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Proton-induced fission of iridium

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The isotopic distribution of rubidium produced in the proton induced fission of iridium has been measured at 68, 85, and 100 MeV incident beam energies using an on-line mass spectrometer with a surface ionization ion source. The deduced neutron multiplicities are substantially higher and show a steeper energy dependence compared to similar measurements done with a heavy ion beam in this mass region and in the same range of excitation energies. This might confirm an effect of the angular momentum of the fissioning nucleus on the fragment deexcitation mechanism.

[NUCLEAR REACTIONS, FISSION ^{191,193}Ir(p,f), $E_p=68$, 85, 100 MeV; measured independent relative yields of Rb. Deduced total number of neutrons emitted in symmetric fission.]

I. INTRODUCTION

The fission characteristics of heavy nuclei in the actinide region have been extensively studied. The experimental observations and results of calculations have been discussed in recent reviews.[1-7] Although the fission of lighter nuclei ($A<200$) has been investigated for some time,[8] much less information is available, particularly from radiochemical methods, because of the very small fission cross sections. The fission barriers of these nuclei are high, which makes the fission process much less probable than particle evaporation. In spite of the experimental difficulties, these lighter nuclei are well suited for the investigation of the fission mechanism at higher excitation energies. As calculations show,[9] the most significant contribution to the fission yield is from the completely equilibrated excited nucleus. The fission cross section of the survivors of particle evaporation is very small. The fission process, therefore, selects events with excitation energies very close to the full energy of the incident beam. Because of the low-angular momentum involved, the proton induced fission complements the heavy ion induced fission.

The earliest studies of the mass distribution in the fission of these nuclei were made by radiochemical techniques.[10-13] In later studies the kinetic energies and mass distributions for the fission of various target-projectile systems were studied using semiconductor detectors.[13-21] Fission track detectors have been successfully used to measure the fission cross sections of some lighter nuclei.[16,22-25] In recent years, the mass spectrometric method has been used in the fission studies.[26] This method allows the selective detection of individual isotopes of alkali elements and has recently been adapted for In and Ga isotopes.[27] Reisdorf *et al.*[28] used this method to study the fission reaction ^{158}Gd(^{22}Ne,Rb)Rb. They found that the average numbers of neutrons emitted per fission event (ν_T) were significantly lower than predicted by evaporation calculations on the basis of the excitation energy of the compound system, assuming zero angular momentum transfer to the fission fragments. One reason for this discrepancy was suggested to be the high intrinsic spin of the fission fragments favoring gamma emission over neutron evaporation. Another explanation (presented as less probable) would be a possible shell effect on the total kinetic energy due to the $N=50$ fragment shells at symmetric fission.

If the first explanation is valid, then in the fission of the same compound system, formed by proton bombardment, no such discrepancy should be observed, since, in the latter case, the angular momentum of the fissioning nucleus is much smaller for the same excitation energy. The same reaction would also allow an evaluation of the second hypothesis. Ideally, this measurement should be conducted with the same experimental technique used by the Orsay-Dubna group.[28]

The ideal system to investigate would be $^{181}Ta(p,f)Rb$. However, because of the extremely low cross sections involved, no reasonable counting statistics could be obtained. Iridium was chosen because of the higher fission cross sections and because the compound nuclei are still very close to the ideal case. The ν_T values obtained can be reasonably explained by the conventional, zero angular momentum process, without any need to include shell effects. For a given excitation energy, the ν_T value is significantly higher than the corresponding quantity observed with the heavy ion beam.

II. EXPERIMENTAL PROCEDURE

The measurements were done using an on-line mass spectrometer facility. As the cross sections for the fission of nuclei in this mass region are very small (a few mb), a high efficiency target ion source assembly[27] designed by the authors (Fig. 1) was used. The basic difference between the ion source used in the present study and the one used by the Orsay-Dubna group[26] is that we used separate systems for controlling the temperatures of the ionizer foil and the target oven. The efficiency was thus increased by a substantial factor, with an appreciable decrease in the level of natural isotope contamination.

The targets were prepared by depositing natural iridium metal powder on one side of graphite disks, 12.6 mm in diameter and 28 mg/cm^2 thick. The average target thickness was 8 mg/cm^2. Twenty-five disks were used to make a target assembly. In the target-oven, the graphite disks were separated from each other by 0.3 mm thick graphite washers. The upper segment of each disk was removed to make a flat platform to accommodate the ionizer foil and its supporting frame (horn).

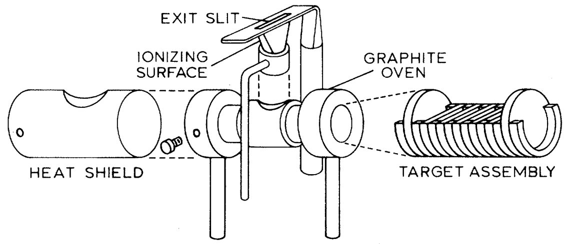

FIG. 1. Target ion source used in the present work.

The oven containing the target disks was placed in the external beam of the McGill synchrocyclotron in such a way that the proton beam traversed the target disks in a direction perpendicular to their plane. The oven was heated to 1600°C by the Joule effect, with an ac current. The fission fragments are stopped in the graphite disks and the alkali elements come out by the thermal diffusion process. While passing through the horn these alkali elements are ionized by the surface ionization mechanism. The isotopes of Rb are very selectively ionized at temperatures in the vicinity of 1000°C. The diffusion time for Rb was found to be 85 ms. The accelerating voltage was 5 kV modulated with a triangular sweep whose amplitude was adjusted to scan a five mass range in each cycle. The data accumulation was done in four groups of 1 s duration, following an irradiation of 0.4 s. The data in the last group were used for background subtraction. To obtain the relative yields of different isotopes, the measured number of counts in a given mass peak was corrected for the mass discrimination response of the spectrometer, diffusion characteristics, and radioactive decay. A detailed description of the method has been presented in a previous paper.[29]

III. RESULTS AND DISCUSSION

The isotopic distributions were measured at incident proton energies of 100, 85, and 68 MeV. Figure 2 shows a mass spectrum of Rb isotopes at 100 MeV proton energy. The relative yields of Rb

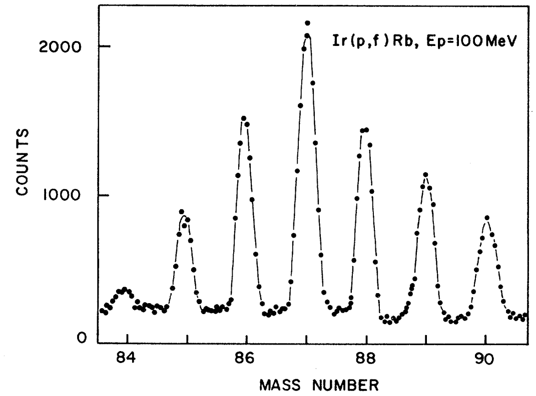

FIG. 2. Mass spectrum of Rb isotopes produced in the proton induced fission of metallic iridium at 100 MeV incident proton energies.

isotopes are shown in Fig. 3. The proton energies indicated are corrected for attenuation in the target. Two sets of data were taken. In one case, a slight contamination of ^{232}Th permitted a determination of the relative fission cross section as a function of energy, using the known[29] yields of Rb isotopes in the proton induced fission of ^{232}Th. The second measurement was done with another source-target assembly constructed with extreme care to avoid any contamination by more fissile materials. After subtracting the ^{232}Th contribution from the first set of data, excellent agreement was obtained with the second measurement (clean target). If one assumes the total (integrated) Rb cross sections to be a constant fraction of the total fission cross sections over the range of incident proton energies used, they can be compared to the same quantities obtained with computer program ALICE. This code incorporates the hybrid model of Blann[30] and takes into account precompound emission of particles. As can be seen from Fig. 4, the observed trend is very well reproduced.

The observed general characteristics of the isotopic distributions are as expected. With decreasing energy, the centroid of the distribution shifts towards the heavier mass side and the width of the distribution decreases.

The average mass of the Rb fission fragments ⟨Rb⟩ after neutron emission was taken to be the centroid of the fragment isotopic distribution. The average total number of neutrons, ν_T, emitted per fission event was evaluated assuming that the N/Z ratio of the fission fragments for the exact symmetric charge split, $Z=39$, was the same as for Rb ($Z=37$). This assumption is consistent with the unchanged charge distribution (UCD) hypothesis which holds for exactly symmetric fission.[31] Thus

$$\nu_T = A_{\mathrm{CN}} - 2\frac{\langle \mathrm{Rb} \rangle}{37}39 , \tag{1}$$

where A_{CN} is the mass of the compound nucleus.

Since we used natural iridium as the target, the value of ν_T depends on the assumption made about the relative fission cross sections of ^{191}Ir and ^{193}Ir. If one assumes equal fission cross sections for the two isotopes, then the target mass has to be taken as the atomic mass of the natural iridium. The ν_T values thus obtained are shown in column 5 of Table I and in Fig. 5. On the other hand, in a radiochemical study of the alpha particle induced fission cross section for ^{185}Re was a factor of 2.5 larger than the cross section for ^{187}Re. To examine the variation of ν_T with energy we have assumed that $\sigma_f(^{191}\mathrm{Ir})/\sigma_f(^{193}\mathrm{Ir})=2.5$ in our calculation of the total average number of neutrons (column 6, Table I).

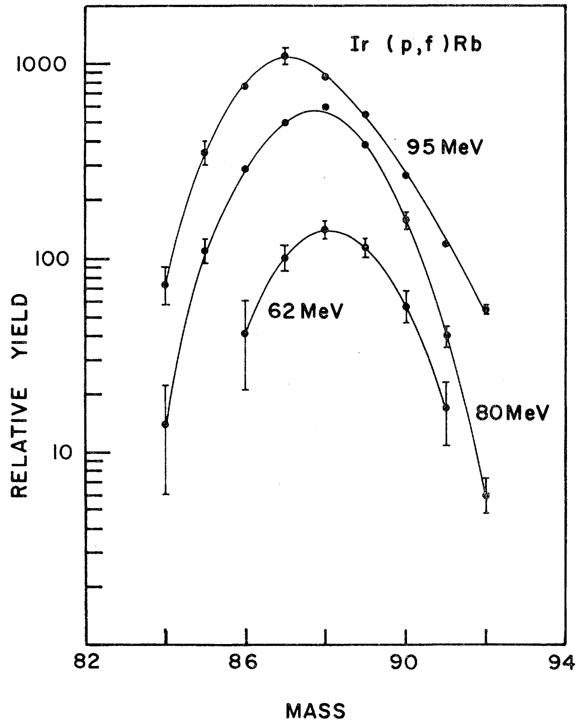

FIG. 3. Independent yields of Rb isotopes at 95, 80, and 62 MeV incident proton energies.

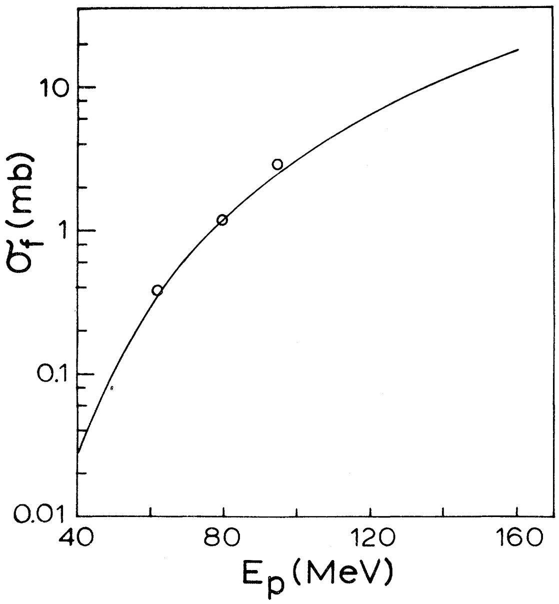

FIG. 4. Calculated (continuous line) and measured (circles) variation of relative fission cross section of iridium with incident proton energy.

TABLE I. The measured centroids and the widths of the isotopic distributions of Rb, and the deduced values of $\bar{\nu}_T$ in the fission reaction Ir(p,f)Rb.

E^*_{CN} (MeV)	$\langle A \rangle$	FWHM	$E^*_{F_1}+E^*_{F_2}$ (MeV)	$\bar{\nu}_T$ obtained by using equal σ_f for ^{191}Ir and ^{193}Ir	$\bar{\nu}_T$ (corrected for relative σ_f)
100	87.37±0.02	3.52±0.03	86	9.06±0.04	8.59±0.04
85	87.72±0.05	3.37±0.10	71	8.33±0.11	7.86±0.11
67	88.13±0.24	3.3 ±0.2	53	7.46±0.51	6.95±0.51

The excitation energy of the fission fragments (E^*_{F1} and E^*_{F2}) was calculated using the relation

$$E^*_{F1}+E^*_{F2}=E_{\mathrm{c.m.}}-T_{\mathrm{KE}}+Q_0 , \tag{2}$$

where $E_{\mathrm{c.m.}}$ is the center of mass energy in the entrance channel, T_{KE} is the total kinetic energy of the fission fragments, and Q_0 is the energy released in the fission event assuming the primary fragments to be produced in their ground states. The value of T_{KE} was obtained from the relation[32]

$$T_{\mathrm{KE}}=0.1071\frac{Z^2}{A^{1/3}}+22.2\ \mathrm{MeV} . \tag{3}$$

The Q_0 values were calculated by using the ground state masses as given by Wapstra *et al.*[33]. The results of these calculations are given in Table I. Charged particle emission from the excited compound nucleus and the fission fragments was neglected. This assumption is reasonable as the contribution to the fission cross section comes mainly from nuclei with a narrow range of excitation energies close to compound excitation. In Fig. 6, we show the relative contribution to fission from residues of prefission emission (including precom-

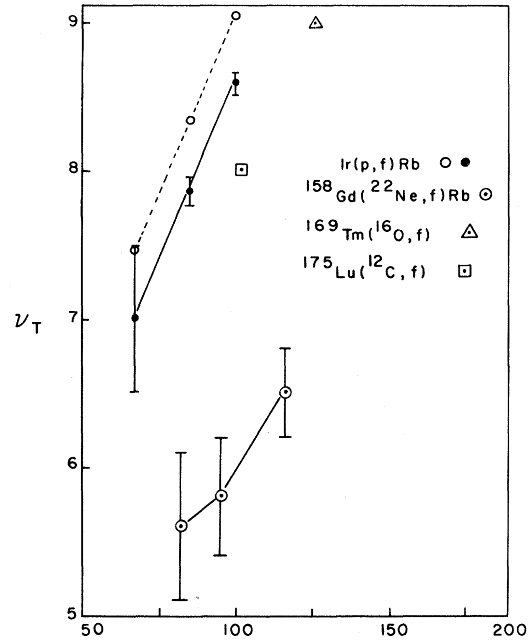

FIG. 5. Variation of the total number of neutrons per fission event ($\bar{\nu}_T$) with incident proton energy. The results obtained in Refs. 15 and 28 are also included for comparison. The broken line joining open circles shows $\bar{\nu}_T$ with equal σ_f for ^{191}Ir and ^{193}Ir.

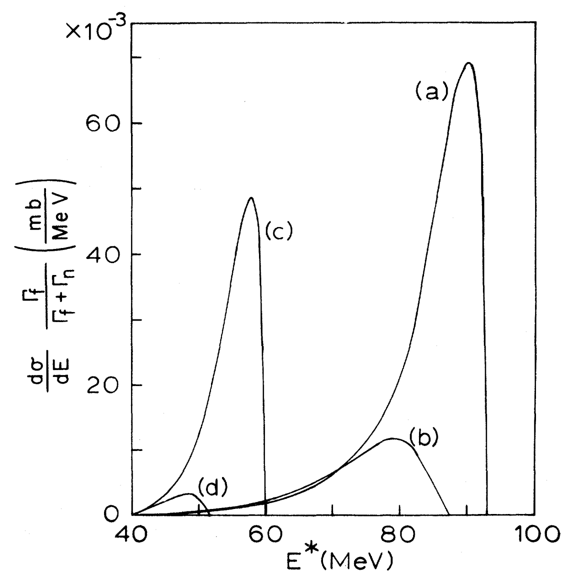

FIG. 6. Relative contribution to fission cross section from nuclei formed after precompound emission. The curves a and b are for residues of neutrons and protons, respectively, at 95 MeV incident energy. The curves c and d represent the contributions from neutron and proton emission residues, respectively, at 62 MeV proton energy.

pound emission). The results have been obtained by compounding excitation energy spectra of particle emission residues, calculated with code ALICE, with the expression for $\Gamma f/\Gamma n$ presented by Huizenga and Vandenbosch.[34] It can be seen that fission following proton emission is negligible at 62 MeV incident proton energy, and still much smaller than fission following neutron emission at 95 MeV. A more detailed account of these calculations is to be presented in a forthcoming publication.[9]

The total numbers of neutrons emitted per fission event are compared in Fig. 5 with those obtained in the fission of compound nuclei in this mass region produced by heavy ion beams. The contrast between the results reported by the Orsay-Dubna group and those obtained in this work is significant. As mentioned before, Reisdorf *et al.* suggested that lower ν_T values observed in heavy ion-induced fission could be attributed to higher angular momenta of fission fragments resulting in enhanced γ emission. The ν_T values obtained in this work are in good agreement with those calculated by Reisdorf *et al.* for zero angular momentum. An alternate explanation proposed by the Orsay-Dubna group was shell effects on the total kinetic energy of fragments. Results obtained in the present work do not seem to indicate such effects. Thus the interpretation based on the assumption that in heavy ion-induced fission a large fraction of the total angular momentum is shared among fission fragments, seems reasonable. More experimental data on neutron and γ multiplicities would be needed for a complete understanding of the reaction mechanism. A more direct comparison to Orsay-Dubna data would be provided by the yet to be measured reaction Ta(p,f)Rb.

Also shown in Fig. 5 are the results reported by Viola and Sikkeland.[15] They used surface barrier detectors to measure the mass distribution in the fission of ^{187}Ir and ^{185}Ir formed by irradiating ^{175}Lu and ^{169}Tm targets with 125 MeV ^{12}C and 166 MeV ^{16}O beams, respectively. Their deduced ν_T values are much closer to our values. However, it should be cautioned that their ν_T values are the average of all possible charge splits and may not be directly comparable to our values obtained from symmetric fission events. It would be of interest to make a more systematic study of this important quantity using both methods.

In a recent theoretical work Savelev *et al.*[35] obtained a linear relationship between the number of neutrons emitted in a fission event and the excitation energy of fission fragments, on the basis of the quasiparticle excitation model. Their equation can be written as

$$\bar{\nu}_T = \nu_0 + \nu_1 \bar{E}_t \ . \tag{4}$$

In the case of the neutron induced fission of ^{235}U, they found $\nu_0 = 0.393$ and $\nu_1 = 0.111$ for a range of fragment excitation energies from 16 to 50 MeV. It is rather remarkable that using the above equation for ν_T we would obtain reasonable agreement with our data. The implication of this agreement is that each neutron emission reduces the excitation energy by about 9 MeV over a wide range of fissioning species.

Although ν_T values appear to increase linearly in the energy range considered, the slope of the curve is low compared to the one expected for the 9 MeV per neutron assumption. This is indicative of the beginning of saturation effects. A reason for this behavior is that particle emission prior to fission increases at higher incident projectile energies, yielding fissioning nuclei with lower excitation energies. Also at higher incident proton energies the assumptions that fission from a compound nucleus predominates and that charged particle emission is negligible, cease to be valid.

IV. CONCLUSION

The proton-induced fission of natural iridium has been studied using an on-line mass spectrometer. The average numbers of neutrons per fission event increase linearly with bombarding energy and are substantially higher for a given excitation energy than the values obtained by a similar technique in the same mass region using a heavy ion projectile. For a quantitative comparison with theory, further data on absolute cross sections of fission and neutron evaporation would be useful. A similar measurement of the proton-induced fission of tantalum would be interesting, albeit much more difficult since σ_f would be about one order of magnitude lower than with an Ir target.

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