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Abstract. Prompt γ rays from the neutron-induced fission of ^{235}U have been studied using the GEANIE spectrometer situated at the LANSCE/WNR "white" neutron facility. Gamma-ray production cross sections for 29 ground-state-band transitions in 18 even-even fission fragments were obtained as a function of incident neutron energy, using the time-of-flight technique. Independent yields were deduced from these cross sections and fitted with standard formulations of the fragment charge and mass distributions to study the transition from asymmetric to symmetric fission. The results are interpreted in the context of the disappearance of shell structure at high excitation energies.

INTRODUCTION

The properties of fragments produced in the fission of actinide nuclei provide a window into the dynamics of the fission process. Because the fission process unfolds on a very short time scale (e.g. $\sim 10^{-21}$ s from saddle to scission), the study of fission fragments and emitted neutrons is usually the only experimental tool available to gain insight into the fission mechanism. A successful theory of nuclear fission would shed light on many areas of study, such as the structure and behavior of well-deformed nuclei in general, and the stabilizing mechanisms at play in the formation of superheavy elements. In addition, fission has played a unique role in the population and study of neutron-rich nuclei, and it provides an important tool for the production of radioactive nuclear beams [1].

In the induced fission of actinide nuclei, the target nucleus is initially excited by an incident beam of charged particles, photons or neutrons. The nucleus may then reduce its excitation energy by emitting "pre-scission" neutrons. Eventually, the fission channel becomes more favorable, and scission occurs. The nascent fragments are accelerated by their mutual Coulomb repulsion and may themselves emit "post-scission" neutrons. Properties of the "primary fragments", defined as the fragments produced by the fission process before they emit neutrons, are directly related to the state of the fissioning system at the moment of scission. Experimentally, only the "secondary fragments", remaining after neutron emission from the primary fragments, can be observed due to the time scale involved (e.g $10^{-18} - 10^{-17}$ s from formation of the primary fragments to the onset of neutron evaporation). After neutron emission, the secondary fragments are left in an excited state which may further decay by emitting γ rays. Following γ emission, the fragments in their ground or isomeric states are still neutron rich, and will usually β-decay until they reach the valley of stability.

The excitation energy of the fissioning system plays an important role in the dynamics of the fission process. Asymmetric division into a light and heavy fragment is thought to result from shell effects [2], whereas symmetric division is consistent with a classical liquid-drop picture of the fissioning nucleus. Thus, fission-fragment yields studied as a function of excitation energy are an experimental measure of the vanishing of shell structure.

The data presented in this paper consist of cross sections for well-characterized, ground-state-band transitions in even-even ^{235}U(n,f) fission fragments, acquired using the GEANIE spectrometer at the LANSCE/WNR "white" neutron source. These cross sections are used to extract fission-fragment yields as a function of the excitation energy of the fissioning nucleus, thereby affording a unique opportunity to follow the transition from asymmetric to symmetric fission in the n+^{235}U reaction. A more detailed account of these results can be found in references [3] and [4].

EXPERIMENTAL DETAILS

A "white" neutron spectrum is produced at the LANSCE/WNR facility through a spallation reaction induced by a pulsed 800-MeV proton beam incident on a natural tungsten target. The proton beam is bunched into 625-μs trains of micropulses 1.8 μs apart produced at a typical frequency of 100 Hz, resulting in a 6% duty cycle for the present data set. The GErmanium Array for Neutron-Induced Excitations (GEANIE) consists of 11 Compton-suppressed planar detectors, 9 Compton-suppressed and 6 unsuppressed coaxial detectors, all situated at a distance of $\approx$ 14 cm from the scattering sample located at the spectrometer focal point. The planar detectors are grouped at mostly forward and backward angles, while the coaxial detectors are located about 90° with respect to the beam direction. The efficiency of the array has been calibrated through a series of source measurements, supplemented by detailed modeling [5] using the transport code MCNP [6]. The GEANIE spectrometer is located 20.34 m downstream from the spallation target. A fission chamber [7], placed in the beam 1.86 m upstream from the GEANIE spectrometer, serves as a neutron flux monitor. Neutron energies are determined by the time-of-flight (TOF) technique.

The present data were acquired over a total of 8 days using a 0.617(44) g/cm^2, 93.2%-enriched ^{235}U metal. In all, 4.7×10^8 and 1.6×10^8 prompt, single-and-higher-fold γ-ray events were recorded in the planar and coaxial detectors, respectively, for all neutron energies. A total of 1.4×10^7 $\gamma-\gamma$ coincidence counts were acquired in the E_n = 1-20 MeV range.

DATA ANALYSIS

In the analysis of the single-fold data, peaks are identified primarily through their γ-ray energies. An accurate γ-ray energy calibration for the singles data was obtained using in-beam peaks with precisely known energies. A background subtraction was applied to the singles γ-ray data, using a spectrum gated on TOFs in-between micropulse bursts. The combination of polyethylene absorbers placed in the beam and the background subtraction served to reduce beam "wrap-around" effects, caused by the natural spread in neutron energies of the beam. Gamma-ray excitation functions with respect to incident neutron energy were generated from these background-subtracted data in equal-TOF steps. The coincidence data were also calibrated using in-beam transitions, and used to confirm γ-ray assignments and to search for possible contaminant peaks. Angular-distribution effects could not be systematically extracted for all the transitions of interest. However, based on those γ rays for which angular distributions could be measured, the correction due to unobserved γ rays with a non-isotropic distribution was estimated to be at most 15%.

A reduced set of γ rays was obtained for the fragment-yield analysis by subjecting the GEANIE data to a series of increasingly demanding constraints. Initially, a total of 206 γ rays were identified as known transitions in 56 distinct fission-fragment nuclei, based on the analysis of coincident data. For 146 of those γ rays, the corresponding full-energy peak in the single-fold data could be fitted and an excitation function could therefore be extracted. Discarding those γ rays that showed evidence of a strong contaminant in the coincidence data, from which they could not be separated using the energy resolution of the singles data, further reduced the number of viable γ rays to 64. From these, a subset of forty $2^+_1 \rightarrow 0^+_1$, $4^+_1 \rightarrow 2^+_1$ and $6^+_1 \rightarrow 4^+_1$ transitions in 22 even-even nuclei was selected. These ground-state band transitions were preferentially chosen because it has been shown in the case of spontaneous fission of ^{252}Cf [8], that the γ-ray decay of excited even-even fission fragments proceeds, for the most part, through the low-lying members of the ground-state band. Finally, the γ-ray yields were compared to radiochemical data at E_n = 14 MeV, evaluated by James *et al.* [9] and tabulated in [10]. Keeping in mind limitations in the data due to angular-distribution effects and decay paths which bypass the ground-state band, a final set of 29 γ rays in 18 fragments was selected. The excitation functions for these 29 transitions were subsequently normalized to the James *et al.* evaluation at E_n = 14 in order to obviate any remaining concerns with the overall scale of the yields.

MODEL

The fission-fragment mass yield distribution is usually described in terms of a five-Gaussian fit of the form [9]:

$$M(A) = \frac{N_1}{\sqrt{2\pi}\sigma_1}\left[e^{-\frac{(A-\bar{A}-D_1)^2}{2\sigma_1^2}} + e^{-\frac{(A-\bar{A}+D_1)^2}{2\sigma_1^2}}\right] + \frac{N_2}{\sqrt{2\pi}\sigma_2}\left[e^{-\frac{(A-\bar{A}-D_2)^2}{2\sigma_2^2}} + e^{-\frac{(A-\bar{A}+D_2)^2}{2\sigma_2^2}}\right] + \frac{N_3}{\sqrt{2\pi}\sigma_3}e^{-\frac{(A-\bar{A})^2}{2\sigma_3^2}} \quad (1)$$

where A is the secondary fragment mass number, $\overline{A}$ is the mean mass of the distribution, and N_i, σ_i, and D_i are the parameters of the i^{th} Gaussian function. Subscripts 1 and 2 in Eq. 1 are associated with two distinct asymmetric fission modes, while subscript 3 corresponds to the symmetric fission channel. The form of this equation is constrained by requirements of symmetry about the centroid $\overline{A}$, and the Gaussian-function scales N_1, N_2, and N_3 are related by the mass-conservation condition $2N_1 + 2N_2 + N_3 = 2$. The fission-fragment charge distribution is given by the "fractional independent yield" modeled by by Wahl [11, 12] using the modified Gaussian form:

$$FIY(A,Z) = F(A,Z)N(A)\int_{Z-1/2}^{Z+1/2} dZ' \frac{1}{\sqrt{2\pi}\sigma_z} e^{-\frac{Z'-Z_p(A)}{2\sigma_z^2}} \tag{2}$$

where $F(A,Z)$ is a correction factor for odd-even effects, $N(A)$ is a normalization factor taken to ensure summation to 1 for each mass, and $Z_p(A)$ and σ_z determine the Gaussian charge distribution before the correction for odd-even effects. For many actinide nuclei, including ^{235}U, $F(A,Z)$ and $N(A)$ are essentially unity in neutron-induced fission for $E_n > 0.4$ MeV [9]. The most probable charge $Z_p(A)$ has been parameterized by Wahl [12] as a piecewise-linear function of the the secondary fragment mass number A.

The product of Eqs. 1 and 2 gives the secondary fragment distribution at a given incident neutron energy. In order to obtain a smooth, consistent variation of the distribution over a wide neutron-energy range, the parameters in Eqs. 1 and 2 are given simple low-order polynomial dependences on E_n, with the polynomial coefficients to be determined by fitting the GEANIE yields. Relating the incident neutron energy to the excitation energy of the fissioning system requires additional physics. In the simplest model, complete fusion between projectile and target can be assumed, with an initial excitation energy $E_x^{(0)} = E_n^{(c.m)} + Q_{CN}$, where $E_n^{(c.m.)}$ is the neutron energy in the center-of-mass frame, and Q_{CN} is the Q value for compound nucleus formation. Pre-scission neutrons are then emitted in a statistical evaporation process, with fission competing with neutron emission at every step. A more accurate description of the process should allow for the emission of high-energy pre-compound neutrons, before the further statistical evaporation of neutrons. In the present work, pre-compound effects have been included by using the reaction code ALICE [13], adapted [14] to generate the distribution of residual nuclei and their excitation energies immediately after pre-compound neutron emission. The subsequent statistical evaporation of neutrons was reproduced by using empirical estimates of Kozulin *et al.* [15] for the neutron multiplicity as a function of the mass and excitation energy of the fissioning nucleus, and assuming and average energy ΔE = 8.5 MeV removed by each statistical neutron. Due to the spread in excitation energy of the residual nuclei following pre-compound emission, it is difficult to associate a well-defined excitation energy for the fissioning nucleus at neutron energies much higher than 85 MeV.

Before proceeding to the analysis of the transition from asymmetric to symmetric fission as a function of E_x, it is worth noting that the fissioning system cannot be readily identified with a specific uranium isotope, since scission can be preceded by the emission of any number of pre-scission neutrons. Therefore, the properties of the fission channel extracted here pertain to an ensemble of nuclei described only by their excitation energy.

RESULTS AND DISCUSSION

The parameters in Eqs. 1 and 2 were given low-order polynomial dependences on neutron energy E_n, producing a total of 17 free parameters. The fits were performed over the energy range E_n = 3.7-84.4 MeV. The set of 29 γ rays over the 40 neutron-energy steps in the fitted range provided 1160 data points, from which 7 obvious outliers were removed. The remaining 1153 data points were fitted to the product of Eq. 1 and 2 with a $\chi^2/\nu = 3.96$. Experimental mass-yield data and the corresponding fitted curves are shown in Fig. 1 at selected neutron energies. The quality of the fits deteriorates with increasing neutron energy, but the trend is clear. By E_n = 50.0 MeV, the symmetric fission channel represents $\approx 72\%$ of the total fission yield. Fits with alternate parameterizations were also tested (see [3]), but did not produce a significant improvement in the results. Furthermore, the fitting range could not be extended beyond E_n = 3.7-84.4 MeV without a noticeable degradation in the quality of the fit, or a substantial increase in the number of free parameters.

The fits displayed in Fig. 1 are combined with the conversion from neutron energy to excitation energy discussed in the previous section to produce a map of the transition from asymmetric to symmetric fission as a function of excitation energy of the fissioning system. The deduced probability of symmetric fission, calculated as $N_3/2$ in Eq. 1, is plotted in Fig. 2. The solid circles represent the fitted values at the discrete experimental energies with appropriate uncertainties from the fit, and the horizontal bars represent the standard deviation of the energy distribution (and not the uncertainty on the energy itself).

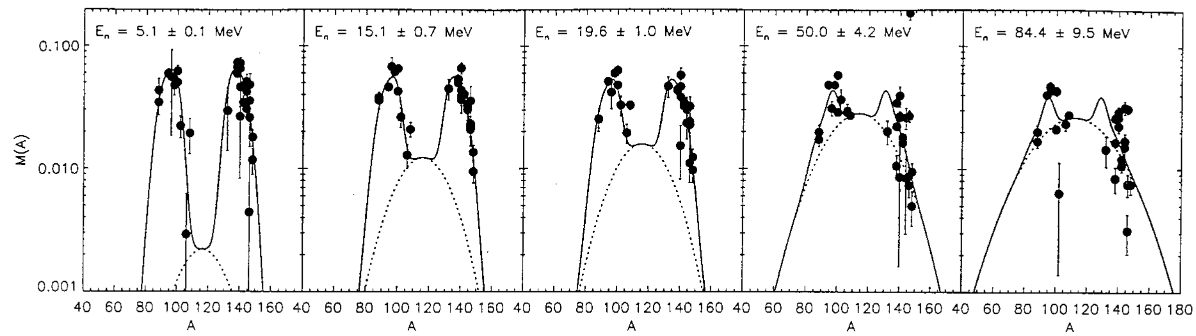

FIGURE 1. Mass yields fitted to the GEANIE data using Eqs. 1 and 2, and shown at selected neutron energies.

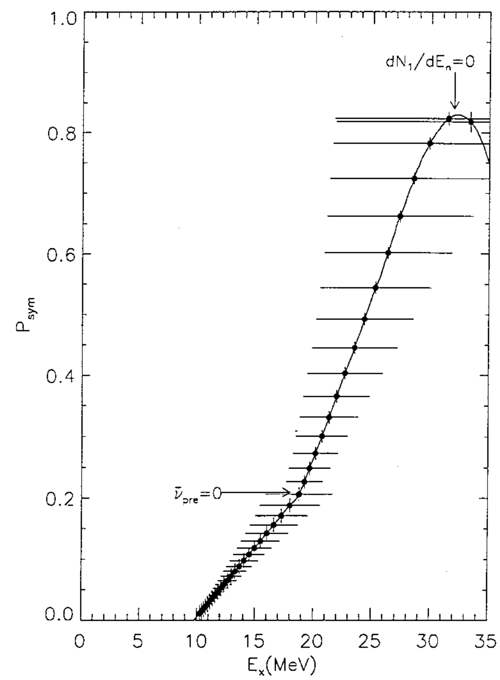

FIGURE 2. Deduced probability P_{sym} of symmetric fission as a function of internal excitation energy of the fissioning nucleus.

This plot must be interpreted within the limitations of the GEANIE data, and of the fitting model. The behavior of the curve is suspect at two points in particular: near E_x = 18.8 MeV, below which there are no data on pre-scission neutron multiplicities which are taken as identically zero, and near E_x = 32.0 MeV, where poor statistics in the GEANIE data and the limited number of free parameters in the model cause the Gaussian strength parameter N_1 in Eq. 1, to start increasing again for $E_n > 73.7$ MeV, instead of remaining small. Nevertheless, the plot shows the strength of the symmetric-fission channel starting to increase near E_x = 14 MeV (defined quantitatively here as the excitation energy for which P_{sym} = 10%) and, as suggested by visual extrapolation, becoming dominant with P_{sym} = 90% somewhere near E_x = 35 MeV. Thus, the transition from mostly asymmetric to mostly symmetric fission, which can be attributed to the washing out of shell effects, occurs over a relatively modest span of $\sim$ 20 MeV of excitation energy.

CONCLUSION

Yields for 29 ground-state-band transition in 18 even-even fission fragments were extracted from GEANIE data for the ^{235}U(n,f) reaction. In order to eliminate some systematic uncertainties, the data were normalized to the accepted yields at E_n = 14.0 MeV. The data were then fitted with a model composed from the product of well-established formulations for the fragment charge and mass distributions. As a result, the evolution of the symmetric- and asymmetric-mass division peaks could be followed as a function of the internal excitation energy of the fissioning nucleus. Based on these trends, and allowing for limitations of both data and model, the symmetric fission channel could be seen to start increasing near $E_x \sim 14$ MeV, becoming dominant within ~ 20 MeV of excitation energy. This behavior has been identified with the systematics of the disappearance of shell structure as a function of excitation energy in the fissioning system.

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REFERENCES

1. von Egidy, T., Kienle, P., Koster, U., Habs, D., Gross, M., Kester, O., Maier, H. J., and Thirolf, P. G., *Acta Phys. Slov.*, **49**, 107–116 (1999).
2. Wilkins, B. D., Steinberg, E. P., and Chasman, R. R., *Phys. Rev. C*, **14**, 1832–1863 (1976).
3. Younes, W., Becker, J. A., Bernstein, L. A., Garrett, P. E., McGrath, C. A., McNabb, D. P., Nelson, R. O., Johns, G. D., Wilburn, W. S., and Drake, D. M., Transition from asymmetric to symmetric fission in the ^{235}U(n,f) reaction (2001), submitted to Phys. Rev. C.
4. Younes, W., Becker, J. A., Bernstein, L. A., Garrett, P. E., McGrath, C. A., McNabb, D. P., Nelson, R. O., Johns, G. D., and Wilburn, W. S., Fission-fragment yields following the ^{235}U(n,f) reaction with $1 \leq E_n$(Mev) ≤ 250, Tech. rep., LLNL (2001), manuscript in preparation.
5. McNabb, D. P., Archer, D. E., Becker, J. A., Bernstein, L. A., Garrett, P. E., Hauschild, K., McGrath, C. A., Younes, W., Devlin, M., Drake, D. M., Johns, G. D., Nelson, R. O., and Wilburn, W. S., Uncertainty budget and efficiency analysis for the ^{239}Pu(n,2nγ) partial reaction cross-section measurements, Tech. Rep. UCRL-ID-139906, LLNL (1999).
6. Briesmeister, J. F., A general monte carlo code for neutron and photon transport, Tech. Rep. LA-7396-M-Rev.2, LANL (1986).
7. Wender, S. A., Balestrini, S., Brown, A., Haight, R. C., Laymon, C. M., Lee, T. M., Lisowski, P. W., McCorkle, W., Nelson, R. O., Parker, W., and Hill, N., *Nucl. Instrum. and Methods Phys. Res. A*, **336**, 226–231 (1993).
8. Wilhelmy, J. B., Cheifetz, E., Jared, R. C., Thompson, S. G., Bowman, H. R., and Rasmussen, J. O., *Phys. Rev. C*, **5**, 2041–2060 (1972).
9. James, M. F., Mills, R. W., and Weaver, D. R., *Prog. Nucl. Energy*, **26**, 1–29 (1991).
10. James, M., and Mills, R., Tech. Rep. AEA-TRS-1015, 1018 and 1019, UKAEA, BNF, PLC and Nuclear Electric (1993), MAT # 9240, June 1993; data retrieved from the ENDF database.
11. Wahl, A. C., Ferguson, R. L., Nethaway, D. R., Troutner, D. E., and Wolfsberg, K., *Phys. Rev.*, **126**, 1112–1127 (1962).
12. Wahl, A. C., *Phys. Rev. C*, **32**, 184–194 (1985).
13. Blann, M., Code alice 85/300, Tech. Rep. UCID-20169, LLNL (1984).
14. Chadwick, M. B. (2000), private communication.
15. Kozulin, E. M., Rusanov, A. Y., and Smirenkin, G. N., *Phys. At. Nucl.*, **56**, 166–174 (1993).

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