AMERICAN JOURNAL OF PHYSICS VOLUME 37, NUMBER 6 JUNE 1969

Measurement of Neutron-Induced, Fission-Fragment Energy Spectra in the Advanced Laboratory*

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(Received 29 July 1968; revision received 2 December 1968)

Fission experiments can be profitably performed in the Advanced Laboratory. Measurement of the fragment energy spectrum from a fissile source using a surface barrier detector demonstrates several characteristics of the fission process. Experimental difficulties in such measurements are discussed, and typical results are presented for neutron-induced fission with sources of natural uranium and those enriched in ^{235}U. Also shown are the results with the spontaneous fissile source ^{252}Cf.

INTRODUCTION

Few other discoveries have had as large and as complicated an impact upon humanity as has fission. In two classic papers, the first published in January 1939, Hahn and Strassman[1,2] showed that when uranium is bombarded with neutrons there are formed radio elements with about half of the atomic number of uranium. Meitner and Frisch[3] correctly interpreted these findings as the division, or fission, of the excited uranium nucleus into two fragments of intermediate mass. Frisch[4] and independently Anderson *et al.*,[5] demonstrated the great amount of energy released. Booth, Dunning, and Slack[6] published an early plot of the energy distribution of uranium fission fragments obtained using an ion chamber–linear amplifier technique. Sparberg[7] and Graetzer[8] have published accounts of some of this work, and an earlier review was published by Turner.[9] A thorough review of all published information on fission up until 1963 is given in the work edited by Hyde.[10] An interesting series of invited papers at the 1967 Washington Meeting of the American Physical Society[11] discussed the early work of fission and some of its implications.

Although approximately 30 years have elapsed since the discovery of fission, and some 27 since the first self-sustained chain reaction of Chicago on 2 December 1942, fission remains a fruitful area of research. During the twenty-fifth anniversary year of the discovery of fission, 1964, more than 600 articles directly relating to the subject were in *Nuclear Science Abstracts*. Many more articles could have been cited that were indirectly a result of fission. It is thus of historical importance and current interest to devise fission experiments which can be done in the advanced laboratory. Not only are such experiments likely to be stimulating to the students, but they lead naturally to a study of current literature. Unlike many areas of physics, undergraduate students can read typical fission articles in research journals with profit. It is also hoped that such experiments will give students a sense of the excitement of a research frontier, and experience with current literature and techniques in nuclear physics.

I. EQUIPMENT

Although the early work cited previously involving the detection of fission fragments and the measurement of the energy released was done with ionization chambers, we have chosen to work with a semiconductor solid-state analog, the surface barrier detector. Measurement of fission spectra, while essentially similar to the measurement of the energy spectra of other charged

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* Supported in part by the National Science Foundation Undergraduate Research Participation Program.

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[1] O. Hahn and F. Strassmann, Naturwiss. **27**, 11 (1939).

[2] O. Hahn and F. Strassmann, Naturwiss. **27**, 89 (1939).

[3] L. Meitner and O. R. Frisch, Nature **143**, 239, 471 (1939).

[4] O. R. Frisch, Nature **143**, 276 (1939).

[5] H. L. Anderson, E. T. Booth, J. R. Dunning, E. Fermi, G. N. Glasoe, and F. G. Slack, Phys. Rev. **55**, 511 (1939).

[6] E. T. Booth, J. R. Dunning, and F. G. Slack, Phys. Rev. **55**, 981 (1939).

[7] Ester B. Sparberg, Amer. J. Phys. **32**, 2 (1964).

[8] Hans G. Graetzer, Amer. J. Phys. **32**, 9 (1964).

[9] L. A. Turner, Rev. Mod. Phys. **12**, 1 (1940).

[10] Earl K. Hyde, *Nuclear Properties of the Heavy Elements, Fission Phenomena* (Prentice-Hall, Inc., Englewood Cliffs, N. J., 1964).

[11] Bull. Amer. Phys. Soc. **12**, 567 (1967).

particles, requires certain different instrumental characteristics and techniques. We have attempted to present sufficient information so that the nonspecialist can perform interesting fission experiments, the sophistication of which will largely depend upon the instrumentation available.

Silicon surface barrier detectors are essentially reversed-biased silicon *p–n* junctions of large area and with thin entrance windows. The latter is usually an evaporated gold layer on the surface of the detector approximately 200 Å thick. The reader is referred to the current literature and manufacturer's manuals for detailed discussions of these devices, and only a few references[12–14] of the many available are cited. A surface barrier detector has a sensitive depth or depletion region, which depends upon the amount of reversed bias voltage applied. For typical detectors, bias voltages of 10–100 V will provide a depletion region deep enough for alpha particles with energies up to 10 MeV, and fission fragments with energies of more than 100 MeV. A charged particle entering the depletion region creates free electron–hole pairs in the silicon. Approximately 3.5 eV is required for each electron–hole pair formed, and the collection of these electrons at the positive terminal of the device gives a quantity of negative charge which is a measure of the energy deposited in the depletion region. If the depletion region is sufficiently thick that the particle loses all of its energy within that region, this charge is a measure of the total kinetic energy of the particle.

Since it is the charge collection that is important in the measurement of energy, it is convenient to use charge-sensitive preamplifiers with semiconductor detectors. The amplifier system used was a commercial version of the Oak Ridge National Laboratory Q-2069C amplifier system designed by Fairstein and Blankenship.[15] The complete electronic system used is shown in block diagram form in Fig. 1. This system is convenient, but more elaborate and expensive than necessary for the fission experiments described. Virtually any charge-sensitive preamplifier and pulse amplifier system could have been used. A dc voltmeter is used to monitor the detector bias voltage, and an ac vacuum-tube voltmeter monitors the detector noise. The oscilloscope is used to view the pulses sent to the pulse–height analyzer. These latter instruments, while not necessary, are particularly helpful in experiments involving long counting times as they permit a check of the noise level of the detector and routine checks of the system while the experiment is in progress.

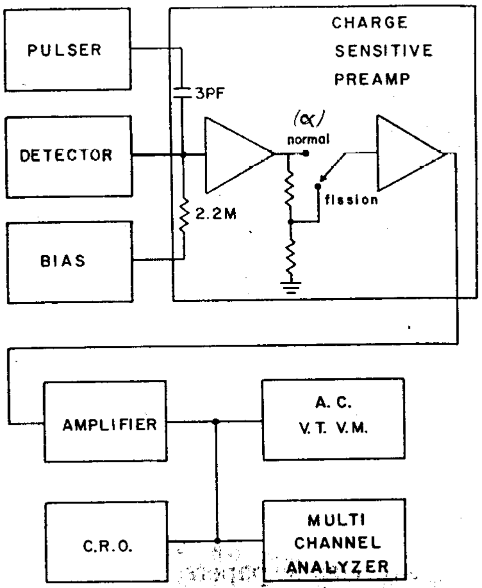

FIG. 1. Block diagram of electronic system used.

Since the charge-sensitive preamplifier used was specifically designed for alpha particle spectroscopy, it had more gain than desired for fission pulses, and was modified by the installation of a gain switch. This allowed the gain to be appropriate for alpha particle work in the "normal" position, but reduced it by a factor of about 30 in the "fission" position. (This gain reduction must be made sufficiently early in the pulse amplification system so that the large fission pulses do not cause saturation and a nonlinear response in later stages of the amplifier.)

Owing to the light-sensitive characteristics of silicon surface barrier detectors and the short range of fission fragments in air, it is desirable to use a metal vacuum chamber. It is also convenient in the calibration process and in the neutron-

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[12] G. Dearnaley and C. D. Northrop, *Semi Conductor Counters for Nuclear Radiations* (John Wiley & Sons, Inc., New York, 1963).

[13] F. S. Goulding, Nucleon. **22**, 54 (1964).

[14] Ralph T. Overman, Laboratory Manual A, (1967) ORTEC Inc., 101 Midland Road, Oak Ridge, Tennessee 37830.

[15] Model 240 Amplifier System (ORNL-Q-2069C) (Fairport Instruments, Inc., 270 Midway Lane, Oak Ridge, Tennessee 37830).

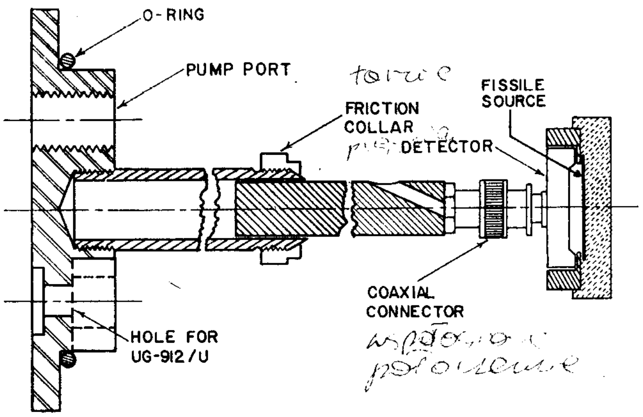

FIG. 2. Fissile source holder which is inserted into the horizontal port of a neutron howitzer and evacuated with an ordinary fore pump.

induced fission experiments, if the same chamber and mount are used for all experiments. Figure 2 shows the probe assembly used in these experiments. It was inserted into a horizontal port of a neutron howitzer,[16] and the port evacuated with an ordinary fore pump. The vacuum seal was made with an "O" ring. While the particular design was for the locally available howitzer, it is believed the idea could be adapted to other types, or a special vacuum chamber constructed if a particular howitzer port will not support a vacuum. It would probably have been better for the detectors to use an LN_2 or other cold trap, but the long counting times necessary made this inconvenient, and no traps were used. The neutron source was removed from the howitzer during the calibration and ^{252}Cf spontaneous fission portions of the experiment.

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II. DETECTOR CALIBRATION

For a given energy particle, the amount of charge collected from within the depletion region (and hence the pulse height from the amplifier) will depend to some extent upon the bias voltage used. Figure 3 shows the pulse–height spectrum resulting from the spontaneous fission of ^{252}Cf as a function of bias voltage. The detector is an ordinary surface barrier detector (i.e., not one specifically manufactured for fission work). Note the shift to larger pulse heights for higher bias voltages. At first the peak-to-valley ratio increases due to improved charge collection, but soon becomes worse because of charge multiplication effects

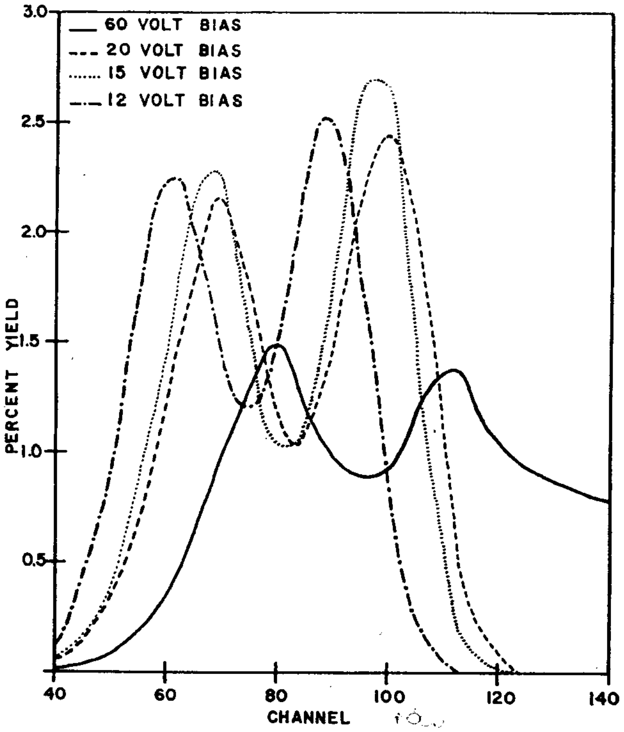

FIG. 3. Spontaneous fission pulse-height spectra of ^{252}Cf taken with an ordinary surface barrier detector at various bias voltages.

within the detector. If such ordinary surface barrier detectors are to be used, it would be helpful if the optimum bias voltage could be determined with a thin ^{252}Cf or ^{235}U source. At least one company now manufactures detectors[17] specifically designed for fission and heavy ion work, and these do not show charge multiplication effects. Figure 4, again a ^{252}Cf spectrum, was taken with a preproduction detector of this type.

All spontaneous or slow-neutron fission spectra show the same general double-peaked shape. Since momentum must be conserved in the fission process, and the majority of the mass is contained in two fragments, these must be oppositely directed. In a noncoincidence experiment with one detector, only one of these fragments can be detected from any particular event. Since in an ideal detector there is an equal probability of detecting the light and the heavy fragments, the characteristic double peak is observed, the lighter fragments having the larger energies. Symmetric fission is rare for both spontaneous and

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[16] Model NR-2, U. S. Nuclear Corp., P. O. Box 208, Burbank, California.

[17] ORTEC 7900 Series Heavy Ion Detectors (Oak Ridge Technical Enterprises Corp., P. O. Box C, Oak Ridge, Tennessee 37831).

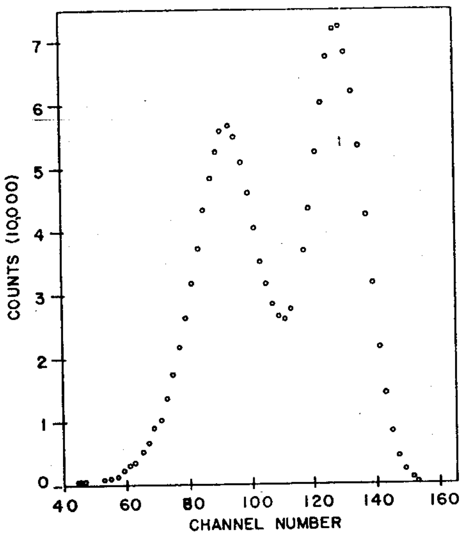

FIG. 4. Spontaneous-fission pulse-height spectrum of ^{252}Cf taken with an ORTEC heavy ion detector at 85-V bias.

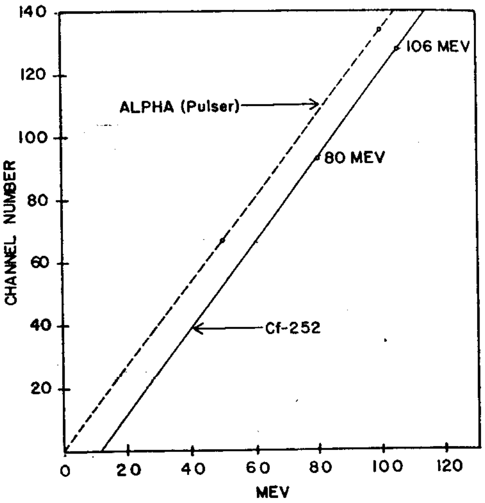

FIG. 5. Approximate ORTEC heavy-ion detector calibration plot showing the alpha calibrated pulser line and the more correct energy vs pulse-height calibration obtained using Whetstone's (Ref. 18) values of 80.01 and 105.71 MeV for the most probable energies of the heavy and light fragment groups, and the pulse-height spectrum of Fig. 4.

slow neutron fission, but becomes much more probable in high-energy fission. Since the fragments are always produced in pairs, the total count or area under the two peaks should be approximately equal. In experimental situations, there are small differences due to the finite thickness of the source and detector window, as the fragment range is a function of the fragment energy, mass, and charge. This results in a relative loss of some of the heavy fragments as far as the detection process is concerned.

Unfortunately, precise energy calibration of the detector is complicated by an effect dependent upon the mass of the fragment involved, as shown by Williams, Kiker, and Schmitt.[18] They showed that each fragment mass number has a linear pulse-height to energy relationship, but the slope and intercept are slightly different for each mass number. Research by Moak, Dabbs, and Walker[19] shows that this effect is related to the direction the detected particle enters the silicon detector relative to the 110 crystal axis. For the purposes of this experiment, these relatively minor differences will be ignored.

To a first approximation one can assume a detector to behave in the same manner for fission fragments as it does for alpha particles. The response to alpha particles can be determined experimentally and the results extrapolated upward to the higher energies resulting from fission. Figure 5 shows such an approximate calibration plot. The "equivalent alpha" line is the one based upon this same response assumption. The equivalent alpha line was obtained using the 6.112-MeV alphas from ^{252}Cf. Obviously, any available, thin, alpha source could be used if the alpha energies are easily distinguishable. To accomplish this calibration, the "pulse-height" dial was set to the alpha energy, 6.112 units. The "normalize" dial was then adjusted to give a pulser peak at precisely the same spectral position as the alpha source under the identical bias conditions. These measurements are best made with the preamplifier gain switch in the "normal" position to achieve the best signal-to-noise ratio. This switch is then placed in the low-gain or fission position, and precision attenuators on the pulser are switched to multiply the pulser pulse height by a factor of 20. The pulse-height dial now spans linearly the alpha-particle charge input equivalent of 0–200 MeV, and the equivalent alpha line is experimentally determined by taking the pulse-height spectra for two or more pulser dial settings.

An improved detector-calibration technique can be accomplished using a ^{252}Cf fissile source and the results of Whetstone's[20] time-of-flight measure-

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[18] C. W. Williams, W. E. Kiker, and H. W. Schmitt, Rev. Sci. Instrum. **35**, 11116 (1964).

[19] C. D. Moak, W. T. Dabbs, and W. W. Walker, Bull. Amer. Phys. Soc. **11**, 101 (1966).

[20] S. L. Whetstone, Jr., Phys. Rev. **131**, 1232 (1963).

ments, which give the most probable energy of the heavy fragment group for ^{252}Cf fission as $\langle E_H \rangle =$ 80.01 MeV, and $\langle E_L \rangle = 105.71$ MeV for the light group. For this improved calibration we will assume the quoted values of $\langle E_H \rangle$ and $\langle E_L \rangle$, and that the detector response is linear and independent of fragment mass. These results are also presented in Fig. 5. Note the presence of the pulse–height defect in the calibration, only a portion of which is attributed to energy losses in the detector window. Also note that the fission-fragment calibration line is approximately parallel to the equivalent alpha line. In the absence of a thin ^{252}Cf or ^{235}U source, a better approximation than the alpha line could be constructed by drawing a line parallel to that line, but with an X axis intercept of about 12 MeV to account for the total pulse–height defect of a typical detector.

III. NEUTRON-INDUCED FISSION

Figure 6 shows the results using the simplest and cheapest sample available, an ordinary uranium metal disk. The disk was placed as close as practical to the detector without touching the sensitive surface. This sample was then infinitely thick as far as the range of the fission fragments were concerned. Since the fissions with slow neutrons are due to the ^{235}U present, the count rate is slow with the 5 Ci Pu–Be neutron source used in these experiments. However, an overnight count is sufficient to show the general features of the spectrum. The neutron source can be removed to show the zero background of the high-amplitude fission pulses, and the necessity of uranium can be demonstrated by substitution of other metal disks. Thus, it is relatively easy to demonstrate the existence of fission, and to show the maximum energy given the light fragments to be approximately 115 MeV.

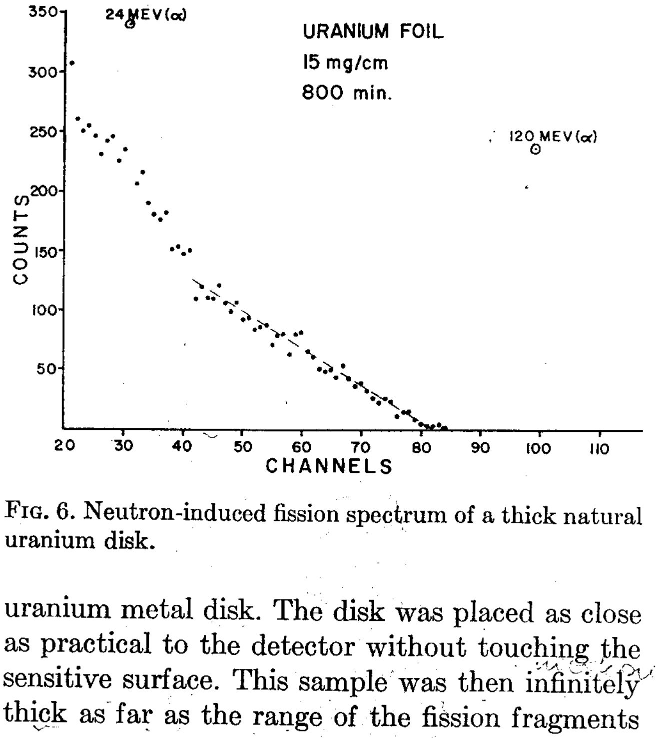

FIG. 6. Neutron-induced fission spectrum of a thick natural uranium disk.

No double hump is apparent in Fig. 6. Redmond, Klongersmith, and Anno,[21] and Kahn, Harman, and Forgue[22] have shown that this is to be expected for thick sources. Figure 7 is adapted from this work, and shows the predicted energy spectra for ^{235}U sources of different thickness. The extrapolation to the maximum energy is compatible with their results.

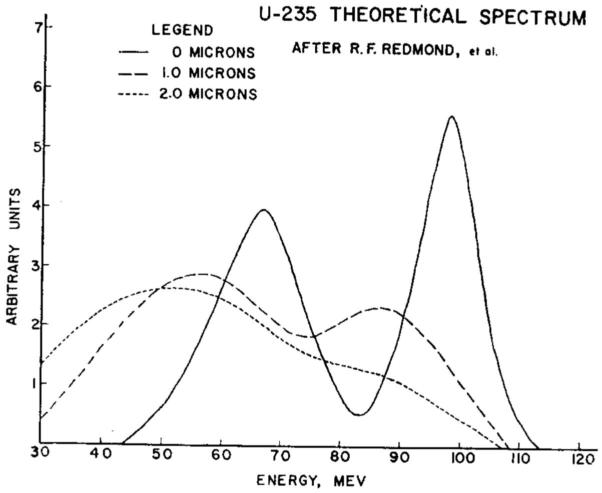

FIG. 7. Theoretical fission energy spectra for sources of various thicknesses. Adapted from Redmond *et al.*[21]

Figures 8, 9, and 10 show the results using progressively thicker "thin" sources of uranium enriched in ^{235}U. While it is possible to buy the ^{235}U nitrate solution and prepare fissile sources using the method described by Redmond, Klongersmith, and Anno,[21] this is difficult and probably not worth the trouble if they can be obtained some other way. The enriched uranium-dioxide films used in this experiment were obtained from Kahn[23] and probably could be made commercially available for approximately \$50–\$150 each, de-

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[21] R. F. Redmond, R. W. Klongensmith, and J. N. Anno, J. Appl. Phys. **33**, 3383 (1962).

[22] Steve Kahn, Randall Harman, and Vernon Forgue, Nucl. Sci. Engineering **23**, 8 (1965).

[23] Steve Kahn, Aerojet-General Nucleonics, San Ramon California, private communication.

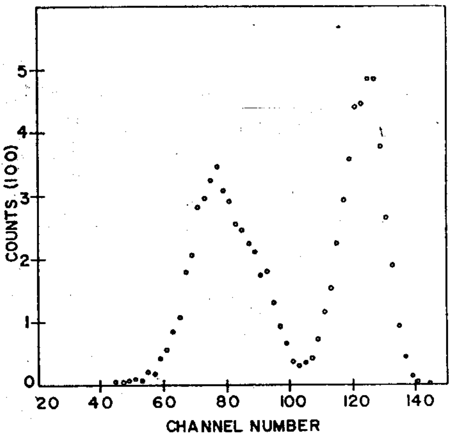

FIG. 8. Neutron-induced fission spectra for $^{235}UO_2$ source of thickness 0.029 μ. Approximately a two-day count.

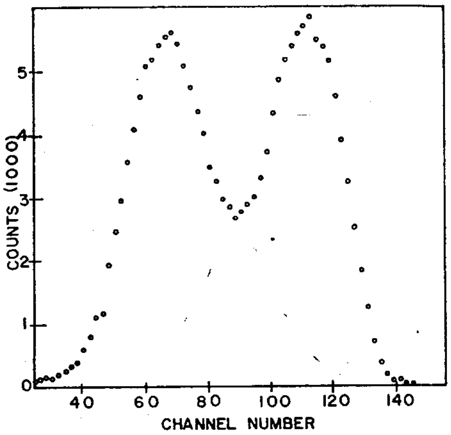

FIG. 10. Neutron-induced fission spectra for $^{235}UO_2$ source of thickness 0.71 μ. Approximately a two-day count.

pending upon the thickness. These sources are prepared by vacuum vapor deposition on steel substrates. The substrates were 1.5-cm-diam 304 L–stainless-steel disks and were masked to provide a centered 1.27±0.013 cm deposition surface. The precision of the the thickness values quoted is ±4%.

IV. RESULTS

It is possible to deduce something about the mass yield from these data. Certainly the initial momentum of the nucleus and thermal neutron can be neglected in most calculations. If we also neglect the momentum of any prompt neutrons, we can write from momentum conservation

$$M_H V_H = M_L V_L. \quad (1)$$

Since the mass of the fragments is high, even though the energies seem large the fragments are nonrelativistic, with velocities of the order of $(0.8\text{–}1.6) \times 10^9$ cm/sec. This gives values of $\beta = v/c$ between 0.027 and 0.053. Thus, we can neglect relativistic effects. Squaring (1), dividing by 2, and rearranging, we have the approximate relation

$$M_L E_L = M_H E_H. \quad (2)$$

Since the mass number of the fissile source is known, using the same assumptions as before, we have

$$M_L + M_H = 236 \quad (3)$$

for the $^{235}U + n$ reaction. These relations, along with the measurement of the most probable fragment energies from Figs. 8–10 can be used to obtain the most probable mass ratio. Thus, since $M_H/M_L = E_L/E_H$ for all values,

$$\text{Most probable mass ratio} = \langle M_H \rangle / \langle M_L \rangle,$$
$$= \langle E_L \rangle / \langle E_H \rangle. \quad (4)$$

Equations (3) and (4) can then be combined to give the most probable light-fragment mass $\langle M_L \rangle$ and heavy-fragment mass $\langle M_H \rangle$. The equivalent of Eq. (3) for the ^{252}Cf case is

$$M_L + M_H = 252. \quad (5)$$

The most probable total fragment kinetic energy can be calculated by merely adding the values of $\langle E_L \rangle$ and $\langle E_H \rangle$, or

$$\langle E_{\text{Kin}} \rangle = \langle E_L \rangle + \langle E_H \rangle. \quad (6)$$

Table I shows the relative number of counts recorded in each peak for Figs. 6–10. The peak for

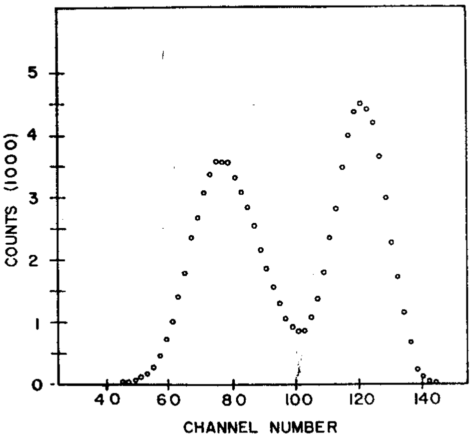

FIG. 9. Neutron-induced fission spectra for $^{235}UO_2$ source of thickness 0.42 μ. Approximately a two-day count.

TABLE I. Comparison of integrated counts in heavy and light ion peaks.

Source	Source thickness (μ)	Counts in heavy ion peak	Counts in light ion peak	Approx. counting time (h)	counts/h
^{252}Cf	···	1 556 000	1 611 000	···	···
^{235}U	0.029	8344	8259	48	11 927
^{235}U	0.42	96 974	94 435	41	11 115
^{235}U	0.71	147 018	174 808	40	11 332

TABLE II. Experimental results.

Fissile source	Thickness μ	Most probable energies (MeV) $\langle E_L \rangle$	$\langle E_H \rangle$	$\langle E_{\mathrm{Kin}} \rangle$	Most probable masses (amu) $\langle M_H \rangle / \langle M_L \rangle$	$\langle M_H \rangle$	$\langle M_L \rangle$	Ref. or Fig.
^{252}Cf	···	105.71	80.01	185.7	1.334	143.61	108.39	Whetstone[a]
^{235}U	···	99.4	68.2	176.68	1.46	140.07	95.93	Milton and Fraser[b]
^{235}U	0.029	102	69	171	1.48	141	95	Fig. 8
^{235}U	0.42	101	69	170	1.49	141	95	Fig. 9
^{235}U	0.71	94	61	155	1.54	143	93	Fig. 10

[a] Reference 20.

[b] Reference 24.

the light fragment was assumed to be symmetrical in order to obtain the shapes in the region of overlap.

For each source the relative counts are nearly the same for both the light and heavy peaks. Where there is a statistical difference, the total count in the heavy peak is generally slightly smaller. The decrease in the relative counts in the heavy ion peak results from the short range of the fission fragments in matter, with the heavy fragments having a larger energy loss in the source and detector window. This is particularly noticeable for the case of the 0.71-μ ^{235}U source. The number of fission events per unit time is also shown to be proportional to the amount of ^{235}U present as shown in the last column.

In Table II we have tabulated the measured values of $\langle E_L \rangle$, $\langle E_H \rangle$ and the calculated values of $\cdots\langle E_{\mathrm{Kin}} \rangle$, $\langle M_H \rangle / \langle M_L \rangle$, $\langle M_H \rangle$, $\langle M_L \rangle\cdots$for the various sources used in this experiment. The energies quoted for ^{252}Cf were used as the basis of the calibration line, and the ^{235}U results are due to Milton and Fraser.[24] Considering the simplicity of the calibration techniques and the low flux, the results of this experiment compare very reasonably with experimental values from the literature.

If only a single $^{235}UO_2$ source is to be used, a source thickness of approximately 0.4 μ makes a satisfactory compromise between good energy measurements, reasonable counting times, and a clean double-humped spectrum with a fairly high peak-to-valley ratio.

The Atomic Energy Commission has made available at no charge some plutonium samples which are a mixture of ^{238}Pu and ^{239}Pu. These are thin sources, with alpha activities of about 2×10^4 dpm. Figure 11 shows a spectrum obtained with one of these sources for a seven-day counting period. Obviously, there is not sufficient plutonium in these sources to permit their use for fission-frag-

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[24] J. C. D. Milton and J. S. Fraser, Can. J. Phys. **40**, 1626 (1962).

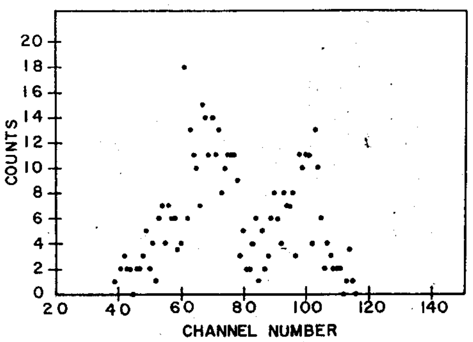

FIG. 11. Pulse-height spectrum for one of the plutonium sources obtained from the Atomic Energy Commission. A seven-day count.

ment energy measurements with reasonable counting times, but they can be used to show the existence of fission in plutonium. It may be possible to obtain these sources without the Special Nuclear Materials License required for the possession of the Pu–Be neutron source, or ^{235}U.

As there is a major program underway to produce sizeable quantities of the transuranic elements using the Oak Ridge HFIR reactor, it is expected that ^{252}Cf and other sources will soon be more readily available and less expensive. The simplest fission experiments are done with such spontaneous fissile sources. It should be noted however that sources such as ^{252}Cf require an AEC Byproduct Materials License.

These are not the only experiments which could be carried out with the equipment described. Students of unusual ability may wish to attempt a measurement of the fission cross section in ^{235}U, or investigate the energy losses suffered by fission fragments in passing through matter. Such experiments are considerably more difficult than those described, and were not attempted. Research articles have been presented in several cases where such work is reported.

It would also be very interesting to experimentally obtain the total mass yield as a function of mass number. However, this is not possible without using coincidence techniques. This is a result of the fact that the total fragment kinetic energy is not constant, but depends strongly on the heavy-fragment mass number. In the case of thermal neutron fission of ^{235}U, this total kinetic energy varies from approximately 176 MeV for a heavy-mass number of 130, to 152 MeV for a heavy-mass number of 150. Tsukuda[25] and Hogg and Lokan[26] have described analog circuits for obtaining the mass yield from a coincidence measurement of the two fragment energies. These measurements are based upon the solution of Eqs. (2) and (3) for M_H or M_L giving the approximate relations.

$$M_H=236E_L/(E_H+E_L);\quad M_L=236E_H/(E_H+E_L). \qquad (7)$$

While such an experiment would be interesting, it is believed that the additional cost and complexity of the equipment, as well as the necessity for thin and therefore fragile ^{235}UO$_2$ sources, make this impractical.

In case the facilities for experiments using semiconductor devices are not available, fission experiments of the type described by Price and Walker[27–29] Fleischer *et al.*,[30] and Cieslak, Piekarz, and Zakrzewski[31] might be possible and interesting. A fissile source close to certain material, such as cleaved mica, will leave tracks in the mica which can be observed with an electron microscope. If the mica is subjected to attack by a 20% solution of HF, these tracks can be enlarged to the point that they are visible under an optical microscope. A variety of experiments could be done with this technique, using a neutron source and various fissile sources.

It is believed that experiments of the type described are exciting to the students, illustrate current techniques in nuclear physics, and are sufficiently fundamental to be worth doing. After all, there are not many 100 MeV experiments you can do in a typical advanced laboratory.

ACKNOWLEDGMENTS

We wish to thank Dr. John Neiler of the Oak Ridge Technical Enterprises Corporation for the

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[25] M. Tsukuda, Nucl. Instr. Meth. **25,** 265 (1965).

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[27] P. B. Price and R. M. Walker, Nature **196,** 732 (1962).

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[30] R. L. Fleischer, P. B. Price, G. M. Symes, and D. S. Miller, Science **143,** 349 (1964).

[31] E. Cieslak, J. Piekarz, and J. Zakrzewski, Nucl. Instrum. Meth. **39,** 224 (1966).