γ-ray Energy Spectra and Multiplicities and Fission Fragment A and Z distributions from the Neutron-induced Fission of ^{239}Pu using STEFF.

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Abstract

An experiment is proposed to address the NEA high-priority request for better information on ^{239}Pu(n,f) prompt γ-ray emission. The experiment will use STEFF, a 2E2v device, to measure important observables in fission: γ and fission fragment energies, γ-ray multiplicity, and fission fragment mass and atomic number distributions, as a function of incident neutron energy.

Introduction and Background

Since the 1960's there have been many measurements of fission yield and fragment kinetic energy distributions for spontaneous and neutron-induced fission of actinide nuclei [1]. These measurements have been performed by a variety of methods, including both radiochemical analyses and event-by-event fragment-detection experiments. The event-by-event measurements may be broadly separated into different categories according to the technique used to obtain the fragment mass (A): $2E$ measurements, in which both fission fragment energies are measured; $2v$ measurements, in which time-of-flight measurements are made for both fragments; Ev measurements, in which the energy and the time-of-flight is measured for one fragment and the other fragment is typically undetected; specific charge separator measurements (such as those performed using Lohengrin) in which mass, energy and charge-state distributions are obtained. These approaches have their own strengths and limitations. Since the parent fissioning nucleus has effectively zero momentum in the lab frame either the measurement of two energies or two velocities gives the experimentalist the mass ratio of the fragments, provided that neutron evaporation is ignored. The $2v$ and $2E$ methods utilise this approach and therefore suffer from a inherent limitation in accuracy of the measured mass distribution due to undetected neutron evaporation [2]. The $2E$ approach has an advantage in that it can be performed in very close geometry with a windowless double-gridded ion chamber, yielding good energy resolution and high efficiency, but nevertheless does not give atomic number sensitivity due to uncertainty over the fragment velocities. Mass spectrometers and Ev devices do not make assumptions about neutron evaporation. However, since these measurements are made on one fragment only there is no direct handle on the number of neutrons evaporated in a given event. Also, while Lohengrin gives unsurpassed mass resolution, the relatively small solid angle requires the use of very high fission rates in the target. This, and the few microseconds of flight time to the focal plane, prevents the use of γ-ray detectors close to the target in experiments that measure the correlation between prompt γ radiation and the mass and kinetic energy of the fission product; *isomeric* γ-rays may of course be studied at the spectrometer focal plane. None of the above event-by-event methodologies make a direct measurement of the atomic number (Z) of either of the fission fragments, to which, by contrast, radiochemical measurements are sensitive. The STEFF spectrometer, see figure 1, is a $2E2v$ device with Z sensitivity, i.e. it measures the energy and time-of-flight of both fission fragments to obtain A (to ± 4 u), in addition to the specific energy loss of the fragments in isobutane (for Z determination to an accuracy of a few percent). The measurement of the kinetic energy for both fragments allows for the determination of the excitation energy $E^\star$ distribution and the segmentation of the anodes allows for measurement of the direction (θ, ϕ) of the fission axis (to an accuracy $\pm 2^\circ$). In experiments to date γ-ray detection is performed in an array of 12 NaI scintillators around the target. STEFF has been used with thermal neutrons at the ILL [3] and in the EAR2 station at n_TOF. Here we are asking for time for a follow up to the first n_TOF experiment which looked at ^{235}U(n,f) [4]. In this experiment we will use a new array of 6 $LaBr_3$ scintillators to overcome the inherent problems with high rates observed in the NaI scintillators observed in the previous experiment. In the proposed experiment measurements of the γ-ray total energy spectrum and γ-ray multiplicities in conjunction with the measurement of A, Z, E, θ, ϕ and $E^\star$ for the ^{239}Pu(n,f) reaction. This allows us to perform a complete analysis of the fission process to within the above-mentioned resolution of STEFF.

Physics Case

Prompt γ-ray distributions are of significant importance to the development of fast-reactors. About 10% of the total energy released in the reactor core is in the form of γ radiation, and of this 40% is emitted before β decay and 30% comes from the later decay of the fission products. Since the other sources of γ radiation (radiative capture and inelastic scattering reactions) are smaller in their contribution and considered to be better known, the details of the prompt flash is the major uncertainty in calculating the γ heating in a reactor. In a fast reactor, the γ heating of the fuel-free assemblies is significant, due to the relatively easy propagation of the γ radiation and is a major contribution to the total energy release in uranium oxide or mixed-oxide cores. Much of the data that contributes to current nuclear data libraries regarding the total γ-ray energy, multiplicity and spectrum shape dates from the early 1970's (e.g. [5, 6, 7, 8, 9]) and shows significant variations (on the level of 15% in the measured total energy [10]). The NEA high-priority request list has identified, amongst other isotopes, the need for more information on ^{239}Pu prompt γ-ray distributions, energies and multiplicities. Additionally, a recent WPEC report on the evaluation of ^{239}Pu highlights possible differences in average neutron multiplicities for different resonances [11]. Despite their importance in developing new nuclear technologies and significant experimental efforts, the A and Z distributions of fission fragments for many actinides are still relatively sparse. In the case of ^{239}Pu, fission yields up to 6 MeV and at 14 MeV using a series of quasi mono-energetic neutron beams [12]; Mass distributions in the thermal-neutron-energy region with the Lohengrin separator [13]; prompt γ-ray singles, sum and multiplicities [8, 14] from thermal up to 30 keV neutrons; and total kinetic energy [15] have all been measured in recent experiments.

Several measurments of prompt γ-rays from the ^{239}Pu(n,f) reaction have been made, the most recent of which that has been included in an evaluation were performed in the 1970's and covered only thermal neutron energies [6, 9]. Two additional measurements cover low and high neutron energy but a restricted energy range for γ-rays [5, 7]. Two recent measurements reported the energy for singles and sum γ-rays, and multiplicities [14, 8]. One study, limited to neutron energies between thermal and 30 keV, reported a higher average γ-ray sum energy than previous work and discrepancies in the low γ-ray energies for singles spectra. The more recent study showed that the prompt γ-rays from incident thermal neutrons are not responsible for the additional heating in reactors [14]. Despite these efforts significant gaps in the experimental data remain, for example little is known of the γ-ray spectra above an incident neutron energy of 30 keV. As pointed out by Madland, the symmetric fission component increases with incident neutron energy, therefore the corresponding larger Q-values for this component should have an effect on the γ sum energy and multiplicities.

Fission product yields as a function of incident neutron energy are important information when developing new nuclear technologies. In the case of ^{239}Pu a number of studies have been performed to investigate mass distributions at thermal energies (see for example [13, 16]) and more recently in the MeV region [12]. These studies are important as it is not always possible to reproduce these yields with phenomenological models. For ^{239}Pu a positive slope is found for high-yield fission products, contrary to ^{235}U [12]. The most recent work employed quasi-monoenergetic neutrons between 0 and 6MeV and at 14 MeV leaving sizeable gaps in the data at the point where second chance fission becomes possible [12].

There has been interest in the large variation of fission-neutron multiplicities as a function of incident-neutron energy in the resolved resonance region [11]. These fluctuations are larger for resonances having $J^\pi = 1^+$ due to this channel having a smaller number of fission transition states compared to the other S-wave resonance states with $J^\pi = 0^+$. The different average fission widths for the two types of states are thought to result in different probabilities for the $(n, \gamma F)$ two-step fission process. The emitted γ-ray takes away an average of 1 MeV, energy that is not then available for neutron emission. We would like to investigate whether this effect of lower excitaion energies is observed in the prompt γ multiplicities. We propose to measure mean γ-ray multiplicities as a function of energy accross the resolved resonance region and make a comparison of the multiplicities for $J^\pi = 0^+$ and $J^\pi = 1^+$ resonances and the off-resonace data.

Experimental and Technical Details

STEFF was employed to investigate fission fragment distributions and γ-rays from the ^{235}U(n,f) reaction using EAR2 at n_TOF [3, 17]. Alongside the primary physics case this experiment was able to evaluate the performance of the STEFF detectors in the presence of the n_TOF γ-ray flash. These detectors included the start/stop detector, NaI scintillators, LaBr$_3$ scintillator (1" × 1.5"),

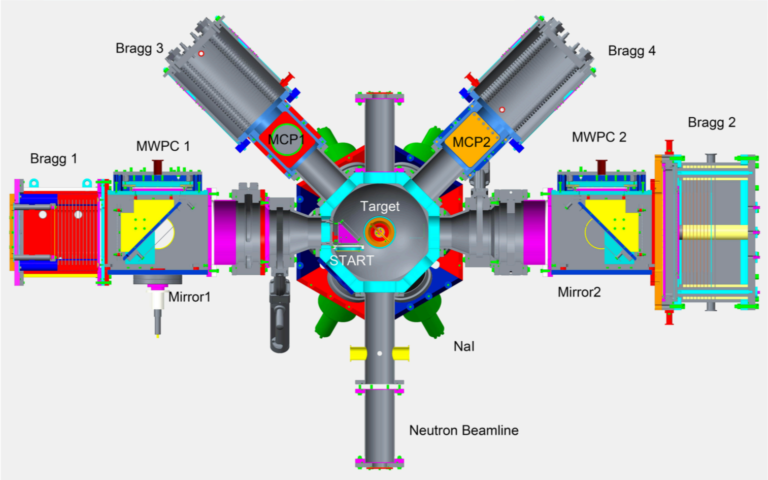

Figure 1: STEFF in the new configuration for EAR2 shown as a section in a vertical plane through the centre-line of the spectrometer. The neutron beam travels from bottom to top in the figure. The TOF arms contain the START detector and two STOP detectors. The short arms have STOP detectors (MCP1 and MCP2) but no STARTs.

and new short flight arms. The results from this experiment are the basis of a PhD thesis [17], a second PhD thesis is awaiting examination.

Our intention is to use STEFF in EAR2 to measure prompt γ-ray singles and sum energies, multiplicities, along with fission fragment energies, masses and atomic numbers. The small collimator will be employed with a thin target corresponding to 30 $\mu g/cm^2$ of ^{239}Pu. STEFF has a solid angle of coverage of 0.134 sr for fission fragments, using a fragment-efficiency of 50% results in a detection rate of approximately 0.5 fissions per pulse over a time interval of 10 ms. Such a set up with a total of 6×10^{18} protons results in around 3×10^5 detected fission events.

The upper neutron energy limit for this measurement is due to the saturation of the γ-ray detectors from the 'γ flash' effect. In order to compensate for the effect of the prompt γ-ray flash two of the large-volume (5" × 4") NaI detectors will be replaced by six (two clusters of 3) smaller volume (1" × 1.5") $LaBr_3$ scintillators. These scintillators are faster than NaI, by an order of magnitude, and will recover more quickly and cope better with the higher background rates at shorter neutron TOF. The analysis of data from the previous experiment demonstrated that with the $LaBr_3$ detectors it is possible to resolve γ-rays at higher neutron energy (shorter TOF) allowing measurments through the resonance region.

In summary an experiment is proposed to address the NEA high-priority request for better information on ^{239}Pu(n,f) prompt γ-ray emission. The experiment will use STEFF to measure γ -ray and fission-fragment energies, γ-ray multiplicity, and fission fragment mass and atomic number distributions, as a function of incident neutron energy.

References

[1] F Goennenwein. *Mass, charge, and kinetic energy of fission fragments* . inis.iaea.org, 2000.

[2] James Terrell. Neutron Yields from Individual Fission Fragments. *Phys. Rev. Lett.*, 127(3):880–904, August 1962.

[3] E Murray, A G Smith, A J Pollitt, J Matarranz, I Tsekhanovich, T Soldner, U Köster, and D C Biswas. Measurement of Gamma Energy Distributions and Multiplicities Using STEFF. *Nucl. Data Sheets*, 119:217–220, May 2014.

[4] A.G. Smith. γ-ray energy spectra and multiplicities from the neutron-induced fission of ^{235}u using steff. INTC-P-405, 2014.

[5] D M Drake. Gamma-ray production cross sections for 1- and 2-MeV neutron interactions with 235U and 239Pu. Technical Report LA5048, Los Alamos Laboratory, 1972.

[6] Frances Pleasonton. Prompt γ-rays emitted in the thermal-neutron induced fission of 233U and 239Pu. *Nucl. Phys. A*, 213(2):413–425, October 1973.

[7] V Polhorský, J Kliman, J Krištiak, A A Bogdzel, N A Gundorin, A B Popov, and U Gohs. Prompt gamma-ray emission from fission of239Pu by resonance neutrons I. *Czech. J. Phys.*, 43(8):783–788, August 1993.

[8] J L Ullmann, E M Bond, T A Bredeweg, A Couture, R C Haight, M Jandel, T Kawano, H Y Lee, J M O'Donnell, A C Hayes, I Stetcu, T N Taddeucci, P Talou, D J Vieira, J B Wilhelmy, J A Becker, A Chyzh, J Gostic, R Henderson, E Kwan, and C Y Wu. Prompt γ-ray production in neutron-induced fission of 239Pu. *Phys. Rev. C*, 87(4):208, April 2013.

[9] V V Verbinski, Hans Weber, and R E Sund. Prompt Gamma Rays from 235U(n,f), 239Pu(n,f) ans Spontanious Fission 252Cf. *Phys. Rev. C*, 7(3):1173–1185, March 1973.

[10] G Rimpault, A Courcelle, and D Blanchet. Needs for accurate measurements of spectrum and multiplicity of prompt γ emitted in Fission. Technical report.

[11] C De Saint-Jean. Co-ordinated Evaluation of Plutonium-239 in the Resonance Region. Technical Report NEA/NSC/WPEC/DOC(2014)447, April 2017.

[12] A Tonchev, M Stoyer, J Becker, R Marcri, C Ryan, S Sheets, M Gooden, C Arnold, E Bond, T Bredeweg, M Folwer, G Rusev, D Vieira, J Wilhelmy, W Tornow, C Howel, C Bhatia, M Bhike, F Krishichayan, J Kelley, B Fallin, and S Finch. Energy Evolution of the Fission-Product Yields from Neutron-Induced Fission of 235U, 238U, and 239Pu: An Unexpected Observation. Technical Report LLNL-PROC-717402, Lawrence Livermore National Laboratory, January 2017.

[13] Y K Gupta, D C Biswas, O Serot, D Bernard, O Litaize, S Julien-Laferrière, A Chebboubi, G Kessedjian, C Sage, A Blanc, H Faust, U Köster, A Ebran, L Mathieu, A Letourneau, T Materna, and S Panebianco. Fission fragment yield distribution in the heavy-mass region from the Pu239(nth,f) reaction. *Phys. Rev. C*, 96(1):014608, July 2017.

[14] A Gatera, T Belgya, W Geerts, A Göök, F J Hambsch, M Lebois, B Maróti, A Moens, A Oberstedt, S Oberstedt, F Postelt, L Qi, L Szentmiklósi, G Sibbens, D Vanleeuw, M Vidali, and F Zeiser. **Prompt-fission γ -ray spectral characteristics from ^{239}Pu(n_{th}, f)** . *Phys. Rev. C*, 95(6):064609, June 2017.

[15] D G Madland. Total prompt energy release in the neutron-induced fission of 235U, 238U, and 239Pu. *Nucl. Phys. A*, 772(3-4):113–137, June 2006.

[16] C Schmitt, A Guessous, J P Bocquet, H G Clerc, R Brissot, D Engelhardt, H R Faust, F Gönnenwein, M Mutterer, H Nifenecker, J Pannicke, C H Ristori, and J P Theobald. Fission yields at different fission-product kinetic energies for thermal-neutron-induced fission of 239Pu. *Nucl. Phys. A*, 430(1):21–60, November 1984.

[17] S Warren. *Development of the STEFF detector for the neutron Time Of Flight facility (n TOF), CERN.* . PhD thesis, University of Manchester.