PHYSICAL REVIEW C **89**, 051604(R) (2014)

Excitation energy dependence of the total kinetic energy release in ^{235}U(n,f)

R. Yanez, L. Yao, J. King, and W. Loveland
Department of Chemistry, Oregon State University, Corvallis, Oregon 97331, USA

F. Tovesson and N. Fotiades
Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA
(Received 24 January 2014; revised manuscript received 6 May 2014; published 27 May 2014)

The total kinetic energy release in the neutron induced fission of ^{235}U was measured (using white spectrum neutrons from the Los Alamos Neutron Science Center) for neutron energies from $E_n = 3.2$–50 MeV. In this energy range the average post-neutron total kinetic energy ($E_{\rm TKE}$) release drops from 167.4 ± 0.7 to 162.1 ± 0.8 MeV, exhibiting a local dip near the second chance fission threshold. The values and the slope of the $E_{\rm TKE}$ vs E_n agree with previous measurements but disagree (in magnitude) with systematics. The variances of the $E_{\rm TKE}$ distributions are larger than expected; and apart from the structure near the second chance fission threshold, the distributions are invariant for the neutron energy range from 11 to 50 MeV. We also report the dependence of the total excitation energy in fission ($E_{\rm TXE}$) on neutron energy.

DOI: 10.1103/PhysRevC.89.051604 PACS number(s): 25.85.Ec, 25.60.Pj, 25.70.Jj

Most of the energy released in the nuclear fission process appears in the kinetic energy of the fission fragments. The "Viola systematics" [1] describe this total kinetic energy ($E_{\rm TKE}$) using the equation

$$E_{\rm TKE} = (0.1189 \pm 0.0011)\frac{Z^2}{A^{1/3}} + 7.3(\pm 1.5)\ \text{MeV}. \quad (1)$$

The first term in this equation represents the Coulomb repulsion between the deformed fragments at scission. The second additive term can be interpreted as representing the fact that the fission fragments are in motion at scission. The value of this term is consistent with some trajectory calculations of long-range α-particle emission in fission [2].

The deformed scission point fragments will contract to their equilibrium deformations and the energy stored in deformation will be converted into internal excitation energy. Thus we can define a related quantity, the total excitation energy ($E_{\rm TXE}$) in fission, as

$$E_{\rm TXE} = Q - E_{\rm TKE}, \quad (2)$$

where Q is the mass-energy release. One quickly realizes that these quantities depend on the mass split in fission which in turn, at low excitation energies, may reflect the fragment nuclear structure. The $E_{\rm TXE}$ is the starting point for calculations of the prompt neutron and γ emission in fission, the yields of β-emitting fission fragments, reactor anti-neutrino spectra, etc. As such, it is a fundamental property of all fissioning systems and sadly not very well known.

As a practical matter, one needs to know the dependence of the $E_{\rm TKE}$ and $E_{\rm TXE}$ on neutron energy for the neutron induced fission of technologically important actinide fissioning systems like ^{233}U(n,f), ^{235}U(n,f), and ^{239}Pu(n,f). The first question we might pose is whether the $E_{\rm TKE}$ should depend on the excitation energy of the fissioning system. Does the energy brought in by an incident neutron in neutron induced fission appear in the fragment excitation energy or does it appear in the total kinetic energy? In a variety of experiments, one finds that increasing the excitation energy of the fissioning system does not lead to significant increases in the $E_{\rm TKE}$ of the fission fragments or changes in the fragment separation at scission [3]. However, there may be more subtle effects that render this statement false in some circumstances. For example, we expect, on the basis of the Coulomb energy systematics given above, that the $E_{\rm TKE}$ will be proportional to changes in the fission mass splits which in turn can depend on the excitation energy.

For the technologically important reaction ^{235}U(n,f), Madland [4] summarizes the known data [5–7] with the following equations:

$$\langle T_f^{\rm tot}\rangle = (170.93 \pm 0.07) - (0.1544 \pm 0.02)E_n\ \text{(MeV)}, \quad (3)$$

$$\langle T_p^{\rm tot}\rangle = (169.13 \pm 0.07) - (0.2660 \pm 0.02)E_n\ \text{(MeV)}, \quad (4)$$

where E_n is the energy of the incident neutron and $T_f^{\rm tot}$ and $T_p^{\rm tot}$ are the average total fission fragment kinetic energy (before neutron emission) and the average fission product kinetic energy after neutron emission, respectively. These quantities are related by the relation

$$\langle T_p^{\rm tot}(E_n)\rangle = \langle T_f^{\rm tot}(E_n)\rangle\left[1 - \frac{\overline{\nu_p}(E_n)}{2A}\left(\frac{\langle A_H\rangle}{\langle A_L\rangle} + \frac{\langle A_L\rangle}{\langle A_H\rangle}\right)\right]. \quad (5)$$

These data show a modest decrease in $E_{\rm TKE}$ with increasing excitation energy for the neutron energy interval $E_n = 1$–9 MeV. There is no clearly identified changes in the $E_{\rm TKE}$ values near the second chance fission threshold, a feature that is important in semi-empirical models of fission such as represented by the GEF code [8].

In this Rapid Communication, we report the results of measuring the total kinetic energy release in the neutron induced fission of ^{235}U for neutron energies $E_n = 3.2$–50 MeV. The method used for the measurement is the 2E method, i.e., measurement of the kinetic energies of the two coincident fission products using semiconductor detectors. The time of

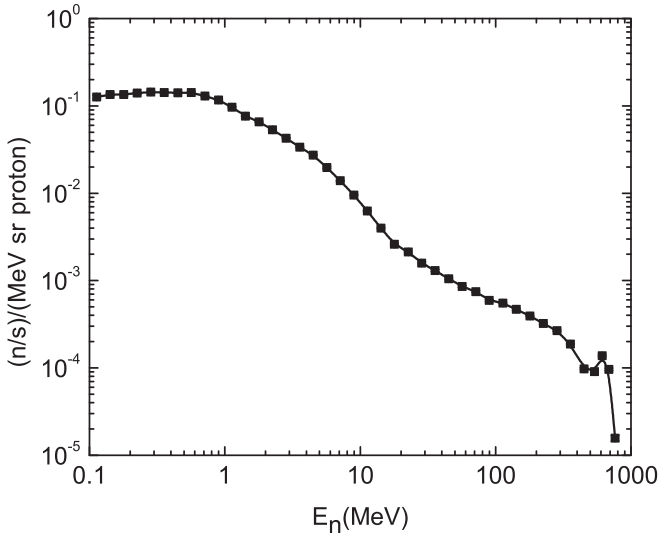

FIG. 1. Calculated neutron spectrum in the 15R beam area [11]. To convert the ordinate to units of neutrons/second, one must multiply the plotted values by the solid angle (sr) of the fission target, the proton beam intensity, and the width of the neutron energy bin (MeV).

flight of the neutrons inducing fission was measured, allowing deduction of their energy. The details of the experiment are discussed, the experimental results are presented, a comparison of the results with various models and theories is made, and conclusions are summarized.

This experiment was carried out at the Weapons Neutron Research Facility (WNR) at the Los Alamos Neutron Science Center (LANSCE) at the Los Alamos National Laboratory [9,10]. "White spectrum" neutron beams were generated from an unmoderated tungsten spallation source using the 800 MeV proton beam from the LANSCE linear accelerator. The experiment was located on the 15R beam line (15°-right with respect to the proton beam). The calculated white spectrum at the target position is shown in Fig. 1 [11]. The proton beam is pulsed allowing one to measure the time of flight (energy) of the neutrons arriving at the experimental area. The proton beam intensity was typically 1.8 μA.

The neutron beam was collimated to a 1 cm diameter at the entrance to the experimental area. At the entrance to the scattering chamber, the beam diameter was measured to be 1.3 cm. A fission ionization chamber [12] was used to continuously monitor the absolute neutron beam intensities. The ^{235}U target and the Si PIN diode fission detectors were housed in an evacuated, thin-walled aluminum scattering chamber. The scattering chamber was located ∼3.1 m from the collimator, and ∼11 m from the neutron beam dump. The center of the scattering chamber was located 16.46 m from the production target.

The ^{235}U target consisted of a deposit of $^{235}UF_4$ on a thin C backing. The thickness of the ^{235}U was 175.5 μg ^{235}U/cm^2 while the backing thickness was 100 μg/cm^2. The isotopic purity of the ^{235}U was 99.91%. The target was tilted at 50° with respect to the incident beam.

Fission fragments were detected by two arrays of Si PIN photodiodes (Hamamatsu S3590-09) arranged on opposite sides of the beam. The area of the individual PIN diodes was 1 cm^2. The distance of the detectors from the target varied with the angle from 2.60 to 4.12 cm. The coincident detector pairs were at approximately 45, 60, 90, 115, and 135°. The α particle energy resolution of the diodes was 18 keV for the 5475 keV line of ^{241}Am.

The time of flight of each interacting neutron was measured using a timing pulse from a Si PIN diode and the accelerator RF signal. Absolute calibrations of this time scale were obtained from the photofission peak in the fission spectra and the known flight path geometry.

The energy calibration of the fission detectors was done with a ^{252}Cf source. We used the traditional Schmitt method [13]. Some have criticized this method especially for PIN diodes. However, with our limited selection of detectors, we were unable to apply the methods of Ref. [14] to achieve a robust substitute for the Schmitt method.

The measured fragment energies had to be corrected for energy loss in the $^{235}UF_4$ deposit and the C backing foil. This correction was done by scaling the energy loss correction given by the Northcliffe-Schilling energy loss tables [15] to a measured mean energy loss of collimated beams of light and heavy ^{252}Cf fission fragments in 100 μg/cm^2 C foils. The scaling factor that was used was a linear function of mass using the average loss of the heavy and light fission fragments as anchor points. The correction factors at the anchor points were 1.24 and 1.45 for the heavy and light fragments, respectively. Similar factors were obtained if the SRIM code [16] was used to calculate dE/dx. These large deviation factors from measured

TABLE I. Measured E_{TKE} release for ^{235}U(n,f).

Mean neutron energy (MeV)	Width of neutron energy bin (MeV)	$\overline{E_{\mathrm{TKE}}}$ (MeV)	Uncertainty ($\overline{E_{\mathrm{TKE}}}$) (MeV)
3.70 ± 0.01	0.95	167.4	0.7
4.66 ± 0.02	1.1	165.7	0.8
5.82 ± 0.02	1.3	167.7	0.8
7.20 ± 0.02	1.4	166.5	0.8
8.92 ± 0.03	2.1	166.2	0.8
11.87 ± 0.06	4.0	165.1	0.7
16.83 ±0.09	6.0	163.4	0.7
24.24 ± 0.12	4.5	162.9	0.7
34.15 ± 0.15	11.0	161.5	0.8
45.04 ± 0.17	10.0	162.1	0.8

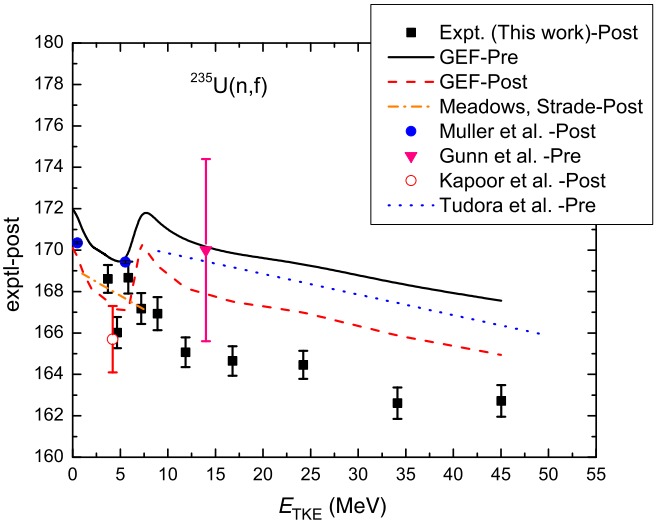

FIG. 2. (Color online) E_{TKE} release data for $^{235}\mathrm{U}(n,f)$.

to calculated fission fragment stopping powers have been observed in the past [17], and represent the largest systematical uncertainty in the determination of the kinetic energies.

The measured average post-neutron emission fission product total kinetic energy release for the $^{235}\mathrm{U}(n,f)$ reaction (Table I) is shown in Fig. 2 along with other data and predictions [18–20]. The uncertainties in the E_{TKE} values reflect the statistical uncertainties in the data analysis and not the systematic errors in the 2E measurement. Other studies [7,21] have suggested a systematic error in this type of measurement of ∼200 keV. These estimates take into account the uncertainties associated with energy loss in the sample and the detector calibration. Other work [22] has suggested that the method of how the prompt neutron correction is applied to the data can change the absolute value of the mean E_{TKE} by an amount of $0.038E_n$ (MeV). The evaluated post-neutron emission data from Madlund [4] are shown as a dashed line while the individual pre-neutron emission measurements of [7] are shown as points. The point at $E_n = 14$ MeV is the average of the data in [18] and [20]. The slope of the measured E_{TKE} release (this work) is in rough agreement with the previous measurements [4] at lower energies. Also shown are the predictions of the GEF model [8]. GEF is a semi-empirical model of fission that provides a good description of fission observables using a modest number of adjustable parameters. The dashed line in Fig. 1 is a semiempirical equation ($E_{\mathrm{TKE}} = 171.5 - 0.1E*$ for $E* > 9$ MeV) suggested by Tudora *et al.* [23]. Qualitatively the decrease in E_{TKE} with increasing neutron energy reflects the increase in symmetric fission (with its lower associated E_{TKE} release) with increasing excitation energy. This general dependence is reflected in the GEF code predictions with the slope of our data set being similar to the predictions of the GEF model but with the absolute values of the E_{TKE} release being substantially less. [The GEF model is known to overestimate the E_{TKE} release in $^{235}\mathrm{U}(n_{\mathrm{th}},f)$ by 1.5 ± 0.5 MeV [8]].

In Fig. 3, we show some typical E_{TKE} distributions along with Gaussian representations of the data. In general, the E_{TKE}

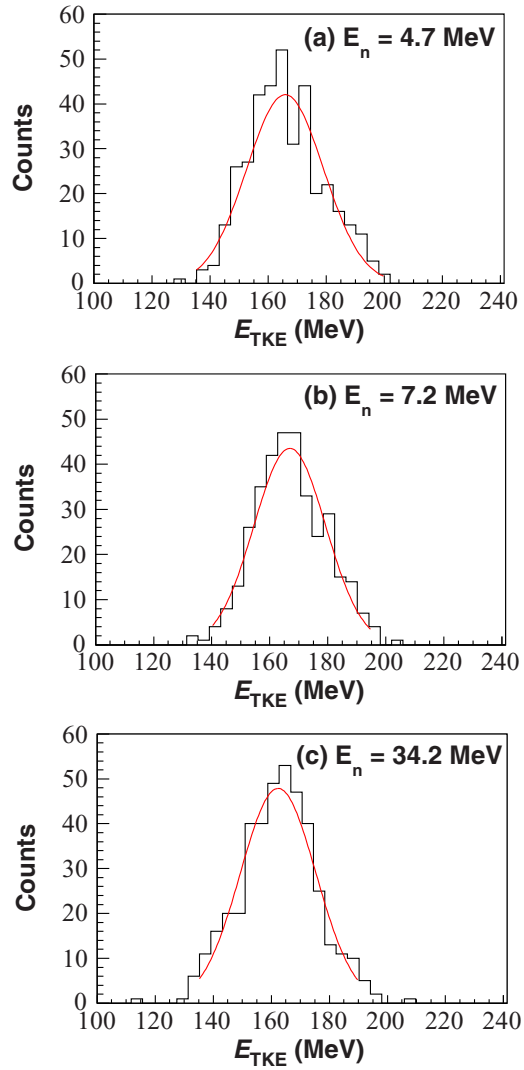

FIG. 3. (Color online) Typical E_{TKE} distributions for $^{235}\mathrm{U}(n,f)$.

distributions appear to be Gaussian in shape. This is in contrast to previous studies [24,25] which showed a sizable skewness in the distributions.

In Fig. 4, we show the dependence of the measured values of the variance of the E_{TKE} distributions as a function of neutron energy along with the predictions of the GEF model of the same quantity. The measured variances are larger than expected. At low energies (near the second chance fission threshold) the observed variances show a dependence on neutron energy similar to that predicted by the GEF model, presumably reflecting the changes in variance with decreasing mass asymmetry. At higher energies (11–50 MeV) the variances are roughly constant with changes in neutron energy. Models [26] would suggest that most of the variance of the E_{TKE} distribution is due to fluctuations in the nascent fragment separation at scission. The constancy of the variances is puzzling.

Using the Q values predicted by the GEF code, one can make a related plot (Fig. 5) of the E_{TXE} values in the $^{235}\mathrm{U}(n,f)$ reaction. The "bump" in the E_{TXE} at lower neutron energies

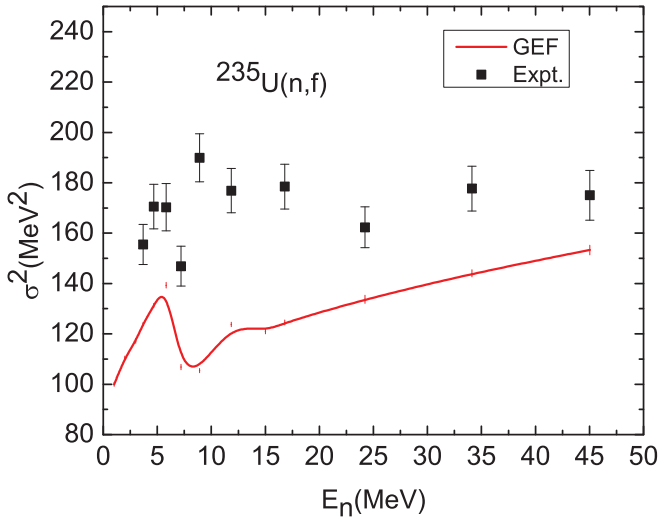

FIG. 4. (Color online) Variance of the E_{TKE} distribution data for $^{235}\mathrm{U}(n,f)$.

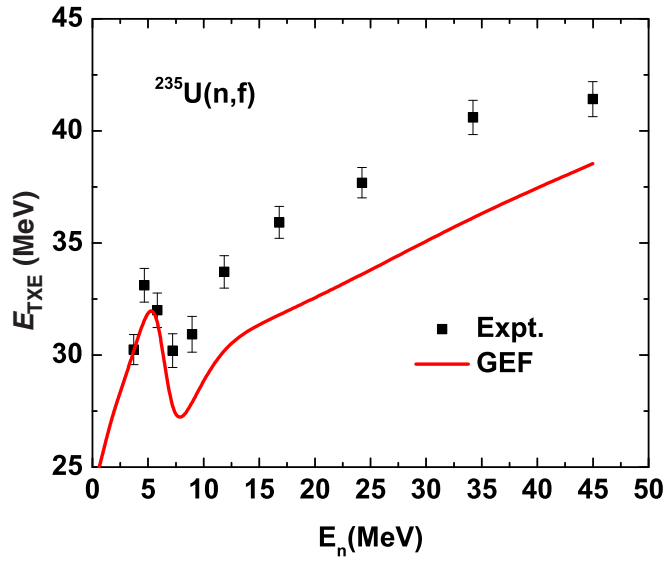

FIG. 5. (Color online) E_{TXE} data for $^{235}\mathrm{U}(n,f)$.

is pronounced and the dependence of the E_{TXE} upon neutron energy agrees with the GEF predictions although the absolute values are larger.

We conclude that (a) for the first time, we have measured the E_{TKE} release and its variance for the technologically important $^{235}\mathrm{U}(n,f)$ reaction over a large range of neutron energies (3.2–50 MeV), (b) the dependence of the E_{TKE} upon E_n seems to agree with semi-empirical models although the absolute value does not, and (c) understanding the variance and its energy dependence for the E_{TKE} distribution remains a challenge.

This work was supported in part by the Director, Office of Energy Research, Division of Nuclear Physics of the Office of High Energy and Nuclear Physics of the US Department of Energy under Grant DE-FG06-97ER41026. W.L. thanks the Department of Energy's Institute for Nuclear Theory at the University of Washington for its hospitality and the Department of Energy for partial support during the completion of this work. This work has benefited from the use of the Los Alamos Neutron Science Center at the Los Alamos National Laboratory. This facility is funded by the US Department of Energy under DOE Contract No. DE-AC52-06NA25396.

[1] V. E. Viola, K. Kwiatkowski, and M. Walker, Phys. Rev. C **31**, 1550 (1985).
[2] M. Rajagopalan and T. D. Thomas, Phys. Rev. C **5**, 2064 (1972).
[3] R. Vandenbosch and J. R. Huizenga, *Nuclear Fission* (Academic, New York, 1973).
[4] D. G. Madlund, Nucl. Phys. A **772**, 113 (2006).
[5] Ch. Straede, C. Budtz-Jorgensen, and H. H. Knitter, Nucl. Phys. A **462**, 85 (1987).
[6] J. W. Meadows and C. Budtz-Jorgensen, ANL/NDM-64, 1982 (unpublished).
[7] R. Müller, A. A. Naqvi, F. Käppeler, and F. Dickmann, Phys. Rev. C **29**, 885 (1984).
[8] The GEF model is described by K.-H. Schmidt, B. Jurado, and C. Amouroux, http://hal.in2p3.fr/in2p3-00976648 while the GEF code is available at http://www.khs-erzhausen.de/GEF.html.
[9] P. W. Lisowski, C. D. Bowman, G. J. Russell, and S. A. Wender, Nucl. Sci. Eng. **106**, 208 (1990).
[10] P. W. Lisowski and K. F. Schoenberg, Nucl. Instrum. Methods Phys. Res., Sect. A **562**, 910 (2006).
[11] http://wnr.lanl.gov/newwnr/About/Beam.shtml.
[12] S. A. Wender *et al.*, Nucl. Instrum. Methods Phys. Res., Sect. A **336**, 226 (1993).
[13] H. W. Schmitt, W. E. Kiker, and C. W. Williams, Phys. Rev. **137**, B837 (1965).
[14] S. I. Mulgin, V. N. Okolovich, and S. V. Zhudanov, Nucl. Instrum. Methods Phys. Res., Sect. A **388**, 254 (1997).
[15] L. C. Northcliffe and R. F. Schilling, At. Data Nucl. Data Tables **7**, 233(1970).
[16] J. F. Ziegler, M. D. Ziegler, J. P. Biersack, Nucl. Instrum. Methods Phys. Res., Sect. B **268**, 1818 (2010).
[17] G. N. Knyazheva *et al.*, Nucl. Instr. Meth. Phys. Res. B **248**, 7 (2006).
[18] S. R. Gunn, H. G. Hicks, H. B. Levy, and P. C. Stevenson, Phys. Rev. **107**, 1642 (1957).
[19] S. S. Kapoor, D. M. Nadkarni, R. Ramanna, and P. N. Rama Rao, Phys. Rev. **137**, B511 (1965).
[20] P. C. Stevenson, H. G. Hicks, J. C. Armstrong, Jr., and S. R. Gunn, Phys. Rev. **117**, 186 (1960).

[21] A. A. Naqvi, F. Käppeler, F. Dickmann, and R. Müller, Phys. Rev. C **34**, 218 (1986).
[22] A. Al-Adili, F.-J. Hambsch, S. Pomp, and S. Oberstedt, Phys. Rev. C **86**, 054601 (2012).
[23] A. Tudora, G. Vladuca, and B. Morillon, Nucl. Phys. A **740**, 33 (2004).
[24] U. Brosa, S. Grossman, and A. Muller, Phys. Rep. **197**, 167 (1990).
[25] P. P. Dyachenko, B. D. Kuzminov, and M. Z. Tarasko, Sov. J. Nucl. Phys. **8**, 165 (1969).
[26] S. Grossmann, U. Brosa, and A. Muller, Nucl. Phys. A **481**, 340 (1988).