Parameter Optimization and Uncertainty Analysis of FREYA for Spontaneous Fission

J. Van Dyke¹, L. A. Bernstein²,³, R. Vogt⁴,⁵

¹Physics Department, University of California, Berkeley, CA 94720
²Nuclear Science Division, Lawrence Berkeley National Laboratory, Berkeley, CA 94720
³Nuclear Engineering Department, University of California, Berkeley, CA 95616
⁴Nuclear and Chemical Sciences Division, Lawrence Livermore National Laboratory, Livermore CA 94550
⁵Physics Department, University of California, Davis, CA 95616

(Dated: September 18, 2018)

Background: The complete event fission simulation code FREYA is used to study correlations in fission. To make the best possible simulations, FREYA one must find the optimized values of the five physics-based parameters which characterize the events produced by FREYA. So far this has been done only empirically or by brute force computational techniques. Purpose: We seek to develop a method of parameter optimization for FREYA that would include uncertainty quantification and an analysis of the correlations between the parameters. Methods: We focus on spontaneous fission as a simpler test case of the method. We first check the results of previous optimizations for 252Cf(sf) and then go on to develop a simulated annealing approach to optimize the parameters. Although 252Cf(sf) has the most measured observables, we are able to apply the technique to all spontaneously fissioning isotopes in the current version of FREYA. Results: We find optimal values of the five FREYA parameters, with uncertainties, for all isotopes studied. Our results are compared with available data. The parameter values are physically reasonable and in accord with intuition, even in cases where the data for optimization are sparse to non-existent. Conclusions: The simulated annealing method provides a way to determine the best parameters for the phenomenologically successful fission model FREYA. We have developed an algorithm that can be updated to include additional isotopes or more data sets for the current isotopes as they become available.

I. INTRODUCTION

Though nuclear fission has influenced society in significant ways, the fission process itself is still not understood in great detail. Nevertheless, we can produce a complete, fully-correlated, physically-consistent description of fission. The Fission Reaction Event Yield Algorithm (FREYA) fission model is designed to serve this purpose in a physically-complete fashion with a relatively modest computational footprint. While our main focus here is on 252Cf(sf), we also present optimized parameters for all spontaneously fissioning nuclei in the event-by-event simulation code FREYA. FREYA generates samples of complete fission events, including the full kinematic information for the two product nuclei, as well as the emitted neutrons and photons. It was designed to quickly generate large numbers of events. FREYA is one of the most time-efficient and effective methodologies. We make a full statistical analysis, including variances and covariances. In this paper we find the best possible set of the input parameters for describing all the spontaneous fission data for each isotope.

In Sec. II, we describe the parameters we optimize in FREYA. Sec. III discusses the numerical methods used to perform the optimization, while Sec. IV identifies the data employed in the fits. Sections V and VI provide the results and their interpretation, as well as a comparison between the resulting parameter values to those previously used. We compare results to the data for specific 252Cf(sf) observables in Sec. VII. Comparisons to other isotopes can be found in the supplemental material.

II. FREYA PARAMETER DESCRIPTION

In this section we briefly discuss the process of nuclear fission as implemented in FREYA. We also identify and provide a physical interpretation of the five parameters required by FREYA.

The fission process begins when a specified initial compound nucleus splits into two fragment nuclei, typically one light and one heavy, which we denote by L and H respectively for each fragment pair. The corresponding Q-value is given by \( Q = M_0c^2 - M_Lc^2 - M_Hc^2 \) for spontaneous fission. The fragment yields as a function of fragment mass and the total kinetic energy of the fragments, TKE, as a function of heavy fragment mass, \( A_H \), are sampled from data. From the fission Q-value and the sampled TKE we determine the total excitation energy at scission, \( E_{\text{sc}}^* \), by energy conservation. The excitation energy \( E_{\text{sc}}^* \) is available for both statistical, \( E_{\text{stat}} \), and rotational, \( E_{\text{rot}} \), excitation of the fragments. These two
quantities are related by:

\[ \dot{E}^*_\text{sc} = Q - \dot{T}\text{KE} = \dot{E}_\text{stat} + \dot{E}_{\text{rot}}. \]  

(1)

The level density parameter\(^1\) \(a \approx A_0/e_0\) [3], for some constant \(e_0\), determines a “scission temperature” \(T_{\text{sc}}\) from the relation:

\[ \dot{E}^*_\text{sc} = a T_{\text{sc}}^2. \]  

(2)

This \(e_0\) is the first parameter required by FREYA, and is usually around 10/MeV [6]. Note that, while Eq. (2) relates \(a\) to the scission temperature, the level density parameter \(a\) is also employed for all neutron emission during the fission process.

In addition to the mean angular momenta of the fragment given by the overall rigid rotation around the scission axis, there are also fluctuations around this value attributed to the wriggling and bending modes [9] that contribute to \(\dot{E}_{\text{rot}}\). The relative degree of these fluctuations is given by

\[ \dot{E}_S = c S T_{\text{sc}}. \]  

(3)

The ratio of the fluctuation temperature \(\dot{E}_S\) to the scission temperature \(T_{\text{sc}}\), \(c_S\), is our second parameter. It is clear that this must be non-zero. If it were zero, there would be no fluctuations and the only angular momentum present in the fragments would be that dictated by the rigid rotation before scission. In the case of spontaneous fission, this would mean that the fragments have no angular momentum, which is not the case. The default value used in the most recently published version of FREYA is \(c_S = 0.87\) [6,7]. See 9 for more details on the addition of angular momentum to FREYA.

The statistical excitation energy, \(\dot{E}_{\text{stat}}\), is initially partitioned as \(\dot{E}_{\text{stat}} = \dot{E}^*_L + \dot{E}^*_H\), where the \(\ast\) indicates that the statistical excitation is initially partitioned according to the level density parameters. This would only be completely accurate if the fragments were in mutual thermal equilibrium. However, since we know that the light fragment emits more neutrons on average, we modify the partition via the third parameter, \(x\):

\[ \dot{E}^*_L = x \dot{E}^*_L, \quad \dot{E}^*_H = \dot{E}_{\text{stat}} - \dot{E}^*_L. \]  

(4)

assumed to be greater than 1. A value around 1.1 – 1.3 is typically found [7,8].

As noted in Eq. (2), the average fragment excitation energy is related to the temperature by \(\dot{E}^*_i \propto T_i^2\). The associated variance of this excitation is given by

\[ \sigma^2 \dot{E}_i = e \dot{E}^*_i T_i. \]  

(5)

\[ \begin{array}{|c|c|c|c|c|}
\hline \text{\(e_0/\text{MeV}\)} & x & c & c_S & d\text{TKKE MeV} \\
\hline 7 - 12 & 1.0 - 1.5 & 1 - 3 & 0.5 - 1.5 & -5 - 5 \\
\hline
\end{array} \]

Table I: Ranges of parameters considered in the optimization.

There is thus an energy fluctuation \(\delta E^*_i\) on each of the two fragments which is sampled from a normal distribution of variance \(2 \epsilon_i \dot{E}^*_i T_i\). In particular, the excitation energies of each fragment are adjusted as \(E^*_i = \dot{E}^*_i + \delta E^*_i\). The factor \(c\), our fourth parameter, controls the truncation of the normal distribution at the maximum available excitation. It primarily affects the neutron multiplicity distribution and was assumed to take a value \(c \sim 1\). We maintain energy conservation by

\[ \text{TKKE} = \text{TKKE} - \delta E^*_L - \delta E^*_H. \]  

(6)

Finally, to ensure reproduction of the measured average neutron multiplicity \(\bar{\nu}\), we allow the value of the average total kinetic energy to shift by a small amount \(d\text{TKKE}\). The measured data have often unquantified systematic uncertainties or, in some cases, low statistics.

The ranges considered for these parameters can be found in Table I. While the range for \(c\) is listed as 1 – 3, for some isotopes we allow this range to expand. Since this parameter controls the width of the neutron multiplicity distribution, for isotopes which are known to have a comparatively narrow distribution, we allow the parameter to vary below 1 to 0.8. In addition, for isotopes with a comparatively wide distribution relative to their average multiplicity we allow \(c\) to be as large as 4.

We note that there are two detector-based photon-related parameters in FREYA: \(g_{\text{min}}\), the minimum detected photon energy, and \(t_{\text{max}}\), the length of the time measurement. Because these are unique to each measurement, they are not counted as tunable parameters. They do however have some effect on the photon multiplicity and energy per photon [11]. The fits use the values of \(g_{\text{min}}\) and \(t_{\text{max}}\) appropriate for the data included in the fits.

III. COMPUTATIONAL METHODS

For each set of five parameters, we generate sets of 1,000,000 events. The output from the generated events contains the full kinematic information for the fragments and the emitted neutrons and photons. We use this kinematic information to calculate physical observable which are then compared to measured data. In this study, the quantities we extracted included the average neutron multiplicity, \(\bar{\nu}\); the second and third moments of the neutron multiplicity, \(\nu_2\) and \(\nu_3\) respectively; the neutron multiplicity distribution, \(P(\nu)\); the average neutron multiplicity as a function of the total kinetic energy, \(\nu(\text{TKKE})\), and as a function of fragment mass, \(\nu(A)\); the neutron energy spectrum, \(N(E)\); the average photon multiplicity, \(\bar{N}_\gamma\); the photon multiplicity distribution, \(P(N_\gamma)\); and the average energy per photon \(\bar{E}_\gamma\).
The second and third moments of the neutron multiplicity are defined as

\[ \nu_2 = \langle \nu (\nu - 1) \rangle , \quad \nu_3 = \langle \nu (\nu - 1) (\nu - 2) \rangle . \]  

(7)

After calculating these observables from the FREYA output, they are compared with available experimental data and evaluations. Unfortunately, not all of these observables are available for many of the isotopes of interest, as we discuss later. More information on the sources and quality of these data can be found in Sec. [IV].

The FREYA output is compared to the data, and for each observable the reduced \( \chi^2 \) uncertainty, \( \chi_0^2 \) is calculated as

\[ \chi_0^2 = \frac{1}{n-5} \sum_{i=1}^{n} \frac{(O_i - E_i)^2}{\sigma_i^2} \]  

(8)

where \( i = 1, \ldots, n \) runs over the bins of the distribution; \( O_i \) is the value of the observable returned by FREYA for the given bin; \( E_i \) is the experimental result; and \( \sigma_i \) is the experimental uncertainty on \( E_i \). The reduced \( \chi^2 \) for the observable is found by dividing the sum over all bins by the number of degrees of freedom, \( n - 5 \), for the five physics-based parameters we are fitting. For single valued observables such as \( \tau \), we simply take \( \chi_0^2 = (O - E)^2 / \sigma^2 \).

The total \( \chi^2 \) is the sum over all observables where data are available,

\[ \chi^2 = \sum_{O} \chi_0^2 . \]  

(9)

This total \( \chi^2 \) is treated as the return value of an objective function. In Fig. 1 we plot the following: first we take the sum of the reduced \( \chi^2 \) values for a linear combinations of the parameters. Then we find the lowest such value, and plot the ratio of all of the values to this value. These plots show us merely one particular two dimensional projection of the five-dimensional parameter space. The particular linear combination of parameters was chosen by-eye to best illustrate the nature of the parameter space we are working in.

Preliminary work in Ref. [7] used a grid-search method where every potential combination of parameters was tested. However, in a five-dimensional space with a reasonably fine mesh, the grid search technique is unwieldy and very computationally intensive. We have therefore also used alternative methods and confirmed that our alternate methodology agreed with the earlier used grid search approach used earlier.

Fig. 1 shows that the objective function displays many local minima which are neither global minima, nor physically relevant. Therefore, we cannot employ a simple algorithm, such as gradient descent, because it can easily fall into such local minima. We have instead employed the so-called simulated annealing method [12]. The motivation for such an algorithm is to inject a certain amount of randomness into the process to allow for the procedure to occasionally jump in a seemingly “worse” direction in order to move out of a potential local minimum and eventually find the global solution. We provide a rough description of the algorithm now.

The simulated annealing algorithm first generates a random solution, calculates its cost using an objective function, generates a random neighboring solution, calculates the cost of this new solution with the same objective function, and then compares these costs using an acceptance probability function. The acceptance probability is calculated by comparing the difference of the two costs with the so-called temperature, \( T \). The parameter \( T \) is initially equal to unity, and is decreased to a new value, \( T' \), after each iteration of the algorithm by employing a scale factor \( \alpha \),

\[ T' = \alpha T . \]  

(10)

The factor \( \alpha \) is usually greater than 0.8, and is always less than 1. The temperature allows for the algorithm to become less stochastic as the number of iterations is.
increased. The value returned by the acceptance probability function is then compared to a randomly generated number to determine whether the new solution is accepted. As a result, when the algorithm compares the costs of these two solutions, there is a certain probability that, even if the new solution is worse, it still might be accepted. This helps prevent the algorithm from sinking into a local minimum. The process is repeated until an acceptable solution is found.

In our particular situation, the solutions consist of values of the 5 parameters and the objective function is the corresponding value of the $\chi^2$ uncertainty from Eq. (9). We define the acceptance probability function of two uncertainties, e.g. $\chi_0$ and $\chi_1$, for two different parameter sets as

$$\exp \left( \frac{\chi_0 - \chi_1}{\chi_0 T} \right)$$

where $T$ is the temperature defined in Eq. (10). Overall, this optimization procedure proved to be the most successful. Gradient descent was successful when the initial guess was guided according to physical intuition. However, simulated annealing was able to determine the global solution without this external help. We also investigated the robustness of this algorithm with respect to the factor $\alpha$ used to lower the temperature. The solution was found relatively reliably for all values of $\alpha$ between 0.85 and 1. Below $\alpha = 0.85$ the process was still largely successful, but not to the same degree. After finding the general range of the global solution, our simulated annealing algorithm then completes a grid search in a small region surrounding our potential solution to obtain the final minimum with high precision.

\section*{IV. DATA EMPLOYED IN THE OPTIMIZATION}

As noted in Sec. III, all of the optimization procedures rely on an objective function which computes how closely the FREYA output reproduces available experimental data. We now discuss in some detail the source and quality of the data for $^{252}$Cf(sf). Though the available data for $^{252}$Cf(sf) are quite extensive, we still were cautious in our selection to avoid fitting to out-of-date or low quality data. We fit the $^{252}$Cf(sf) parameters to all eight observables mentioned in the previous section.

We note that $^{252}$Cf(sf) is the only isotope we consider that has data available for all observables used in the optimization. Thus the parameters for $^{252}$Cf(sf) are the most constrained out of all the fits performed.

The observed neutron multiplicity distribution $P(\nu)$ is taken from Ref. [13], an evaluation of the prompt neutron multiplicity distributions for the spontaneous fission of a number of isotopes. This consensus resource combines all of the reliable direct sources of data available at this time. We also employ this evaluation for the average neutron multiplicity as well as the second and third moments of $P(\nu)$ and $P'(\nu)$ itself. We have chosen to use $\bar{\nu}$ in addition to the actual distribution because the parameters in FREYA are capable of shifting $\bar{\nu}$ explicitly, as well as changing the shape of $P(\nu)$, as described in Sec. II.

Note that for the optimization, we use the square root of the uncertainty given in the evaluation since when we used the reported uncertainty, it was so low it dominated the optimization.

We have used the data from Ref. [14] for the neutron multiplicity as a function of fragment mass, $\nu(A)$. See Ref. [14] for more experimental specifics. While Ref. [14] also measures $\nu(A)$, these data are not used in the optimization procedure. We do however compare FREYA to this result in Sec. VII. These two data sets are very similar, so the decision between them was largely inconsequential in terms of the fit.

We take the neutron multiplicity as a function of TKE from Ref. [16]. Since these data are from 1988, this set is rather dated. However, this work includes a thorough and honest statistical analysis of the results which yields reliable uncertainties. While Ref. [15] includes a more recent measurement of $\nu(TKE)$, it agrees within uncertainties with Ref. [16] except in regions where the TKE is very low or very high and is thus of low statistical significance. We compare the FREYA results to both data sets in Sec. VII.

The prompt fission neutron energy spectrum is taken from the Mannhart evaluation [17]. While also somewhat dated, it is a well-established evaluation. We have used the non-smoothed data, which are presented as the ratio to a Maxwellian distribution of temperature $T = 1.32$ MeV. We have multiplied the evaluation by the Maxwellian at the center of each energy bin to obtain the prompt fission neutron spectrum directly. There is also a smoothed version of this spectrum which we have not used here because the non-smoothed version provides an uncertainty while the smoothed spectrum does not. The disadvantage of using this version is the fact that there is a slight kink around 0.1 MeV in the Mannhart spectrum.

Finally, the photon multiplicity distribution, average photon multiplicity, and average photon energy are taken from Ref. [18], measured in 2012 using the DANCE array. See Ref. [18] for more details on this analysis.

We now briefly discuss the data used in fitting the other spontaneously fissioning isotopes in FREYA. Far fewer data are available for these. The optimizations for $^{240}$Pu(sf) and $^{242}$Pu(sf) were completed using the neutron multiplicity distribution, average neutron multiplicity, average photon multiplicity, and average photon energy. The neutron multiplicity distribution and its moments were taken from Ref. [13]. Indeed, evaluations from Ref. [13], available for all spontaneously fissioning isotopes included in FREYA so far, were at times the only data available. The average photon multiplicity and energy for $^{240}$Pu(sf) and $^{242}$Pu(sf) both come from Ref. [19]. These data, taken in 2016, are the most recent of all the data used in the optimization. There is also a
The neutron multiplicity distribution, average neutron multiplicity, and second and third moments of the distribution for $^{240}$Pu(sf) are also available from Ref. [13]. We also fit to the neutron multiplicity as a function of fragment mass, take from Ref. [21]. These data only have uncertainties for some values of $A$. These uncertainties are around 0.15, so we took this to be the default uncertainty for the values of $\nu(A)$ without one. We have done this because some value of uncertainty is required for the calculation of $\chi^2$. Finally we use the neutron spectrum from Ref. [22]. Reference [22] also has a neutron spectrum for $^{242}$Pu(sf) but we choose not to use it in the optimization due to quality issues.

The only available data for $^{238}$U(sf) and $^{238}$Pu(sf) are the neutron multiplicity distribution, the average neutron multiplicity, and the second and third moments of the distribution from Ref. [13].

### V. FIT RESULTS

We have confirmed the previous $^{252}$Cf(sf) fit results [7] within a reasonable margin, produced uncertainties, and calculated correlation matrices for the parameters. In Table II we list our optimized parameter values for $^{252}$Cf(sf). These results are consistent with the default values based on physical intuition given in Sec. [1].

The optimized values for $^{252}$Cf(sf) from the preliminary optimization in Ref. [7] are shown in Table II. There is some difference between our results and those of Ref. [7] because we employ some different data sets, as well as a slightly different optimization scheme, as described in Sec. [III]. While some preliminary work was also done for $^{240}$Pu(sf), we provide the first complete analysis for this isotope, as well as the other spontaneously fissioning isotopes in FREYA.

We calculate the probability as a function of the $\chi^2$ uncertainty, as well as the expectation values according to

$$P(\vec{y}) = \left(\frac{\chi^2(\vec{y})}{\nu}ight)^{\nu/2-1} e^{-\chi^2(\vec{y})},$$

$$\langle y_i \rangle = \int y_i P(\vec{y}) d^5\vec{y},$$

$$\langle y_i y_j \rangle = \int y_i y_j P(\vec{y}) d^5\vec{y},$$

where $\vec{y}$ denotes the 5-dimensional vector containing the 5 parameter values. We integrate over the parameter ranges. Here $n$ is the number of degrees of freedom for all observables. The variance and covariance of the parameters are defined as

$$\sigma^2_{y_i} = \langle y_i^2 \rangle - \langle y_i \rangle^2, \quad \sigma_{y_i y_j} = \langle y_i y_j \rangle - \langle y_i \rangle \langle y_j \rangle.$$

The correlation matrices in Table III are readily calculated as

$$\rho_{ij} = \frac{\sigma_{y_i y_j}}{\sigma_{y_i} \sigma_{y_j}}.$$

While Eqs. (12)-(16) provide analytic definitions of these quantities, we have numerically calculated the results in Table III using a Hessian. In particular, we construct a function representing the logarithm of the probability of $\vec{y}$ and then calculate the Hessian matrix at the optimal point using the parameter uncertainties from Table II. The negative of the inverse of this matrix is then the covariance matrix. We use Eq. (16) to extract the correlation coefficients displayed in the tables.

In Fig. I we present contour plots of the $\chi^2$ uncertainty for different values of the parameters. We vary the linear combinations listed on the axes and fix all parameters which are not listed to their central values. These plots can be interpreted as surfaces in the higher dimensional space which gives us a particular uncertainty for any choice of 5 parameters. As previously described, the particular linear combinations of parameters was based on physical intuition in order to best illustrate the nature of the parameter space we are working in. The linear combination of parameters on the axes in Fig. I was determined by eye according to the contour plots of the individual parameters. We do not show the variance as an error bar in the lower plot, because it effectively fills the entire displayed range.

As discussed in greater detail in Sec. III, employing a grid search will always find the proper solution by testing every possible combination, whereas the alternative
optimization methods attempt to “climb” around these contours in order to find the point of minimum uncertainty. These plots show that there is not always a clear “valley” of minimal uncertainty, and a simple grid approach is very likely to fall into a local minimum.

It is worth addressing the size of the $\chi^2$ uncertainty in our results. Our $\chi^2$ is summed over the uncertainty estimate for each bin of the data sets and evaluations we fit to. While this value is very large, this should not suggest that our fit is low quality, see Sec. III.

While our main focus is on $^{252}$Cf(sf), we also completed the same analysis for $^{238}$U(sf), $^{238}$Pu(sf), $^{240}$Pu(sf), $^{242}$Pu(sf), and $^{244}$Cm(sf), the other spontaneously fissioning isotopes currently included in FREYA 6. These results are listed in Table IV. A comparison of the fits for these isotopes to the data and evaluations used in the fits are available in the supplemental information.

While we have determined uncertainties on the parameter values for these isotopes, obtaining reliable correlation matrices for them is difficult. Table IV which also lists the number of data sets and evaluations used in our fits, makes this obvious. If only a single evaluation is available, as is the case for $^{238}$U(sf) and $^{238}$Pu(sf), it is difficult to say, without other constraints, how changing one parameter with respect to the others would affect the correlation.

VI. INTERPRETATION

In this section we develop a physical interpretation for the parameter values obtained in Sec. V. We have treated all spontaneously-fissioning isotopes in FREYA individually, with all five parameters allowed to vary independently regardless of how many data sets are available to constrain them. This is not unreasonable because we do not generally expect the parameters to have the same value for all fissioning systems.

The parameters $c$ and $d\text{TKE}$, which influence $P(\nu)$ and $\tau$, are perhaps best constrained because evaluations of $P(\nu)$ and the values of its moments are available for all isotopes in FREYA. Indeed, for some cases, these are the only available parameter constraints. Because the shape of $P(\nu)$ and its moments affect both $c$ and $d\text{TKE}$, and given that $\tau$, $\nu_2$ and $\nu_3$ vary considerably from isotope to isotope, we can expect $c$ and $d\text{TKE}$ to vary independently as well. We might expect the largest range of variation for these as well.

The other three parameters ($e_0$, $x$, and $c_S$) have fewer data available to constrain them. The average photon multiplicity and energy per photon can be used to guide the value of $c_S$ for $^{240}$Pu(sf), $^{242}$Pu(sf), and $^{252}$Cf(sf). We have $\nu(A)$ data to constrain $x$ for $^{244}$Cm(sf) and $^{252}$Cf(sf). Finally, we have used spectral data for $^{244}$Cm(sf) and $^{252}$Cf(sf) which provides a partial constraint on $c_0$. We remark that it is only partial because all parameters influence the prompt fission neutron spectrum. While $c_0$ is directly related to the temperature, see Eq. 2 and thus the slope of the prompt fission neutron spectrum, the parameters $c_S$, $x$, and $c$ are also related to the temperature, at least indirectly. Recall that $c$ sets the level of thermal fluctuations, Eq. 3; $x$ controls the sharing of excitation energy between fragments, initially related to the level density, Eq. 4; and $c_S$ is related to the scission temperature, Eq. 5. Thus these parameters all also influence the spectrum.

We expect $c_S$ to be less than unity while we expect $x$ and $c$ to be larger than unity. Since $e_0$ is related to the level density parameter, we expect a value of $8 - 12$ /MeV from other work 23. We may also expect $d\text{TKE}$ to vary considerably to make up for a lack of other constraints on the parameters aside from $\tau$ and also, because, on some cases the input data used for TKE($A_H$) have either large uncertainties based on low-statistical samples, or no uncertainties given. An examination of the results in Table IV can give us insight into how well the optimization procedure met our expectations.

The parameter values for the spontaneously fissioning isotopes in FREYA 2.0.2 6 were obtained in a far more empirical fashion. The values for $^{252}$Cf(sf) were taken from Ref. 7, obtained by a grid search procedure. Universal values were then assumed for $c_S$ and $e_0$. (We note that while $e_0$ was fixed to the $^{252}$Cf(sf) value from Ref. 7 for neutron induced fission, the value $e_0 = 10.0724$ /MeV was retained from FREYA version 1.0 [5] for the other spontaneously fissioning isotopes.) While one can reasonably assume that $e_0$ has a universal value since the nuclear level densities are related to nuclear structure and not reaction dependent, $c_S$ was fixed for expedience. The $x$ parameter for $^{240}$Pu(sf) in FREYA 2.0.2 was fixed from experimental analysis of neutron-neutron correlations in Ref. 8, an observable not used in this optimization because it requires full analysis of the detector setup in each case. However, these correlations exhibit strong sensitivity to $x$ [10]. For other spontaneously fissioning isotopes, it was taken to be $\sim 1.2$.

The parameter $c$ was fixed via examination of $P(\nu)$. Finally, $d\text{TKE}$ was tuned to $\tau$ after the other parameter values were fixed. The work in this paper is the first to make a full optimization of all parameters for all isotopes. It is interesting to compare how well this empirical approach compares with the numerical optimization performed in the current paper.

As already noted, we do not expect $c$ and $d\text{TKE}$ to be independent of isotope. As can be seen in Table IV they are not. The values obtained for $c$ are driven entirely by $P(\nu)$ and its moments. In the cases where $c$ is large, $c > 3$, $^{240}$Pu(sf) and $^{242}$Pu(sf), it is because despite the low average neutron multiplicity, $P(\nu)$ is broader than might be expected for low $\tau$. In such cases, the range of $c$ needs to be increased to match the higher moments of the multiplicity distribution, $\nu_2$ and $\nu_3$. There is also one exception to the expectation that $c \geq 1$, $^{238}$U(sf). In this case, the evaluated $P(\nu)$ is actually more peaked than a distribution with $c = 1$ for the same $\tau$, requiring the fluctuations to be reduced to achieve agreement with
the evaluated $P(\nu)$ and its moments. Note, however, that, within uncertainties, $c$ is still compatible with unity in this case. The values of $c$ in FREYA 2.0.2 were 0.92, 1.91, 3, 3.4 and 1.34 for $^{238}$U(sf), $^{238}$Pu(sf), $^{240}$Pu(sf), $^{242}$Pu(sf) and $^{244}$Cm(sf) respectively, in addition to the value of 1.18 found in Ref. [7] for $^{252}$Cf(sf). These empirical guesses are very close to the results obtained from our current optimization based on evaluations that explicitly constrain $c$.

Next, as indicated, we expect $dTKE$ to vary from case to case, independent of isotope. Ideally $dTKE$ should be zero with a perfect model along with high statistics input yields and TKE ($A_H$). This is indeed the case for $^{252}$Cf(sf), a well-measured standard with a high spontaneous fission rate. $dTKE$ is small for $^{252}$Cf(sf): $dTKE = 0.525$ MeV here and 0.52 MeV in Ref. [7]. We now compare our optimized $dTKE$ values for the other isotopes studied here with those in FREYA 2.0.2 [6]: $dTKE = -1.345$ MeV, $-1.366$ MeV, $-3.071$ MeV, $-1.600$ MeV, and $-4.35$ MeV for $^{238}$U(sf), $^{238}$Pu(sf), $^{240}$Pu(sf), $^{242}$Pu(sf) and $^{244}$Cm(sf) respectively. These values are in rather good agreement with those found in our optimization. We note that the large range in $dTKE$ values is expected and does not affect the physical interpretation of the parameters.

It is notable that these values are, in contrast to that for $^{252}$Cf(sf), all negative and the absolute values are considerably larger. A negative value for $dTKE$ indicates that the reported TKE ($A_H$) distribution is too high, reducing the overall available excitation energy for neutron emission. However, the measured fission rates are much lower for other isotopes and large fluctuations exist in the data. In most of these cases, the number of fission events measured was small so that not many events go into each $A_H$ bin. FREYA samples the yields and TKE ($A_H$) directly from measured fission fission fragment data, often with undefined or unquantified systematic uncertainties. Thus the input TKE ($A_H$) in FREYA in these cases are based on low statistics, sometimes without uncertainties on the data, and with unknown systematic errors. Introducing $dTKE$ is a way to correct for these unknowns as well as offering a means to compensate for any remaining, unquantified, physics effects.

Previously, the value of $c_S$ was fixed at 0.87 for every isotope of FREYA. We can see that our results for $^{252}$Cf(sf) agree with this but the $^{240}$Pu(sf) result is somewhat larger. We generally find that the spin temperature is close to the scission temperature, resulting in fragment spins close to the maximum available rotation. We note that there is some correlation between $c_S$ and $dTKE$. While it may be especially weak for $^{252}$Cf(sf), it could be responsible for the differences observed between the values of $dTKE$ in FREYA 2.0.2 [24] and Table IV since changing $c_S$ changes $E_{rot}$ which, in turn, modifies $E_{stat}$, thus ultimately affecting $dTKE$. Increasing $c_S$, as for e.g. $^{240}$Pu(sf) to 0.908 from 0.87, increases $E_{rot}$. Thus for a fixed scission energy $E_{sc}$ then, $E_{stat}$ is reduced, decreasing the energy available for neutron emission. To keep $\nu$ fixed, absent other variation, $dTKE$ has to decrease. This is seen in Table IV as $dTKE$ is now $-3.219$ MeV instead of $-3.07$ MeV. Similar reductions of $dTKE$ can be seen for increased $c_S$ in the other cases studied.

The parameter $x$ controls the distribution of statistical excitation energy between the fragments after scission. It is well established that $x$ is greater than unity based on $\nu(A)$ data and previous measurements of the average

| $c_0$/MeV | $x$ | $c$ | $c_S$ | $dTKE$ MeV | # Data Sets | # Evaluations |
|----------|-----|-----|-------|------------|-------------|--------------|
| $^{238}$U(sf) | 10.391 | 1.220 | 0.939 | 0.899 | -1.375 | 0 | 1 [13] |
| $^{238}$Pu(sf) | 10.521 | 1.232 | 1.968 | 0.893 | -1.408 | 0 | 1 [13] |
| $^{239}$Pu(sf) | 10.750 | 1.307 | 3.176 | 0.908 | -3.219 | 1 [19] | 1 [13] |
| $^{242}$Pu(sf) | 10.018 | 1.144 | 3.422 | 0.911 | -1.662 | 1 [19] | 1 [13] |
| $^{244}$Cm(sf) | 10.488 | 1.239 | 1.391 | 0.906 | -4.494 | 2 [21, 22] | 1 [13] |
| $^{242}$Cf(sf) | 10.429 | 1.274 | 1.191 | 0.875 | 0.525 | 4 [13, 16, 18] | 1 [13, 17] |

Table IV: Results of the optimization for all spontaneously-fissioning isotopes modeled by FREYA. The best fit values of the five parameters, $y$, and their associated standard deviations, $\sigma_y$, are given for each isotope. In addition, the number of data sets and evaluations used for each isotope are indicated, along with the references for these data. Note that in the case of Ref. [13], the evaluation gives the result for multiple observables: $P(\nu)$, $\nu_2$, $\nu_3$, $\nu_4$. 
neutron multiplicities from the light and heavy fragments respectively. The previous values of $x$ [24] generally assumed $x \sim 1.2$ aside from the value of 1.27 established in Ref. [7] for $^{252}$Cf(sf) and the 1.3 found for $^{240}$Pu(sf) based neutron-neutron angular correlation data [8]. The estimates of $x \sim 1.1 - 1.2$ for the other spontaneously fissioning isotopes were borne out by our independent fits. Despite the fact that the $x$ range was $1 < x < 1.5$ in all the fits, with $\nu(A)$ data only available for $^{244}$Cm(sf) and $^{252}$Cf(sf), the optimized values are very similar to the default of $x \sim 1.2$ assumed previously.

The values of $e_0$ in Table IV are remarkably similar, between 10/MeV and 10.75/MeV for all isotopes, despite the wide range, $7 < e_0 < 12$/MeV. This is particularly striking because, of the observables considered, the prompt fission neutron spectrum shows any direct dependence on $e_0$, even though it also depends on every other parameter. For example, increasing $c_S$ gives more available excitation energy to neutron emission which could, in principle, increase the average energy per neutron rather than increasing the number of neutrons and thus change the slope of the prompt fission neutron spectrum. Giving a larger share of the excitation energy to the light fragment would also influence the average neutron emission, see Eq. (2), increasing $e_0$ while keeping the average neutron multiplicity fixed forces the temperature to increase. Since $c_S$ is related to the thermal fluctuations in the decaying nucleus, if the temperature increases, then the fluctuations can also increase so that $c_S$ has to decrease to compensate. The correlation between $e_0$ and $c_S$ is the strongest of all, near $+1$, implying that $c_S$ must increase when $e_0$ increases. Again, increasing $e_0$ can imply an increase in temperature and a probability of greater neutron emission. To compensate, $c_S$ needs to increase to give more rotational energy to the fragments and more photon emission to keep the neutron emission fixed. There is also a relatively strong, negative, correlation between $e_0$ and $dTKE$. A higher fragment temperature could either lead to increased neutron emission or emission of higher energy neutrons. If fewer neutrons are emitted with higher average energy, then $dTKE$ would need to decrease to compensate to increase the total excitation energy to increase neutron emission.

There is a relatively weak correlation between $x$ and $c$. Since $x$ adjusts how the statistical excitation energy is divided between the fragments, it primarily affects $\nu(A)$ whereas $c$ adjusts the width of the multiplicity distribution $P(\nu)$. A moderate positive correlation is seen between $x$ and $c_S$. If $x$ is increased to give more energy to the light fragment, the average neutron multiplicity can be expected to increase. Thus $c_S$ must increase to take more rotational energy and keep the neutron multiplicity constant. The correlation between $x$ and $dTKE$ is small and positive so that, if $x$ increases neutron emission, then $dTKE$ must increase to compensate and reduce the total excitation energy.

A moderate, negative correlation is seen between $c$ and $c_S$. If $c$ increases, $\nu$ will decrease so that, for $\nu$ to be maintained, the rotational energy, and thus $c_S$, has to decrease. On the other hand, the correlation between $c$ and $dTKE$ is moderate but positive. If neutron emission increases with increasing $c$, then to increase the total excitation energy to compensate, $dTKE$ has to increase also to decrease the neutron multiplicity. Finally, there is a relatively large negative correlation between $c_S$ and $dTKE$. If $c_S$ is increased, the fragment spin and thus rotational energy increases, taking energy away from that available for statistical neutron emission. Thus $dTKE$ has to decrease to give more total excitation energy to the fragments and maintain the value of $\nu$.
aren't calculating an average, we instead take this variance to be $1/\sqrt{N}$ where $N$ is the number of observed events in that bin. (b) Ratio of calculated values to evaluation results.

VII. COMPARISON TO DATA

We now use the optimized parameters presented in Sec. V to generate a set of one million FREYA events, and compare this to the data used in our optimization, along with some data which were not included. We present the direct comparisons as well as ratios of the calculated to experimental values (C/E). It is important to note that throughout the section, any uncertainties given on the results from FREYA arise from calculating the variance of the result. In cases where there is no relevant variance to calculate, we instead use $1/\sqrt{N}$, where $N$ is the relevant event multiplicity in the bin of a distribution. Especially in this case, this 'uncertainty' should not be compared to the uncertainty on the experimental data.

As can be seen in Figs. 2 and 3 we reproduce the neutron probability distribution $P(\nu)$ within very low uncertainty in both cases. As discussed in Sec. IV the neutron multiplicity distributions are well established for both of these isotopes. Even where we do differ from the result, the uncertainties on C/E are still compatible with unity. We also reproduce the average neutron multiplicity in Table V to one or two decimal points, effectively the regime in which we can accurately interpret the FREYA results. We also note that the neutron multiplicity moments have improved with the new set of parameters over those from Ref. [7]. The moments are important for criticality studies. Note that the uncertainty on the FREYA calculation is a calculation of the variance of the result, and should therefore not be interpreted as the range of values we should expect FREYA to return.

The prompt fission neutron spectrum is compared to the Mannhart evaluation in Fig. 4. We show the results on a logarithmic scale on the $y$ axis in (a) and (b) as well as the $x$ axis in (c) and (d). These particular scales allows us to get a good sense of the behaviour of the spectrum in both the low energy regime from 0 to 1 MeV, in (a) and (b), as well as the high energy range from 1 to 12 MeV, in (c) and (d). FREYA reproduces this distribution with high accuracy in the low energy range except for the slight kink around 0.1 MeV present in the non-smoothed version of the Mannhart evaluation, which we employ because it includes uncertainties. There is a more significant deviation in the high energy range, but the range of uncertainty in C/E is consistent with unity for neutron energies above 7 MeV. It is also important to note that
Figure 4: (Color online) (a) Neutron energy spectrum for $^{252}\text{Cf}(sf)$ from FREYA as well as the Mannhart evaluation [17] with logarithmic scale on the $x$ axis. (b) Ratio of calculated values to evaluation results. A logarithmic scale is used on the $x$-axis in both cases. In (c) and (d) the same results are shown now with a logarithmic scale on the $y$-axis, and a linear scale on the $x$-axis in order to highlight the difference and uncertainty at high energy.

The uncertainties are extremely large in this high-energy region for both the experimental data and the FREYA output. We note that the FREYA uncertainties in the high energy tail of the spectrum can be reduced by generating a larger number of events while the uncertainties on the evaluation cannot.

The FREYA results differ more significantly from the data on the neutron multiplicity as a function of fragment mass, as seen in Fig. 5. This is expected since $x$ is single valued and our fit employs the mass region $105 < A < 145$. Even though we have only used this well-behaved region for our optimization procedure, it is important to note that the result is still within the uncertainty on C/E, meaning that this result is still statistically successful. We can also compare to experimental data not used in the optimization. In Fig. 6 we show a more recent data set for $\nu(A)$ which also agrees well with FREYA in the fit region. Note that the uncertainty on the FREYA calculation is a calculation of the variance of the result, and should therefore not be interpreted as the range of values we should expect FREYA to return.

The results for the neutron multiplicity as a function of TKE in Fig. 5 are particularly successful for $160 < \text{TKE} < 190 \text{ MeV}$. In the region of low TKE, we see far fewer fragments, so the results in this region are less reliable. Similarly, as we move to higher TKE, while the results begin to differ more, the uncertainty on C/E typically contains unity since there are also fewer events

| Moment Evaluation | FREYA | C/E |
|-------------------|-------|-----|
| $^{238}\text{U}(sf)$ |       |     |
| $\nu_1$ | $1.98 \pm 0.03$ | $2.0 \pm 0.94$ | $1.01 \pm 0.27$ |
| $\nu_2$ | $2.8743 \pm 0.1411$ | $2.87 \pm 3.37$ | $1.0 \pm 1.37$ |
| $\nu_3$ | $5.6219 \pm 0.481$ | $2.83 \pm 9.81$ | $1.0 \pm 11.71$ |
| $^{239}\text{Pu}(sf)$ |       |     |
| $\nu_1$ | $2.19 \pm 0.07$ | $2.17 \pm 1.15$ | $0.99 \pm 0.27$ |
| $\nu_2$ | $3.8736^a$ | $3.85 \pm 4.35$ | $0.99 \pm 1.26$ |
| $\nu_3$ | $5.417^b$ | $5.25 \pm 10.97$ | $0.97 \pm 4.1$ |
| $^{240}\text{Pu}(sf)$ |       |     |
| $\nu_1$ | $2.154 \pm 0.005$ | $2.22 \pm 1.25$ | $1.03 \pm 0.33$ |
| $\nu_2$ | $3.7889 \pm 0.029$ | $4.26 \pm 4.88$ | $1.12 \pm 1.66$ |
| $\nu_3$ | $5.2105 \pm 0.1402$ | $6.53 \pm 13.3$ | $1.25 \pm 6.51$ |
| $^{241}\text{Cm}(sf)$ |       |     |
| $\nu_1$ | $2.149 \pm 0.008$ | $2.12 \pm 1.19$ | $0.99 \pm 0.3$ |
| $\nu_2$ | $3.8087 \pm 0.036$ | $3.79 \pm 4.51$ | $0.99 \pm 1.4$ |
| $\nu_3$ | $5.3487 \pm 0.036$ | $5.36 \pm 12.13$ | $1.0 \pm 5.14$ |
| $^{242}\text{Cf}(sf)$ |       |     |
| $\nu_1$ | $2.71 \pm 0.01$ | $2.7 \pm 1.16$ | $1.0 \pm 0.18$ |
| $\nu_2$ | $5.941 \pm 0.0188$ | $5.95 \pm 5.46$ | $1.0 \pm 0.84$ |
| $\nu_3$ | $10.112 \pm 0.175$ | $10.17 \pm 16.78$ | $1.0 \pm 2.75$ |
| $^{243}\text{Cf}(sf)$ |       |     |
| $\nu_1$ | $3.757 \pm 0.01$ | $3.74 \pm 1.3$ | $1.0 \pm 0.12$ |
| $\nu_2$ | $11.951 \pm 0.0188$ | $11.94 \pm 8.79$ | $1.0 \pm 0.54$ |
| $\nu_3$ | $31.668 \pm 0.175$ | $31.84 \pm 39.94$ | $1.01 \pm 1.59$ |

$^a$No uncertainty reported.

$^b$No uncertainty reported.

Table V: Average neutron multiplicity and the second and third moments of the neutron multiplicity distribution for all six isotopes in FREYA compared with the evaluations in Ref. [13]. Note that the uncertainty on the FREYA calculation is a calculation of the variance of the result, and should therefore not be interpreted as the range of values we should expect FREYA to return.
Table VI: Average photon multiplicity and average energy per photon for all six isotopes in FREYA compared with experimental data (when available). The data for $^{252}$Cf(sf) comes from Ref. [18] while the data for $^{240}$Pu(sf) and $^{242}$Pu(sf) both come from Ref. [19]. Note that the uncertainty on the FREYA calculation is a calculation of the variance of the result, and should therefore not be interpreted as the range of values we should expect FREYA to return.

| Isotope | $N_\gamma$ (MeV) | $\overline{\nu}$ (MeV) | $\chi^2$ | C/E |
|---------|------------------|------------------|--------|-----|
| $^{238}$U(sf) | 8.2 ± 0.4 | 6.6 ± 2.48 | 0.8 ± 0.09 | — |
| $^{240}$Pu(sf) | 6.72 ± 0.07 | 6.61 ± 2.43 | 0.98 ± 0.13 | — |
| $^{242}$Pu(sf) | 6.72 ± 0.07 | 6.61 ± 2.43 | 0.98 ± 0.13 | — |
| $^{244}$Cm(sf) | 7.07 ± 2.56 | — | — | — |
| $^{244}$Cf(sf) | 8.14 ± 0.4 | 7.71 ± 2.8 | 0.95 ± 0.12 | — |

$^a$From Ref. [19].
$^b$From Ref. [19].
$^c$From Ref. [19].
$^d$From Ref. [19].
$^e$From Ref. [19].
$^f$From Ref. [19].

with high TKE. We also show the more recent data, not used in the fit, in Fig. 6. FREYA actually agrees better with this new data at large TKE because $\nu$ (TKE) \(\rightarrow 0\) at large TKE.

As explained in Sec. [14] the parameters, especially $c$, have a high level of control over the shape of the neutron multiplicity distribution. This is, however, not the case for the photon multiplicity distribution: there is no parameter that has direct control over the width of this distribution as there is for $P(\nu)$. The shape generated by FREYA in Fig. 7 is narrower than the data. There is an estimated uncertainty of ±1 in the detected photon multiplicity [25]. If we adjust the FREYA output to account for multiple scattering [25], the width becomes broader. As we can see in Fig. 7 after adjusting the FREYA output for multiple scattering, the agreement of FREYA with the data is considerably improved. As is evident, the uncertainty on C/E is compatible with unity in the high multiplicity range. The average photon multiplicity is also closely recreated. These results can be found in Table VI along with the average energy per photon.

Figure 5: (Color online) (a) Neutron multiplicity as a function of fragment mass for $^{252}$Cf(sf) along with experimental data from Refs. [14] [15]. As discussed in Sec. [14] we use the Dushin data in the fit, and provide the Göök data [15] for comparison. Note that the uncertainty on the FREYA calculation is a calculation of the variance of the result, and should therefore not be interpreted as the range of values we should expect FREYA to return. (b) Ratio of calculated result from FREYA to the experimental results.

VIII. CONCLUSIONS

We have performed a numerical optimization of the 5 physics-based parameters in FREYA for all spontaneously-fissioning isotopes so far included. The fits, using simulated annealing to find a global minimum, which agree with our physics intuition, are also in rather good agreement with the empirical values in FREYA 2.0.2 [6].

The parameters provide good agreement with the data where they are available. We will next apply the fitting procedure we have developed here to neutron-induced fission.

Acknowledgments

We wish to acknowledge helpful conversations with Andrew Nicholson, Jorgen Randrup, Jerome Verbeke, and Patrick Talou. The computational work was done with the Savio cluster using the faculty computing allowance provided by Berkeley Research Computing. This work was supported by the Office of Nuclear Physics in the U.S. Department of Energy’s office of Science under contracts No. DE-AC52-07NA27344 (RV) and DE-AC02-05CH11231 (LAB), as well as by the U.S. Department of
Figure 6: (Color online) (a) Neutron multiplicity as a function of total kinetic energy for $^{252}$Cf(sf) compared to experimental data from Refs. [15, 16]. As is discussed in Sec. [14] we use the Budtz-Jorgensen data [16] for the fit, and provide the the Göök data [15] for comparison. Note that the uncertainty on the FREYA calculation is a calculation of the variance of the result, and should therefore not be interpreted as the range of values we should expect FREYA to return. (b) Ratio of calculated FREYA result to the experimental results.

[1] J. Randrup, R. Vogt, Calculation of fission observables through event-by-event simulation, Phys. Rev. C 80, 024601 (2009).
[2] R. Vogt, J. Randrup, D. A. Brown, M. A. Descalle, and W. E. Ormand, Event-by-event evaluation of the prompt fission neutron spectrum from $^{239}$Pu(n,f), Phys. Rev. C 85, 024608 (2012).
[3] R. Vogt and J. Randrup, Event-by-event study of photon observables in spontaneous and thermal fission, Phys. Rev. C 87, 044602 (2013).
[4] R. Vogt and J. Randrup, Event-by-event study of neutron observables in spontaneous and thermal fission, Phys. Rev. C 84, 044621 (2011).
[5] J. Verbeke, R. Vogt, and J. Randrup, Fission Reaction Event Yield Algorithm, FREYA – For event-by-event simulation of fission, Comp. Phys. Comm. 191, 178 (2015).
[6] J. Verbeke, R. Vogt, and J. Randrup, Comp. Phys. Comm. 222, 263 (2018).
[7] R. Vogt, A. Nicholson, J. Randrup, I. Gauld, and S. Croft, Uncertainty Quantification with the Event-by-Event Fission Model FREYA, Proc. 1st ANS Advances in Nucl. Nonpro. Tech. and Policy, Santa Fe, NM, 2016, LNL-CONF-690741.
[8] J.M. Verbeke, L.F. Nakae, and R. Vogt, Neutron-Neutron Angular Correlations in Spontaneous Fission of $^{252}$Cf and $^{240}$Pu Phys. Rev. C 97, 044601 (2018).
[9] J. Randrup and R. Vogt, Refined treatment of angular momentum in the event-by-event fission model FREYA, Phys. Rev. C 89, 044601 (2014).
[10] R. Vogt and J. Randrup, Neutron angular correlations in spontaneous and neutron-induced fission, Phys. Rev. C 90, 064623 (2014).
[11] R. Vogt and J. Randrup, Improved modeling of photon observables with FREYA, Phys. Rev. C 96, 064620 (2017).
[12] S. Kirkpatrick, C.D. Gelatt and M.P. Vecchi, Optimization by Simulated Annealing, Science 220, 4598 (1983).
[13] P. Santi and M. Miller, Reevaluation of Prompt Neutron Emission Multiplicity Distributions for Spontaneous Fission Nuclear Science and Engineering 160, 190 (2008).
[14] V. Dushin, F.-J. Hambsch, V. Jakovlev, V. Kalinin, I. Kraev, A. Laptev, D. Nikolaev, B. Petrov, G. Petrov, V. Petrova, V. Petrova, A.S. Vorobyev, et al., Facility for neutron multiplicity measurements in fission, Nuclear Instruments and Methods in Physics Research A 516, 539 (2004).
[15] A. Göök, F.-J. Hambsch, and M. Vidali, Prompt neutron multiplicity in correlation with fragments from spontaneous fission of $^{252}$Cf, Phys. Rev. C 90, 064611 (2014).
[16] C. Budtz-Jorgensen and H.-H.Knitter, Simultaneous Investigation of Fission Fragments and Neutrons in $^{252}$Cf(sf) Nucl. Phys. A 490, 307 (1988).
[17] W. Mannhart, Evaluation of the $^{252}$Cf(sf) Fission Neu-
Evidence for the stochastic aspect of prompt γ emission in spontaneous fission et al., Phys. Rev. C 85, 021601 (2012).

[19] S. Oberstedt, A. Oberstedt, A. Gatera, A. Göök, F.-J. Hambsch, A Moens, G Sibbens, D Vanleeuw, and M. Vidal, Impact of low-energy photons on the characteristics of prompt fission γ-ray spectra Phys. Rev. C 93, 054603 (2016).

[20] Z.A. Aleksandrova, V.I. Bol’shov, V.F. Kuznetsov, G.N. Smirenkin, M.Z. Tarasko, Spectra of the Prompt Neutrons Arising from the Spontaneous Fission of 252Cf, 244Cm, and 240Pu Atomnaya Energiya 36, 282-285 (1973).

[21] R. Schmidt and H. Henschel, Comparison of the Spontaneous Fission of 244Cm(sf) and 252Cf(sf), Nucl. Phys. A 395, (1983).

[22] L.M. Belov, M.V. Blinov, N.M. Kazarinov, A.S. Krikovkhatskiy, and A.N. Protopopov Spectra of Fission Neutrons of 244Cm(sf), 242Pu(sf), and 239Pu(sf), Yadernye-Fizicheskie Issledovaniya Reports 6, (1968).

[23] R. Capote et al., RIPL – Reference Input Parameter Library for Calculations of Nuclear Reactions and Nuclear Data Evaluations, Nucl. Data Sheets 110, 3107 (2009).

[24] J. M. Verbeke, J. Randrup and R. Vogt, Fission Reaction Event Yield Algorithm: FREYA 2.0.2 User Manual, LLNL report LLNL-SM-705798.

[25] A. Chyzh, private communication.

Figure 7: (Color online) (a) Gamma multiplicity distribution for 252Cf(sf) along with experimental data from [18] before correcting for multiple scattering. (b) Ratio between the calculated values from FREYA and the experimental data. (c) Gamma multiplicity distribution for 252Cf(sf) along with experimental data from [18] after correcting for multiple scattering. (d) Ratio between the calculated values from FREYA and the experimental data.