Comparative measurement of prompt fission γ-ray emission from fast-neutron-induced fission of ^{235}U and ^{238}U

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Prompt fission γ-ray (PFG) spectra have been measured in a recent experiment with the novel directional fast-neutron source LICORNE at the ALTO facility of the IPN Orsay. These first results from the facility involve the comparative measurement of prompt γ emission in fast-neutron-induced fission of ^{235}U and ^{238}U. Characteristics such as γ multiplicity and total and average radiation energy are determined in terms of ratios between the two systems. Additionally, the average photon energies were determined and compared with recent data on thermal-neutron-induced fission of ^{235}U. PFG spectra are shown to be similar within the precision of the present measurement, suggesting that the extra incident energy does not significantly impact the energy released by prompt γ rays. The origins of some small differences, depending on either the incident energy or the target mass, are discussed. This study demonstrates the potential of the present approach, combining an innovative neutron source and new-generation detectors, for fundamental and applied research on fission in the near future.

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I. INTRODUCTION

Prompt γ emission in nuclear fission is one of the least understood parts of the fission process. Its study can give information on the partition of energy and angular momentum, and on the competition between neutron and γ emission during fission fragment deexcitation. Furthermore, information on prompt fission γ-ray emission has practical applications in nuclear reactors. Indeed prompt fission γ emission significantly contributes to the total reactor core γ-ray spectrum (∼40%) and γ heating effects, particularly at the core periphery, and it is thus linked to reactor safety. Currently, γ-heating effects are underestimated [1] and there is a need for improved prompt γ emission data. Such measurements are at the top of the OECD/NEA high priority request list [2,3]. In recent years the characterization of prompt fission γ rays has become an important topic of research with experimental efforts underway to measure their characteristics such as spectral shapes, average multiplicities and distributions, average energy per quantum and total energy, in thermal and epithermal induced fission for a range of actinide nuclei [4–10]. However, up until now almost no data exist for prompt γ-ray emission characteristics from fast-neutron-induced fission, even though most of the generation IV reactor concepts will involve such neutron energies [11]. The lack of data is due to the experimental difficulties associated with such measurements, requiring an intense neutron source, since fission cross sections are typically three orders of magnitude lower for fast neutrons as compared to thermal neutrons. Furthermore, shielding the γ detectors from the source neutrons is difficult, and the thermal-neutron background must be kept extremely low due to the associated larger fission cross section.

These difficulties have been overcome with the development of the LICORNE directional neutron source at the Tandem accelerator of the ALTO facility of the IPN Orsay. LICORNE can provide high-flux, naturally focused neutrons with energies from 0.5 to 4 MeV, typical of those found in innovative fourth generation reactors [11]. LICORNE is based on the use of an intense ^{7}Li beams and the $p(^{7}\mathrm{Li},n)$ reaction [12,13]. The major advantages for prompt fission γ measurements are that the natural directionality ensures that no γ detector shielding is required for protection from source neutrons, and that the thermal-neutron background in

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the experimental hall is extremely low. At conventional, direct-kinematics-based facilities, the source neutrons are emitted isotropically in 4π. The flux at the target is thus considerably smaller; source neutrons will directly hit the γ detectors, and multiple neutron scatterings from the walls produce a significant thermal-neutron background that can dominate the fission rate. This paper reports on the first experiment carried out with the LICORNE neutron source after its recent successful commissioning. The measurement consists of a comparative characterization of prompt fission γ-ray emission from fast-neutron-induced fission of ^{235}U and ^{238}U. Such a simultaneous measurement ensures identical experimental conditions, considerably reducing systematic errors.

II. EXPERIMENTAL SETUP

The experiment was carried out at the Tandem accelerator of the ALTO facility of the IPN Orsay where a ^{7}Li beam was used to bombard rotating hydrogen-rich polypropylene target discs of 8 cm in diameter and 20 μm thickness. The bombarding energy was 15 MeV at a maximum intensity of 100 nA. Neutrons are produced in the $p(^{7}\mathrm{Li},n)$ reaction within a cone centered at 0° with respect to the beam axis and with a maximum opening angle of 20° with a flux up to 10^7 neutrons per second. The average neutron energy was $\bar{E}(n) =$ 1.7 MeV, constrained by the kinematics of the reaction. Fission fragments were detected with a twin Frisch-grid ionization chamber (IC), filled with P10 counting gas at a pressure of 1.2 atm. A neutron flux up to 3×10^5 neutrons/s cm^2 was reached at the target position in the IC at 15 cm from the source. Two targets of ^{235}U and ^{238}U of around 300 μg/cm^2 thickness and 6.5 cm diameter were placed back to back at the central cathode. Fission fragments were tagged by the pulse height signals from the coincident fragments at the common cathode and separate anodes for each target. Figure 1 gives a schematic view of the layout of the detection setup used for this experiment. Coincident fission γ rays were detected in 14 large hexagonal BaF_2 crystals of 10 cm diameter and 14 cm length and three cylindrical $LaBr_3$ scintillator detectors of dimensions 5.01 cm diameter and 5.01 cm length (not shown on the figure). $LaBr_3$ scintillators are recently developed detectors which have both excellent energy resolution and timing properties and are ideal for characterizing the lower part of the PFG spectra. The BaF_2 crystals have a larger geometrical coverage and higher intrinsic efficiency for detecting the highest energy γ rays, but have a lower energy resolution. Both types of detector have subnanosecond time resolution—around 300 and 800 ps for $LaBr_3$ and BaF_2, respectively. In this paper only BaF_2 spectra are presented since we were unable to fully exploit the $LaBr_3$ properties due to difficulties in unfolding detector responses for spectra with low counting statistics. However, results using $LaBr_3$ detectors in a recent high-statistics experiment will be reported in a future publication.

A. Neutron/γ discrimination

Prompt neutron/γ discrimination in the BaF_2 and $LaBr_3$ scintillators was achieved via the time of flight technique with the combined time resolution of the ionization chamber and BaF_2 detector being 4.2 ns FWHM. Neutron/γ discrimination for neutrons with energies up to 10 MeV was thus possible.

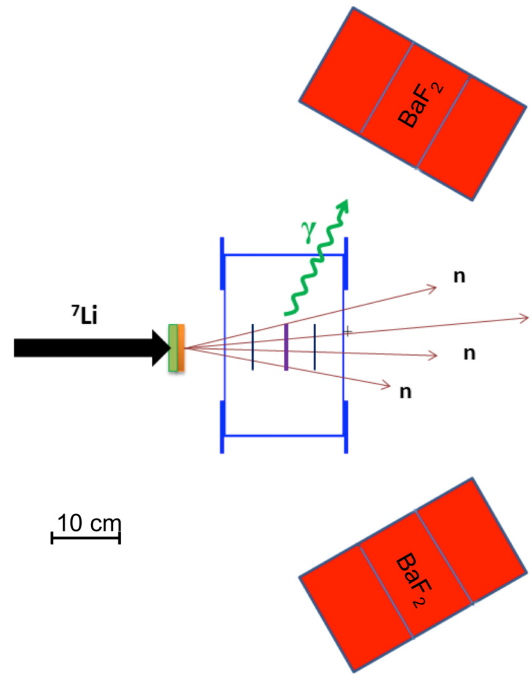

FIG. 1. (Color online) Schematic view of the experimental setup used to measure prompt fission γ-ray characteristics drawn to scale. The ionization chamber to tag the fission is drawn in blue. The three lines at the center of the IC represent anode, target position, and anode, respectively, left to right. The arrows show schematically the opening of the neutron cone.

The γ detectors were calibrated in energy with ^{137}Cs, ^{60}Co, ^{152}Eu, ^{232}Th, and AmBe sources to obtain data points over a large energy range from 121 keV to 4.4 MeV. Spectra were subsequently binned with nonlinear energy bin sizes which were chosen to be the same size as the intrinsic resolution of the detector as a function of energy.

B. Thermal-neutron background with LICORNE

With conventional isotropic neutron sources produced in direct kinematics, measurement of prompt fission γ rays for fissile nuclei is more difficult to perform. It requires heavy shielding of the γ detectors or a good collimation [14] of the neutron beam which decreases the available neutron flux and thus must be compensated for with a very intense primary beam. Furthermore, without collimation, a strong residual thermal-neutron field in the experimental hall can be found. Even low thermal-neutron fluxes can be the dominant contribution to the total fission rate in a fissile target according to the comparatively larger cross section.

The LICORNE experimental hall has 50 cm thick concrete interior walls about 7 m apart and a concrete floor but no interior roof, allowing neutrons to escape into the large accelerator facility building which has a volume ~100 times greater. This and the naturally directional neutron emission keeps the thermal-neutron flux in the target area very low. No detectable excess of fission in the ^{235}U run was observed in the fission rate ratio for ^{235}U and ^{238}U when compared with the expected ratio from the known neutron spectrum and the measured target masses, which were (12.1 ± 0.6) mg and (9.5 ± 0.5) mg, respectively. In addition, Monte Carlo N-particle (MCNP) simulations of the experimental hall showed that even in the most unfavorable experimental conditions, the total thermal-neutron-induced fission rate of ^{235}U is smaller than 1% of the total.

III. RESULTS

A. Data analysis

During the first experiment discussed here (around 36 h beam exposure), 1.2×10^5 fission events were recorded to disk. Results from subsequent experiments after the recent upgrade of LICORNE with a hydrogen gas target which produces higher neutron fluxes, will be reported in future publications. Fission fragments were discriminated from the target's intrinsic α-particle activity via gating on cathode and anode pulse heights and relative time differences. Prompt fission γ-ray selection is performed via the time of flight between the IC cathode and the γ detector. To obtain detector response matrices, a detailed MCNP5 [15] simulation of all the components of the experimental setup was carried out. Detector responses to monoenergetic photons of energies between 0 and 10 MeV were obtained via simulating emission from the ^{235}U and ^{238}U targets at the center of the IC for each detector energy bin. Response matrices were then created from the ensemble of the resulting simulated detected spectra.

The PFG spectra of ^{235}U and ^{238}U were then determined correcting the measured spectra in a similar manner to the procedure described in [7]. Since the intrinsic efficiencies for detection of single monoenergetic γ rays of the BaF_2 crystals are very large, the size of the required spectral corrections from response unfolding is relatively small and the absolute value of associated systematic errors is also small. Statistical errors are thus the dominant source of uncertainty in the emission spectra.

The PFG emission spectra from fast-neutron-induced fission of ^{235}U and ^{238}U from the BaF_2 detectors are shown in Fig. 2. The spectral shapes are similar to previous observations for thermal-neutron-induced fission [7] with two major exponential spectral components below and above 2 MeV with differing slopes. Fits of an exponential function to the high-energy tails of the spectra are also shown in the figure. Since the threshold of the BaF_2 detectors was rather high (around 400 keV) and there were problems associated with dead time in the experiment, a renormalization was necessary. We were thus unable to extract reliable average γ multiplicities per fission in this experiment. However, average photon energies could be extracted, since these depend only on the spectral shape. It was also possible to perform precise comparisons of the $^{235}\mathrm{U}/^{238}\mathrm{U}$ ratios of average multiplicity, average photon energy, and total radiation energy release per fission, and hence investigate potential differences between the two nuclides studied simultaneously under identical experimental conditions. The relative values of these $^{235}\mathrm{U}/^{238}\mathrm{U}$ ratios for the mean multiplicity, mean photon energy, and total energy release are reported in Table I. All three ratios are consistent with unity and thus the main

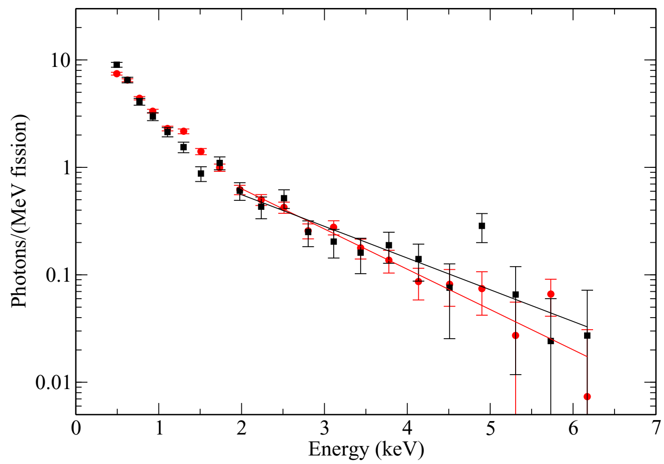

FIG. 2. (Color online) Comparison of the prompt fission γ-ray emission spectra, measured with BaF_2, for fast-neutron-induced fission of ^{235}U (red circles) and ^{238}U (black squares).

spectral characteristics are similar to within the precision of the measurement. While comparing average photon energies with those extracted in previous measurements, the detection threshold can play an important role. The presence of an unavoidable finite threshold implies that the reliable extraction of average spectral quantities, such as multiplicity and energy, requires a correction accounting for the missing low-energy γ rays. Recent studies [16] show that the part below 500 keV can have a great impact on the prompt γ spectral data, and that corrections are indispensable to arriving at meaningful average spectral quantities. In our case, the BaF_2 detector threshold was similar to that used in recent published data from the DANCE array [9,10]. In this work we attempted to correct below threshold by extrapolation according to the method detailed in Refs. [5,16]. The corresponding large relative uncertainties of around 35% are reflected in the total error in the measured values. The average energy per photon was measured to be (0.76 ± 0.04) MeV and (0.77 ± 0.06) MeV for fast-neutron-induced fission of ^{235}U and ^{238}U, respectively. These values compare to thermal-neutron-induced fission of ^{235}U at (0.85 ± 0.02) MeV from [5]. Then, threshold effects appear to be a reasonable explanation for the discrepancy between our values and Billnert's measurement [4] with the DANCE data [9]. However, in this work, we do not have the means to discuss if this effect is completely due to a threshold effect. Nonetheless, for energy applications, contributions from the lowest energy γ rays are relatively unimportant due to the very high attenuation in the reactor core medium compared with the higher energy γ rays—higher

TABLE I. Comparison of spectral characteristic ratios between ^{235}U and ^{238}U.

$^{235}\mathrm{U}/^{238}\mathrm{U}$ ratio	Measured value
Mean photon multiplicity	1.008 ± 0.090
Mean photon energy	0.976 ± 0.040
Total energy	1.003 ± 0.070

TABLE II. Fitted exponential parameters to the high-energy tails (above 2 MeV) of the emitted the PFG spectra from the three different fission reactions.

Fission reaction	Fitted exponential parameter (MeV^{-2})
$^{235}U(n_{th}, f)$	-0.940 ± 0.017
$^{235}U(n, f)$	-0.86 ± 0.09
$^{238}U(n, f)$	-0.68 ± 0.10

than 2 MeV—which penetrate the farthest. Figure 2 shows a comparison of the fast-fission-induced PFG data from ^{235}U obtained with LICORNE to that obtained in thermal-induced fission [5]. The spectral shapes are again similar even though an extra 1.73 MeV is given to the compound nucleus before fission. For ^{238}U the average extra excitation energy is slightly higher at 2.07 MeV due to the fission reaction threshold, which is not present for ^{235}U.

B. Comparison of high-energy tails

Finally, we can make a comparison of fits to the high-energy component of the PFG spectra from fast-neutron-induced fission of ^{235}U and ^{238}U and thermal-induced fission of ^{235}U from [5]. The results are shown in Table II. As Table II shows, the PFG spectrum is slightly harder for $^{238}U(n, f)$ than for $^{235}U(n, f)$, and the spectrum from $^{235}U(n_{th}, f)$ is the least hard. Yet the statistical significance of this effect is low, but will be studied in greater detail in planned future experiments.

IV. DISCUSSION

Changes in the prompt γ emission spectra for fast-neutron-induced fission as a function of incident neutron energy will occur for two different reasons. First, the extra energy brought into the compound nuclear system via the neutron will change the fission yields, with the higher excitation energies favoring more symmetric fissions and populating masses more and more strongly in the valley at around $A \sim 120$ [17]. Second, more excitation energy is available for both neutron and γ emission from the excited fission fragments. It has been shown that the total kinetic energy (TKE) of the fragments is rather insensitive to changes in the incident neutron energy [18]. The competition between neutron and γ emission as a function of incident neutron emission is an open question. Very recently, a strong and renewed theoretical effort is underway with several fission codes being developed by different groups to predict all fission observables such as yields, prompt emission spectra, fragment TKEs, etc. [19–22], and importantly, their correlations. The results from this experiment, as can be seen in Fig. 3, seem to indicate that much of the excess energy in the compound nucleus in fast-neutron-induced fission ends up in the emitted neutrons. However, measurements with higher statistics are needed to understand better the partition of energy in the fission process and particularly how the fragments share their energy and angular momentum and how the emission proceeds as a function of incident neutron energy and fragment mass.

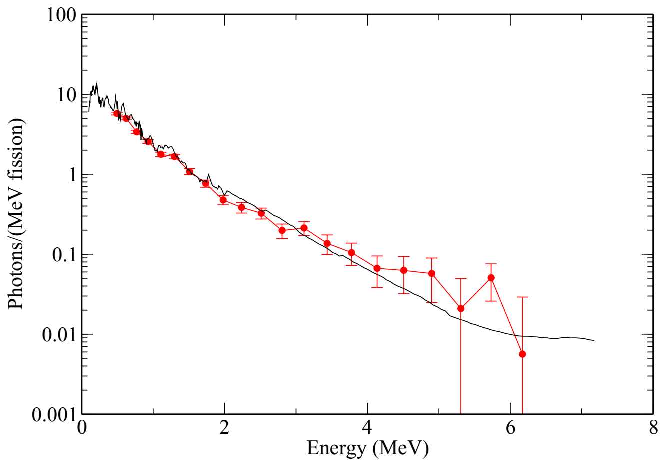

FIG. 3. (Color online) Comparison of PFG emission spectra from fast-neutron-induced fission of ^{235}U (red circles) from this experiment measured with BaF_2 detectors with spectra from the thermal-induced fission of ^{235}U (black line) from a previous experiment [4].

V. CONCLUSION

Comparative prompt fission γ-ray characteristics for fast-neutron-induced fission of two key nuclei, ^{235}U and ^{238}U, have been measured for the first time. The results are important both for understanding the competition between neutron and γ emission in fission and the γ heating phenomena in Gen IV reactors. This type of experimental investigation is particularly challenging due to the low cross sections (three orders of magnitude lower than thermal-induced fission) and the difficulty in providing a uniquely fast-neutron field with negligible thermal component. Both these technical difficulties have been recently solved by the development of the LICORNE inverse kinematics neutron source. γ detectors can thus be placed around the targets to be irradiated without being blinded by neutrons from the source. In the first LICORNE experiment the relative shapes of the PFG from fast-neutron-induced fission of ^{235}U and ^{238}U are observed to be similar within the experimental error bars. In addition, the spectral shapes for fission of ^{235}U induced with either thermal or fast neutrons do not indicate a large difference. The exponential components of the spectra above 2 MeV have been measured and the $^{235}U(n, f)$ and $^{238}U(n, f)$ appear to be slightly harder spectra than that for thermal-induced fission of ^{235}U, within the limit of the statistical significance of the measurement. These results suggest that the extra energy input—on average 1.73 MeV for ^{235}U and 2.07 MeV for ^{238}U—into the compound nucleus does not manifest itself via significant changes in the γ-ray spectral shapes. The increases in neutron multiplicities as a function of incident neutron energy are well known and measured, although the changes in neutron spectra are less well studied. More investigations are needed due to the almost complete lack of experimental data on prompt γ-ray emission in fission as a function of the incident neutron energy. The recently developed LICORNE directional neutron source, combined with new-generation scintillators for γ-ray detectors, provide powerful tools with which to further carry out more precise studies.

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