Spontaneous fission of uranium*

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1. INTRODUCTION

N. Bohr and J. A. Wheeler[1] pointed to the possibility of fission of uranium with a half-life of the order of 10^{22} years. The calculation was carried out for the abundant isotope U^{238} using the formula for the penetration of the particle through the potential barrier. The width of the barrier was taken equal to the radius of the fragment, which is an assumption the validity of which can be checked only by experiment.

The question of the possibility of spontaneous fission of uranium was studied experimentally by Libby[2] who based his work on the assumption that in the process of spontaneous fission neutrons should be emitted just as in the case of the fission of uranium under the action of neutrons. Libby attempted to detect the neutrons originating in the process of spontaneous fission of uranium nuclei using a BF_3 counter sensitive to slow neutrons. On the basis of the negative result of the experiment one could establish the lower bound for the decay half-life of uranium ($T > 10^{14}$ years).

2. THE EXPERIMENTAL METHOD

In the experiments described below the method was used which was proposed by Frisch[3] for recording the processes of fission of nuclei. An ionization chamber with plates covered by a layer of uranium oxide is connected to a linear amplifier adjusted in such a manner that the α-particles emitted by uranium are not recorded by the system; but the pulses from fragments which greatly exceed the pulses from α-particles fire the output thyratron and are recorded by the mechanical relay. The thickness of the working layer of uranium oxide and the area of the plates of the commonly used ionization chambers (2 plates of 30 mm diameter) gave the possibility of establishing approximately the same lower limit on the decay half-period for spontaneous fission as in Libby's experiments. In order to increase the sensitivity of this method it was necessary to increase the working surface of uranium oxide. For this a specially constructed ionization chamber was made in the form of a multi-layer plane capacitor with a total area of 15 plates of 1000 cm^2 (Fig. 1). The plates separated from each other by 3 mm were covered by a layer of uranium oxide of 10–20 mg/cm^2. The collecting potential was 360 V. In order to reduce the microphone effect the ionization chamber together with the first tube of the amplifier was placed on a double damper consisting of lead slabs resting on rubber cushions.

In testing this chamber for recording the fragments it turned out that the special features of its construction also imposed special requirements on the amplifier. The large number of pulses from the α-particles required a considerably greater resolving ability of the amplifier than in working with chambers of the usual type. The superposition of pulses from individual α-particles in the case of the usual resolving power of the amplifier might give pulses comparable in magnitude with those which are obtained from fragments. In order to increase the resolving power of the system the grid of the first tube was grounded through a resistance of 10^5 Ohm. In the case of a self-capacitance of the chamber of 150 $\mu\mu$F and leakage resistance of 10^5 Ohm the time of charging up the capacitance amounted to 10^{-5} s. The pulse on the anode of the first tube had the shape shown in Fig. 2. The usual shape of the pulse without the leakage resistance is shown by the dotted line on the same diagram. A further increase in the resolving ability of the amplifier is attained by reducing the transition capacitance between the first and the second tubes down to 10 $\mu\mu$F (instead of the usual 100–1000 $\mu\mu$F). The inclusion of this capacitance shifted the frequency passband towards higher frequencies. This sharpened the pulse even more, since the amplifier admitted only the high-frequency components of the decomposition of the voltage pulse into a Fourier series. The narrowing of the frequency pass-band by the amplifier gave in addition a number of other advantages, specifically it allowed one to reduce to a minimum the low-frequency flicker-effect and the microphone noise of the ionization chamber. The optimum value of the transition capacitance between the first and the second tubes was chosen experimentally by including between the tubes a capacitance bank and choosing such a capacitance for which the ratio between the pulses from fragments and the amplifier noise would be a maximum.

An increase in the capacitance of the ionization chamber and the process of sharpening the pulse by switching in a leakage resistance greatly decreased the amplitude of the voltage from particles so that the pulses from the α-particles were of the same order of magnitude as the Johnson- and the shot-noises of the first tube. The pulses from the fragments exceeded the background of the amplifier; in order to make this excess more noticeable the last tube of the 6-F-5 amplifier was shifted by grid bias into the nonlinear region, after which the pulses from the fragments exceeded by a factor of 4–5 the background from the superposition of the pulses from the α-particles on the variations of the voltage brought about by the Johnson- and the shot-noises. The overall amplification coefficient was $\sim 10^7$.

The pulses from the output of the amplifier were applied to a cathode-ray oscillograph for visual observation and to a tube counting circuit of the usual type for recording the number of pulses with the aid of a mechanical relay.

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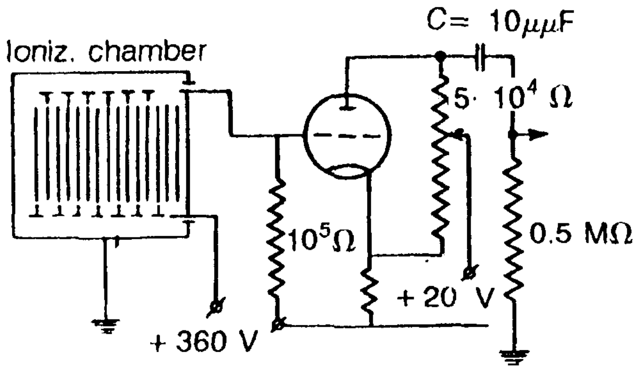

FIG. 1.

When the linear amplifier was operated in the nonlinear region special requirements were imposed on the constancy of the gain coefficient. In our experiments in the course of 5–10 hours of operation the gain coefficient varied by no more than 1–2%. The constancy of the gain coefficient was monitored by a calibrating device. The power supply of the anode and filament circuits was provided by storage batteries with a constant buffer recharge.

3. RESULTS OF MEASUREMENT AND CONTROL EXPERIMENTS

The developed methodology enabled one to observe the processes of uranium fission under the action of neutrons. The sensitivity of the chamber, as should have been expected, was by a factor 30–40 greater than the sensitivity of chambers of the usual construction. The amplifier together with the ionization chamber adjusted for recording the fragments gave approximately one pulse per minute from a 1 mCi of (Rn+Be)-fast neutrons when the source is brought right up against the chamber.

In the very first experiments with the amplifier adjusted to counting the fragments success was achieved in observing the spontaneous (in the absence of a neutron

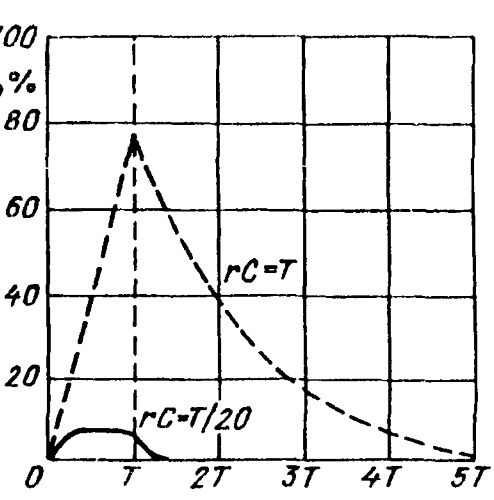

FIG. 2.

source) pulses both on the relay and the oscillograph. The number of such pulses was not very great (6 per hour) and therefore it is easy to understand that this phenomenon could not have been observed using chambers of the usual type. Although as far as their shape was concerned the observed pulses were quite similar to the pulses from fragments, it was necessary to carry out a number of control experiments in order to confirm the reality of the existence of uranium fission under these conditions. The origin of the observed pulses could have been explained:

1) by the recording by the amplifier of external oscillations;

2) by the superposition of pulses from individual α-particles;

3) by the existence of regions of gas amplification in individual regions of the ionization chamber;

4) by the random discharges on the surface of the uranium oxide.

Special experiments showed that none of the above causes could serve to explain the effect. The chamber with the plates without the uranium oxide did not produce a single pulse during 5 hours. This showed that the observed spontaneous pulses were due to the presence of uranium oxide on the plates of the chamber, and not to the reception of external oscillations.

In order to check the second assumption regarding the origin of these pulses a chamber was assembled in which in place of the uranium oxide thorium oxide was deposited on the plates with an addition of Po in such an amount that the total ionization current produced by the α-particles would exceed by a factor of two the ionization current in the uranium chamber. In the course of 10 hours of work only three kicks of the relay were observed. In these experiments there was no complete equivalence of the conditions of operation of the chamber due to the difference in the surface of the uranium oxide and the thorium oxide. Therefore another control experiment was performed in which into the uranium ionization chamber a thorium emanation was introduced in such an amount that the products of its decay would produce an ionization current brought about by α-particles twice as big as from the α-particles from the uranium itself. In this experiment we did not observe an increase of the effect, from which it follows that the random coincidences of the α-particles were excluded by the resolving ability of the amplifier.

From this same experiment it follows that in the chamber there are no regions of gas amplification. The active deposit had to be uniformly distributed over the plates of the chamber. Therefore, in the case if there had existed such regions of gas amplification, the introduction of the active deposit which increases the number of ionizing particles in these regions should have led to an increase in the number of pulses being studied. An increase in the voltage across the chamber from 360 to 500 V did not give any appreciable increase in the effect, which also excludes the possibility of gas amplification. The covering of the uranium oxide by a bronze foil of thickness of 1 μm, which guarantees good conductivity of the surface, led to a decrease both in the size and in the number of spontaneous

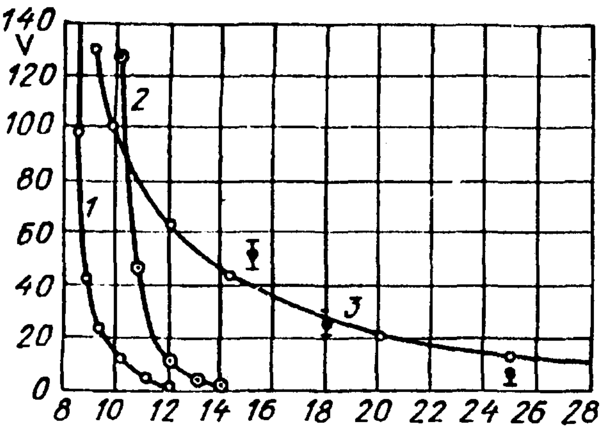

FIG. 3.

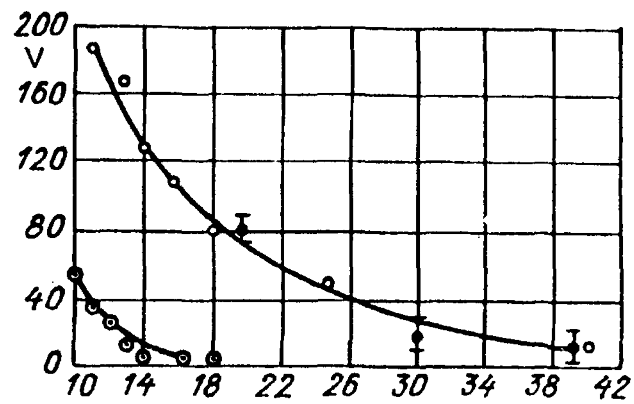

FIG. 4.

pulses, but in the same ratio there was a decrease in the number and the magnitude of the pulses from the fragments of uranium due to neutrons from the ampoule of (Rn+Be). Thus, none of the four subsidiary causes put forth to explain the effect actually occurs. It should also be noted that in the process of carrying out the experiments the measurements were made in three different chambers always with the same results. This excludes the possibility of explaining the observed spontaneous pulses by defects in construction of the chamber.

However, in order to verify finally that the spontaneous pulses are obtained as a result of the fission of uranium nuclei a curve was plotted of the distribution of the pulses with respect to their magnitude. A dependence of the number of spontaneous pulses on the bias on the grid of the first tube of the circuit controlling the mechanical relay was observed. The graphs are shown in Fig. 3. The number of counts of the mechanical relay was plotted along the vertical axis and the negative bias in volts on the grid of the tube was plotted along the horizontal axis. Curve *1* was obtained in the absence of the neutron source and represents the count of the number of α-particles against the background of the Johnson-noise. Curve *2* represents the same dependence in the case of an additional loading of the chamber by an active deposit of ThEm. Curve *3* was recorded with a neutron source and gives the distribution of fragments with respect to the magnitude under the conditions of nonlinear operation of the apparatus. The nature of this curve agrees with the nature of the distribution of spontaneous pulses with respect to magnitude. The figure shows the number of recorded spontaneous pulses per hour in the case of three different biases of the counting circuit. All these control experiments were carried out with an ionization chamber that has a working surface of ~ 1000 cm^2. Subsequently a still greater chamber was constructed with a total surface of 15 plates of 6000 cm^2. In order to increase the mobility of the ions the chamber was filled with dessicated argon. The maximum number of spontaneous kicks which could be observed with the aid of this chamber increased to 25–30 kicks per hour. The distribution curve of these spontaneous pulses with respect to their magnitude is presented in the same coordinates in Fig. 4. The same diagram shows on a different scale the dependence of the number of spontaneous pulses on the bias on the grid of the tube of the counting circuit. On the basis of all these experiments one can conclude that the spontaneous pulses observed by us are indeed caused by fragments from uranium fission.

4. DISCUSSION OF RESULTS

The effect that has been established can be explained by the action of cosmic neutrons on the uranium. The presently available experimental data both on the number and on the energies of the cosmic neutrons at ground level force us to reject this hypothesis. In order for it to be possible to ascribe the observed effect to cosmic neutrons it would be necessary to have a flux of 5 neutrons per cm^2 per sec. This number is much greater than the upper limit on the number both of cosmic and of terrestrial neutrons present in the atmosphere. Moreover, the absence of any noticeable effect in the ThO_2 chamber (3 kicks during 10 hours) convinces us of the impossibility of ascribing the observed effect to cosmic neutrons. From the investigations of Petrzhak and Flerov[4] and of Nikitina and Flerov[5] it follows that the limits for the fission of uranium and thorium are close and amount to approximately 1 MeV. The effective cross section for thorium fission is lower by a factor of 5 than the effective cross section for uranium fission. Consequently, the cosmic neutrons which do not produce an effect in the thorium chamber, cannot explain the effect observed in the uranium chamber.

Uranium fission could have been ascribed to the action of cosmic electrons and mesons. There are no indications whatever that there exists such a mechanism of interaction between electrons and heavy nuclei. But even if one supposes such an interaction to be possible the requirements on the effective cross section for the interaction $\sigma_{\rm inf} \sim 10^{-23}$ cm^2 make such an assumption improbable.

Hoffman ionization bursts could have produced similar large pulses. The number of ionization Hoffman bursts in the large chamber filled with a gas under high pressure is smaller than the effect observed by us. Moreover, in the uranium chamber an ionization burst would not have produced a sufficiently great ionization since the electrons could lose only a small fraction of their energy in the gas of the chamber. Experimentally this is supported by the absence of the effect in a chamber with plates without uranium.

One could have ascribed the observed effect to neutrons which arise from nitrogen and impurities in the uranium oxide under the action of α-particles of uranium itself. The observed 6 kicks per hour correspond to 1/20 mCi neutron activity of the (Rn+Be)-source. In order to obtain such a number of neutrons one needs a number of α-particles that is greater by approximately a factor of 100 than is emitted by uranium. Moreover, it is possible on the basis of Libby's data to reject this hypothesis also. Measurements using the BF_3 counter have shown that 1.5 kg of uranyl nitrate can account for a number of neutrons not exceeding the number of neutrons emitted by a 1/10 mCi (Rn+Be)-source. Since in our experiments the total amount of uranium oxide on the plates amounted to 15 g the neutrons emitted by the uranium oxide could explain only 1/50 of the observed effect.

We tend to think that *the effect observed by us should be ascribed to fragments obtained as a result of spontaneous fission of uranium.*

To clear up the question of the possibility of spontaneous fission of the closest products of uranium decay that are in an excited state following a β-transition we carried out special experiments with a layer of U_3O_8 deposited on the plates of a chamber enriched in UX_1 by a factor of 12 compared with its equilibrium content in U, and did not notice any increase in the effect. Therefore spontaneous fission should be ascribed to one of the unexcited isotopes of U with decay half-lives obtained from an evaluation of our results:

U^{238}—$10^{16}\sim 10^{17}$ years,

U^{235}—$10^{14}\sim 10^{15}$ years,

U^{234}—$10^{12}\sim 10^{13}$ years.

We wish to express our sincere gratitude for guiding this work to Prof. I. V. Kurchatov who outlined all the principal control experiments and who participated very directly in discussing the results of this research.

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[4] K. A. Petrzhak and G. N. Flerov (Zh. Eksp. Teor. Fiz.).
[5] T. I. Nikitinskaya and G.N. Flerov (Zh. Eksp. Teor. Fiz.).

SUPPLEMENT*

Further experiments on the study of the phenomenon of spontaneous fission of heavy nucleus were carried out using the apparatus described above with a somewhat altered method of recording the fragments. The voltage pulses from the Wynn-Williams amplifier were passed to the power amplifier at the output of which a ballistic galvanometer was attached. From the deflection of the galvanometer it was possible to draw conclusions concerning the ionization produced by the fragment in the chamber.

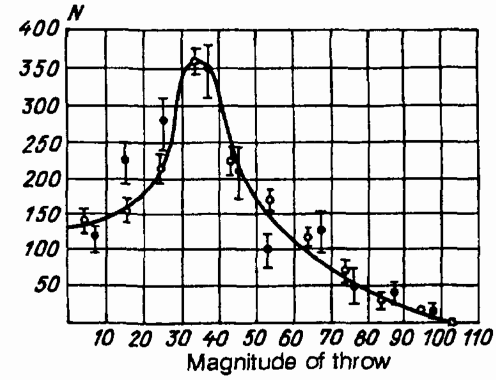

FIG. 5. Dark symbols—number of galvanometer throws of a given amplitude in the presence of a (Rn-Be)-source, light symbols—number of galvanometer throws of a given magnitude in the absence of a source. N is the number of throws.

In spite of the fact that the system consisting of the ionization chamber and the amplifier was nonlinear, this method could be used for comparing the distribution over the magnitudes of the pulses from the fragments obtained as a result of irradiating uranium by (Rn+Be) neutrons and the spontaneously appearing pulses. The coincidence of both curves of Fig. 5 not only is an extra proof of the fragment nature of the spontaneous pulses, but at the same time points to the not such a great difference in a number of characteristics of the fragments obtained in both forms of fission,-the masses of the fragments, the energy and the effective ranges.

A number of experiments was carried out using the ionization chamber on the plates of which thorium oxide was deposited instead of uranium oxide. On the basis of the negative results of experiments and of estimates of the sensitivity of the method one could have determined only the lower bound for the half-life of thorium fission. The half-life decay period turned out to be $2\cdot 10^{18}$ years. These results and also the data on the occurrence of the uranium isotopes U^{238}, U^{235}, U^{234} and the experimentally obtained information on the degree of instability of nuclei as a function of their Z and A enable one to propose a hypothesis that it is one of the lighter isotopes of uranium, apparently U^{234}, that undergoes spontaneous fission.

For a more exact determination of the half-life for the decay of uranium by spontaneous fission a series of experiments was conducted with layers of uranium oxide of different thickness. It turned out that for a layer of uranium oxide with a surface density of 1.4 mg/cm^2 it was possible to assume that all the cases of fission of uranium nuclei were recorded. The half-life for the decay of uranium calculated for all the atoms turned out to be equal to $(4\pm 1)\cdot 10^{16}$ years.

An estimate of the length of effective range of the fragments obtained spontaneously from experiments with layers of uranium oxide of different thickness gave a value of the order of 5 mm of air, which is in good agreement with the measured effective range of fragments under the conditions of the large ionization chamber.

We have carried out a number of experiments at a depth of 50 m underground at one of the stations of the Moscow subway. These experiments were designed to provide final proof of the impossibility of ascribing the effect observed by us to the spontaneous fission of one of the components of cosmic rays. In the underground experiments we obtained results similar to the results obtained previously in Leningrad at sea level. The intensity of all the known components of cosmic rays should have decreased at this depth by not less than a factor of 40. Consequently the absence of a noticeable change in the number of spontaneous acts of decay at this depth once again points to the impossibility of explaining the effect of cosmic rays.

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