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High-energy Neutron-induced Fission Cross Sections of Natural Lead and Bismuth-209

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The CERN Neutron Time-Of-Flight (n_TOF) facility is well suited to measure small neutron-induced fission cross sections, as those of subactinides. The cross section ratios of ^{nat}Pb and ^{209}Bi relative to ^{235}U and ^{238}U were measured using PPAC detectors. The fragment coincidence method allows to unambiguously identify the fission events. The present experiment provides the first results for neutron-induced fission up to 1 GeV for ^{nat}Pb and ^{209}Bi. A good agreement with previous experimental data below 200 MeV is shown. The comparison with proton-induced fission indicates that the limiting regime where neutron-induced and proton-induced fission reach equal cross section is close to 1 GeV.

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I. INTRODUCTION

Data on neutron-induced fission cross sections at intermediate energies are crucial for the development of accelerator-driven systems (ADS). In particular, ^{nat}Pb and ^{209}Bi play a key role since the spallation target will most likely consist of lead or lead-bismuth. In consequence, not only the shape of the neutron spectrum, but also the heating of the target and the radioactivity produced in it will be dominated by the fission of ^{nat}Pb and ^{209}Bi induced by high-energy neutrons.

^{209}Bi(n,f) is recommended as a cross section standard for neutron energies above 50 MeV and new measurements are requested [1]. Its high fission threshold (about 20 MeV) makes possible to use it as a fluence monitor of high-energy neutrons even when a background of low-energy neutrons is present. A smooth behaviour of the fission cross section with neutron energy and the fact that is monoisotopic and a non-radioactive material makes ^{209}Bi a well suited isotope for this purpose.

Despite the importance of these kind of measurements, available experimental data for the neutron-induced fission of subactinides are not abundant. Only recently, with the arising interest on intermediate energy, precise measurements were performed covering a broader energy range up to about 200 MeV, as one can find in Refs. 2 to 5.

The present work, taking advantage of the high intensity neutron beam of the n_TOF facility at CERN, provides a new set of high-precission measurements for the ^{nat}Pb(n,f) and ^{209}Bi(n,f) cross sections.

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II. EXPERIMENTAL METHOD

The experiment was carried out at the CERN-n_TOF (Neutron Time-Of-Flight) facility, where a very intense neutron flux is produced by spallation reactions on a lead target, using a 20 GeV/c proton beam from the Proton Synchrotron at CERN. The cooling water surrounding the spallation target acts as a moderator and produces a neutron flux covering a wide energy range. High-resolution energy measurements are possible due to a short pulse width of 7 ns and the long flight path of 185 m between the spallation target and the experimental area. The data acquisition system is based on Flash-ADCs with a memory large enough to record fissions induced by neutrons with energies ranging from 0.7 eV (16 ms of time-of-flight) up to 1 GeV (700 ns time-of-flight). Up to now, n_TOF is the only neutron facility covering such a wide energy range. More details can be found in [6].

1. Parallel Plate Avalanche Counters (PPACs)

Fission events were detected using a reaction chamber with Parallel Plate Avalanche Counters (PPAC) developed at IPN-Orsay [7], where samples of ^{235}U and ^{238}U were placed as references. The PPAC detectors used in this experiment consist of one central anode between two cathodes under a low-pressure gas filling. The gaps between the electrodes are 3 mm width. Both cathodes of each PPAC are stripped in perpendicular directions, so that the fission fragment trajectory can be reconstructed. Since the PPAC signals are very fast (9 ns in FWHM for the anode signals), the probability of pile-up is very small, making possible to reach energies as high as 1 GeV.

As shown in Fig. 1, the reaction chamber contains

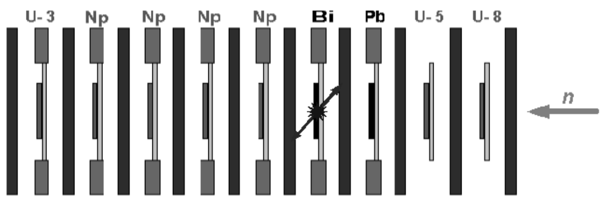

Fig. 1. Schematic view of the PPAC detectors and the samples used in this experiment.

10 PPAC detectors and 9 targets in between, all placed perpendicularly to the direction of the neutron beam, in such a way that both fission fragments are detected in coincidence between two consecutive PPACs.

2. Targets

The total masses, the spatial distributions and the chemical composition of the samples were measured by Rutherford Backscattering Spectroscopy (RBS). For the radioactive ones, α counting was also employed. The thickness and the composition of the backings on which the samples are deposited (mylar for ^{nat}Pb and ^{209}Bi, and aluminium for ^{235}U and ^{238}U) were also measured in order to correct the energy losses of the fission fragments.

III. DATA ANALYSIS

Both fission fragments are emitted in opposite directions and are detected by the closest PPACs within a coincidence window of 10 ns. By imposing this coincidence constraint, most of the background produced by the α emission of the radioactive samples and by the products of other reactions is rejected.

The number of detected fission events per unit interval of incident energy induced in a target is given by: $C(E) = \Phi(E) \cdot N \cdot \sigma(E) \cdot \varepsilon(E)$, where Φ(E) is the time-integrated neutron fluence (in n/cm^2/MeV) for the full measuring time, N is the total number of atoms in the target, σ(E) is the fission cross section of the isotope and ε(E) is the detection efficiency. From this expression, the ratio of the fission cross sections for two samples is:

$$\frac{\sigma_i(E)}{\sigma_j(E)} = \frac{C_i(E)}{C_j(E)} \cdot \frac{\Phi_j(E)}{\Phi_i(E)} \cdot \frac{N_j \varepsilon_j(E)}{N_i \varepsilon_i(E)} . \quad (1)$$

If the two samples involved in this ratio have the same diameter, we can assume that $\Phi_j(E) = \Phi_i(E)$, because MCNP calculations have demonstrated that the neutron flux attenuation through the whole system is lower than 1% [8]. However, the size of the samples is different: ^{235}U and ^{238}U have a well defined diameter of 8 cm, smaller than the beam size, while the samples of ^{nat}Pb and ^{209}Bi are spread over the whole backing surface, which is larger than the beam size, so that the samples are not exposed in the same way to the neutron beam. Using the cathode signals it is possible to map the fission events on the target (as shown in Fig. 2 for ^{209}Bi and

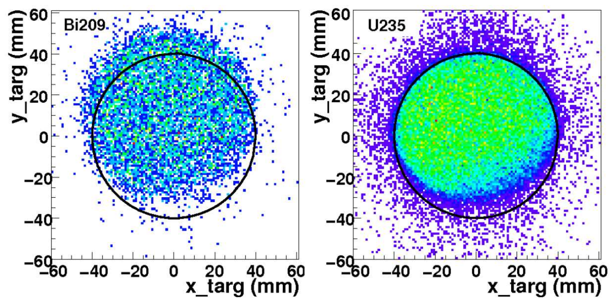

Fig. 2. (Color online) Position distribution of the fission events in the ^{209}Bi and in the ^{235}U samples, compared with the real position of ^{235}U and ^{238}U samples, showed by the black circle.

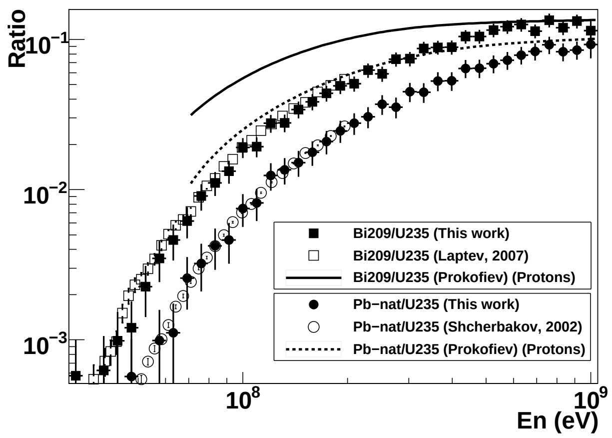

Fig. 3. Neutron-induced fission cross section ratios ^{nat}Pb/^{235}U and ^{209}Bi/^{235}U from this work, compared with data from Laptev [3] and Shcherbakov [4] (open symbols). The lines represent the Prokofiev's systematics [9] for (p,f).

^{235}U) to estimate the effect of the different size of the targets in the ratio of Eq. (1). This is equivalent to setting a ratio of neutron fluences of 1.2. The black circle in Fig. 2 indicates the true location of the ^{235}U and ^{238}U targets.

Differences in backing and sample thicknesses lead to different stopping power of the fission fragments and, therefore, different efficiencies. The ratio of detection efficiencies $\varepsilon_j(E)/\varepsilon_i(E)$ is expected to be very close to unity in the case $\sigma(^{nat}Pb)/\sigma(^{209}Bi)$ since they have identical backings and thicknesses, but this is not the case for the ratios involving the reference samples. To address this problem, a simulation combining Monte-Carlo methods and numerical calculations for the stopping power was done. The ratios $\varepsilon_{U235}(E)/\varepsilon_{Pb}(E)$ and $\varepsilon_{U235}(E)/\varepsilon_{Bi}(E)$ were estimated to 0.85 and to 0.88 when the ^{238}U target is the one used as reference. The effect of the linear momentum transferred (LMT) from the incident neutrons to the nuclei has been found to be much lower than the other components of systematic uncertainty.

IV. RESULTS

We have measured relative cross sections for neutron-induced fission up to 1 GeV and our results do not rely

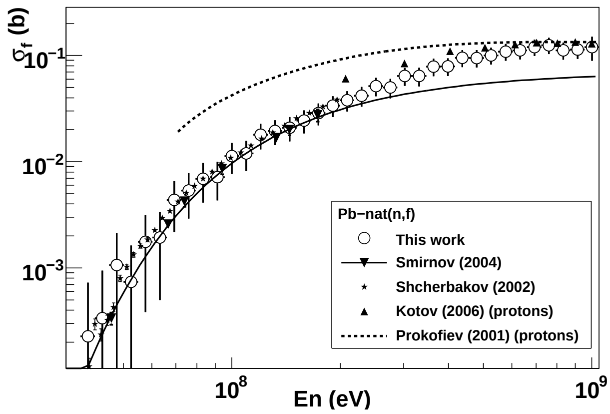

Fig. 4. ^{nat}Pb(n,f) cross section obtained in this work, compared with previous data from Shcherbakov [4] and Smirnov [5]. Data from Kotov [10] and the Prokofiev's systematics [9] for proton-induced fission are also drawn.

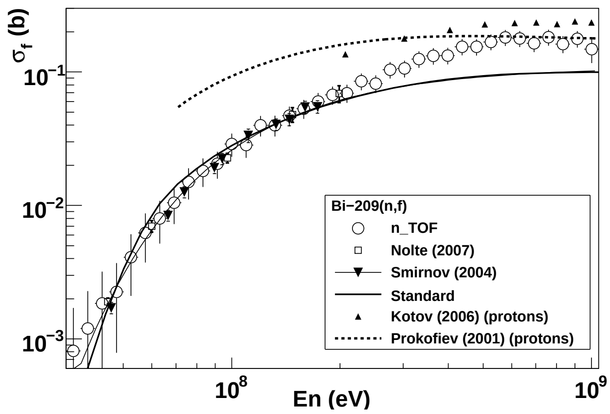

Fig. 5. ^{209}Bi(n,f) cross section obtained in this work, compared with previous data from Nolte [2] and Smirnov [5]. The IAEA standard parameterization [1] and the Smirnov's one [5] do not fit our results above 200 MeV. Data from Kotov [10] and Prokofiev's systematics [9] for (p,f) are also drawn.

on any normalization to other results or evaluations, because the required correction factors have been calculated, as explained above.

Both ratios $\sigma_f(^{nat}\mathrm{Pb})/\sigma_f(^{235}\mathrm{U})$ and $\sigma_f(^{209}\mathrm{Bi})/\sigma_f(^{235}\mathrm{U})$, extended up to 1 GeV for the first time, are presented in Fig. 3. The error bars represent the statistical uncertainties, while the systematic uncertainties are around 10%. Good agreement with the results from Refs. 3 and 4 is found below 200 MeV, which is the maximum neutron energy in the data. Because of the absence of experimental data above this energy, the parameterizations for (p,f) cross sections given in Ref. 9 for the same ratios are also shown.

To obtain the fission cross sections of ^{nat}Pb and ^{209}Bi from these ratios, the ^{235}U(n,f) and ^{238}U(n,f) cross sections given by JENDL/HE-2007 have been used, which is the only evaluation that covers the whole energy range. The final results are shown in Figs. 4 and 5.

We can see that, in both cases, our results are compatible with other measurements below 200 MeV such as those of Refs. 2, 4, and 5. The available parameterizations by Smirnov [5] and the recommended standard ^{209}Bi(n,f) [1] underestimate the present (n,f) cross sections above around 200 MeV.

A comparison of our results with experimental data [10] and systematics [9] for (p,f) cross section shows that the (p,f) and the (n,f) cross sections are compatible only above around 700 MeV.

V. CONCLUSIONS

The ^{209}Bi(n,f) and ^{nat}Pb(n,f) cross sections were measured at the n_TOF facility between threshold and up to 1 GeV for the first time. A fast response PPAC fission chamber was developed to this purpose and the fission fragments were detected in coincidence using PPAC detectors. Samples of ^{235}U and ^{238}U were used as references.

Our data are in good agreement with other experimental values below 200 MeV. Above this energy, the parameterizations by Smirnov [5] and Carlson [1] are not coherent with our experimental results and should therefore be updated by taking the results of this work into account.

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