single crystal pulse-height distribution is given approximately by:

$$N(E)=\epsilon_{ph}(E)\gamma(E)+\int^{\infty}\epsilon_C(E'\to E)\gamma(E')dE' +\epsilon_{pr}(E+2mc^2)\gamma(E+2mc^2)(1-n) +\epsilon_{pr}(E+mc^2)\gamma(E+mc^2)n,$$

where $N(E)$ = measured number of pulses corresponding to an energy loss, E, in the crystal; $\gamma(E)$ = number of photons of energy E incident on the crystal; $\epsilon_{ph}(E)$ = photoelectric efficiency of the crystal corrected for total Compton absorption; $\epsilon_C(E'\to E)$ = Compton efficiency of the crystal for photons of energy, E', to produce Compton recoil electrons of energy, E; $\epsilon_{pr}(E)$ = pair efficiency of the crystal, and n = probability of the crystal recapturing one annihilation quanta. Or, in matrix form, one has $\mathbf{N}=\mathbf{A}\gamma$, where the components of $\mathbf{A}$ are calculable from known theory.[13] The determination of the incident photon spectrum from the pulse-height distribution is equivalent to inverting the matrix $\mathbf{A}$ in an energy space of sufficient dimension to give the desired accuracy. Unfortunately this inversion is difficult owing to the small size of many of the diagonal components (photoefficiencies). For this reason an iterative method is used, the calculation

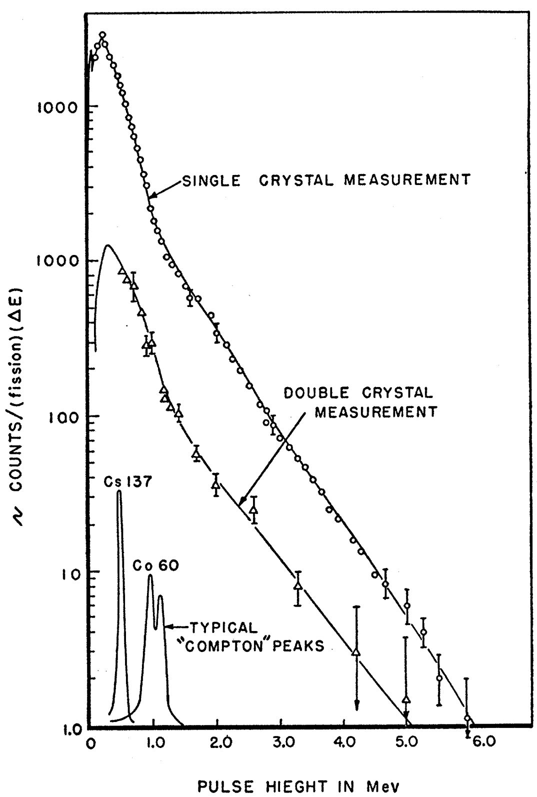

FIG. 2. Measured pulse-height distributions.

[12] L. Magnusson *et al.*, Phys. Rev. **96**, 1576 (1954).

[13] H. A. Bethe and J. Ashkin, *Experimental Nuclear Physics*, edited by Emilio Segrè (John Wiley & Sons, Inc., New York, 1953), Vol. 1.

being carried to the desired accuracy. The energy calibration and absolute normalization are checked against National Bureau of Standards sources. The single-crystal method is most satisfactory at higher energies where the cross sections are not changing rapidly. At lower energies there is considerable error due to the necessary averaging of rapidly varying quantities. In this region a more accurate measurement is obtained by the double-crystal technique. The efficiency of this system is calculated in a straightforward manner[10,11] and the results are again checked against National Bureau of Standards sources. The measured distribution of recoil Compton electrons in the primary crystal is shown in Fig. 2. By combining the results of the single- and double-crystal measurements, the composite spectrum of prompt fission gamma rays is obtained (Fig. 3).

Upon completion of the above measurements the source is transferred to a large precision fission chamber.[5] A thin aluminum window allows the placement of a NaI crystal directly behind the source. The single-crystal measurements are repeated with the recording system arranged to give a one-to-one time and energy correlation between photons and fission fragments. In this manner the absolute spectrum of the emitted photons is determined as a function of the fragment mass ratio. In the analysis of the data the fragment ratio is divided into three equal increments, the symmetric group, the most probable group, and the most asymmetric group. Within the statistical accuracy of the measurement (~8%), the photon emission is found to be independent of the mass ratio. The averaging procedure used precludes a detailed analysis but certainly a general trend would have been detected.

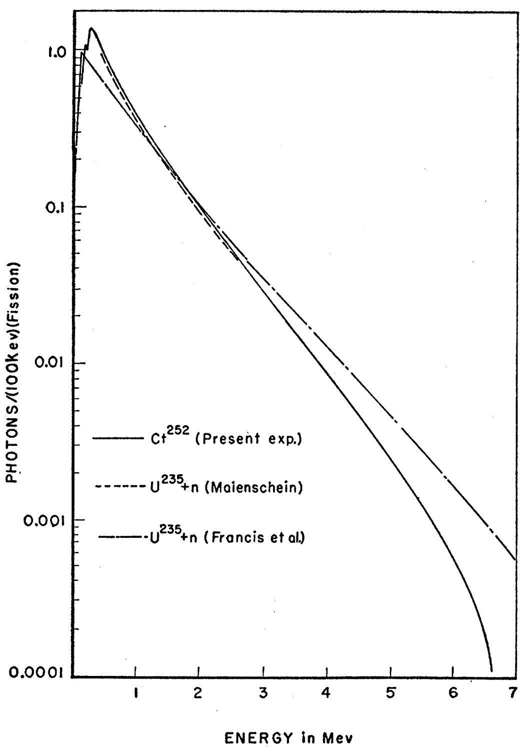

FIG. 3. Photon spectrum from the fission of Cf^{252}.

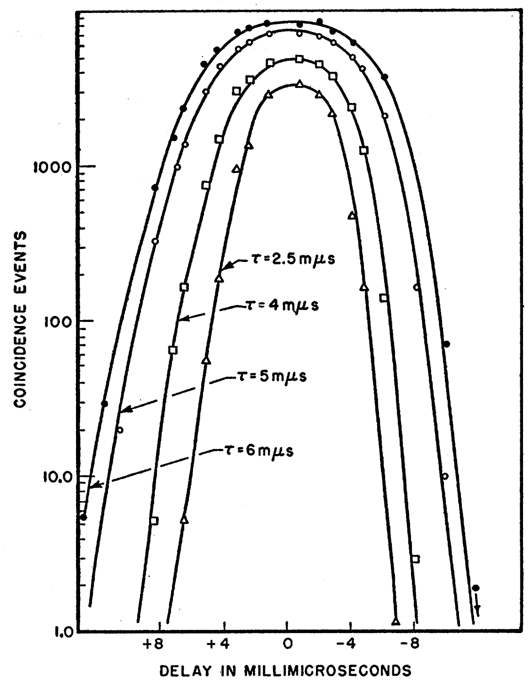

FIG. 4. The time distribution of neutrons and gamma rays from the fission of Cf^{252}.

In conjunction with the spectrum measurements, a search is made for fission neutrons and photons delayed in the order of a millimicrosecond. The slow-fast coincidence system is applied in a straightforward manner with resolving times as short as 2.5 millimicroseconds. Such fast resolving times are possible through the use of the gas scintillator as the fission detector and a plastic scintillator as the neutron-gamma detector. The measured distribution of coincidence events as a function of the time delay between the fission and neutron-gamma detector is shown in Fig. 4. During this measurement the effective centers of the two detectors are 1.8 cm apart. With the exception of a very slight tail, the curve is seen to be symmetrical with respect to time. The resolving times and efficiencies of coincident systems of this type have been shown to be functions of the pulse duration and size.[14] In order to analyze the data, two assumptions are made: first, that the gamma distribution under investigation is assumed to be equivalent to that determined above, and secondly, that the incident neutron distribution is assumed to be given by Watt's expression.[15] These assumptions are not as critical as one would at first suspect, owing to the compensating effect of the

[14] R. Post and L. Schiff, Phys. Rev. 78, 80 (1950).
[15] B. Watt, Phys. Rev. 87, 1037 (1952).

TABLE I. Experimentally determined upper limits for the relative intensity of neutrons and photons emitted in short time intervals after the spontaneous fission of Cf^{252}.

Time increment, ΔT, after fission (millimicroseconds)	Upper limit for relative neutron intensity in interval, ΔT	Upper limit for relative photon intensity in interval, ΔT
1–3	10%	15%
3–5	3%	3%
5–10	3%	3%
10–50	10%	10%
greater than 50	50% or more	50% or greater

increasing sensitivity of the plastic scintillator for lower energy delayed neutrons and gammas. Under the above assumptions, the efficiency of the plastic scintillator for neutrons and gamma rays is calculated. This calculation is verified experimentally by separating the detectors sufficiently to obtain complete velocity resolution of the γ rays and neutrons. From these measurements it is also evident that the small tail noted in Fig. 4 is due to the time of flight of the slower fission neutrons from the fission source-detector to the effective center of the adjacent plastic scintillator. The experimentally determined upper limits for the intensity of photons and neutrons emitted in the order of 10^{-9} second after the fission of Cf^{252} are given in Table I.

DISCUSSION

The characteristics of the prompt photon emission accompanying the fission of Cf^{252} are given in Table II and Fig. 3. For comparison, the photon emission from the fission of U^{235} is also given. As predicted, the photon spectrum from the fission of Cf^{252} is very similar to that from the fission of U^{235}. In fact, the measurements of Maienschein *et al.*, are in good agreement with the present work. All of the results show that considerably more energy is dissipated in prompt photons than is theoretically expected.

The limits set on the presence of delayed neutrons from the fission of Cf^{252} are not in disagreement with the known delayed groups from the neutron-induced fission processes.[16–18] While the fission phenomenon is believed to be relatively slow, prompt emanations will still occur in times five orders of magnitude shorter than the limit of the present technique. Thus any quanta or neutrons in the millimicrosecond range must be of a delayed nature. If one accepts the usual premise that delayed neutron emission follows a fast beta decay, the reason for the absence of delayed neutron groups in these time intervals is obvious. Even for the most favorable hypothetical case of a 10^{-9}-second superallowed transition between mirror nuclei of zero charge,

TABLE II. Characteristics of prompt photons from fission.

Fissioning isotope	Total photons/ fission	Photons/ fission (0.5–2.3 Mev)	Energy loss in photons/ fission (0.5–2.3 Mev)	Total energy loss in photons	Reference
Cf^{252}	10.3	5.0	5.2	8.2 Mev	Present work
$U^{235}+n$	···	5.0	5.1	8.0 extrapolated	a
	7.5	···	···	7.46	b

[a] See reference 4.
[b] See reference 3.

the energy of the beta decay would exceed 15 Mev. The probability of a fragment having this amount of energy plus the binding energy of a neutron is indeed small. If any low-intensity delayed-neutron group does exist in this range, it must be explained by an entirely new mechanism. The absence of millimicrosecond delayed gamma rays is also anticipated. The region of maximum mass yield in the fission of Cf^{252} is well away from nuclei known to emit the $E2$, $M2$, and $M1$ type transitions necessary to fulfill the time requirements.

[16] J. Bendt and F. Scott, Phys. Rev. **97**, 744 (1955).
[17] D. J. Hughes *et al.*, Phys. Rev. **73**, 111 (1948).
[18] T. Snyder and R. Williams, **81**, 171 (1951).