Fission of Nuclei under the Influence of Accelerated Nitrogen Nuclei

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Experiments on fission induced in U^{235}, U^{238}, Bi, Au, Re, and Yb by accelerated nitrogen ions are described. The measurements were conducted with the aid of an ionization chamber. Graphs are presented showing the dependence of the fission cross sections for these nuclei on the nitrogen ion energy in the energy range from 70 to 110 Mev. The results obtained are analyzed from the viewpoint of influence of the parameter Z^2/A and of the excitation energy on the probability of fission.

1. INTRODUCTION

IN RECENT YEARS experiments have been conducted to investigate the fission of nuclei by light particles of high energy. These experiments have established many characteristics of such fission. It has been shown that in general there is symmetrical fission of the nucleus, and that the probability of fission is strongly dependent on the parameter Z^2/A (where Z is the nuclear charge, and A is the atomic weight). Nevertheless, it is difficult to obtain quantitative data on the dependence of the probability of fission on the excitation energy of the nucleus with various values of Z^2/A at high energies, since a cascade process occurs when the nuclei interact with high energy particles. This nuclear cascade process makes the nuclear excitation energy appear to be rather small and almost independent of the initial energy of the particle.

Calculations based on the Monte Carlo method and experimental data[1] indicate that a major part of the ejected nucleons in the nuclear cascade are protons. This leads to a large indeterminacy in the value of Z^2/A which is present after the nuclear cascade of the excited nucleus. Furthermore, this indeterminacy grows larger with increasing incident particle energy.

In light of the above it appears that quantitative data concerning the fission process can be obtained by the use of heavy accelerated ions such as nitrogen, oxygen, etc. Nuclear cascades do not appear in reactions produced by heavy incident nuclei, and the excitation energy of the resulting compound nucleus can be accurately determined if the energy of the incident particle is known. Thus, it is possible to change the excitation energy of the compound nucleus over a wide range by varying the energy of the incident particle. Also, the value of the parameter Z^2/A of the excited nucleus can be determined accurately.

Using nuclei with $Z < 83$ for targets, and employing nitrogen or oxygen nuclei as bombarding particles, one can study the fission of nuclei over a wide range of the parameter Z^2/A. It is worth noting that the study of fission in such nuclei as U, Np, Pu, etc., using accelerated heavy ions, is also of interest, since in these cases there can occur several specifically characteristic interactions of accelerated ions with the nuclei. Thus, it is presently well known that heavy ions, accelerated to moderate energies, can be successfully used for Coulomb excitation of the rotational levels of nuclei. When the accelerated heavy ions have sufficiently large energies, it will be found that in a certain number of cases the excitation energy of the nucleus exceeds the fission threshold because of the Coulomb interaction.

We give below experimental results on the fission cross sections of U^{235}, U^{238}, Bi, Au, and Yb under the influence of accelerated nitrogen ions, with respect to the energy of the nitrogen nuclei.

2. EXPERIMENTAL TECHNIQUE

Production of Accelerated Nitrogen Nuclei and Measurement of Their Energy

The accelerated nitrogen ions were produced in a cyclotron having pole pieces 150 cm. in diameter. The cyclotron was designed for the acceleration of ions with charge 5, produced directly in an ion source developed by P. M. Morozov, B. N. Makov and M. S. Ioffe. The nitrogen ion energy was 115 Mev at the final cyclotron radius. The measurements were carried out using the external beam in a cabin 12 meters away from the accelerator. Although no special measures were taken in bringing the

beam out of the cyclotron chamber, a small portion of the accelerated ions of energy 70 to 115 Mev entered the outlet channel. Before the ion beam hit the target, it went through an aluminum foil 6 microns thick (which "stripped" the ions to a +7 charge state) and through a magnetic analyzer. For a given current, the magnetic analyzer would let through only ions of the corresponding fixed energy.

The ion beam intensity impinging on the target varied between 10^5 and 10^6 ions/second, depending on the ionic energy and state.

Recording of Fission Fragments

An ionization chamber was used to record the fission fragments. The nitrogen nuclei were admitted into the chamber through a thin metal foil window. The passage of nitrogen nuclei through the chamber raises the problem of distinguishing fission-fragment pulses from the pulses of nitrogen nuclei. Although the fission-fragment pulses are significantly larger than the nitrogen pulses, accidental superposition of the latter can produce pulses resembling those of fission fragments. A special chamber was therefore designed to permit strong attenuation of the nitrogen pulses. The chamber consisted of two compartments, separated by a thin metal foil (5 microns of nickel) which served as the collecting electrode. Both fission fragments and nitrogen nuclei caused ionization in the first compartment, but only nitrogen nuclei caused ionization in the second compartment. The dimensions of the compartments were so chosen that the nitrogen nuclei gave equal amounts of ionization in the two compartments.

The collecting electrode was at a potential of +200 volts with respect to the chamber casing. A supplementary electrode placed in the second compartment was at a potential of +400 volts, so that an electric field existed in the second compartment of equal magnitude but opposite direction to that in the first compartment. Thanks to this, the potentials induced on the collecting electrode as the result of the ionization caused by nitrogen ions in the first compartment were almost completely offset by the potentials of opposing sign induced by nitrogen ions in the second compartment. This scheme of compensation permitted reliable isolation of the fission-fragment pulses.

The chamber was filled with argon at a pressure of one atmosphere. A schematic drawing showing the construction of the chamber is given in Fig. 1.

The pulses from the chamber were passed to the grid of the first stage of a high-frequency linear amplifier, and after amplification, were recorded by a mechanical counter. A sample of typical results obtained with this chamber is given in Fig. 2.

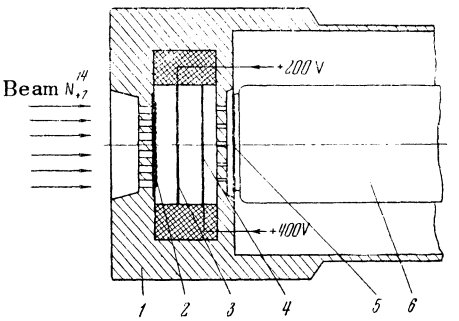


FIG. 1. Ionization chamber. 1 – chamber casing; 2 – investigated target material; 3 – collecting electrode; 4 – supplementary electrode; 5 – layer of ZnS; 6 – photomultiplier tube.

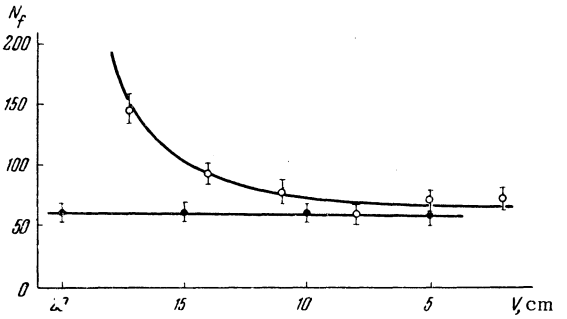


FIG. 2. Dependence of fission pulse count on the discriminator bias. ⊖ – for Re with nitrogen nuclei; ⊖ – for U^{235} with thermal neutrons.

Recording of Nitrogen Ions

In order to determine the absolute fission cross section and to investigate the dependence of the fission cross section on the energy of the nitrogen ions, it is necessary to record the number of nitrogen ions which hit the target. With this aim in mind, the front and back walls of the chamber were made of thin metal foils, permitting the passage of the ion beam. A photomultiplier tube coated with a layer of ZnS was placed in contact with the back wall of the chamber to count the number of nitrogen ions. To decrease the loading on the photomultiplier, a collimator was placed in front of the ZnS. This collimator reduced the incident intensity by a factor of 800. After additional amplification, the pulses from the photomultiplier tube were passed to a counter having a discriminator. The system had a good plateau which stayed steady over a wide range of dis-

criminator settings, and did not change with the passage of time. In a special experiment using alpha particles, it was established that the counting efficiency was practically 100%.

Targets

The targets consisted of discs of either aluminum 14 microns thick or nickel 6 microns thick, coated with a layer of the material being investigated (the diameter of the coated area was 30 millimeters). The gold and bismuth targets were prepared by diffusion of the metal in vacuo onto an aluminum foil; the targets of uranium and of a natural isotopic mixture of rhenium were diffused onto nickel, and the ytterbium targets were prepared on aluminum by electrolytic means.

The quantity of material on each target was determined by weighing (2 – 4 milligrams); in the case of U^{238} it was also determined by the emitted alpha-activity. The results by weighing in the case of U^{238} agreed satisfactorily with the results obtained by measuring alpha activity.

Experiments with the Internal Beam

The principal experiments were conducted with the external cyclotron beam. However, experiments were also carried out with the internal beam for U^{238}. In this case only the relative change of the fission cross section with respect to the energy of the nitrogen nuclei was studied. Fission fragments leaving the irradiated layer of uranium were gathered on tantalum plates which were positioned at an angle of 150 degrees with respect to the direction of the nitrogen ion beam. After a certain irradiation time, the beta-activity of the tantalum plates was measured. This beta-activity was proportional to the fission cross section. (It was assumed that for the energy interval 70 to 110 Mev of the nitrogen ions, the spectrum and angular distribution of the uranium fission fragments did not change materially. The agreement between results obtained with the internal and with the external beam supported this assumption.)

The thin uranium target (~0.6 mg/cm^2) was attached directly to the current collector. This permitted the determination of the integral of the nitrogen ion beam current over the time of irradiation. The irradiation was carried out at a radius of 67 cm., and the measurement of the energy was accomplished with the aid of a nickel foil placed in front of the target. The experiments showed that the tantalum collecting plate gathers beta-active nuclei as well as fission fragments; these beta-active nuclei do not appear to be fission products, and are of only short duration. Apparently they are produced by the interaction of nitrogen ions with nickel, and reach the tantalum collectors by scattering. To avoid such contamination, the tantalum collectors were covered with nickel foils 2 microns thick. To exclude any activity being generated in the tantalum collecting plate by neutrons, an identical background plate was installed.

3. EXPERIMENTAL RESULTS

As a result of the experiments there were obtained data on the fission cross sections of U^{235}, U^{238}, Bi, Au, Re and Yb for various nitrogen ion energies. These data are presented graphically in Figs. 3 and 4.

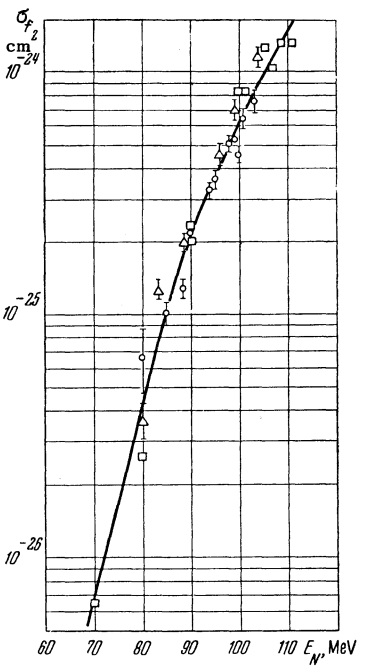


FIG. 3. Dependence of the fission cross section on energy of the nitrogen nucleus: Δ – U^{238} with external beam; □ – U^{238} with internal beam; O – U^{235} with external beam.

The statistical errors of the measurements are shown on the graphs. Naturally, the absolute values of the cross sections are found with less accuracy, owing to errors in measuring the amount of material on the targets, in the counting efficiency for the nitrogen ions, etc. Analysis of the effect of these factors on the accuracy of the absolute val-

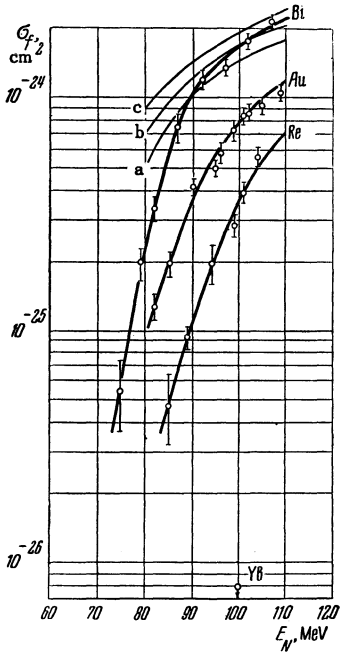


FIG. 4. Dependence of the fission cross section on the nitrogen nucleus energy for Bi, Au, Re and Yb.

ues of the fission cross sections shows that the maximum error does not exceed 15%.

4. DISCUSSION OF RESULTS

To analyze the experimental data one must know the mechanism of interaction between the nitrogen ions and the heavy nuclei; namely, one must know whether the process involves basically complete fusion of the nitrogen and target nuclei, or whether it involves particulate penetration of the target nucleus by the nitrogen nucleus. The first case leads to a highly excited state of the target nucleus, with a definite excitation energy and a known value of the parameter Z^2/A. In the second case, the excitation energy and the parameter Z^2/A are indeterminate, so that interpretation of the results is more complicated.

Experimental investigations[2,3] of the interactions of multi-charged accelerated ions with heavy elements show that the basic process involves complete fusion of the interacting nuclei. From analysis of our experimental results we have concluded that the fission process is preceded by the formation of a highly excited nucleus with a known value of the excitation energy and of the parameter Z^2/A.

In the determination of the nuclear excitation energy, we took account of the nuclear recoil energy and of the change in the nuclear binding energy between the initial and final states of the nucleus. The latter correction was computed by the Weizsäcker formula

$$-E_B(Z, A) = -14.0\,A + 13.1\cdot A^{2/3} + \frac{77.3}{A}\left(\frac{A}{2} - Z\right)^2 + 0.584\,\frac{Z^2}{A^{1/3}} + \begin{cases} +34 & \text{for } Z \text{ odd, } A \text{ even} \\ -A^{3/4} & \text{for } Z \text{ even, } A \text{ even} \\ 0 & \text{for } A \text{ odd} \end{cases}$$

and amounted to 12, 17, and 22 Mev for rhenium, gold, and bismuth respectively.

Experimental investigation of the interaction of accelerated nitrogen ions with U^{235} and U^{238} nuclei showed that non-fission reactions occur with significantly lower probability than do fission reactions (by a factor of about 100). Therefore one can state that the curve given in Fig. 3 for U^{235} and U^{238} represents the dependence of the cross section for compound nucleus formation on the nitrogen ion energy. Since the probability of formation of the compound nucleus depends on the size of the Coulomb barrier, comparison of the curves for U^{235} and U^{238} indicates that the Coulomb barriers for these two nuclei are identical within the limits of experimental error.

The given results permit an estimate of the upper limit of the cross section for uranium fission resulting from Coulomb excitation by interaction with accelerated nitrogen ions. For a nitrogen ion energy of 70 Mev, this value is 7×10^{-27} cm.

The experimental results for Bi, Au, Re and Yb (Fig. 4) can be analyzed in order to clarify the influence on the probability of fission of the parameter Z^2/A and of the excitation energy which results from the nuclear reaction. The fission cross section is related to the probability of fission by the formula

$$\sigma_f = \sigma_c \cdot W_f, \qquad (1)$$

where σ_f is the fission cross section, σ_c is the cross section for formation of the compound nucleus, and W_f is the probability of fission. It can be seen from the formula that it is necessary to know both the cross section for fission of the excited nucleus and the cross section for formation of the compound nucleus in order to determine the probability of fission. Experimental investigation of the products of non-fissioning interactions of nitrogen nuclei with bismuth[4] show that for bismuth, as for uranium, the

fission cross section practically coincides with the cross section for formation of a compound nucleus. For incident particle energies $E > 1.2\,E_{exc}$ (where E_{exc} is the nuclear Coulomb barrier energy) the cross section for formation of a compound nucleus agrees[5] to within 15% with the cross section calculated by the formula

$$\sigma_c \approx \pi (R_1 + R_2)^2 \left[1 - (E_B / E)\right], \qquad (2)$$

where R_1 is the radius of the target nucleus and R_2 is the radius of the nitrogen nucleus. Moreover, the Coulomb barrier of the nucleus is found by the formula

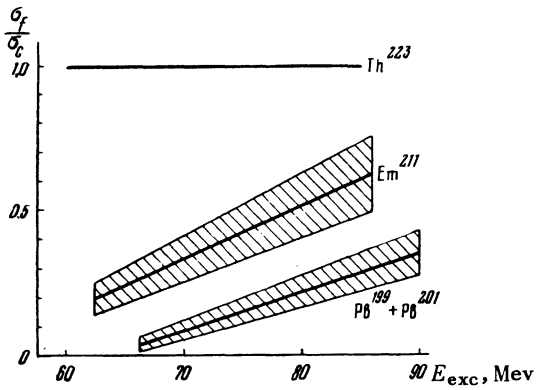
$$E_{exc} = \frac{Z_1 Z_2}{r_0 \left(A_1^{1/3} + A_2^{1/3}\right)} \frac{(A_1 + A_2)}{A_1}, \qquad (3)$$

where Z_1 is the charge of the target nucleus, Z_2 is the charge of the nitrogen nucleus, A_1 is the atomic weight of the target nucleus and A_2 is the atomic weight of the nitrogen nucleus.

Fig. 4 shows the curves *a*, *b*, and *c*, calculated by formula (2) for Bi with values of r_0 equal to: 1.5×10^{-13} cm for curve *a*; 1.55×10^{-13} cm for curve *b*; and 1.6×10^{-13} cm for curve *c*. It is obvious that the theoretical curve calculated with $r_0 = 1.55 \times 10^{-13}$ cm best fits the experimental data for energies above 90 Mev.

The cross sections for formation of the compound nucleus for rhenium in the energy region above 85 Mev, and for gold above 90 Mev, were calculated by formula (2) using $r_0 = 1.55 \times 10^{-13}$ cm.

Figure 5 shows the dependence on the excitation energy of the probability of fission for the excited nuclei Th^{223} ($Z^2/A = 36.3$), Em^{211} ($Z^2/A = 35$) and for an isotopic mixture of Pb^{199} and Pb^{201} (average value of $Z^2/A = 33.6$), resulting from the complete fusion of the nitrogen nuclei with the nuclei of Bi, Au, and Re. Figure 6 shows the dependence of the probability of fission for the excited compound nucleus on the parameter Z^2/A with the given values of the excitation energy. It can be seen from the graphs that the probability of fission is strongly dependent on the value of Z^2/A of the excited nucleus.



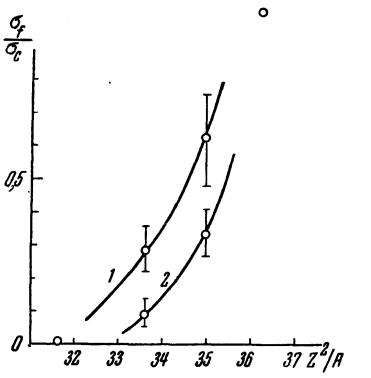


FIG. 5. Dependence of the probability of fission on the excitation energy E_{exc} for nuclei with various values of the parameter Z^2/A.

FIG. 6. Dependence of the probability of fission on the parameter Z^2/A for two different excitation energies: I—86 Mev; II—70 Mev.

The resulting values of the probability of fission fully determine the total probability of fission for the excited nuclei, but do not permit the establishment of the relation between high-temperature and emissive fission; that is, the relation between the number of fissions occurring upon the initial excitation of the nucleus and the number of fissions occurring after the first emission of several neutrons. To establish this relation, further investigations of the properties of strongly excited nuclei is necessary. Such an investigation, in combination with the data on probability of fission for the excited nuclei, will permit a more detailed study of the dependence of the relation Γ_f/Γ_n (Γ_f being the fission width and Γ_n being the neutron width) on the excitation energy and on the parameter Z^2/A.

We are indebted to Academician I. V. Kurchatov for many valuable comments made during discussion of the present work. To a group of associates led by Iu. M. Pustovoi we give our thanks for good cyclotron operation during our experiments.

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Structure of Superconductors. X Thermal, Microscopic and X-ray Investigation of the Bismuth–Palladium System

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A more accurate determination of the phase composition of alloys containing 43 – 54% by weight Pd is made, and the behavior of these alloys at low temperatures is explained. It is established that increase in the minimum interatomic distances in superconducting compounds of bismuth with palladium leads to an increase in T_c.

INVESTIGATION OF THE ALLOYS of bismuth with palladium has been undertaken in connection with the study of superconductivity, in view of the fact that certain alloys of this system become superconductors at low temperatures[1–4]. In this system superconductivity is thus manifested by compounds formed by two non-superconducting elements: bismuth and palladium. From previous microscopic and X-ray investigations of bismuth–palladium alloys, as well as from determinations of the melting temperatures for a series of these alloys. a tentative fusion diagram for the bismuth–palladium system has been constructed.[5] The crystalline structures of α- and β-Bi_2Pd and of BiPd have been determined,[6–9] and the low-temperature behavior of variously heat-treated alloys has been explained.[4,5,10] The present study was undertaken with the object of determining whether there is any connection between the atomic-crystalline structure of these alloys and their superconducting properties and with the further object of determining more precisely the phase composition of certain alloys and explaining their behavior at low temperatures.

THERMAL ANALYSIS

Chemically pure bismuth and palladium were employed in the preparation of the alloys; melting was accomplished in a resistance oven, in quartz capsules under an argon atmosphere. The heating and cooling curves were recorded with the aid of an automatic electronic potentiometer EPP-09 to temperatures of 200 – 400° C. The weights of the alloy samples ranged between 0.5 and 2 g. One curve having been recorded, the requisite amount of bismuth or palladium was added to the alloy; the latter was maintained for some time at an elevated temperature, and then the next curve was taken. Curves were recorded in this manner over a certain concentration interval (10 – 20% by weight), following which the alloy was replaced by a freshly-prepared mixture of bismuth and palladium, to avoid the accumulation of errors in the concentration.

The results obtained in this thermal analysis are represented graphically in Fig. 1. As palladium is added to the bismuth, the liquefaction temperature falls along *AB*. Beginning with ~ 5% by weight Pd, the liquefaction curve rises progressively along *BC*, and at 20.3% by weight Pd reaches a transition point at a temperature of 497°; at this temperature the compound Bi_2Pd liquefies with decomposition. The next branch of the liquefaction curve, *CDE*, beginning at 20.3% by weight Pd, rises as the palladium content of the alloys increases and reaches its peak at 33.8% by weight Pd, corresponding to the compound BiPd. Further addition of palladium