PHYSICAL REVIEW VOLUME 82, NUMBER 4 MAY 15, 1951

The Emission of Short-Range Charged Particles in the Slow Neutron Fission of Uranium

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(Received January 15, 1951)

A coincidence counting method has been employed to investigate short-range charged particles produced in the slow neutron fission of U^{235} and U^{233}. In the case of U^{235}, charged particles with ranges up to 8 mm of air were observed with a frequency of one in 76 ± 8 fissions. The maximum initial specific ionization of these particles was ten times that of α-particles from uranium, which suggests that some of the particles have masses considerably greater than that of the α-particle. The mass distribution could not be determined from the experimental data. Additional experiments proved that the observed particles were not nuclear recoils produced by fission fragments in their passage through the source and counter gas or fission fragments scattered by the backing material of the source. In the fission of U^{233}, particles were observed with ranges up to 7.4 mm of air with an abundance of one in 72 ± 6 fissions.

I. INTRODUCTION

WHEN fission of uranium occurs, the compound nucleus occasionally disintegrates into three or more charged particles instead of into the usual two fragments. Several cases of quadripartition have been seen in photographic plates,[1] and it has been reported that, in one out of every 250,000 fission events, division into three particles of comparable mass takes place.[2,3] In about 0.2 percent of fission events, an energetic α-particle with a range up to 50 cm of air is emitted in addition to the two main fragments; this mode of fission has been discussed in detail by many authors.[4] Still more often, in roughly one percent of all fission events, a charged particle with a range less than one cm of air is emitted along with the two main fragments. The present paper deals exclusively with these short-range particles.

The occasional emission of short-range charged particles in the fission of U^{235} was first detected by Cassels *et al.*,[5] using a pair of proportional counters in coincidence. They reported that, assuming isotropic emission, approximately 4 percent of fission events gave rise to a short-range charged particle that was tentatively identified as an α-particle of about 1-Mev energy. Green and Livesey,[6] searching for these particles in uranium loaded photographic emulsions, observed that there were more short spurs branching from the fission track near the origin of the event than were to be expected from calculations on the number of nuclear recoils which should be produced by the fast moving fission fragments. They concluded that, in one percent of fission events, a short-range ternary particle was emitted, preferentially at 90° to the main fission track, with a range up to 8 mm of air. Having determined the angular distribution, they corrected the abundance of Cassels *et al.* to 1 in 90 fission events, thus bringing the two experiments into good agreement. On the other hand, Tsien and his collaborators[7,8] observed particles with the same abundance but with ranges up to 3 cm of air. The mean angle of emission of the particles was found to be 70° with respect to the lighter of the two main fragments.

Investigation of the short-range particles by means of photographic plates containing uniformly dispersed uranium, aside from being laborious, has two disadvantages. Firstly, ignorance of the exact origin of a fission event makes it difficult to distinguish short ternary tracks from nuclear recoils, and secondly, the short ranges of the particles in the emulsion do not allow an accurate estimation of their masses. For these reasons coincidence counting methods were used in the present investigation. Even so, special experiments were necessary to demonstrate that the observed particles were not nuclei recoiling from the fission fragments. A preliminary account of this work has already been given.[9]

II. APPARATUS AND METHOD

The distribution in range of the charged particles was studied by means of the apparatus shown in Fig. 1. A thin source of uranium S, less than 0.1 mg/cm^2 in thickness, was placed in a chamber containing a pair of proportional counters F and P, the whole chamber being filled with methane to a suitable pressure. A collimated beam of slow neutrons from the Chalk River heavy water pile produced fissions in the source, and coincidences were sought between fission fragments detected in F and charged particles in P. The fission counter, generally operated at a gas multiplication of about 10, detected roughly 10 percent of all fissions occurring in the source and was prevented from recording particles of less than one cm range by a thin Nylon film stretched across its entrance. Wire grids across the entrances to

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[1] Tsien, Ho, Chastel, and Vigneron, Phys. Rev. **71**, 382 (1947).

[2] Tsien, Ho, Vigneron, and Chastel, Nature **159**, 773 (1947).

[3] L. Rosen and A. M. Hudson, Phys. Rev. **76**, 181(A) (1949).

[4] K. W. Allen and J. T. Dewan, Phys. Rev. **80**, 181 (1950). This paper contains complete references to earlier work on the subject.

[5] Cassels, Dainty, Feather, and Green, Proc. Roy. Soc. (London) **A191**, 428 (1947).

[6] L. L. Green and D. L. Livesey, Trans. Roy. Soc. (London) **A241**, 323 (1948).

[7] Tsien, Ho, Chastel, and Vigneron, J. phys. radium **8**, 165, 200 (1947).

[8] Tsien San-Tsiang, J. phys. radium **9**, 6 (1948).

[9] J. T. Dewan and K. W. Allen, Phys. Rev. **76**, 181(A) (1949).

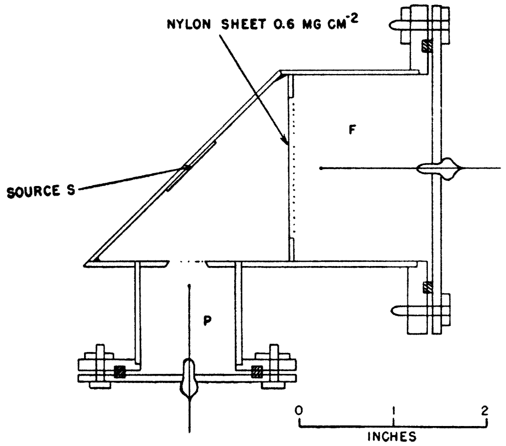

FIG. 1. Apparatus for range measurements.

both counters insured that ions produced in the source region by a particle entering one counter would not be dragged into the other counter, thereby causing a spurious coincidence.

The proportional counter P was operated at a gas multiplication of about 20 and was calibrated in energy by means of α-particles from a thin source of U^{233} placed at S. The initial ionization of these α-particles was about 0.1 Mev/mm. Using this calibration, the initial ionization produced by fission fragments traversing P at a very low pressure (2.2 cm of methane) was found to be 5.5 Mev/mm, which is in good agreement with the value of Sherr and Peterson[10] although rather lower than that reported by some authors. Hence, the counter response was reasonably linear over the desired working region of 0.1 to 5.5 Mev/mm. The solid angle of detection for P, as determined by counting a source of α-particles of known strength, was 1/250 of 4π-steradians.

The method of the experiments was to study the dependence of the true coincidence rate between F and P on the pressure in the apparatus and on the energy spent in counter P, relative to that spent by natural α-particles from the source at the same gas pressure. It will be evident that the counter P, when set to detect particles of low specific ionization and range less than 1 cm of air, counted natural α-particles and fission fragments from the source S. This gave rise to a large random coincidence rate, and long counting periods at relatively low intensity were necessary to obtain significant results. It soon became apparent, however, that most of the short-range particles had specific ionizations considerably greater than α-particles but smaller than those of fission fragments. Figure 2 is a typical bias curve of pulses from P. Natural α-particles produced pulses up to 2 v in height; short-range fission particles gave pulses up to 23 v, while all pulses above 23 volts came from normal fission fragments traversing P. It was seen from the flatness of this bias curve that a very considerable improvement in the ratio of true to random coincidences could be obtained by recording only coincidences between pulses from F and those pulses from P lying between 2 and 30 v. This region included all of the short-range particle pulses but excluded normal fission fragments which could only contribute to the random coincidence rate (since F did not detect short-range particles). An anticoincidence circuit was therefore arranged in such a manner that the large pulses from P (greater than 30 v) momentarily deadened the output of the coincidence mixer and prevented the recording of undesired random coincidences. In this way, the ratio of true to chance coincidences was increased by about a factor of ten without the loss of any true coincidences, since the slight increase in dead time due to the anticoincidence arrangement had a negligible effect at the relatively low counting rates employed. Figure 3 is a block diagram showing the arrangement of the electronic circuits. The anticoincidence discriminator level was normally set at 30 v.

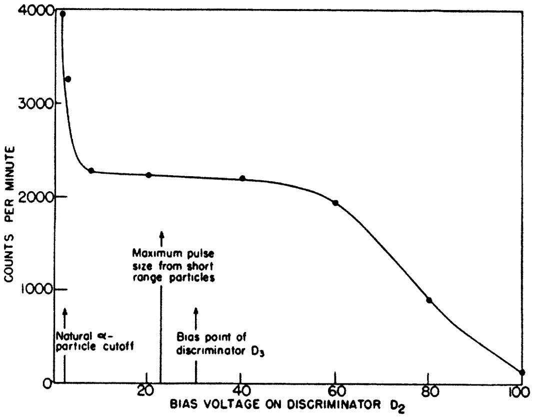

FIG. 2. Bias curve of pulses from the proportional counter P.

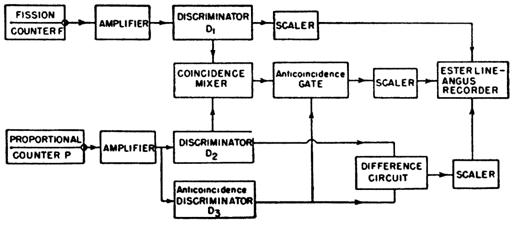

FIG. 3. Block diagram of the electronic arrangement.

The apparatus was made completely automatic and self-recording. A single Esterline-Angus chart recorded, after suitable scaling, the number of coincidences, the number of fissions counted in F, and the *difference* in the number of pulses passed by discriminators D_2 and D_3. By means of a step switch, the bias level of discriminator D_2 could be preset to a number of desired values between 2 and 20 volts. At any setting, the information was recorded for a given number of fissions,

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[10] R. Sherr and R. Peterson, Rev. Sci. Instr. **18**, 567 (1947).

generally about 5×10^6; then, the step switch automatically set the bias at the next preset level and the process was repeated. A shift in base line of the Esterline-Angus chart accompanied each change in bias setting. In this manner, complete bias curves could be run and recycled without attention to the apparatus.

III. RESULTS FOR U^{235}

To investigate the range distribution of the particles, the pressure in the counter system was varied from 2.2 cm (the lowest convenient working pressure) to about 16 cm Hg in a number of steps. At each pressure the bias of discriminator D_2 was varied from 2 to 20 v, as described above, and a curve of coincidence rate, after correcting for casual coincidences, was plotted against bias. Figure 4 shows a typical curve obtained at a pressure of 3.7 cm of methane, and it is apparent that practically all of the particles ionize more heavily than do the 5 Mev α-particles from the uranium source. Each of the curves so obtained was extrapolated back to zero bias in order to obtain the total abundance of short-range particles entering P at that pressure. Plotting these extrapolated abundances against pressure resulted in the range distribution curve of Fig. 5. The equivalent ranges shown are those which particles must have in order to reach the proportional counter at the corresponding pressures. It is evident that the maximum range of the particles is 8 mm of air, but it is difficult to deduce much about the actual range distribution of the observed particles because of lack of information concerning their specific ionizations. The coincident rates which were obtained in our system have been corrected on the basis of Green and Livesey's[6] angular distribution to give the abundances plotted as ordinates on Fig. 5. Extrapolation to zero pressure results in a total abundance of 1 in 76±8 fissions for all short-range particles.

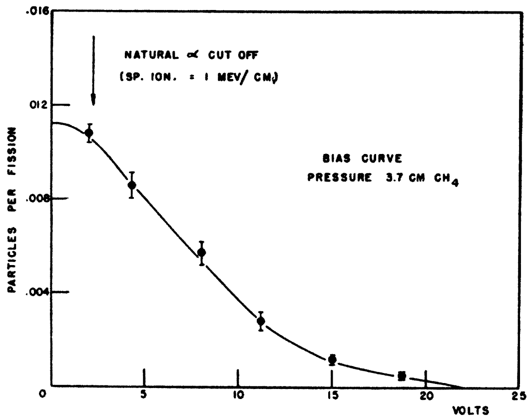

FIG. 4. Typical bias curve of coincident pulses.

Figure 6 (similar to Fig. 4) shows the bias curve of coincident pulses taken at a pressure of 2.2 cm of methane, which was the lowest pressure at which the counter operated satisfactorily. At this pressure the equivalent length of counter P was only 0.6 mm of air, so that this curve is effectively the distribution of *initial* specific ionization of the particles. The maximum specific ionization is 1.02 Mev/mm, which is 10 times greater than that produced by the uranium α-particles from the source, and 4.5 times greater than the maximum possible specific ionization of an α-particle of any range. Thus, at least some of the particles must have masses appreciably greater than that of the α-particle.

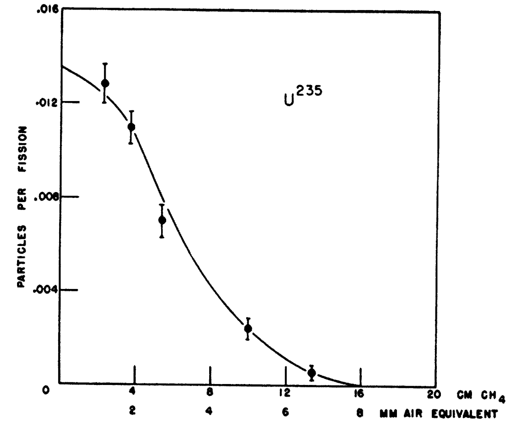

FIG. 5. Range distribution of short-range particles from U^{235}.

There are three plausible mechanisms which would give rise to short-range particles which are not true ternary fission fragments. Firstly, the observed particles may be recoil atoms projected into the counter P by the fission fragments as they pass through the uranium source or gas surrounding the source; secondly, they may be spurious coincidences caused by imperfect shielding of the source region by the two wire grids; and thirdly, they may be fission fragments back scattered into P by the platinum backing of the source S. The first two hypotheses were tested by placing a thin Nylon film, just sufficiently thick to stop particles of 8 mm range, directly over the source S. The dotted

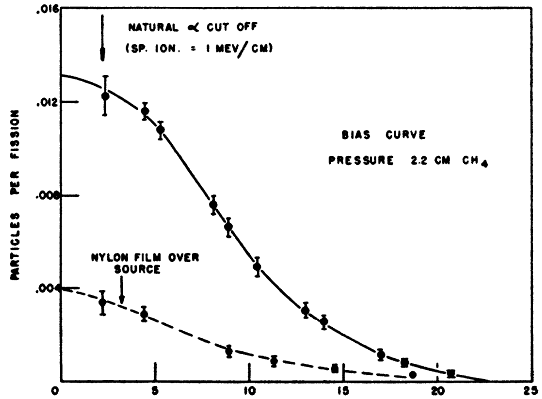

FIG. 6. Initial specific ionization of the coincident pulses.

curve of Fig. 6 shows the resultant coincident bias curve. The large decrease in coincidence rate caused by the Nylon foil rules out the possibilities of imperfect grid shielding and recoils from the counter gas. Furthermore, it is convincing evidence for the absence of source recoils. For, if the source did produce observable recoils, the Nylon film would stop them but would itself be expected to give rise to a greater number; firstly, because it contained many more light atoms than did the source; and secondly, because the slowing down of the fission fragments by the Nylon would increase the probability of such recoils. From the relatively small number of recoils produced from the Nylon layer, we concluded that a negligible number would be produced in the source and counter gas.

Calculations based on the Rutherford scattering formula indicated that less than 0.2 percent of the observed coincidences should be caused by fission fragments back scattered into P by the platinum backing of the source. However, experimental verification was obtained by replacing the uranium source on platinum by one on aluminum backing. This caused no change in the coincidence rate per fission nor in the shape of bias curve obtained. As it is impossible for fission fragments to be deflected sufficiently by aluminum nuclei to enter the counter P, none of these coincidences could be due to back scattering. Scattering of aluminum nuclei into P is a possible process, but further calculations showed this effect to be at least 50 times smaller than would be required to account for the coincidence rate. Independently of this calculation, however, it is highly improbable that the scattering of aluminum nuclei into the counter would reproduce exactly the back scattering of fission fragments from platinum.

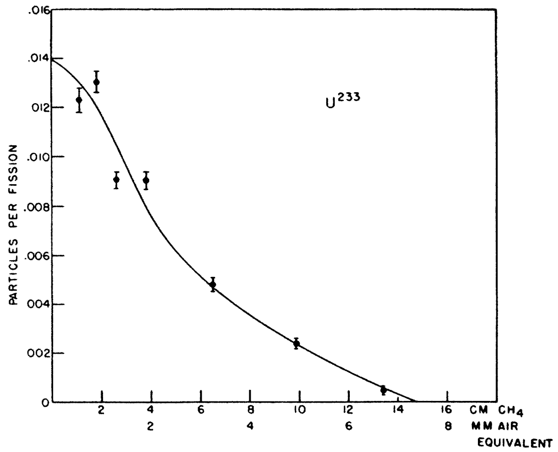

FIG. 7. Range distribution of short-range particles from U^{233}.

These auxiliary experiments show that the contribution to the coincidence rate arising from recoil atoms and back scattered fission fragments is very small, and no correction has therefore been applied. We feel confident in concluding that the true coincidences observed were due to short-range particles produced directly in the fission process.

IV. RESULTS FOR U^{233}

The emission of short-range particles from U^{233} was investigated in the same manner as for U^{235} and similar results were obtained. Figure 7 shows the resulting range distribution curve.

The maximum range was 7.4 mm, and the abundance one in 72±5 fissions, on the assumption that the angular distribution is the same as that for U^{235}. The maximum initial specific ionization of the particles was slightly lower than that found in the fission of U^{235}, although still considerably greater than that of α-particles.

V. NATURE OF THE PARTICLES

The experiments described above show that in approximately 1.3 percent of fission events in U^{235} short-range charged particles considerably more massive than α-particles are emitted. The range distribution of the particles measured in the present experiments is in excellent agreement with that obtained by Green and Livesey, as also is the abundance obtained using these authors' data on the angular distribution. The greater maximum range, *viz.* 3 cm air, obtained by Tsien *et al.* suggests that some nuclear recoils may have been included in their data, although this is difficult to reconcile with their abundance, which is in good agreement with the present work. Green and Livesey concluded that the short-range particles were probably α-particles, although they pointed out the difficulty of estimating their masses owing to their very short ranges in the emulsion.

It is not possible to obtain the mass distribution of the particles from the present data because only the range distribution and maximum specific ionization have been measured. Photographic plate evidence is against the existence of particles with mass numbers greater than about 12, while the present measurements suggest that most of the particles are heavier than α-particles. Owing to the rather meager data on range-energy relations for particles heavier than α-particles, it is not possible to estimate masses accurately. If it is assumed that the maximum initial specific ionization corresponds to the maximum range, then an estimated mass of 13±4 is obtained,[11] although there seems to be little justification for this assumption. It is clear, however, that a different process is involved in the emission of short-range particles than that concerned with the emission of long range α-particles.

It is a pleasure to acknowledge the interest shown by Dr. B. W. Sargent and Dr. B. Pontecorvo concerning these experiments. The cooperation of the pile operating staff, under the direction of Mr. D. D. Stewart and Mr. G. M. James, is greatly appreciated; thanks are also due Dr. J. G. MacHutchin for the preparation of the sources of fissile material.

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[11] This mass was misquoted as 12±1 in a recent publication. [E. W. Titterton and T. A. Brinkley, Phil. Mag. **41**, 500 (1950).]