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Emission of Neutrons by Uranium*

W. H. ZINN, *City College, The College of the City of New York, New York*
LEO SZILARD, *Pupin Physics Laboratories, Columbia University, New York, New York*
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Fast neutrons emitted by uranium under the action of thermal neutrons were studied by using a radium-beryllium photoneutron source. The background due to the primary neutrons can be neglected since only a few of the photoneutrons are sufficiently fast to be counted. Data are obtained concerning the energy spectrum of the uranium fission neutrons by recording photographically by means of a linear amplifier and cathode-ray oscillograph the pulses due to helium atoms projected in an ionization chamber. Visual inspection of the record gives an upper limit of the spectrum of 3.5 Mev. The number of neutrons emitted is estimated by analyzing the pulse distribution of hydrogen atoms projected by uranium neutrons in an ionization chamber filled with hydrogen and argon. The number found is brought into relationship with the number of fissions, observed under comparable conditions, in an ionization chamber lined with a thin film of uranium oxide containing a known amount of uranium. In this way it is found that about 2.3 neutrons are emitted per fission. The method used would permit a greater accuracy in the determination of this number than the actual accuracy obtained in the present experiments. This number, together with the fission cross section and the cross section for radiative capture gives the number of neutrons produced for each thermal neutron absorbed in uranium.

WE reported[1] some time ago that fast neutrons are emitted—apparently instantaneously—from uranium under the action of thermal neutrons and we found, as a rough estimate, an average of two neutrons per fission process. This result was obtained by counting the helium recoil nuclei which the fission neutrons project in a helium-filled ionization chamber. The emission of neutrons in the fission of uranium was independently discovered by von Halban, Joliot and Kowarski[2] as well as by Anderson, Fermi and Hanstein,[3] who observed an increase of the thermal neutron density in water in the presence of uranium. Others[4] have investigated the same phenomenon.

Before this "instantaneous" emission had been observed, Roberts, Meyer and Wang[5] discovered a delayed emission of neutrons from uranium which apparently follows a beta-transformation of a half-life period of twelve seconds. We had found that the instantaneous emission was very much stronger than the delayed emission and we assumed that it corresponds to a direct ejection of neutrons from the uranium fragments, without being preceded by a beta-transformation, and that accordingly the time delay involved is far too small to be measured by the usual

* Publication assisted by the Ernest Kempton Adams Fund for Physical Research of Columbia University.
[1] L. Szilard and W. H. Zinn, Phys. Rev. **55**, 799 (1939).
[2] H. von Halban, F. Joliot and L. Kowarski, Nature **143**, 470 (1939).
[3] H. L. Anderson, E. Fermi and H. B. Hanstein, Phys. Rev. **55**, 797 (1939).
[4] G. P. Thomson, J. L. Michiels and G. Parry, Nature **143**, 760 (1939); G. von Droste and H. Reddeman, Nat. Wiss. **20/21**, 371 (1939).
[5] R. B. Roberts, R. C. Meyer and P. Wang, Phys. Rev. **55**, 510 (1939).

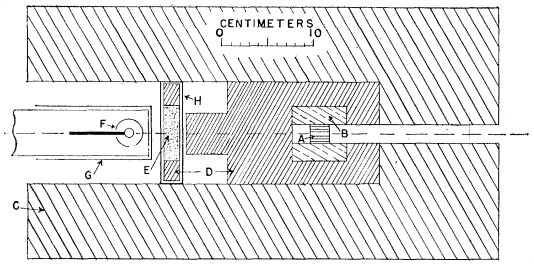


FIG. 1. Arrangement for the observation of the emission of fast neutrons from uranium. *A*—radium; *B*—beryllium block; *C*—paraffin wax; *D*—lead; *E*—uranium cell; *F*—spherical ionization chamber; *G*—cadmium sheet cap; *H*—cadmium sheet shield.

methods. This assumption was based on the arguments that it would be very difficult to explain the great abundance of the instantaneous neutron emission without assuming direct ejection and that no hard beta-rays were observed which should be expected to be present if the neutron emission followed a very short-lived beta-transformation. From direct experimental evidence, however, we could not exclude a delay smaller than one-tenth of a second. Gibbs and Thomson[6] have now shown by direct experiments that the delay is smaller than one-thousandth of a second and this appears to leave little doubt as to a direct ejection of neutrons.

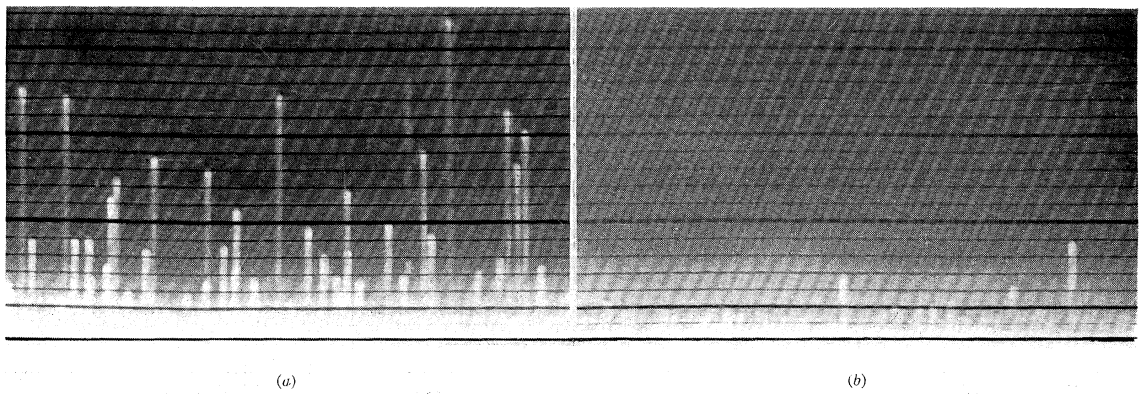
In the present experiments helium recoils were used for investigating the energy distribution of the fission neutrons, but hydrogen recoils were used for estimating the number emitted per fission.

The experimental arrangement is shown in Fig. 1. The source of thermal neutrons was about one gram of radium, *A*, placed in the center of a beryllium block, *B*, and surrounded by a paraffin cylinder, *C*. Fast neutrons emitted under the action of the thermal neutrons by about 430 grams of uranium metal enclosed in the cell *E*, were detected by the spherical ionization chamber, *F*. The pulses from the chamber were fed into a linear amplifier and were made visible by means of a cathode-ray oscillograph. A camera with a moving film was used to obtain a photographic record of the pulses appearing on the oscillograph screen.

Two such records are shown in Fig. 2; one was obtained in the absence and the other in the presence of the cadmium sheet shield, *H*. The shield *H* completely surrounds the uranium and shuts it off from most of the thermal neutrons, leaving a background of pulses which is partly due to particularly fast photoneutrons from the source, and partly due to fission neutrons from the uranium emitted under the action of the few thermal neutrons which pass through the cadmium shield. This background amounts to less than one pulse per minute.

The ionization chamber, which was filled with 10 atmospheres of hydrogen and 8 atmospheres of argon, contained a small amount of nitrogen. By removing the cadmium cap, *G*, from the chamber the nitrogen atoms in the chamber can be exposed to the action of thermal neutrons and

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



(*a*) (*b*)

FIG. 2. (*a*) Oscillograph record of the fast neutrons from uranium. Cadmium sheet shield, *H* of Fig. 1, absent. Thermal neutrons falling on the uranium in the cell *E*. Ionization chamber filled with 10 atmospheres of hydrogen and 8 atmospheres of argon. (*b*) Record obtained with the cadmium shield *H* shutting off the thermal neutrons from the cell *E*.

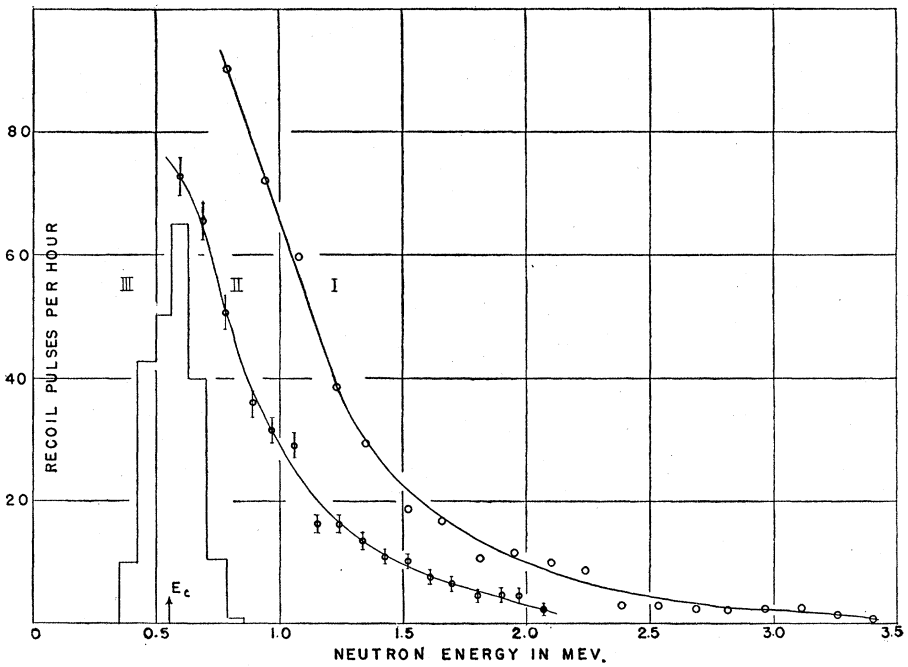


FIG. 3. Curve I: Pulse distribution due to helium recoils. Ionization chamber filled with 10 atmospheres of helium and 10 atmospheres of argon. Curve II: Pulse distribution $P(E)$ due to hydrogen recoils. Ionization chamber filled with 10 atmospheres of hydrogen and 8 atmospheres of argon. Curve III: Pulse distribution due to protons emitted, under the action of thermal neutrons, by a small amount of nitrogen in the chamber filled with 10 atmospheres of helium and 10 atmospheres of argon.

will then emit protons of about 0.6 Mev energy.[7] The pulses due to these protons were recorded and their distribution is shown in curve III of Fig. 3. This curve shows a sharp maximum which should correspond to an energy of about 0.6 Mev, and therefore this curve was used for calibrating the ionization chamber.

In order to find from the observed number of hydrogen recoils the number of neutrons which pass through the chamber it is necessary to know something about the energy distribution of the fission neutrons. This knowledge is required for two reasons. First, the scattering cross section of hydrogen is a function of the neutron energy; secondly, the observed pulse distribution of the hydrogen recoils is cut off at a certain energy E_c, which in this case was 0.55 Mev, in order to avoid the counting of pulses in the region which is affected by the gamma-ray background. Neutrons which have an energy below this cut-off energy, E_c, do not contribute to the recorded pulse distribution and their number has to be determined from the shape of the neutron spectrum, provided this spectrum is known.

If the time required for the collection of ions in the chamber were short compared with the time constant of the amplifier, the size of the pulses recorded by the oscillograph might be considered a fair measure of the energy which the recoil proton loses in the chamber. Even so, the size of the pulses cannot be considered a measure of the initial energy of the recoil protons if these lose only part of their energy in the chamber and are stopped by the walls. Therefore, if $R(E)dE$ is the number of recoil protons having an initial energy between E and $E+dE$, and if $P(E)dE$ is the number of recoil protons which lose in the chamber an amount of energy between E and $E+dE$, these two functions will be rather different in the high energy region where the range of the recoil protons cannot be neglected in comparison with the linear dimensions of the chamber. For the hydrogen-argon filled chamber

[7] J. Chadwick and M. Goldhaber, Proc. Camb. Phil. Soc. **31**, 612 (1935); T. W. Bonner and W. M. Brubaker, Phys. Rev. **49**, 778 (1936); M. S. Livingston and H. A. Bethe, Rev. Mod. Phys. **9**, 344 (1937).

which was used the two functions can be expected to coincide very nearly in the region of the cut-off energy E_c, and can be expected to differ widely for energies above 1.5 Mev.

For this reason helium recoils (which have about $\frac{1}{10}$ the range of recoil protons for equal neutron energies) had to be used instead of hydrogen recoils in order to find the upper end of the energy spectrum of the fission neutrons. Curve I, in Fig. 3, shows the pulse distribution of helium recoils obtained with 10 atmospheres of helium and 10 atmospheres of argon in the chamber. This curve shows that the spectrum of the fission neutrons extends to about 3.5 Mev. Though the existence of a small number of high energy neutrons such as reported by von Halban, Joliot and Kowarski,[8] is not inconsistent with our result, the number of neutrons having energies above 4 Mev appears to be too small to have much bearing on our estimate of the total number of fission neutrons.

Since the calibration of the chamber, which we performed by means of protons, is not entirely satisfactory for correlating the size of the pulses due to helium recoils with the energy of the helium recoils, the helium-argon filled chamber was also calibrated by means of D+D neutrons of 2.5 Mev energy. The two calibrations coincided within the limits of the experimental error.

An estimate of the number of fission neutrons should be based on a count of hydrogen recoils rather than helium recoils since the scattering cross section of helium has a sharp maximum[9] for neutrons of about 1.0 Mev energy, and helium is therefore not suitable for the purposes of a quantitative estimate. It can be shown that, if the neutron-proton scattering is spherically symmetrical in the system of the center of gravity, the number of neutrons $N(E)dE$ which pass through the chamber, and which have an energy between E and $E+dE$, is given by:

$$N(E)=-\frac{E}{H\sigma(E)}\frac{dR(E)}{dE},$$

where $\sigma(E)$ is the scattering cross section of the proton and H is the number of hydrogen atoms in the ionization chamber. From this we derive, for the total number of neutrons, N, passing through the chamber:

$$N=\int_0^\infty N(E)dE=\frac{\alpha}{\sigma_{Av}H}\left[E_cR(E_c)+\int_{E_c}^\infty R(E)dE\right],$$

where α is the ratio of the total number of neutrons to the number of neutrons which have an energy in excess of E_c, and σ_{Av} is an average scattering cross section of the proton, the value of which has to be determined from the energy spectrum of the neutrons. Since we have:

$$\int_0^\infty R(E)dE=\int_0^\infty P(E)dE$$

and since, for the reasons stated above, we have with good approximation:

$$R(E)\backsimeq P(E) \quad \text{for} \quad E\leq E_c,$$

we can express N in terms of $P(E)$ instead of $R(E)$. We then have:

$$N=\frac{\alpha}{\sigma_{Av}H}\left[E_cP(E_c)+\int_{E_c}^\infty P(E)dE\right].$$

Let it now be assumed for the sake of argument that all the neutrons are emitted from a moving uranium fragment which has a mass number of about 120 and a kinetic energy of about 100 Mev. If all the neutrons were emitted from such a moving fragment with a single energy E_0, the energy distribution of the neutrons in the

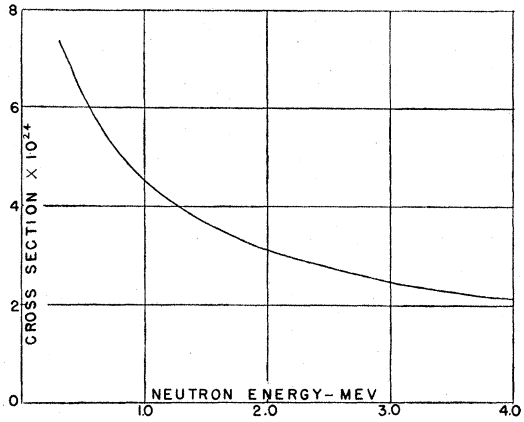


FIG. 4. Neutron-proton cross section as a function of neutron energy according to current theory.

[8] H. von Halban, F. Joliot and L. Kowarski, Nature **143**, 939 (1939).
[9] H. Staub and W. E. Stephens, Phys. Rev. **55**, 131 (1939).

laboratory reference system would stretch from:

$$E_{\min}=(0.9-E_0^{\frac{1}{2}})^2 \text{ Mev}$$

to:

$$E_{\max}=(0.9+E_0^{\frac{1}{2}})^2 \text{ Mev}.$$

It is easy to see that the neutrons should be uniformly distributed in this energy interval if their distribution is spherically symmetrical in the center of mass system. One obtains accordingly:

$$\alpha=(E_{\max}-E_{\min})/(E_{\max}-E_c)$$

and

$$\frac{1}{\sigma_{Av}}=\frac{1}{E_{\max}-E_c}\int_{E_c}^{E_{\max}}\frac{dE}{\sigma(E)}.$$

Using for $\sigma(E)$ the curve shown in Fig. 4 which has been theoretically derived,[10] the following values of α/σ_{Av} are obtained for various values of $E_{\max}$.

$E_{\max}$	2.0 Mev	3.0 Mev	4.0 Mev
α/σ_{Av}	$0.316\times10^{+24}$	$0.353\times10^{+24}$	$0.388\times10^{+24}$

The variation of α/σ_{Av} with $E_{\max}$ is so slight because of the manner in which both α and σ_{Av} decrease with rising $E_{\max}$.

The value of the expression:

$$E_cP(E_c)+\int_{E_c}^{\infty}P(E)dE$$

was found from the observed pulse distribution (curve II of Fig. 3) to be 13.7 pulses per minute. Since the number H of hydrogen atoms in the chamber was $H=6.9\times10^{+21}$ the number of neutrons passing through the chamber is

$$N=1.98\times10^{-21}(\alpha/\sigma_{Av}) \text{ per minute.}$$

If the cadmium cap G is removed the number of thermal neutrons reaching the uranium cell is increased and the number of fast neutrons passing through the ionization chamber is increased by the same factor. This factor was found to be 1.22 by filling the ionization chamber with pure hydrogen and then counting the hydrogen recoils giving rise to pulses above a certain arbitrarily set level, both in the presence and absence of the cadmium cap. Thus the number of neutrons N^* which pass through the chamber in the absence of the cadmium cap, G, is

$$N^*=2.415\times10^{-21}(\alpha/\sigma_{Av}) \text{ per minute.}$$

From N^* the number, K, of neutrons emitted per minute by the uranium was calculated by taking into account the geometrical factors, including the variation of the thermal neutron density within the uranium cell. K is thus found to be:

$$K=8.25\times10^{-19}(\alpha/\sigma_{Av}) \text{ per minute.}$$

In order to obtain the number of neutrons emitted per fission it is necessary to compare K with the number of fissions, L, which occur in the uranium under the conditions of this experiment. For this purpose both the ionization chamber and the uranium cell were removed and a parallel plate ionization chamber lined with a thick layer of uranium oxide was placed in the position previously occupied by the uranium cell. The number of fissions produced in this chamber was observed and found to be 19 per minute. The chamber was then calibrated by comparing the number of fissions obtained from the thick uranium oxide layer with the number of fissions obtained in the same chamber at the same thermal neutron intensity from a thin layer of uranium oxide containing 1.4 mg of uranium. The calibration was carried out by using a particularly strong neutron source, so as to obtain a sufficiently large number of counts from the thin layer. The ratio of the fission counts from the thick layer and from the thin layer was found to be 29.2, from which it is concluded that 196,000 fissions per minute should take place in the uranium cell containing 427.7 grams of uranium. This would be the number of fissions if the density of the thermal neutrons were not reduced in the uranium cell due to the absorption of such neutrons in uranium. We estimate that the average density of thermal neutrons within the cell is reduced by a factor of 0.715. The number of fissions L actually taking place within the cell is therefore

$$L=140{,}000 \text{ per minute.}$$

In order to estimate the reduction of the average thermal neutron density within the uranium cell leading to the factor of 0.715, we first explored the anisotropy of the thermal neutron radiation near the uranium cell by means

[10] J. Schwinger and E. Teller, Phys. Rev. **52**, 286 (1937).

of a rhodium indicator, and then calculated the thermal neutron density within the uranium by assuming the distribution of thermal neutrons to be the same as would result from the superposition of two parallel thermal neutron beams, one directed away from the source and the other towards it, and having an intensity ratio of 3 to 1. We assume exponential absorption for these two beams within the uranium and an exponent corresponding to a half-value thickness in uranium of 14 g per cm^2.

The number of neutrons emitted per fission is

$$K/L=5.9\times10^{-24}(\alpha/\sigma_{Av}).$$

This number should be increased by perhaps 10 percent in order to correct for the fact that $P(E)$ does not exactly coincide with $R(E)$ even for $E\leq E_c$. The magnitude of this correction was estimated by comparing for D+D neutrons of 2.5 Mev energy the observed pulse distribution $P(E)$ with the calculated distribution $R(E)$ in the region of the low recoil energies. Making this correction one finds for ρ, the number of neutrons per fission

$$\rho=6.5\times10^{-24}(\alpha/\sigma_{Av}).$$

Using for α/σ_{Av} the value 0.353×10^{-24} which corresponds to $E_{max}=3$ Mev rather than to the actually observed upper limit of the fission neutron spectrum, one finds

$$\rho=2.3.$$

Since the fission neutrons hardly will be emitted with a single energy E_0, too high a value for α/σ_{Av} would be obtained if the observed value of the upper limit of the energy spectrum were used for E_{max}. In any case the error introduced by the uncertainty of the actual energy distribution of the fission neutrons should be small since one finds for

$$E_{max}=2 \text{ Mev} \quad \rho=2.0$$

and for

$$E_{max}=4 \text{ Mev} \quad \rho=2.5.$$

More serious, however, may be a number of experimental inaccuracies which might conceivably add up to give a considerable error.

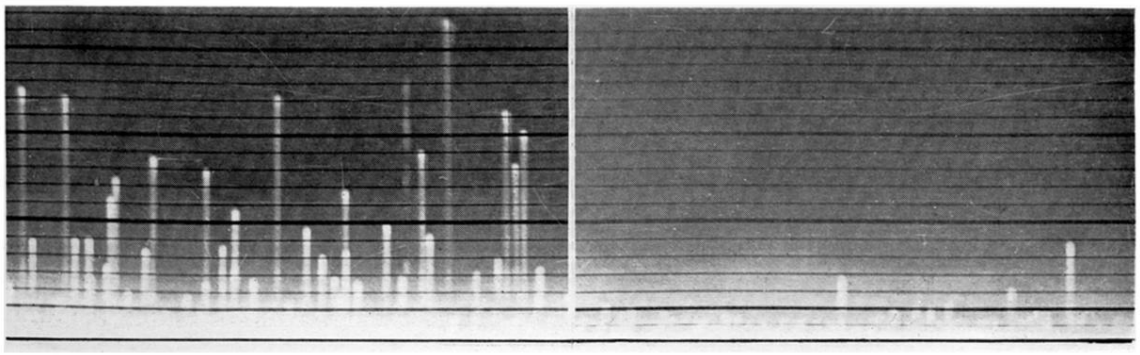
The interest in the number of neutrons emitted per fission arose out of its obvious importance from the point of view of the possibility of nuclear chain reactions. At present we have the following set of values: number of neutrons per fission, 2.3; fission cross section,[11] 2.0×10^{-24} cm^2; cross section for radiative capture[12] 1.3 or 1.2×10^{-24} cm^2. According to these values, the number of neutrons emitted by uranium per thermal neutron absorbed should be 1.4, which agrees with the value of 1.5 recently obtained by another method by Anderson, Fermi and Szilard.[13] Too much significance should not be attributed to this agreement, since the values given above are subject to fairly wide experimental errors.

If required the present experiments could be repeated with greater accuracy since the method used is quite capable of being applied with greater precision. Moreover, it gives the number of neutrons per fission independently of the value of the fission cross section which enters into the method used by von Halban, Joliot and Kowarski. These authors report[14] a value of 3.5 ± 0.7 neutrons per fission.

It should be mentioned that it appears to be essential for the method presented here to work with a low background count. The background is due to the primary neutrons and can be kept small by using a photoneutron source. We did not find it possible to obtain quantitative results by using neutrons from radon-beryllium sources or from the D+D reaction on account of the high background count due to the primary neutrons.

We are indebted to Dr. G. N. Glasoe for suggesting the method of obtaining the photographic records and for much valuable advice in this connection, and to Dr. E. T. Booth for determining by means of an alpha-particle count the uranium content of the thin uranium sheet which we used for purposes of calibration. Also, we wish to thank the Department of Physics of Columbia University for the laboratory facilities placed at our disposal and the Association for Scientific Collaboration for enabling us to obtain the radium used in this experiment.

[11] H. Anderson, E. Booth, J. Dunning, E. Fermi, G. Glasoe and F. Slack, Phys. Rev. **55**, 511 (1939).
[12] H. v. Halban, L. Kowarski and P. Savitch, Comptes rendus **208**, 1396 (1939); H. L. Anderson and E. Fermi, Phys. Rev. **55**, 1106 (1939).
[13] H. L. Anderson, E. Fermi and L. Szilard, Phys. Rev. **56**, 284 (1939).
[14] H. von Halban, F. Joliot and L. Kowarski, Nature **143**, 680 (1939).



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