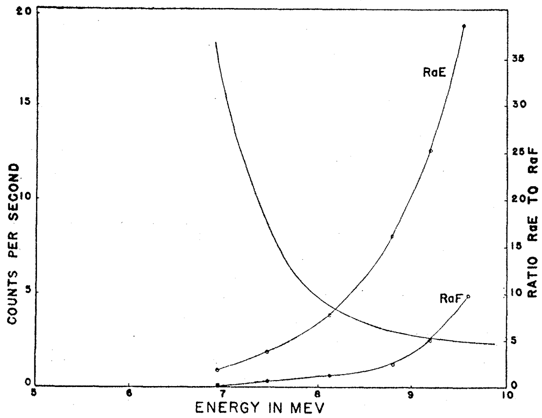

FIG. 3. Activity of targets for various energies of bombarding particles.

number (N_0) of radium E atoms formed. Thus

$$N_0 = N \Big/ \frac{\lambda_1}{\lambda_2 - \lambda_1}[e^{-\lambda_1 t} - e^{-\lambda_2 t}],$$

where λ_1 and λ_2 are the decay constants for the parent and daughter substances, respectively. The maximum activity is attained twenty-five days after bombardment.

The results obtained from the bismuth targets bombarded by deuterons of various energies are shown collectively in Fig. 3. A practical threshold appears for the formation of radium F at about 6.5 Mev. The ratio of the radium E to radium F is shown by the scale on the right side of the figure. At 10 Mev the yield of radium E is still almost five times that of radium F. If the deuteron in every case entered the nucleus then this ratio should be slightly less than unity since it should be easier for the neutron to leave than for the proton. It thus seems that at these energies the deuteron has a low probability of entering the nucleus as a whole. It must obey an Oppenheimer-Phillips[2] process, in which the deuteron in the external field of the nucleus yields a neutron and a proton and only the neutron enters. The entrance barrier for the deuteron must be considerably above 10 Mev. The cross section (σ) for each process may be calculated. At 10 Mev these values are 9×10^{-29} cm^2 and 4×10^{-28} cm^2 for radium F and radium E, respectively.

This investigation is made possible by the support of the Horace H. Rackham Fund.

[2] J. R. Oppenheimer and M. Phillips, Phys. Rev. **48**, 500 (1935).

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The Distribution in Energy of the Fragments from Uranium Fission

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The ionization produced by the fragments from uranium fission was measured. The number-energy curve shows two peaks corresponding to 65 and 98 Mev. The ionization produced by the two fragments simultaneously was measured by using a thin foil coated with U. The number-energy curve has one peak at 159 Mev and a half-width of 30 Mev.

I. INTRODUCTION

SHORTLY after the discovery of the fission of uranium by Hahn and Strassmann[1] a number of experiments[2–5] were performed in which the kinetic energy liberated in the process was measured. The method used in these experiments was the comparison of the ionization produced by the single fission fragments with that produced by α-particles of known energy. The agreement between these experiments was not very good. In each of the above experiments at least two maxima in the number-ionization curve were found, and the sum of the energies corresponding to these maxima was about 125 Mev according to v. Droste, Haxel, and Booth,

[1] O. Hahn and F. Strassmann, Naturwiss. **27**, 11 (1939).
[2] W. Jentschke and F. Prankl, Naturwiss. **27**, 134 (1939).
[3] G. v. Droste, Naturwiss. **27**, 198 (1939).
[4] O. Haxel, Zeits. f. Physik **112**, 681 (1939).
[5] E. T. Booth, J. R. Dunning and F. G. Slack, Phys. Rev. **55**, 981 (1939).

Dunning, and Slack, and 160 Mev according to Jentschke and Prankl.

Recently more direct experiments on the average energy liberated in the fission process have been made by Henderson,[6] who measured the heat evolved in a sample of U_3O_8 by a known number of fissions. His preliminary value is 175 Mev. This result is in good agreement with that of Jentschke and Prankl, and also with the theoretical predictions of Bohr and Wheeler.[7]

The experiments here described were undertaken in order to measure directly the total ionization produced in a single fission process and to attempt to reduce the discrepancy among some of the earlier measurements.

II. Experimental

The measurement of the ionization produced by the fission fragments was made with the ionization chamber shown in Fig. 1. It was filled with air at atmospheric pressure. The chamber consisted of a brass pillbox 12 cm in diameter and 4.5 cm deep, which had in its median plane a brass electrode which supported the U samples. The dimensions were chosen so that the fragments of longest range[8] (2.2 cm) had their entire path in the chamber. The pillbox served as the high voltage electrode and was connected to a rectifier which supplied 5000 volts. The central electrode served as collector and was connected to the linear amplifier. In order to reduce electrical disturbances the collector lead was surrounded by a grounded shield where it passes through the high voltage electrode. The linear amplifier used had six high gain stages and a power stage. Strong negative feedback was used in all stages except the first, with the result that the gain of the amplifier remained constant within 1 percent over times of several days. The pulses were recorded photographically by means of a torsion type mechanical oscillograph of the type described by Dunning,[9] which has a natural frequency of 1600 cycles/sec. and a sensitivity of 0.7 ma/mm at 1 m.

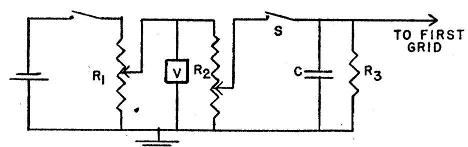

FIG. 2. Circuit of artificial pulse generator. $R_1=1000$-ohm w.w. potentiometer, $R_2=1600$ ohms tapped at 400, 800, 1200 ohms, $R_3=1$ megohm, $C=0.01$ μf, $S=$micro-switch.

In order to standardize the amplifier and oscillograph over the large range required for an extrapolation from α-particle to fission energies the artificial pulse generator whose circuit is shown in Fig. 2 was built. The action of the circuit is as follows: A known voltage, which can be varied by means of the voltage divider R_1 and the tapped voltage divider R_2, is impressed on the condenser C by closing the switch S. The voltage across C is transmitted to the grid of the first amplifier tube through a small, lucite-insulated condenser permanently mounted in the amplifier box. The successful action of the generator depends on sharp positive action of S. A micro-switch[10] was found to give excellent results. The pulses transmitted through the amplifier had wave forms very similar to those produced by ionizing particles in the chamber. Complete calibrations from this instrument were made at several points on each record.

The energy calibration was made by comparing the artificial pulses with the pulses produced by introducing into the chamber long range α-particles from ThC′. For this purpose the U

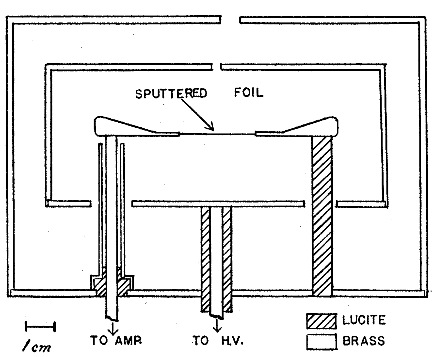

FIG. 1. Schematic diagram of the ionization chamber.

[6] M. C. Henderson, Phys. Rev. **56**, 703 (1939).
[7] N. Bohr and J. A. Wheeler, Phys. Rev. **56**, 426 (1939).
[8] E. T. Booth, J. R. Dunning and G. N. Glasoe, Phys. Rev. **55**, 982 (1939).
[9] J. R. Dunning, Rev. Sci. Inst. **5**, 387 (1934).
[10] Manufactured by Micro-Switch Corporation, Freeport, Illinois.

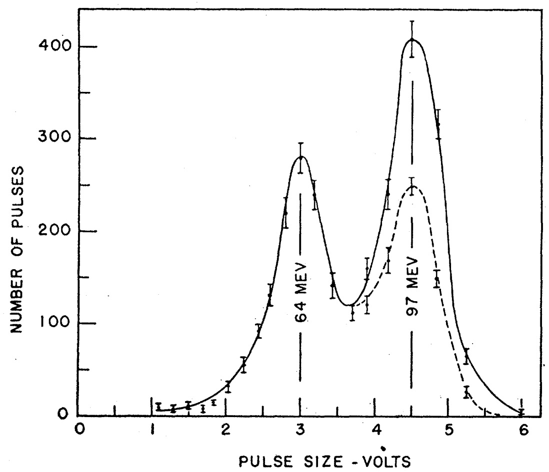

FIG. 3. Number-ionization curve for single fission fragments.

sample and its support were removed from the chamber, and replaced by a solid brass collector having identical dimensions. The α-particles were shot into the chamber through a diaphragm and the holes shown in Fig. 1, at an angle of 60° with the plane of the collector. The portion of the range spent in the chamber was defined by the distance between the top of the high voltage electrode and the collector. The calibrations were made from two different portions of the α-particle range. The values found were the same within 1 percent and yielded the result that a one-volt standard pulse corresponded to 21.4 Mev. Calibrations made before and after the fission experiments gave the same result. All fission results were measured in terms of standard pulses and converted into energy units by the above correspondence.

In order to check the validity of this method of extrapolation to very high energies the reduction in gain of the amplifier, made in order to reduce the size of the fission pulses, was found by measurement of the ratios of the voltage dividers used as attenuators. The attenuation factor thus found agreed with the ratio of the standard pulses necessary to produce equal oscillograph deflections at each gain setting within 2 percent. We believe, however, that the standard pulse method is more accurate than the attenuation method, since the latter involves some assumptions concerning the linearity of the amplifier.

The U samples used were of two kinds. For the observation of single fission fragments both sides of a sheet of 1 mm Al were covered with thin, quite uniform[11] layers of U metal by cathode sputtering.[12] The Al sheet was then mounted between two recessed brass sheets to form the collector of the ionization chamber. The brass pieces each had a 3.3-cm hole in the center, so that an area of about 8 cm^2 of U-covered Al was exposed on each side. The surface density of the U was determined by counting the α-particles; its value was 0.12 mg/cm^2, corresponding roughly to a stopping power of 0.3 mm of air.

The simultaneous observation of the ionization due to both fission fragments requires that the U sample be sputtered on a foil of stopping power small compared with the sum of the ranges of the two fragments (about 3.7 cm). This requirement was met by using Al foil of 0.18 mg/cm^2 (stopping power 1.1 mm) as a support for the U layer. Attempts to use this foil unsupported were unsuccessful because of its extreme fragility. The method of support which proved satisfactory was the following. Sixty No. 33 holes spaced on 0.15-inch centers in a hexagon pattern were punched in a sheet of 2 mil Al with a special die. The 0.18-mg/cm^2 Al foil was mounted on this with thin shellac, to form a composite foil, which was then sputtered with U. During the sputtering process the composite foil was covered with a 1-mm Al sheet in which was punched a hole pattern identical with the one in the 2-mil sheet. The patterns were made to coincide by means of 3 locating holes in each sheet. In this way U was allowed to fall only on the thin parts of the composite foil, i.e., those not covered by the 2-mil pierced sheet. Foils made in this way were quite strong mechanically and could be easily handled. They were mounted between the brass plates mentioned above to form the collector of the ionization chamber. The surface density of U was

[11] Estimated from the uniformity of the interference color over the surface.

[12] We are indebted to Drs. Rentschler and Marden of the Westinghouse Company for furnishing the uranium rods used in the sputtering process. A description of the sputtering technique by C. C. Van Voorhis has appeared in Rev. Sci. Inst. 11, 77 (1940).

determined in the same way as before and had the same value, 0.12 mg/cm^2.

The ionization chamber was put below a D_2O ice target which was bombarded by 50–100 μa of 350-kv deuteron molecules from th treansformer-rectifier set in this laboratory. On account of the geometry of the target chamber, larger fission yields could be secured by not slowing down the 3-Mev neutrons from the D – D reaction, in spite of the smaller value of the cross section at this energy. Due to the large number of recoil nuclei in the ionization chamber, a considerable statistical ionization background was present during the measurements.

III. Results

The results of several sets of observations of the single fragments are plotted in Fig. 3. In this plot the abscissae are the standard pulse voltages corresponding to the deflection intervals on the photographic records, and the ordinates are the numbers of pulses ending in these intervals. This method of plotting provides a linear energy scale, but, on account of the fact that the recording oscillograph characteristic becomes somewhat nonlinear for large deflections, it distorts the vertical scale. For large deflections the response of the oscillograph falls off, thus compressing a larger energy interval into the standard deflection interval (1 mm) used in measuring the records. This results in fewer plotted points (with correspondingly larger ordinates) per energy interval. A correction is made for this effect by dividing the ordinates of the points by numbers proportional to the size of the associated energy intervals. The corrected curve is shown as a dashed line in Fig. 3. It is seen from Fig. 3 that there are two peaks, at 3

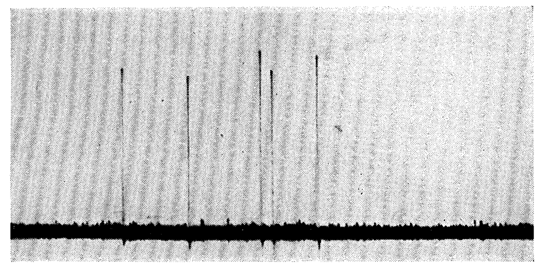

Fig. 5. Record showing 150-Mev pulses from pairs of fission fragments.

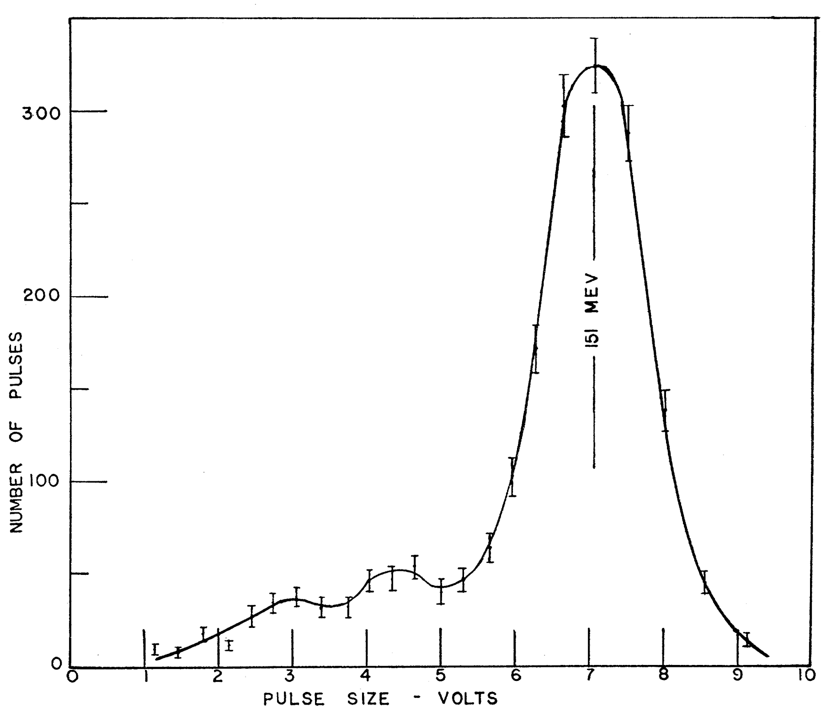

Fig. 4. Distribution of ionization due to both fission fragments.

and 4.5 volts, respectively, corresponding to energies of 64 Mev and 97 Mev. The position of the peaks was unchanged from one run to the next, within the limits of error. The total number of counts plotted is about 2500. Addition of the individual points under each peak shows that the number of counts in each is the same within 3 percent. The half-width of each peak is about 18 Mev. There are some individual pulses corresponding to energies as high as 110 Mev, although their number is small.

The values given are subject to a correction for absorption in the U layer. Under the rough assumption that the energy loss per unit path length is constant over the entire range, its value can be estimated by dividing the measured energies of the peaks in the number-ionization curve by the corresponding ranges.[8] This yields a result of 4.4 Mev per mm of air and therefore about 1 Mev, somewhat less than the accuracy of the experiments, should be added to the energy value of each peak.

The data resulting from a similar set of observations of the total ionization, with the thin Al foil, are plotted in Fig. 4. A total of 1900 pulses was evaluated. There is a fairly sharp maximum at 7.05 volts, corresponding to an energy of 151 Mev. The correction for nonlinearity of the oscillograph would make this peak slightly sharper. As the correction is small no reduced plot is given. Fig. 5 shows a sample record of 150-Mev pulses. Since the Al foil plus the U layer have a combined stopping power of about 1.5 mm, the correction for absorption in

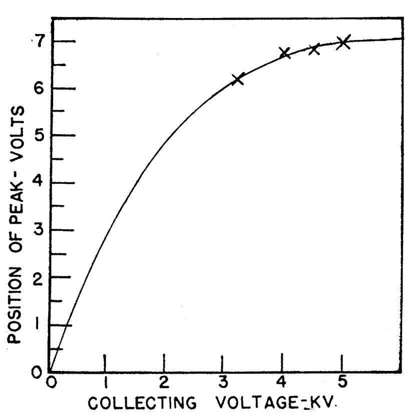

FIG. 6. Saturation curve.

this case is somewhat larger, about 8 Mev,[13] which brings the corrected energy to 159 Mev. The half-width of the peak is 32 Mev. The two small peaks in this plot correspond exactly to the single-fragment peaks, and probably arise from small amounts of U which were accidentally deposited on the 2-mil Al support. There are a number of pulses corresponding to 190 Mev and a very few having energies as high as 200 Mev. In order to find out whether the shape of the distribution curve was influenced by a possible uneven deposit of the U layer, some experiments were carried out with a second foil. The distribution curve was the same in each case.

The saturation conditions in the ionization chamber were investigated by making a set of measurements with various collecting voltages, statistically sufficient only to locate the standard pulse voltage corresponding to the maximum of the number-ionization curve. The resulting saturation curve is shown in Fig. 6. The numerical values were roughly as follows. For 3000 volts the value was 6.2 volts, 13 percent below that for 5000 volts. For 4000 volts it was 6.75 volts, only 4 percent below the 5000-volt value. Since this latter is a 20-percent change in voltage, it seems that the chamber was fairly well saturated at 5000 volts.

The reproducibility of the results was such that the value of the 151-Mev peak never shifted by our standard deflection interval (3 percent) from run to run. The presence of the background of statistical ionization mentioned above tends to broaden the peaks, but as it is symmetric about zero deflection, it has no effect on the position of the maximum. This was shown experimentally by the fact that different runs, made with widely different values of ion current in the deuteron beam, gave the same results. Statistical ionization and thickness of the foil influence, however, the width of the peaks somewhat. We estimate that not more than 5 Mev of the half-width is due to these effects. There may be other effects which are responsible for part of the observed widths. It is possible that the values found for the ionization should

[13] An estimation of the distribution in energy loss among the fragments indicates that the mean energy loss in the foil corresponds to about 1.2 times its stopping power.

be slightly increased because of incomplete saturation. It is also possible that the rough assumptions used in our foil corrections may introduce some error into the value of the total ionization peak. Taking these factors into account, we believe that our values, as measures of ionization, are accurate to ±5 percent.

IV. Discussion

The validity of any quantitative correlation between the ionization produced by fission fragments and their energy is, of course, extremely questionable, since one knows so little about the mechanism of ionization by highly charged particles of high velocity. Some information concerning the mean energy required for the creation of an ion pair by a fission fragment can be got from a comparison of the results of ionization measurements with values obtained by independent methods. The only such result available is that of Henderson,[6] whose preliminary heating experiments give 175 Mev±10 percent, for the energy release per fission. His value includes the heating effect of short-lived β-emitters, which is of the order of 5 Mev,[14] yielding a value of about 170 Mev for the kinetic energy alone. This agrees within experimental error with our value of 159 Mev, which is found under the assumption that the mean ionization energy is the same for fission fragments as it is for α-particles. More precise comparison must await the results of refined heating experiments now being performed by Dr. Henderson.

Our ionization values are in good agreement with those of Jentschke and Prankl,[2] and somewhat above those found by v. Droste,[3] Haxel,[4] and Booth, Dunning and Slack.[5] We were unable to observe the fine structure reported by v. Droste. The ratio of abscissae of the single-fragment peaks, 98 to 65, is in agreement with that found by Jentschke and Prankl and by Booth, Dunning and Slack.

The corrected ionization due to the entire fission (159 Mev) and the sum of the values of the single-fragment peaks (65+98=163 Mev) agree within experimental error. This fact, together with the equality of the numbers of counts in the two energy groups of Fig. 3, suggests the hypothesis that the pairs of fragments corresponding to the two peaks may be associated with single fission processes.

Under this assumption the energy ratio of the two peaks in Fig. 3 (98 : 65) is inversely proportional to the mass ratio of the fission fragments. If the disintegrating nucleus is U^{239} the masses of the most probable fragments are 96 and 143. The latter is in a mass region in which available

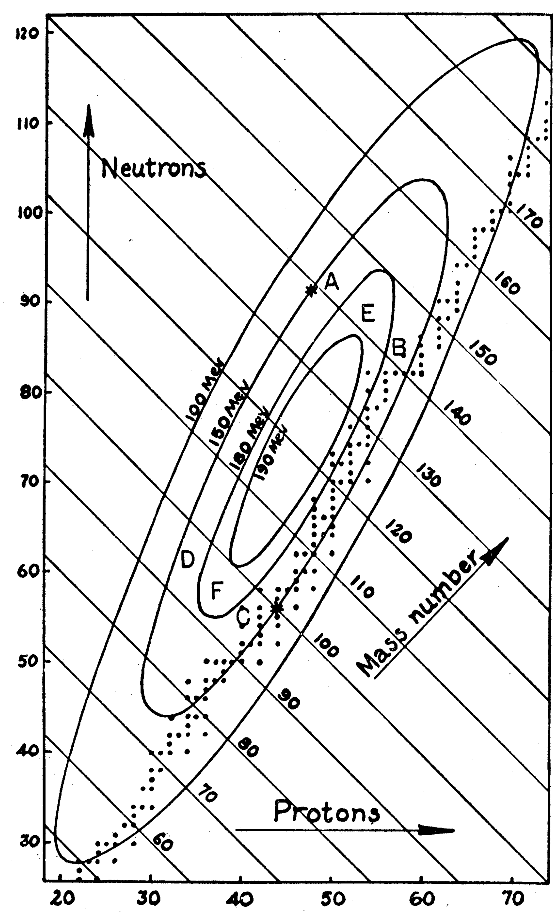

FIG. 7. (After N. Bohr and J. A. Wheeler) The difference in energy between the nucleus $_{92}U^{239}$ in its normal state and the possible fragment nuclei $_{44}Ru^{100}$ and $_{48}Cd^{139}$ (indicated by the crosses in the figure) is estimated to be 150 Mev as shown by the corresponding contour line. In a similar way the estimated energy release for division of U^{239} into other possible fragments can be read from the figure.

[14] H. H. Barschall, W. T. Harris, M. H. Kanner and Louis A. Turner, Phys. Rev. 55, 989 (1939).

chemical evidence has already placed some of the strongly β-active fission fragments.*

Figure 7 is a reproduction of Fig. 1 in the paper of Bohr and Wheeler.[7] If one assumes that the energy values found in these experiments represent true absolute values, then it follows that the most probable mode of fission leads to nuclei which lie in a region near the intersection of the line corresponding to mass 143 with the 160-Mev contour ellipse. There are two such intersections (A,B) on the diagram. The regions C and D represent the lighter fragments corresponding to A and B, respectively. To account for the observed radioactivity of both light and heavy fragments, one must assume that nuclei in all four regions are produced, since B and C lie in the region of the stable isotopes. This would indicate that the most probable fragments have neutron-proton ratios very different, both from each other and from that of the original U. This seems rather unlikely, however, as the fragments might be expected, on statistical grounds, to lie along a line corresponding to the U neutron-proton ratio, i.e., the principal axis of the contour ellipses.

Shifts of about 10 percent in the values of the energies reported here could move the position of the most probable fragment nuclei to the regions around points E and F, which correspond to radioactive nuclei having the expected neutron-proton ratio. Shifts of this kind could be accounted for if our correlation of ionization and energy were inaccurate. They could also be due to an additional energy release unobservable in these experiments, i.e., emission of γ-rays or neutrons during the fission process. This possibility is substantiated by the work of Zinn and Szilard[15] who observed that about two neutrons, having energies up to 3.5 Mev, are emitted per fission. It was further found by Gibbs and Thomson[16] that the bulk of the neutrons are emitted less than 10^{-3} sec. after the fission. This makes it plausible that the neutrons observed by Zinn and Szilard are associated directly with the fission process.[17] These results indicate that about 15 Mev has to be added to our 160 Mev value for comparison with the theory. In addition, we are informed by Professor Wheeler that the calculated values of Fig. 7 may be in error by as much as 10 Mev.

V. Acknowledgments

These experiments were performed under the direction of Professor Rudolf Ladenburg, to whom we wish to express our sincere appreciation of his continued interest and encouragement. We wish also to thank Professor J. R. Dunning for his help and advice in the construction of the recording oscillograph, Dr. C. C. Van Voorhis for the sputtering of the foils, and Professors L. A. Turner and J. A. Wheeler for many helpful discussions. The high voltage equipment used in this work was purchased with the aid of a grant from the Rockefeller Institute.

* *Note added in proof.*—In a recent paper O. Hahn (Ann. d. Physik (5) **36**, 368 (1939)) gives two well-established examples for modes of fission: $U^{239} \rightarrow Kr^{88} + Ba^{151}$ and $U^{239} \rightarrow Xe^{139} + Sr^{100}$. Assuming 160 Mev for the total energy liberated in the fission process one obtains from these masses single-fragment energies of 59, 101, 67 and 93 Mev. These values lie almost symmetrically on both sides of the maxima in Fig. 3. As two peaks which are only 8 Mev apart would not have been resolved, particularly if they have any appreciable natural width, the mass-values quoted by Hahn are well compatible with our measurements.

[15] W. H. Zinn and L. Szilard, Phys. Rev. **56**, 619 (1939).

[16] D. F. Gibbs and G. P. Thomson, Nature **144**, 202 (1939).

[17] For a more complete discussion of this point see L. A. Turner, Rev. Mod. Phys. **12**, 1 (1940).

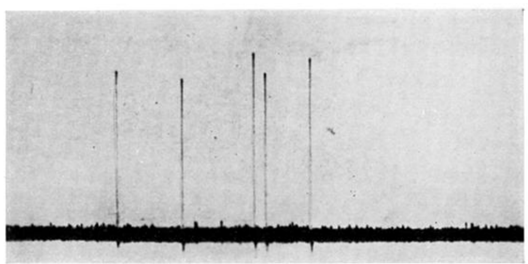

FIG. 5. Record showing 150-Mev pulses from pairs of fission fragments.