



LAWRENCE  
LIVERMORE  
NATIONAL  
LABORATORY

# Calculation of $^{239}\text{Pu}$ fission observables in an event-by-event simulation

R. Vogt, J. Randrup, J. Pruet, W. Younes

April 1, 2010

Nuclear Data 2010  
Jeju, South Korea  
April 26, 2010 through April 30, 2010

## **Disclaimer**

---

This document was prepared as an account of work sponsored by an agency of the United States government. Neither the United States government nor Lawrence Livermore National Security, LLC, nor any of their employees makes any warranty, expressed or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States government or Lawrence Livermore National Security, LLC. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States government or Lawrence Livermore National Security, LLC, and shall not be used for advertising or product endorsement purposes.

# CALCULATION OF $^{239}\text{Pu}$ FISSION OBSERVABLES IN AN EVENT-BY-EVENT SIMULATION

R. VOGT<sup>\*1,2</sup>, J. RANDRUP<sup>3</sup>, J. PRUET<sup>1</sup>, and W. YOUNES<sup>1</sup>

<sup>1</sup>Lawrence Livermore National Laboratory, Livermore, CA 94550, USA

<sup>2</sup>Physics Department, University of California at Davis, Davis, CA 95616, USA

<sup>3</sup>Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA

\*Corresponding author. E-mail : vogt2@llnl.gov

*Received*

*Accepted for Publication*

---

The increased interest in more exclusive fission observables has demanded more detailed models. We describe a new computational model, FREYA, that aims to meet this need by producing large samples of complete fission events from which any observable of interest can then be extracted consistently, including any interesting correlations. The various model assumptions are described and the potential utility of the model is illustrated. As a concrete example, we use formal statistical methods, experimental data on neutron production in neutron-induced fission of  $^{239}\text{Pu}$ , along with FREYA, to develop quantitative insights into the relation between reaction observables and detailed microscopic aspects of fission. Current measurements of the mean number of prompt neutrons emitted in fission taken together with less accurate current measurements for the prompt post-fission neutron energy spectrum, up to the threshold for multi-chance fission, place remarkably fine constraints on microscopic theories.

---

**KEYWORDS** : Fission Models, Covariances

## 1. INTRODUCTION

Nuclear fission remains important to society at large due to its many practical applications, including energy production and security. For example, reactors and other critical systems demand that neutron growth be known to about the 0.1% level for model simulations to be reliable. In such cases, scattering experiments are insufficiently accurate, requiring reliance on more inclusive, higher statistics integral critical assembly experiments.

Recently, efforts have been made to develop methods for detecting concealed nuclear material. These applications place new demands on fission models by attempting to exploit specific information carried by particles resulting from fission. Thus there is a need for a fission model that can describe for particle correlations on an event-by-event level. Such a description, employing a model incorporating the relevant physics with a few key parameters, compared to the pertinent data through a statistical analysis, presents a potentially powerful tool for bridging the gap between current microscopic models and important fission observables and for improving estimates of the relatively gross fission characteristics important for applications. This approach uses readily measured observables to constrain our understanding of the microscopic details of fission.

Relatively recently, Lemaire *et al.* [1] implemented a Monte Carlo simulation of statistical decays of fission fragments by sequential neutron emission for spontaneous

fission of  $^{252}\text{Cf}$  and thermal fission of  $^{235}\text{U}$ . That work demonstrated how such simulations, in conjunction with experimental data and models of the relevant physics, can be used to predict the neutron spectrum and to validate and improve the underlying physics models.

We have implemented a similar Monte Carlo-based approach and applied it to calculate sequential neutron emission from the  $^{240}\text{Pu}$  compound nucleus created from the reaction  $n + ^{239}\text{Pu}$ . The  $^{239}\text{Pu}$  fission cross section changes significantly for  $0.5 \leq E_n \leq 5.5$  MeV, making applications very sensitive to incident neutrons in the few MeV region.

We have adapted the recently developed fission event generator FREYA [2] to calculate the production and decay of fission fragments. We use maximum-likelihood analysis to estimate properties of the emitted fission neutrons and their correlation coefficients. The detailed statistical analysis presented here is essential for developing a more quantitative understanding of fission and thus obtaining better evaluations of fission data for various applications.

FREYA follows each fission event from the birth of the excited fragments through their decay via prompt neutron emission until the fragment excitation energy is below the neutron separation energy. It also includes the subsequent gamma emission from each fragment, albeit in a rather preliminary way.

## 2. MODEL DESCRIPTION

We assume binary fission of the compound nucleus, e.g.  $^{240}\text{Pu}$ , with mass  $A_c$  and charge  $Z_c$  formed by incident neutrons of energy  $E_n$  on an actinide with mass  $A_c - 1$ , e.g.  $^{239}\text{Pu}$ . The identities of the hot, excited fission fragments are obtained by sampling the mass and charge of the light,  $L$ , and heavy,  $H$ , fragments from fission fragment distributions, ensuring mass and charge. The fission  $Q$  value is determined from the mass and charge of the fission fragments and subsequently divided into the total fragment kinetic and excitation energies. We make an initial estimate of the total fragment kinetic energy, TKE, by sampling the kinetic energy due to mutual Coulomb repulsion,

$$\text{TKE} = \frac{e^2 Z_L Z_H}{R_L + R_H + s(E_n) d_{LH}(A_H, E_{\text{thermal}})} \quad (1)$$

Here  $Z_i$  and  $R_i$  are the charges and radii of the fragments,  $d_{LH}$  is the tip separation distance between the fragments at scission, extracted from measurements of the TKE as a function of the heavy fragment mass,  $A_H$ , obtained from experiments with thermal neutrons,  $E_{\text{thermal}}$ , and  $s(E_n)$  is an energy-dependent scale factor. The total fragment excitation energy is found using energy conservation,  $\text{TEE} = Q - \text{TKE}$ . This TEE is divided between the light and heavy fragments and translated into a fragment temperature assuming  $E_i^* = a_i T_{LH}^2$  where  $a_i = A_i i / e_0$  and  $e_0$  is an asymptotic level density parameter. Allowing for temperature fluctuations in small systems, we modify the excitation energies by their thermal fluctuations. We adjust the TKE accordingly to retain total energy conservation.

Neutrons are then evaporated from the excited fragments until the excitation energy is too low for further neutron emission. Prompt gamma emission follows after prompt neutron emission ceases. So far, the fission neutron spectrum has been evaluated up to the threshold for neutron emission before fission. Multi-chance fission for values of  $E_n$  greater than a few MeV is in the process of being implemented.

There are significant uncertainties in our overall understanding of fission. To reduce these uncertainties, we need to look at the big picture, not just spectra since other physics processes feed into these spectra. Both the average neutron multiplicity,  $\bar{\nu}$ , and the spectra,  $d\bar{\nu}/dE$ , depend on the physics of the fission process. The multiplicity and the spectra are intimately linked and cannot really be treated separately in a realistic fission model. Thus improvements in the spectral evaluation will come with improved modeling of fission.

Spectral data for thermal neutrons are inconsistent with each other and have large uncertainties in important regions, see Fig. 1, much larger than the constraints on  $\bar{\nu}$  itself. The published spectral data do not extend into the low energy region of less than 100 keV. These data are also not generally available for incident neutrons of more than a few MeV, see Fig. 2, making extrapolation necessary. Measurements of other quantities that could guide model calculations, such as total fragment kinetic energy and neutron multiplicity as

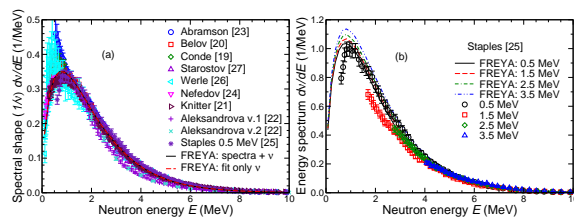
a function of fragment mass, only exist for thermal incident energies,  $E_{\text{thermal}}$ . Modeling complete fission events with FREYA helps fill the gaps in data.

To obtain the best possible agreement between the neutron multiplicity and the neutron spectra, three FREYA parameters have been tuned in two possible scenarios: either to both the spectral data and  $\bar{\nu}$  or to the more accurate measurements of  $\bar{\nu}$  alone [3].

1. The factor  $s(E_n)$  which scales the average fragment tip separation distance,  $d_{LH}$ , obtained from the experimental TKE( $A_H$ );
2. The asymptotic level density parameter,  $e_0$ , which sets the fragment ‘temperature’ for neutron evaporation;
3. The relative excitation of the light and heavy fragments,  $x$ , with  $E_L^* = x a_L \text{TEE} / (a_L + a_H)$ ,  $E_H^* = \text{TEE} - E_L^*$ . Here  $x = 1$  is the equal temperature situation, resulting in the same number of neutrons emitted from both fragments;  $x > 1$  means more neutrons are evaporated from the light fragment than the heavy fragment.

## 3. RESULTS

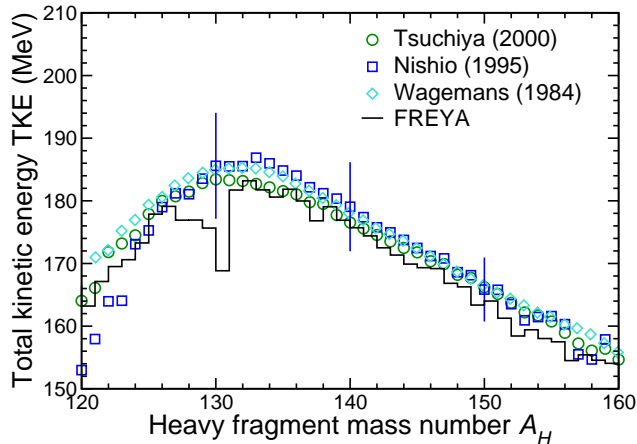
Figure 1 shows the spectral data we have used in our comparison. The uncertainties are not given for all the data shown. In these cases we have assigned a 5% uncertainty to the data, comparable to the typical uncertainties associated with other spectra data. Note that in Fig. 1(a), the discrepancies between the data sets for  $E < 1$  MeV can be rather large. These data were taken at  $E_n < 0.5$  MeV and are not absolutely normalized. To compare the data sets and our results most straightforwardly, we normalize all data sets, as well as our calculations, to unity using a Watt spectrum.



**Fig. 1.** The measured prompt neutron spectra are compared to our fit results. The comparison to the low energy results from Refs. [6, 5, 8, 10, 11, 4, 9, 7, 12] (a) are of the normalized spectral shapes while the results at higher incident neutron energies from Ref. [12] (b) are compared to the spectral distributions.

Figure 1(b) shows data taken over a range of incident neutron energies,  $0.5 \leq E_n \leq 3.5$  MeV. In this case, the data and the calculations are not normalized to unity but to  $\bar{\nu}$  since the multiplicity increases with energy to be able to distinguish between the curves.

Two measurements that provide more detailed information with which to tune models to the underlying physics of fission are shown in Figs. 2 and 3, the total kinetic



**Fig. 2.** The measured average TKE as a function of the mass number of the heavy fragment [15, 14, 13] compared to FREYA.

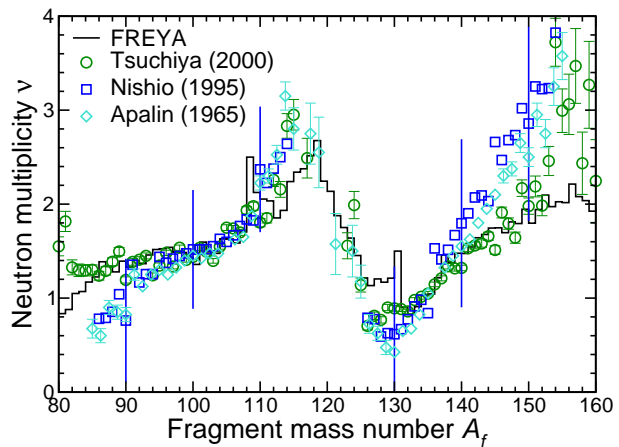
energy as a function of heavy fragment mass,  $\text{TKE}(A_H)$ , and the neutron multiplicity,  $\nu$ , as function of fragment mass, respectively. Both are shown for thermal neutrons incident on  $^{239}\text{Pu}$  and compared to FREYA calculations.

There are no uncertainties shown on the TKE in Fig. 2. The vertical bars indicate the full-width at half-maximum of the TKE distribution at several values of  $A_H$ . Near symmetric fission, the fission fragments are mid-shell nuclei subject to strong deformation, resulting in a large tip separation distance and a low TKE. At  $A_H = 132$ , the heavy fragment is close to a doubly-magic closed shell ( $Z_H = 50$ ,  $N_H = 82$ ) and is resistant to distortions, remaining more spherical, while the light fragment is significantly deformed. This combination results in a smaller tip separation and thus a larger TKE. The model calculations reproduce this behavior rather well.

The multiplicity of the neutrons emitted by the hot fragments as they de-excite has a characteristic sawtooth shape for incident thermal neutrons, see Fig. 3. This follows from the TKE observed in Fig. 2. The peak in  $\nu(A)$  near symmetry is due to the larger excitation energy corresponding to the lower TKE. The drop in  $\nu(A)$  around  $A = 130$  can be attributed to the lower excitation energy of the closed shell nucleus. The sawtooth character of the multiplicity distribution is likely to be reduced as the incident energy increases since the excitation energy increases the overall multiplicity and binary fission produces more mass-symmetric fragments. Unfortunately, no data are available at higher than thermal energies to study the energy dependence of these observables in detail.

#### 4. COVARIANCES

In addition to single particle observables, it is interesting to consider correlations in the spectral strengths at different



**Fig. 3.** The average neutron multiplicity as a function of the fragment mass from Refs. [14, 13, 16] compared to FREYA.

energies. The diagonal elements of the covariance matrix are the squares of the standard deviations in the model calculations. The off-diagonal matrix elements give the covariances between two outgoing energies. The covariance matrix between spectral strengths at different outgoing neutron energies is defined as

$$\tilde{\sigma}(E_k, E_{k'}) = \langle (E_k - \tilde{E}_k)(E_{k'} - \tilde{E}_{k'}) \rangle. \quad (2)$$

For continuous observables, such as spectra, the covariance matrix is singular along the diagonal,

$$\tilde{\sigma}(E_k, E_{k'}) = \tilde{\sigma}_{E_k}^2 \delta(E_k - E_{k'}) + \tilde{\sigma}_{E_k E_{k'}} , \quad (3)$$

where  $\tilde{\sigma}_{E_k}^2$  is the variance in the differential yield at the specified energy  $E_k$ , while  $\tilde{\sigma}_{E_k E_{k'}}$  expresses the correlation between the differential yields at two *different* energies  $E_k$  and  $E_{k'}$ . After the singular part has been removed, the matrix of correlation coefficients is

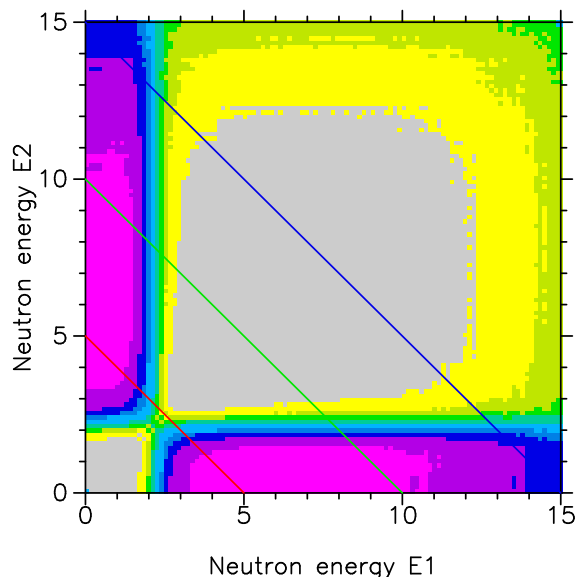
$$C(E_k, E_{k'}) = \tilde{\sigma}_{E_k E_{k'}} / [\tilde{\sigma}_{E_k} \tilde{\sigma}_{E_{k'}}] , \quad (4)$$

is regular. The result for  $E_n = 0.5$  MeV when fitting to  $\bar{\nu}$  alone is shown in Fig. 4. The cross-diagonal lines are lines of constant total energy,  $E_T = E_k + E_{k'}$ . The values of the correlation matrix along these lines are shown in Fig. 5.

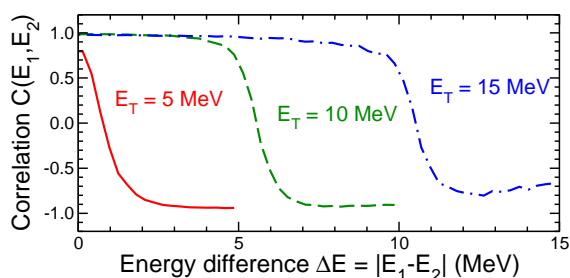
When the model parameters are varied, the spectral shapes tend to pivot around  $E \sim 2$  MeV. Thus, when both neutron energies lie on the same side of this value, the differential changes are in phase and the correlation coefficient is close to unity. The changes are in opposite directions when the two energy values are on opposite sides of the pivot energy and  $C(E_k, E_{k'})$  is close to  $-1$ . The results for constant  $E_T$  in Fig. 5 show this behavior more clearly.

#### 5. SUMMARY

We have shown that the evaluated neutron spectra are strongly influenced by the spectral data,  $\bar{\nu}$ , with its much



**Fig. 4.** The correlation coefficients,  $C(E_1, E_2)$ , for the spectral strength of the evaporated neutrons as obtained from the statistical analysis at  $E_n = 0.5$  MeV fitting to  $\bar{\nu}$  only. Figure 5 shows cuts along the three indicated lines of constant total energy.



**Fig. 5.** The spectral correlation coefficients,  $C(E_1, E_2)$ , along the three lines of constant combined energy indicated in Fig. 4.

smaller associated uncertainty is more important. Improvements in modeling will come from better knowledge of the complicated fission process through microscopic models and high statistics, less inclusive data.

FREYA bridges models and data by addressing complete events with full energy-momentum conservation and correlations between observables. The next step is to include multi-chance fission in FREYA to address higher incident neutron energies.

## ACKNOWLEDGMENTS

This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344 (RV, JP and WY), by Lawrence Berkeley National Laboratory under Contract DE-AC02-05CH11231 (JR) and was

also supported in part by the National Science Foundation Grant NSF PHY-0555660 (RV).

## REFERENCES

- [1] S. Lemaire, P. Talou, T. Kawano, M.B. Chadwick, and D.G. Madland, “Monte Carlo approach to sequential neutron emission from fission fragments”, *Phys. Rev. C* **72**, 024601 (2005).
- [2] J. Randrup and R. Vogt, “Calculation of fission observables through event-by-event simulation”, *Phys. Rev. C* **80**, 024601 (2009).
- [3] R. Vogt, J. Randrup, J. Pruet and W. Younes, “Event-by-event study of prompt neutrons from  $^{239}\text{Pu}(n, f)$ ”, *Phys. Rev. C* **80**, 044611 (2009).
- [4] D. Abramson and C. Lavelaine, “Fission neutron spectra of  $^{235}\text{U}$  and  $^{239}\text{Pu}$ ”, A.E.R.E. Harwell Rep. No. 8636 (1977).
- [5] L.M. Belov, M.V. Blinov, N.M. Kazarinov, A.S. Krivokhatskij, and A.N. Protopopov, “Spectra of fission neutrons of  $^{244}\text{Cm}$ ,  $^{242}\text{Pu}$  and  $^{239}\text{Pu}$ ”, *Yad.-Fiz. Issl. Rep.* **6**, 94 (1968).
- [6] H. Conde and G. During, “Fission neutron spectra, part II, fission neutron spectra of  $^{235}\text{U}$ ,  $^{239}\text{Pu}$  and  $^{252}\text{Cf}$ ”, *Arkiv för Fysik* **29**, 313 (1965).
- [7] B.I. Starostov, V.N. Nefedov, and A.A. Bojcov, “High precision prompt neutron spectra measurement for neutrons from  $^{252}\text{Cf}$ ,  $^{233}\text{U}$ ,  $^{235}\text{U}$  and  $^{239}\text{Pu}$  fission in the energy range 2-11 MeV”, *6<sup>th</sup> All-Union Conf. on Neutron Physics*, Kiev, Ukraine (1983) Vol. 2, p. 290
- [8] H. Werle and H. Bluhm, “Fission neutron spectra measurements of  $^{235}\text{U}$ ,  $^{239}\text{Pu}$  and  $^{252}\text{Cf}$ ”, *J. Nucl. Energy* **26**, 165 (1972).
- [9] V.N. Nefedov, B.I. Starostov, and A.A. Bojcov, “High precision spectra measurements for neutrons arising from the fission of  $^{252}\text{Cf}$ ,  $^{233}\text{U}$ ,  $^{235}\text{U}$  and  $^{239}\text{Pu}$  in the energy range 0.04-5 MeV”, *6<sup>th</sup> All-Union Conf. on Neutron Physics*, Kiev, Ukraine (1983) Vol. 2, p. 285.
- [10] H. Knitter, “Measurement of the energy spectrum of prompt neutrons from the fission of  $^{239}\text{Pu}$  by 0.215 MeV neutrons”, *Atomkernenerg.* **26**, 76 (1975).
- [11] Z.A. Aleksandrova, V.I. Bol Shov, V.F. Kuznetsov, G.N. Smirenkin, and M.Z. Tarasko, “Spectra of secondary neutrons at fission of  $^{235}\text{U}$  and  $^{239}\text{Pu}$  by neutrons with 0.1 MeV energy”, *Atom. Energ.* **38**, 108 (1975).
- [12] P. Staples, J.J. Egan, G.H.R. Kegel, A. Mittler, and M.L. Woodring, “Prompt fission neutron energy spectra induced by fast neutrons”, *Nucl. Phys. A* **591**, 41 (1995).
- [13] C. Tsuchiya, Y. Nakagome, H. Yamana, H. Moriyama, K. Nishio, I. Kanno, K. Shin, and I. Kimura, “Simultaneous measurement of prompt neutrons and fission fragments for  $^{239}\text{Pu}(n_{\text{th}}, f)$ ”, *J. Nucl. Sci. Technol.* **37**, 941 (2000).
- [14] K. Nishio, Y. Nakagome, I. Kanno, and I. Kimura, “Measurement of fragment mass dependent kinetic energy and neutron multiplicity for thermal neutron induced fission of  $^{239}\text{Pu}$ ”, *J. Nucl. Sci. Technol.* **32**, 404 (1995).
- [15] C. Wagemans, E. Allaert, A. Deruytter, R. Barthélémy, and P. Schillebeeckx, “Comparison of the energy and mass characteristics of the  $^{239}\text{Pu}(n_{\text{th}}, f)$  and the  $^{240}\text{Pu}(sf)$  fragments”, *Phys. Rev. C* **30**, 218 (1984).
- [16] V.F. Apalin, Yu.N. Gritsyuk, I.E. Kutikov, V.I. Lebedev, and L.A. Mikaelian, “Neutron emission from  $^{233}\text{U}$ ,  $^{235}\text{U}$  and  $^{239}\text{Pu}$  fission fragments”, *Nucl. Phys. A* **71**, 553 (1965).