γ -ray Energy Spectra and Multiplicities from the Neutron-induced Fission of ²³⁵U using STEFF.

A.G. Smith, T. Wright, J. Billowes, J.Ryan, S.Warren, C. Gurrrero^{\approx}, L. Tassan-Got[⊗], A. Pollitt^{*}, O.Serot[†], I. Tsekhanovich[‡]

The University of Manchester, Manchester, M13 9PL, U.K. ^DCERN Physics Department, CH-1211 Genève 23, Switzerland. /Universidad de Sevilla, Sevilla, Spain [⊗]IPN, Université Paris-Sud, 91406 ORSAY Cedex, France ‡Université de Bordeaux 1, Talence Cedex 33405, France †CEA-Cadarache, DEN/DER/SPRC/LEPh, France ∗ Institut Laue-Langevin, 6 rue Jules Horowitz, 38042 Grenoble Cedex, France.

December 8, 2013

EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH Proposal to the ISOLDE and Neutron Time-of-Flight Committee

Abstract

An experiment is proposed to use the STEFF spectrometer at n_TOF to study fragment- γ correlations following the neutron-induced fission of ²³⁵U. The STEFF array of 12 NaI detectors will allow measurements of the single- γ energy, the γ multiplicity, and the summed γ energy distributions as a function of the mass and charge split, and deduced excitation energy in the fission event. These data will be used to study the origin of fission-fragment angular momenta, examining angular distribution effects as a function of incident neutron energy. The principal application of this work is in meeting the NEA high-priority request for improved γ ray data from 235 U(n, F). To improve the detection rate and expand the range of detection angles, STEFF will be modified to include two new fission-fragment detectors each at 45° to the beam direction.

> Requested Protons: 3×10^{18} on target in one run Experimental Area: EAR2

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 fisison 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-byevent measurements may be broadly separated into different catagories 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-offlight is measured for a one fragment and the other fragment is typically undetected; mass-separator measurements (such as those performed using Lohengrin[ref]) 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 utilize this approach and therfore 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 can be performed in very close geometry with a windowless double-gridded ion chamber, yielding good energy resolution and high effeciency, 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 mumber (Z) of either of the fission fragments, to which, by constrast, radiochemical measurements are sensitive. The STEFF spectrometer 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^* distribution and the segmentation of the anodes allows for measurement of the direction (θ, ϕ) of the fission axis (to an accuracy $\pm 2^{\circ}$). 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 and a preliminary report of those activities is attached to this proposal as a technical appendix. Here we are asking for time for the first experiment in a series in which we will use the array of scintillators to make 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^* of the fission fragments. This allows us to perform a complete analysis of the fission process to within the above-mentioned resolution of STEFF.

Physics Case

The motivation for study of a large set of parameters of the fission process in one experiment lies in the ability to address correlations of relevance to the fission mechanism and to better determine nuclear data of relevance to the nuclear industry. The array of scintillators together with the capability to distinguish between prompt neutrons and prompt γ rays by time-of-flight means that we will measure multiplicity and energy distributions as a function of mass. The 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. This is especially important in the region outside of the core, where heating by prompt and delayed γ rays dominates. 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. [3][4][5]) and shows signicant variations (on the level of 15% in the measured total energy). The NEA high-priority request has prompted recent experiments to adress these uncertainties[6, 7].

On a fundemental level, the data may be used to address the long-standing question as to the origin of the 10 or so units of angular momentum produced in fission. γ -ray multiplicities and angular distributions will be used to give systematic information on the variation of mean spin and alignment with A and E^* . Our measurements of fragment- γ correlations have shown that it is possible to use prompt γ -ray coincidence data to probe the dynamical properties of the fission mechanism itself. It is well known that the nuclear spin is aligned perpendicular to the fission axis, but until recently it has been difficult to quantify this alignment. By the novel approach of using fragment- γ angular correlation data in conjunction with measurements of the feeding distribution of low-spin yrast states as input to a statistical model decay calculation, it has been possible to determine not only the mean spin of a particular fission-fragment at scission, but also the width of the initial m-substate distribution, i.e. the spread in polar angle of the nuclear spin. These results show that there is a significant contribution from $m \neq 0$ states, pointing to the importance of twisting modes in generating fragment spins. This technique is sensitive to γ -ray contamination from the complementary fragments and would greatly benefit from the use of STEFF as a fragment trigger when used in conjunction with Ge detectors. Anisotropy measurements may also be made for the incident-neutron to fissionfragment angular distribution. These are of particular interest in the case of fast-neutron induced fission where the existing data show remarkable differences to corresponding results from proton-induced fission at similar energies (a factor of two difference in the anisotropies for 232 Th fission is observed in the energy range 5-80 MeV as confirmed by recent n TOF measurements[8]). At present, the reason for the difference in the dynamics of neutron-induced and proton-induced fission is poorly understood, but the sensitivity of anisotropy to neutron energy and the marked proton/neutron differences point to the structure of transitional states playing a role, even up to energies of nearly 100 MeV. So far there is very little data on anisotropy as a function of fragment mass or fragment kinetic energy. Accurate knowledge of the anisotropy is also of some importance for converting experimental data into accurate yield measurements and therefore has an immediate application in the area of waste management.

Experimental and Technical Details

Discussions with the n TOF collaboration have concluded that STEFF can be accommodated at the new EAR2 experimental hall with only minor modifications to the supporting structure. These alterations give the opportunity for a redesign so that STEFF will include an extra two short ight arms, each consisting of a timing detector and a Bragg chamber. This means that STEFF will provide clean detection of the fission fragments along 3 possible axes, more than trebling the solid angle of the spectrometer. These new arms have short time-of-flight sections and hence inferior mass resolution, but serve to boost the solid angle and increase the range of angles possible for angular correlation work. The new Bragg detectors may be set at a higher pressure than the main detectors giving the possibility of detection of ternary ssion where the lighter ternary particle is distinguished from the main fission fragments by the digitized pulse shape. The upgrades to STEFF will be completed during the course of 2014.

Our intention is to use STEFF at the EAR2 station, where the 25-fold increase in neutron flux over EAR1 makes it possible to perform fission experiments measuring a greater range of parameters than before. The neutron beam will be collimated such that it covers a 25 cm² area and is incident onto a target of 0.25 mg 235 U, with an active depth of 5 nm. In such a situation the fission reaction rate will be approximately 700 fissions per proton pulse. The modifications to STEFF will increase the geometrical acceptance for fragment detection to a solid angle of 0.134 sr; combined with the intrinsic fragment-efficiency of 50% this results in a detection rate of approximately 5.62 fission per pulse over a time interval of 10ms. The detection rates (divided into several energy windows) are shown in the Table below. Such a set up with a total of 3×10^{18} protons results in around 10⁶ detected fragment- γ events, with around 5×10^5 with time-of-flight information. The upper neutron energy limit of the measurements is primarily limited by the saturation of the NaI scintillators from the ' γ flash' effect. Simulations have shown that, in comparison to EAR1, the γ flash at EAR2 will be significantly reduced[9]; this will be tested during the commissioning of the EAR2 station. There is also a plan to implement a Gated Voltage Divider system for the scintillators of STEFF to further combat the flash. During the γ flash the GVD will remove the voltage (thus electric potential) across several of the photomultiplier tube dynodes, preventing the signal propagating through the detector, whilst still allowing time for the scintillators to recover in time for the faster neutrons[10]. As there will be no experimental data on the intensity of the γ flash at EAR2 until the comissioning phase, it is recognized by the collaboration that there is a small chance that the conditions at EAR2 will preclude the use of the NaI detectors. In this event the experiment will be withdrawn.

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.

References

- [1] F. Gönnenwein. Mass, Charge and Kinetic Energy of Fission Fragments. CRC, 2000.
- [2] James Terrell. Neutron Yields from Individual Fission Fragments. Phys. Rev., $127(3):880-904$, Aug 1962.
- [3] R. W. Peelle and F. C. Maienschein. Spectrum of Photons Emitted in Coincidence with Fission of ²³⁵U by Thermal Neutrons. Phys. Rev. C, $3(1)$:373-390, Jan 1971.
- [4] Frances Pleasonton, Robert L. Ferguson, and H. W. Schmitt. Prompt Gamma Rays Emitted in the Thermal-Neutron-Induced Fission of ²³⁵U. Phys. Rev. C, 6(3):1023– 1039, Sep 1972.
- [5] V. V. Verbinski, Hans Weber, and R. E. Sund. Prompt Gamma Rays from $^{235}U(n, f)$. ²³⁹Pu(n, f), and Spontaneous Fission of ²⁵²Cf. Phys. Rev. C, 7(3):1173-1185, Mar 1973.
- [6] A. Oberstedt, T. Belgya, R. Billnert, R. Borcea, T. Brys, W. Geerts, A. Göök, F.-J. Hambsch, Z. Kis, T. Martinez, S. Oberstedt, L. Szentmiklosi, K. Takàcs, and M. Vidali. Improved values for the characteristics of prompt-fission γ -ray spectra from the reaction $^{235}U(n_{th},f)$. *Phys. Rev. C*, 87:051602, May 2013.
- [7] I. Stetcu, P. Talou, T. Kawano, and M. Jandel. Isomer production ratios and the angular momentum distribution of fission fragments. *Phys. Rev. C*, 88:044603, Oct 2013.
- [8] Tarrio D. Neutron-induced Fission Fragment Angular Distribution at CERN n TOF: The Th-232 Case. PhD thesis, CERN, 2012.
- [9] E. Chiaveri et al. Proposal for nTOF Experimental Area 2 (EAR-2). Technical report, CERN, 2012.
- [10] K.Y. Hara, H. Harada, Y. Toh, and J. Hori. Suppression Using a Gated Photomultiplier Assembled with an LaBr3(Ce) Detector to Measure Fast Neutron Capture Reactions. Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment, $723(0):121 - 127$, 2013 .