SOVIET PHYSICS JETP VOLUME 11, NUMBER 2 AUGUST, 1960

ON THE MECHANISM OF URANIUM FISSION INDUCED BY SLOW NEGATIVE MUONS

G. E. BELOVITSKIĬ, N. T. KASHCHUKEEV, A. MIKHUL, M. G. PETRASHKU, T. A. ROMANOVA, and F. A. TIKHOMIROV

Joint Institute for Nuclear Research; P. N. Lebedev Physics Institute, Academy of Sciences, U.S.S.R.

Submitted to JETP editor August 14, 1959

J. Exptl. Theoret. Phys. (U.S.S.R.) **38**, 404-408 (February, 1960)

Photographic emulsions were employed to study the possibility of uranium fission induced by direct transfer to the nucleus of the energy liberated in the 2p — 1s mesic-atom transition. The upper limit of the probability for fission by this mechanism is ~ 0.01. Uranium fission induced by μ^- mesons is due mainly to nuclear capture of the μ^- meson, the probability of which is of the order of 0.07. Arguments are presented in favor of the notion that the mesic-atom 2p — 1s transition in uranium is in part non-radiative.

THE fission of uranium by slow negative muons can occur in at least two different ways.

1. By nuclear capture of the μ^- meson according to the reaction $p + \mu^- \rightarrow n + \nu$. According to theoretical[1] and experimental[2] data, the average excitation energy of the heavy nucleus in the capture of a slow μ^- meson is on the order of 15 or 20 Mev, enough to fission a uranium nucleus.

2. The fission of uranium by negative muons is possible also by direct transfer of energy to the nucleus in mesic-atom transition of the negative muon from the state 2p into the state 1s, in which an energy of 6.3 Mev is liberated. This fission mechanism was considered in detail in Zaretskiĭ's theoretical paper.[3] According to him, the probability of non-radiative energy transfer to the uranium nucleus in the 2p — 1s transition is ten times greater than the probability of radiative transition. Since the lifetime of the negative muon on the K shell of the nucleus is much longer than the lifetime of the excited nucleus, the nuclear capture of the μ^- meson can be preceded by a breakup of the excited nucleus in some manner or another (fission, emission of a neutron or γ^- quantum, etc). Fission will be possible, however, only if the energy of this transition in uranium is greater than the energy of the fission threshold for the state in which the μ^- meson is on the K shell of the nucleus.

In this method of fission, the μ^- meson is not absorbed by the uranium nucleus, and is captured after fission on the orbit of one of the fragments, as a rule the heavier one. The meson is subsequently either absorbed by the fragment, or else is discarded from the excited fragment because of the internal-conversion mechanism (in a small number of events, a $\mu - e$ decay should be observed at the end of the fragment range). If the second possibility is realized, then the negative muon ejected from the fragment can again cause fission of the next uranium nucleus, i.e., conditions are created for a catalytic fission reaction. The probability of emission of a negative muon from the excited fragment by means of the mechanism of internal conversion was estimated by Zaretskiĭ[3] and was found to be 0.25.

When this investigation was begun, very scanty data on the fission of uranium nuclei by negative muons were published in the literature. John and Fry[4] estimated, on the basis of seven fission events, the fission probability and found it to be 0.07. Galbraith and Whitehouse,[5] who used pure uranium samples irradiated by negative muons from cosmic rays, estimated the upper boundary of the fission probability, found to be < 0.25. These papers, therefore, did not yield any information on the possibility of non-radiative fission (fission due to energy liberated in the non-radiative mesic-atom transition 2p — 1s).

In this connection, we set up experiments to ascertain the existence of such a mechanism of uranium fission. A confirmation of such a process would be the observation of conversion μ^- mesons or of heavy charged particles (p, α) and electrons from $\mu - e$ decay, emerging from the stopping point of the fragment.

1. EXPERIMENTAL PART

To observe the fission of uranium nuclei by slow μ^- mesons we used NIKFI type "R" photographic plates, 200 — 250 μ thick, impregnated with uranyl acetate. The loading and development procedure enabled us to introduce up to 1.5×10^{20} uranium nuclei into a cubic centimeter of emulsion with uniform development of the emulsion over the entire depth. Some of the plates were treated to make the tracks of relativistic particles invisible, while the remainder retained their sensitivity to relativistic particles, a fact that could be monitored by the presence of $\mu - e$ decays. The number of uranium nuclei introduced into the emulsion was determined by counting the α particles from the natural radioactivity of uranium. This quantity, averaged over all the experiments, was found to be 1.7×10^{20} cm^{-3}. The plates were irradiated in the synchrocyclotron of the Joint Institute for Nuclear Research by a beam of slow μ^- mesons, obtained by slowing down the initial negative-pion beam, of energy ~ 160 Mev, in a copper absorber 11.5 cm thick. The admixture of negative pions (~ 1%) was determined from the number of stars with three or more prongs, due to the stopped mesons.

A total of 738 fission events was found, of which 520 were in plates insensitive to relativistic particles, and 218 in relativistic plates. The plates were scanned at an overall magnification of 300×. The fission events detected were analyzed at a magnification of 2000 ×. The accuracy of measurement of the range was ±1 μ. On the basis of 397 fission events, obtained from 271,600 negative muons stopped in the emulsion, we calculated the fission probability P_f of uranium by negative muons, using the formula

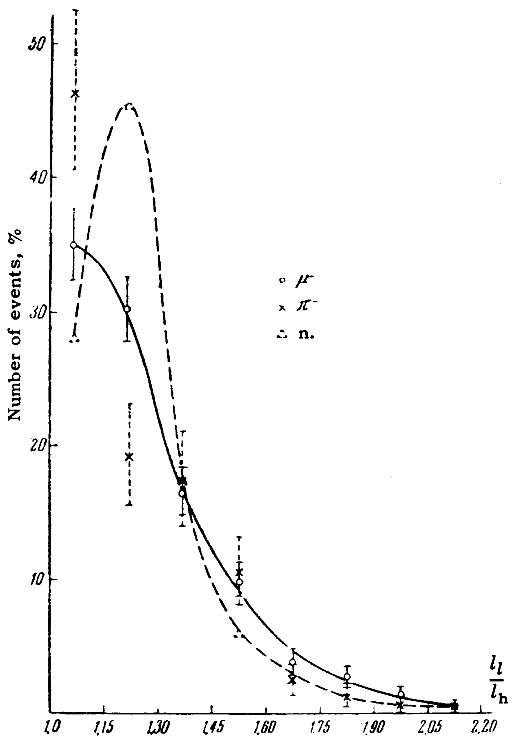
$$P_f = \frac{n_f}{0.4 S_\mu N_U Z_U / (N_U Z_U + \Sigma N_i Z_i)},$$

where n_f is the number of fissions, S_μ the number of stopped μ^- mesons, N_U the number of uranium nuclei, N_i the number of carbon, oxygen, or nitrogen nuclei per cubic centimeter of emulsion, contained in the uranium-impregnated gelatin, Z is the nuclear charge, and the factor 0.4 arises because 40% of the negative muons are stopped in the uranium-containing gelatin (see references 6 and 7). The probability of capture of a negative muon by the uranium was calculated from the well known composition of the NIKFI emulsion and the assumption that the Fermi-Teller law[8] holds, whereby the capture of the μ^- meson by the various nuclei contained in the gelatin is proportional to Z.

The probability of uranium fission by μ^- mesons calculated under these assumptions was found to be 0.070 ± 0.008.

Recent experimental investigations[9,10] have shown that in chemical compounds, such as Al_2O_3, SiO_2, AgCl, UF_4, etc, the capture of negative muons is proportional to the number of atoms in the molecule. If this result is applied to our case (gelatin plus uranium), then a quantity greater than unity is obtained for the fission probability, which is absurd. The foregoing is confirmed also by the results of Galbraith and Whitehouse,[5] who used pure uranium specimens, and who obtained a value less than 0.25 for the fission probability. An even more conclusive deduction is made in reference 11, where the photo method was used, in which it is shown that the capture of negative mesons in the (gelatin plus uranium) medium follows more readily the Fermi-Teller law. Thus, the Fermi-Teller law does not distort the results greatly, if at all.

To obtain information on the excitation energy of uranium nuclei fissioned by negative muons, we measured the range of each fragment. This could be done because the track of the negative muon usually makes it possible to determine the point where the fission occurred.



The diagram shows data on the degree of asymmetry of the fission of uranium induced by negative muons (the abscissas represent the ratio of the ranges of the light and heavy fragments, and the ordinates represent the relative number of such events, in percent). For comparison, the same figure shows analogous data for fission of uranium by slow neutrons[12] and slow negative pions.[12,13]

2. DISCUSSION OF THE FISSION MECHANISM

The experimental data we obtained lead to the conclusion that if the process of non-radiative fission of uranium does indeed take place, its probability is ~ 0.01, i.e., more than one order of magnitude smaller than computed.[3] This statement is based on the following facts:

a) Were the fissions observed due to non-radiative transition, then the emission of heavy (p, α) charged particles from the end of the fragment track would be observed in approximately ten cases, and in eight cases electrons from $\mu - e$ decay would be observed. We did not observe a single event of this kind.

b) In the presence of non-radiative fission, emission of conversion μ^- mesons is possible. If the meson energy is less than 1 Mev, the emission of such a meson can be detected from the $\mu - e$ decay. Approximately ten such events were expected, but not a single one was observed among the 228 fission events found in the relativistic plates.

c) A comparison of the distributions of the light to heavy fragment-range ratio in the fission of uranium by μ^- mesons (see the figure) with analogous data for the fission induced by slow neutrons (excitation energy ~ 6 Mev) and slow negative pions (excitation energy ~ 60 Mev) indicates that uranium is fissioned by μ^- mesons essentially at an excitation energy considerably greater than 6 Mev. Were a noticeable role to be played by non-radiative fission, the character of the fragment-range asymmetry in the fission by μ^- mesons (excitation energy ~ 6 Mev) would be similar to that induced by slow neutrons.

Finally, the difference between the probabilities of fission of Th^{232} and U^{238} (0.018 and 0.07) under the influence of μ^- mesons, as reported in reference 14, also apparently indicates that the fission is not via non-radiative excitation. At an excitation energy of 6.3 Mev, the probability of fission of Th^{232} should not be considerably less than that of uranium.

As shown by later calculations (D. F. Grechukhin, private communication), so small a probability of non-radiative uranium fission by negative muons, compared with that previously obtained,[3] is apparently due to the fact that when the μ^- meson lands on the K shell the potential barrier for fission increases by ~ 1 Mev, which should reduce the fission probability by several orders of magnitude. Thus, the fission of U^{238} by negative muons is apparently due essentially to nuclear capture of the μ^- meson. This results in an excited Pa^{238} nucleus with an excitation spectrum from 0 to 20 Mev.[1,2] The probability of fission of Pa^{238} at such excitation energies can be calculated from the empirical formula:[15]

$$\frac{P_f(\mathrm{Pa}_{91}^{238})}{P_f(U_{92}^{238})} = 1.3\left[\frac{Z^2}{A} - 34.7\right], \qquad (1)$$

where $P_f(Pa^{238})$ and $P_f(U^{238})$ are the fission probabilities of Pa^{238} and U^{238} at equal excitation energies. This formula is applicable to nuclei with $Z \geq 90$ at least at an excitation energy 8 — 12 Mev.

To calculate $P_f(U^{238})$ we used the experimental data on the uranium fission probability under the influence of neutron[16] and gamma rays[17] at excitation energies which obtain in the Pa^{238} when a U^{238} nucleus captures a μ^- meson. The fission probability $P_f(U^{238})$, averaged over the entire excitation spectrum obtained by Kaplan et al.[2] was found to be 0.27 ± 0.02. Substituting this quantity in the above formula, we obtain $P_f(Pa^{238}) \approx 0.03$, which is less than the value 0.07 obtained in our experiments.

Thus, if (1) is correct in our case, then to explain the experimental value of $P_f(Pa^{238})$ it is necessary to assume the existence of another channel, by which fission takes place. As established in our investigation, the transition 2p — 1s of the negative muon into a mesic atom of uranium, if it is non-radiative, does not lead in a noticeable number of cases to uranium fission. The excited nucleus that is produced thereby apparently emits a neutron (binding energy of the last neutron in uranium is 6 Mev). Consequently, this nuclear capture of the negative muon occurs already in U^{237} with formation of Pa^{237} and its subsequent fission.

Using (1) again, we obtain for the probability of Pa^{237} fission a value of 0.08, close to the experimental value. Considering, however, the possible inaccuracy of the formula at excitation energies greater than 12 Mev, and the inaccuracy of the other quantities used in these estimates, we cannot draw a final conclusion regarding the extent to which the transition 2p — 1s is non-radiative.

We have recently become acquainted with the results of reference 18, in which it was established, by measuring the number of γ quanta with energy > 6 Mev per capture of a μ^- meson, that the 2p — 1s transition in the mesic atom of uranium is non-radiative with a probability ~ 0.5, confirming our conclusion regarding the mechanism of uranium fission by μ^- mesons. The presence of a non-radiative transition 2p — 1s and the small fission probability ($P_f = 0.07$) allow us to conclude that when the μ^- meson lands on the K shell of the uranium nucleus, the fission barrier increases by more than 0.2 Mev.

The authors express their gratitude to Professor V. P. Dzhelepov and I. M. Frank for constant interest in the work and for useful discussion, and also to I. Ya. Barit and A. E. Ignatenko for help in the work, and to the laboratory staff for participating in scanning the photographic plates.

Three of us (N. T. Kashchukeev, A. Mikhul, and M. G. Petrashku) are also grateful to V. M. Sidorov for help in this work.

[1] V. A. Wheeler, Revs. Modern Phys. **21**, 133 (1949).

[2] Kaplan, Moyer, and Pyle, Phys. Rev. **112**, 968 (1958).

[3] D. F. Zaretskiĭ, Тр. Второй Международной конф. по мирному использованию атомной энергии. Доклады советских ученых (Second Geneva Conference; Papers by Soviet Scientists), Vol. 1, 1959, p. 462.

[4] W. John and W. F. Fry, Phys. Rev. **91**, 1234 (1955).

[5] W. Galbraith and W. J. Whitehouse, Phil. Mag. **44**, 77 (1953).

[6] O. V. Lozhkin and V. P. Shamov, JETP **28**, 739 (1955), Soviet Phys. JETP **1**, 587 (1955).

[7] W. F. Fry, Phys. Rev. **85**, 675 (1952).

[8] E. Fermi and E. Teller, Phys. Rev. **72**, 399 (1947).

[9] Sens, Swanson, Telegdi, and Jovanovith, Nuovo cimento **7**, 536 (1958).

[10] Backenstoss, Bloch, Chidley, Reiter, Romanowski, Siegel, and Sutton, Bull. Am. Phys. Soc. **4**, No. 4, Sec. T, 1959.

[11] G. E. Belovitskiĭ, JETP **38**, 658 (1960), this issue p. 473.

[12] V. P. Shamov and O. V. Lozhkin, JETP **29**, 286 (1955), Soviet Phys. JETP **2**, 111 (1956).

[13] Belovitskiĭ, Romanova, Sukhov, and Frank, JETP **28**, 729 (1955), Soviet Phys. JETP **1**, 581 (1955).

[14] G. M. Petrashku and A. K. Mikhul, Dokl. Akad. Nauk SSSR **126**, 752 (1959), Soviet Phys.-Doklady **4**, 628 (1959).

[15] Huizenga, Gindler, and Duffield, Phys. Rev. **95**, 1009 (1954); J. R. Huizenga, Phys. Rev. **109**, 484 (1958).

[16] Knight, Smith, and Warren, Phys. Rev. **112**, 259 (1958).

[17] Lazareva, Gavrilov, Valuev, Zatsepina, and Stavinskiĭ, Сессия АН СССР по мирному использованию атомной энергии (заседания отд. физ.-мат. наук). (Session of U.S.S.R. Acad. Sci. on Peaceful Uses of Atomic Energy, Div. of Phys.-Math. Sciences), Acad. Sci. Press, 1955, p. 306.

[18] Blats, Kondrat'ev, Landsberg, Lebedev, Obukhov, and Pontecorvo, Preprint R-448 Joint. Inst. for Nucl. Res., Dubna, 1959.

Translated by J. G. Adashko
89