Half-Lives and Yields of X Rays from Thermal Fission of U^{235} and Pu^{239}†

LYNN BRIDWELL,*‡ MARVIN E. WYMAN, AND BERNARD W. WEHRING*
Nuclear Engineering Program, University of Illinois, Urbana, Illinois
(Received 27 December 1965)

Time and energy characteristics of x rays from fission fragments of thermally fissioned U^{235} and Pu^{239} were investigated. The timing data were obtained electronically using amplitude-conversion techniques while the energy spectra were obtained using NaI(Tl) as the detector. The timing measurements show that about 50% of the x rays have half-lives greater than 4 nsec. This suggests that the dominant or even the entire mechanism for x-ray production is internal conversion of low-energy transitions of the fission fragments. Half-lives obtained from a least-squares fit of the time spectra indicate that the transitions are of the type $E2$ and $M1$. A model for predicting the x-ray energy spectra is given. X-ray yields calculated from the energy spectra were determined to be 0.17 ± 0.02 and 0.43 ± 0.004 x rays per fission for the light- and heavy-group fragments of U^{235}, and 0.15 ± 0.02 and 0.26 ± 0.03 x rays per fission for the light- and heavy-group fragments of Pu^{239}. Results from other experimenters are given for comparison.

I. INTRODUCTION

THE purpose of the experiments which are described here was twofold: first to determine the time characteristics, relative to fission, of K x rays resulting from thermal fission of U^{235} and Pu^{239}, and second to determine the number of K x rays per fission from energy spectral measurements. As was shown by Carter, Wagner, and Wyman,[1] and as reported by Leachman at Geneva,[2] the fission fragments of U^{235} emit characteristic x rays. They obtained both L and K x-ray spectra that resembled the mass yield distribution from fission. If the processes responsible for the x-ray emission can be understood, these x rays provide a means for determining the division of primary nuclear charge in fission.

As pointed out by Leachman, the influence of the following processes must be considered when correlating charge yield to x-ray spectra:

(a) x rays resulting from internal conversion;
(b) fluorescent yield;
(c) the number of atomic vacancies produced by the formation of the fragments.

Process (b), fluorescent yield, being well known from nonfission experiments,[3] was not investigated here. Information about processes (a) and (c) can be obtained from a determination of the time of emission of the x rays with respect to the fission event. If the atomic vacancies were created by the formation of the fragments, the delay times with respect to fission would be characteristic of atomic lifetimes. (Using x-ray spectral data[4] and Heisenberg's uncertainty principle the mean life of a K vacancy was determined to be 10^{-17} sec in $_{74}W$ and 10^{-16} sec in $_{38}Sr$.) If, however, the vacancies were the result of internal conversion, half-lives would be characteristic of multipole nuclear transitions in the range 10^{-10} to 10^{-7} sec.[5,6]

Timing studies on x-ray emission from fission of U^{235} in the nanosecond range have been reported previously.[7–9] Sklyarevskii *et al.*,[7] as well as Shiryayev,[8] used time-of-flight techniques to determine an average single lifetime for the entire photon spectrum. Desi, Lajtai, and Nagy[9] used electronic means of timing and determined that the lifetimes decreased as the photon energy discriminator threshold was increased. Bowman, Thompson, Watson, Kapoor, and Rasmussen,[10] using solid-angle considerations, determined half-lives of the x-ray emitters in fragments of Cf^{252} in the range 0 to 1 nsec.

Numerous investigators have reported energy spectra of x rays emitted within 0.5 μsec of fission of U^{235}.[1,2,7,11–14] Voitovetskii, Levin, and Marchenko[11] measured 0.45 ± 0.15 heavy-fragment x rays per fission. Sklyarevskii, Fomenko, and Stepanov[7] report 0.42 ± 0.12 per fission for the heavy group. Sklyarevskii, Stepanov, and

† The material in this article is based upon a dissertation by one of the authors (LB) submitted in partial fulfillment of the requirements for the doctoral degree at the University of Illinois.

* Former Oak Ridge Institute of Nuclear Studies Graduate Fellow.

‡ Present address: Department of Physics, Murray State College, Murray, Kentucky.

[1] R. E. Carter, J. J. Wagner, and M. E. Wyman, Bull. Am. Phys. Soc. 3, 228 (1958).

[2] R. B. Leachman, in *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy Geneva, 1958* (United Nations, Geneva, 1958), Vol. 15, pp. 665, 331.

[3] C. D. Broyles, D. A. Thomas, and S. K. Haynes, Phys. Rev. 89, 715 (1953).

[4] A. H. Compton and S. K. Allison, *X-Rays in Theory and Experiment* (D. Van Nostrand, Inc., New York, 1935), p. 745.

[5] V. F. Weisskopf, Phys. Rev. 83, 1073L (1951).

[6] A. H. Wapstra, G. J. Nijgh, and R. Van Lieshout, *Nuclear Spectroscopy Tables* (North-Holland Publishing Company, Amsterdam, 1959), p. 71.

[7] V. V. Sklyarevskii, D. E. Fomenko, and E. P. Stepanov, Zh. Eksperim. i Teor. Fiz. 32, 256 (1957) [English transl.: Soviet Phys.—JETP 5, 220 (1957)].

[8] B. M. Shiryayev, FTD-TT-61-175, 25, 1962 (unpublished).

[9] S. Desi, A. Lajtai, and L. Nagy, Acta. Phys. Acad. Sci. Hung. 15, 185 (1962).

[10] H. R. Bowman, S. G. Thompson, R. L. Watson, S. S. Kapoor, and J. O. Rasmussen, Lawrence Radiation Laboratory Report No. UCRL-11902, 1965 (unpublished).

[11] V. K. Voitovetskii, B. A. Levin, and E. V. Marchenko, Zh. Eksperim. i Teor. Fiz. 32, 263 (1957) [English transl.: Soviet Phys.—JETP 5, 184 (1957)].

[12] F. C. Maienschein, R. W. Peelle, and T. A. Love, Atomic Energy Commission Report No. TID-6302, 1959 (unpublished).

[13] V. V. Sklyarevskii, E. P. Stepanov, and B. A. Medvedev, Zh. Eksperim. i Teor. Fiz. 36, 326 (1959) [English transl.: Soviet Phys.—JETP 9, 225 (1959)].

[14] H. Hohmann, Z. Physik 172, 143 (1963).

Medvedev[13] report 0.10 ± 0.03 per fission for the light-fragment group. Hohmann[14] reports $0.2\pm0.06K$ x rays per fission for both the light and heavy fragments combined.

Recently Griffin and Glendenin,[15] and Glendenin, Unik, and Griffin[16] have reported comprehensive investigations into the origin and spectrum of x rays emitted by fission fragments of Cf^{252}. On the basis of timing measurements using both time-of-flight and electronic means, they concluded that the K x rays were the result of internal conversion.

2. EXPERIMENTAL METHOD

The method chosen for making the lifetime measurements was the delayed-coincidence method.[17] A thin plastic detector was used to indicate the time of fission. It was designed to selectively detect fission fragments in a high background of alpha particles and Compton electrons. The x rays were detected by a 1-mm NaI(Tl) crystal covered by a 0.005-in. Be window. Both scintillators were mounted on Amperex 56 AVP photomultipliers. A time-to-amplitude converter operating in the nanosecond region was used to measure the amount of delay between fission-fragment pulses and x-ray pulses. Events were selected on the basis of energy by a conventional coincidence system and the time differences were analyzed by a 400-channel pulse-height analyzer. Details of the detectors and the fast-coincidence system are described elsewhere.[18] Figures 1 and 2 show a schematic diagram of the electronics and the experimental arrangement respectively. The prompt response of the fast system at the heavy-fragment x-ray energy peak was measured by using K x-ray emission following internal conversion of Cs^{137}. For the light-fragment group the response was measured by using L x-ray emission of U^{235} following alpha decay of Pu^{239}. In the fission fragment experiment only x rays near the peaks of the energy spectrum were accepted by the slow coincidence corresponding to the conditions that existed in the measuring of the prompt responses.

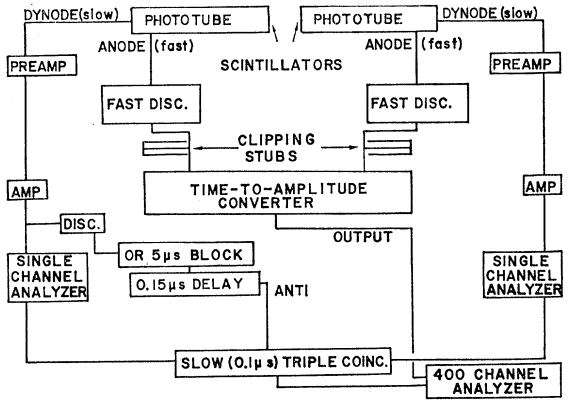

FIG. 1. Electronic block diagram for half-life measurements.

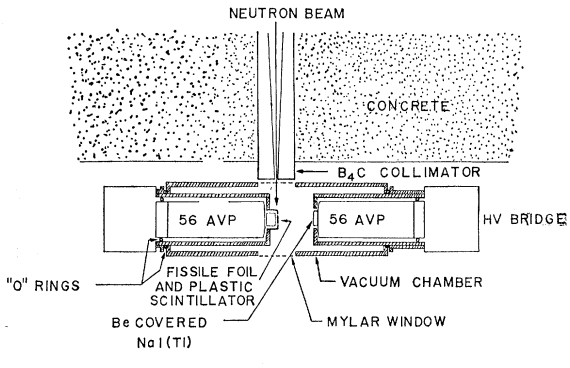

FIG. 2. Experimental arrangement.

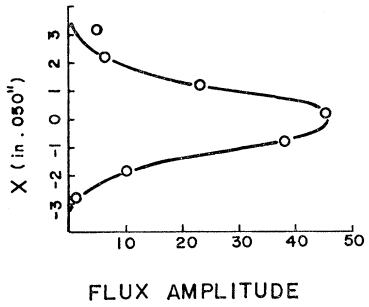

FIG. 3. Flux map across slit of neutron collimator.

Thin oxide films of Pu^{239} and uranium enriched to 93% U^{235} on 0.001-in. Al were used as the fissile sources. A neutron beam from the thermal column of the University of Illinois TRIGA Mark II reactor served as the source of neutrons. Boron in the form of B_4C imbedded in paraffin was used to construct a neutron collimator. The narrow flux profile (see Fig. 3) allowed the NaI(Tl) to be placed within 5 cm of the fissile foil without shifts in phototube gains due to high count rates.

3. RESULTS

A. Time Spectra for Light- and Heavy-Fragment X Rays

Typical results for the light- and heavy-fragment x rays from both U^{235} and Pu^{239} are presented in Figs. 4 through 7. These are representative of the 29 runs that were made during the 100 h of reactor operation. In the Pu^{239} experiments the high alpha count rates and the resulting pulse pile-up in the slow system made it necessary to move the plastic detector 1 cm away from the fissile foil. Time variations resulting from the spread of fragment flight times due to kinetic-energy variations

[15] H. C. Griffin and L. E. Glendenin, Phys. Letters **15**, 153 (1965).
[16] L. E. Glendenin, J. P. Unik, and H. C. Griffin, in *International Symposium on the Physics and Chemistry of Fission, 1965* (International Atomic Energy Agency, Vienna, 1965). Also published as L. E. Glendenin and J. P. Unik, Phys. Rev. **140**, B1301 (1965).
[17] A. Schwarzschild, Nucl. Instr. Methods **21**, 1 (1963).
[18] L. Bridwell and M. E. Wyman, Rev. Sci. Instr. (to be published).

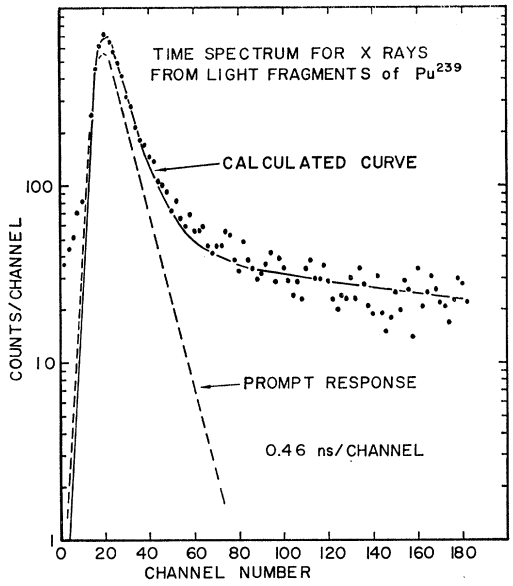

FIG. 4. Time spectrum for light-fragment x rays from fission of Pu^{239}. Prompt response was measured by using the L x ray from U^{235} following alpha decay of Pu^{239}.

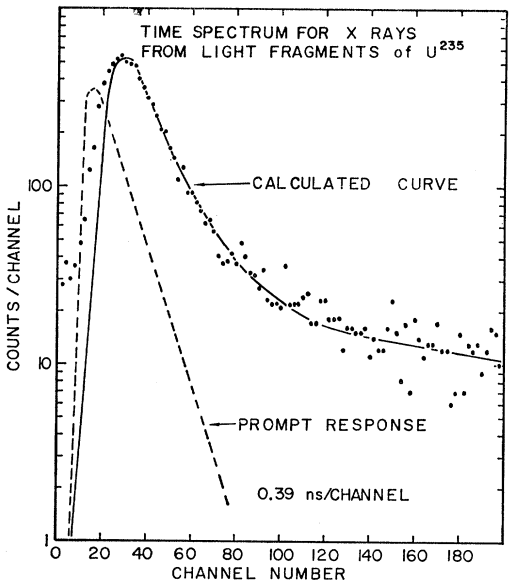

FIG. 6. Time spectrum for light fragment x rays from fission of U^{235}. Prompt response was measured by using the L x ray from Th^{230} following alpha decay of U^{234}.

were reduced by accepting only the more energetic fragments.

The true time spectrum is related to the measured spectrum and the prompt response through the following relationship:

$$F(t)=\int_0^t f(x)g(t-x)\,dx. \qquad (1)$$

The true time spectrum $f(t)$ is composed of a large sum of exponential functions the decay constants and amplitudes of which are to be determined. From the measured data one cannot hope to resolve the individual components. However, the time characteristics can be represented by a greatly reduced number of exponentials. The tail of $F(t)$ was fitted, in the least-squares sense, using a computational scheme developed by Need and Fessler.[19] Preliminary searches were conducted to

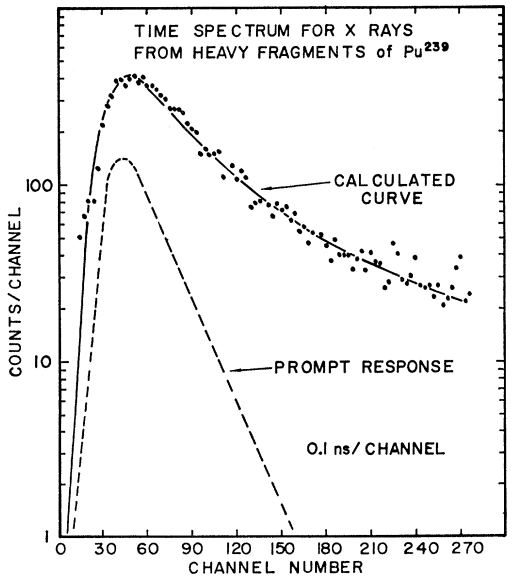

FIG. 5. Time spectrum for heavy fragment x rays from fission of Pu^{239}. Prompt response was measured by using internal conversion of Cs^{137}.

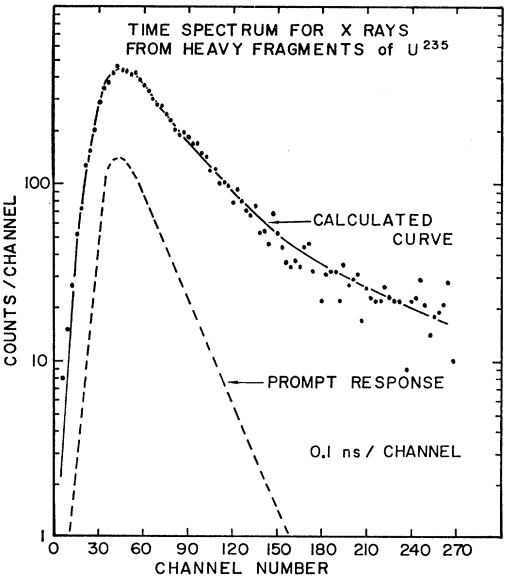

FIG. 7. Time spectrum for heavy-fragment x rays from fission of U^{235}. Prompt response was measured by using internal conversion of Cs^{137}.

[19] J. L. Need and T. E. Fessler, National Aeronautics and Space Administration Report No. NASA TN D-1453, 1962 (unpublished).

TABLE I. Summary of data obtained by least-squares fitting the time spectra of the x rays. Entries under "abundance" are fractional yields for that particular half-life.

Group	Source	Abundance (%)	Half-life (nsec)
Light	U^{235}	59	<4
		22	7
		29	75
	Pu^{239}	47	<4
		5	23
		48	100
Heavy	U^{235}	44	<2.5
		30	5
		26	30
	Pu^{239}	39	<2.5
		32	5
		29	40

TABLE II. Summary of data from the analysis of the x-ray energy spectra.

Group	Source	Yield (x rays/ fission)	Energy of most probable x ray (keV)	Z corresponding to this energy	Ref.
Light	U^{235}	0.17±0.02	16 ±1	40 ±2	This work
		0.10±0.03	16 ±1	40 ±1.5	13
		0.08[a]	16 ±1	40	14
	Pu^{239}	0.15±0.02	17 ±1	41.5±2	This work
Heavy	U^{235}	0.43±0.04	31.5±1	55.5±1	This work
		0.42±0.12	31 ±1.5	55 ±1.5	13
		0.12[a]	30.5±1	54.5	14
	Pu^{239}	0.26±0.03	31.5±1	55.5±1	This work

[a] Reference 14 gives the results for the sum of both the heavy- and light-group x rays as 0.20 x ray/fission. Hohmann's data suggest this as a reasonable division.

find the dominant characteristic half-lives. Components were then added to see if an improvement in χ^2 could be obtained. In general, when χ^2 was equal to the number of degrees of freedom the search was abandoned and the fit was considered to be statistically good. The parameters thus determined were used to construct a true time function [the $f(t)$ of Eq. (1)] which was then folded with the prompt response. The difference in total counts between the calculated and measured results was then added in the form of the prompt response. The solid lines in Figs. 4 through 7 are the result of this fitting procedure. Data thus collected are listed in Table I. Entries under abundance are fractional yields for that species. The half-lives must not be construed as being real, but rather as representing averages in a very complicated radioactive system.

The contribution of higher energy gammas to the prompt component can be estimated. The cross section of NaI(Tl) for Compton scattering at 200 keV (the peak energy[20] of the fission gamma distribution) and a 1-mm detector thickness combine to yield a total efficiency of about 5%. The Compton scattered electrons are distributed approximately equally in energy below the Compton edge. The number that fall within the acceptance window for the x rays is therefore less than 5% since the gamma yield is approximately the same as the x-ray yield.[12]

B. Energy Measurements

Figure 8 shows the energy spectrum for x rays for the fission of U^{235}. A similar curve for Pu^{239} is shown in Fig. 9. For this case the fast-time channels were discarded, the x-ray energy window was widened to include energies from 5 to 45 keV, and the output of the x-ray amplifier was connected to the multichannel analyzer.

The experimental curves approach zero in the regions below 10 keV and above 40 keV. The contribution of

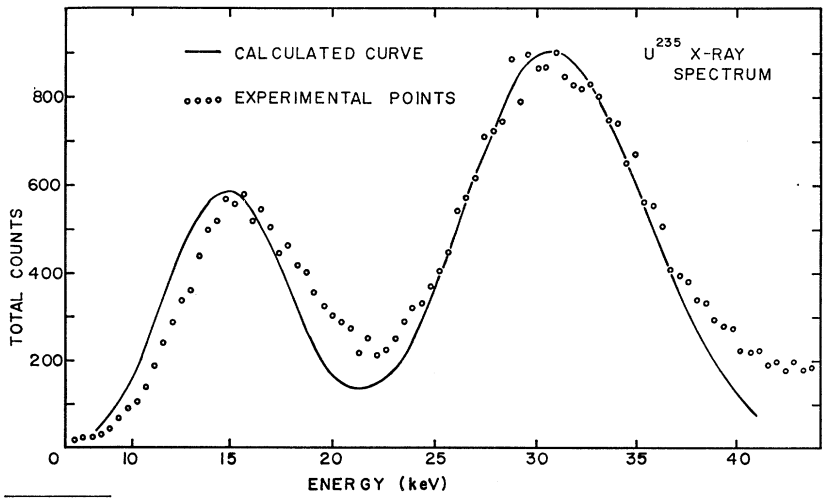

FIG. 8. X-ray energy spectrum for U^{235} (open circles). Solid line is the calculated spectrum using $M1$, $E2$ transitions at 100 keV.

[20] W. J. Price, *Nuclear Radiation Detection* (McGraw-Hill Book Company, Inc., New York, 1958).

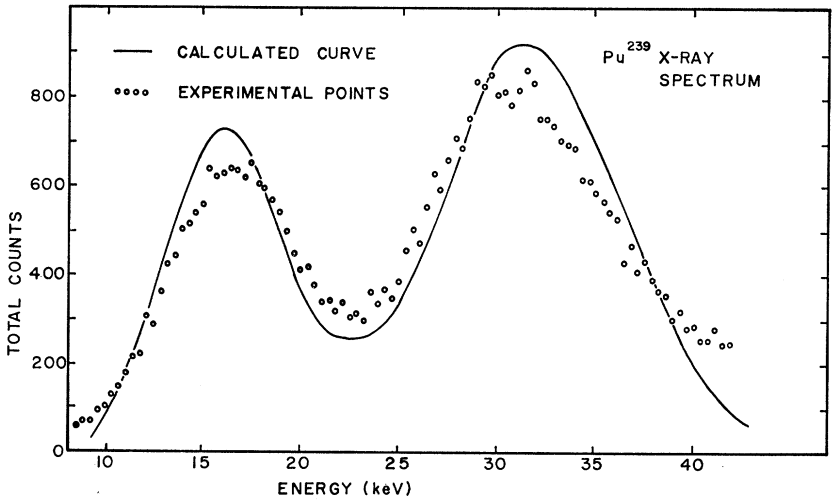

FIG. 9. X-ray energy spectrum for Pu^{239} fission fragments (open circles). Solid line is the calculated spectrum using $M1$, $E2$ transitions at 50 keV.

high-energy gammas to the x-ray energy spectrum can be estimated by using this characteristic. The accidental background was measured and found to be negligible. The coincident-background energy spectrum was assumed to have a linear functional form that intersects the measured curve at the limits stated above. The areas under the assumed curves were computed to be 10 and 12% when compared to the areas under the measured curves for the light-fragment groups for U^{235} and Pu^{239}, respectively. Similarly the background contributions to the heavy fragment were computed to be 15 and 25%. These figures are considered to be upper limits; the true numbers are probably considerably lower.

Data determined from the analysis of the energy spectra are collected in Table II. The area under each peak was determined, corrections for solid angles were made, and the result was normalized to the total number of fissions. Results from other experimenters are listed for comparison.

IV. CALCULATION OF ENERGY SPECTRA

Although x rays from adjacent Z's cannot be separated with the energy detector used in the present experiment, the qualitative shape of the energy spectra and the position of the peaks could be readily observed. Timing measurements indicate that internal conversion is responsible for a significant number of the x rays, and that the types of the nuclear transitions are probably $M1$ and $E2$. If it is assumed that each nuclear species contains the same number of excited states from which internal conversion can take place, and that the transition energy is some $\langle E\rangle_{av}$, the expected observable x-ray energy spectrum can be expressed by the following:

$$M(E)=K\sum_j n_j G(E,E_j)Y(E_j) \times[C_{E2}(\langle E\rangle_{av},E_j)+C_{M1}(\langle E\rangle_{av},E_j)],$$

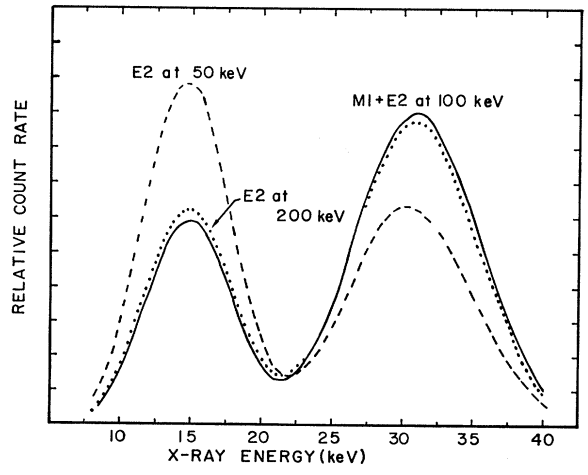

FIG. 10. Calculated x-ray energy spectra for fission fragments of U^{235} for various types and energies of nuclear transitions. Solid line fits measured data best.

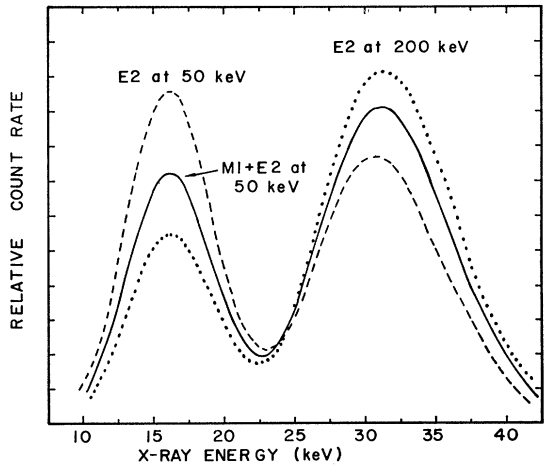

FIG. 11. Calculated x-ray energy for fission fragments of Pu^{239} for various types and energies of nuclear transitions. Solid line fits measured data best.

where K=normalizing constant, n_j=yield from fission of the jth Z value, G=Gaussian resolution function for the NaI(Tl), Y=fluorescent yield, and C=internal-conversion coefficients.

The n_j were obtained by equal charge displacement (ECD), sometimes referred to as "Glendenin's rule," from the mass distributions in Weinberg and Wigner[21] using an average chain length of 3 for U^{235} and 2.5 for Pu^{239}. Values for C were taken from Rose.[22]

The calculated spectra are presented as the solid lines in Figs. 8 and 9. It was found that agreement was best when the U^{235} data were fitted by assuming a mixture of 50% $M1$ and 50% $E2$ transitions at an average energy of 100 keV. Agreement was best when the Pu^{239} data were fitted with the same mixture but at an average energy of 50 keV. To illustrate the sensitivity of the calculated spectrum to the type and energy of transitions used in the calculation, the curves in Figs. 10 and 11 are shown.

[21] A. M. Weinberg and E. P. Wigner, *The Physical Theory of Neutron Chain Reactors* (University of Chicago Press, Chicago, 1958).

[22] M. E. Rose, *Internal Conversion Coefficients* (North-Holland Publishing Company, Amsterdam, 1958).

ACKNOWLEDGMENTS

The authors are grateful to G. P. Beck, Paul Hesselmann, and S. E. Boudreaux for assistance in the use of the University of Illinois reactor facilities; to Ken Watts for constructing the plastic scintillator; to G. Moscatti for the use of his version of the half-life fitting program; and to Argonne National Laboratory for the loan of the Pu^{239} foil.

PHYSICAL REVIEW VOLUME 145, NUMBER 3 20 MAY 1966

Lifetime of the 462-keV Level in Te^{125}†

F. R. METZGER AND R. S. RAGHAVAN*
Bartol Research Foundation of The Franklin Institute, Swarthmore, Pennsylvania
(Received 12 January 1966)

Resonance scattering from Te^{125} has been investigated with Sb^{125} as the source of the exciting gamma radiation. The centrifuge technique was used for the compensation of the recoil energy losses. Appreciable resonance scattering was observed only from the 462-keV $\frac{5}{2}^+$ level. With a branching ratio $\Gamma_0/\Gamma=0.24$, the observed scattering corresponds to a partial width $\Gamma_0=(8.5\pm0.9)\times10^{-6}$ eV for the 462-keV $E2$ ground-state transition. The mean life of the 462-keV level is thus $(1.9\pm0.3)\times10^{-11}$ sec. The absence of resonance scattering from Te^{125} levels reported at 633-, 640-, 652-, and 668-keV excitation energy is consistent with the assignment of spin values $\geq\frac{7}{2}$ to the first two of these states, an upper limit of 0.03% per Sb^{125} decay for the intensity of the 652-keV transition, and a lower limit of 10^{-11} sec for the mean life of the 668-keV ground-state transition.

I. INTRODUCTION

IN the course of a program aimed at the measurement of gamma-ray transition probabilities in odd-mass nuclei and at their comparison with the predictions of the pairing-plus-quadrupole-force model,[1] it was decided to study transitions in Te^{125}. The decay scheme of 2.7-year Sb^{125}, which populates at least eight excited states of Te^{125}, appeared to be well established.[2] It indicated ground-state transitions from states at 462-, 633-, and 668-keV excitation energy, thus making it feasible to perform resonance-fluorescence experiments. In addition to the study of the 462-keV level, which is most prominently populated in the decay of Sb^{125}, the investigation of the 633-keV excited state appeared to be of special interest because of difficulties encountered in the interpretation[2] of early Coulomb-excitation results[3] and the suggestion of a spin assignment of $\frac{3}{2}^+$ made on the basis of γ-γ angular-correlation data.[4]

After completion of our experiments, which succeeded only in exciting the 462-keV level, the results of a study utilizing oriented Sb^{125} nuclei and high-resolution Li-drifted Ge detectors became available.[5] The modifications in the decay scheme suggested by that study readily explain our failure to observe resonance scattering from the 633-, 652-, and 668-keV levels.

†Work supported by the U. S. Atomic Energy Commission. Support in the initial stages came from the National Science Foundation.

* Present address: Institut für Strahlen- und Kernphysik der Universitaet Bonn, Bonn, Germany.

[1] L. S. Kisslinger and R. A. Sorenson, Rev. Mod. Phys. **35**, 854 (1963).

[2] K. C. Mann, F. A. Payne, and R. P. Chaturvedi [Can. J. Phys. **42**, 1700 (1964)], where references to earlier work on the decay scheme of Sb^{125} are given.

[3] L. W. Fagg, E. A. Wolicki, R. O. Bondelid, K. L. Dunning, and S. Snyder, Phys. Rev. **100**, 1299 (1955).

[4] T. Inamura, T. Iwashita, Y. Ikemoto, and S. Kageyama, J. Phys. Soc. Japan **19**, 239 (1964).

[5] N. J. Stone, R. B. Frankel, J. J. Huntziker, and D. A. Shirley, Lawrence Radiation Laboratory Report UCRL-11828, 59, 1965 (unpublished).