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Expanding FREYA Capabilities to Topics Relevant to the Fuel Cycle

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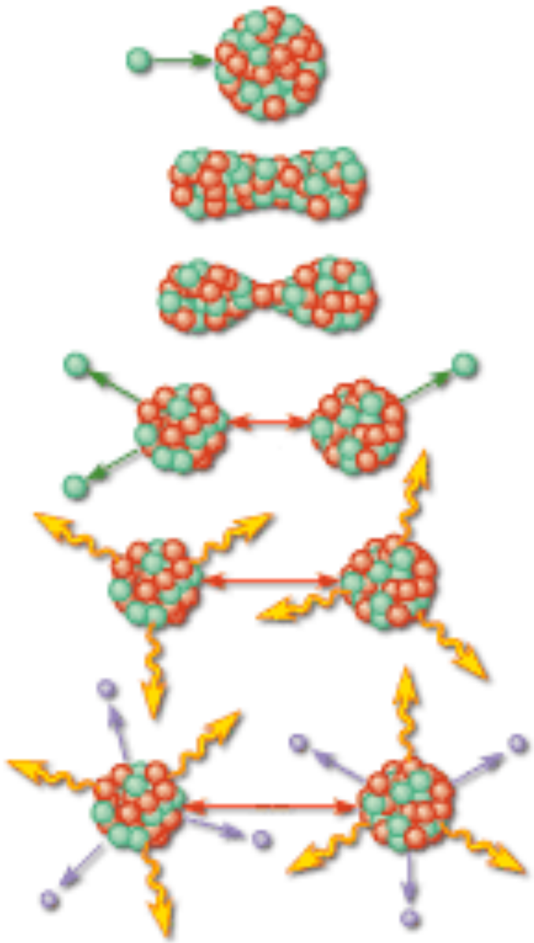
We are developing FREYA (Fission Reaction Event Yield Algorithm) for spectral evaluations and correlation studies

- Collaborative Pu fission spectrum theory and modeling effort at LLNL (D. Brown, E. Ormand, J. Pruet, R. Vogt and W. Younes)
- J. Randrup (LBNL) is collaborator on **FREYA** development
- Papers in Phys. Rev. C **80** (2009): 024601 (JR & RV on **FREYA**) and 044611 (early spectral evaluation, with J. Pruet and W. Younes); another in progress

How FREYA works

- Assume binary fission of compound nucleus with mass A_c and charge Z_c formed by incident neutrons with energy E_n on actinide with mass $A_c - 1$
- Sample mass and charge of light, L, and heavy, H, fragments from fission fragment distributions, conserving mass and charge
- Determine fission Q from fragments, divide Q value between fragment kinetic and excitation energies
- Fix total kinetic energy, TKE, by sampling kinetic energy due to mutual Coulomb repulsion, obtain total excitation energy by conservation, TEE = Q − TKE
- Divide TEE between light and heavy fragments
- Allow for temperature fluctuations in small systems; adjust TKE accordingly to retain total energy conservation
- Evaporate neutrons from each fragment until excitation energy is too low for further neutron emission
- Prompt gamma emission follows after prompt neutron emission ceases



Setting up scission configuration

- Scission configuration assumed to be two spheroidal prefragments with tips separated by distance d
- At scission, prefragments distorted relative to ground state shapes due to mutual Coulomb repulsion
- Two contributions to prefragment excitation energy, the distortion energy δV_i and statistical excitation (heat) Q_i, $E_i^* = \delta V_i + Q_i$
- Distortion moves centers apart, lowering mutual Coulomb repulsion V_{ij}^C
- δV_i depends on the surface energy E_S^0 and Coulomb energy E_C^0, $\delta V = \frac{8}{45}[E_S^0 - \frac{1}{2}E_C^0](\epsilon_{\rm sc}^2 - \epsilon_{\rm gs}^2)$ where $\epsilon_{\rm sc}$ is the fragment deformation at scission and $\epsilon_{\rm gs}$ is the ground state deformation
- Mean excitation energy in the nucleus is $\overline{Q}_i = a_i T_i^2$

Fragment kinetic energy due to Coulomb repulsion

- Average total fragment kinetic energy obtained from Coulomb repulsion between two deformed prefragments

$$V_{ij}^C = e^2 \frac{Z_i Z_j}{c_i + c_j + d} F(x_i, x_j)$$

- $F(x_i, x_j) = 1$ if both fragments spherical, greater than one if one or both are prolate
- F depends on deformation measures x_i and average fragment radius R_i

$$\begin{aligned} x_i^2 &= \frac{c_i^2 - b_i^2}{R_i^2} \\ c_i &= R_i \frac{1 + \epsilon/3}{(1 - 2\epsilon/3)^{2/3}} \quad \text{major axis} \\ b_i &= R_i \frac{1 - 2\epsilon/3}{(1 + \epsilon/3)^{1/3}} \quad \text{minor axis} \end{aligned}$$

- When fragments lose contact, they are accelerated by V_{ij}^C and relax back into equilibrium shapes
- Scission distortion energy converted into extra statistical excitation
- Assume these changes take place before de-excitation begins

Fission Q value

For any given binary fission channel into light and heavy fragments

$$^{A_0}Z_0 \longrightarrow\ ^{A_L}Z_L +\ ^{A_H}Z_H$$

$$\begin{aligned} M_0^* = M_0^{\text{gs}} + E_0^* &= M_L^{\text{gs}} + E_L^* + M_H^{\text{gs}} + E_H^* + V_{LH}^C \\ &= M_L^* + M_H^* + K_{LH} \end{aligned}$$

where M_i^{gs} is ground-state mass of $^{A_i}Z_i$, $i = 0, L, H$, and E_i^* is its excitation, so that the total mass is $M_i^* = M_i^{\text{gs}} + E_i^*$. Ground-state masses are taken from Audi supplemented by calculated masses by Möller *et al.*.

Coulomb repulsion V_{LH}^C is assumed to be fully converted to relative kinetic energy of the two fragments, K_{LH}.

Fission channel Q value is

$$Q_{0\to LH} = M_0^{\text{gs}} + E_0^* - M_L^{\text{gs}} - M_H^{\text{gs}} = K_{LH} + E_L^* + E_H^* \ .$$

Thermal fluctuations distribute heat and kinetic energy

Average total statistical excitation energy of scission configuration, $\overline{Q}$ is

$$\overline{Q} \equiv \overline{Q}_L + \overline{Q}_H = M_0^{\rm gs} + E_0^* - M_L^{\rm sc} - M_H^{\rm sc} - V_{LH}^C$$

where $M_i^{\rm sc} = M_i^{\rm gs} + \delta V_i$ is the mass of the distorted prefragment in the scission configuration. Internal energy $\overline{Q}$ assumed to be partitioned statistically between the two prefragments.

Total excitation energy divided in proportion to the respective heat capacities (level density parameters), $\partial\overline{Q}_i/\partial T_i = 2a_iT_i \propto a_i$.

Prefragments in the scission configuration have a common temperature, $T_{LH} = [\overline{Q}/(a_L + a_H)]^{1/2} = [\overline{Q}_i/a_i]^{1/2}$, so that $\overline{Q}_i = a_iT_{LH}^2$.

Fluctuations given by thermal variances, $\sigma_i^2 = 2\overline{Q}_iT_{LH}$. The fluctuations δQ_i are sampled from normal distributions with variances σ_i^2 so that $Q_i = \overline{Q}_i + \delta Q_i$.

Due to fragment fluctuations, combined statistical excitation, $Q = Q_L + Q_H$, also fluctuates. Must be compensating fluctuation in total kinetic energy,

$$K_{LH} = \overline{K}_{LH} + \delta K_{LH} \;\; \overline{K}_{LH} = V_{LH}^C \;, \quad \delta K_{LH} = -\delta Q_L - \delta Q_H \;.$$

Approximately gaussian distribution of heat in each prefragment,

$$P_i(Q_i) \approx (2\pi\sigma_i^2)^{-\frac{1}{2}} {\rm e}^{-(Q_i - \overline{Q}_i)^2/2\sigma_i^2} \;.$$

Associated total variance is sum of individual variances, $\sigma_Q^2 = \sigma_L^2 + \sigma_H^2$. Energy conservation specifies K_{LH} distribution is gaussian with variance $\sigma_K = \sigma_Q$.

Prompt neutron evaporation from excited fragments

Q value for neutron emission (maximum possible excitation of daughter nucleus)

$$Q_{\rm n} = M_i^{\rm gs} + E_i^* - M_f^{\rm gs} - m_{\rm n}$$

where $M_f^{\rm gs}$ is ground state mass of daughter nucleus

Assume kinetic energy of evaporated neutron, $\epsilon_{\rm n} = p_{\rm n}^2/2m_{\rm n}$, is isotropic in rest frame of emitting nucleus with $v_{\rm n} \propto \sqrt{\epsilon_{\rm n}}$ ($d^3\mathbf{p}_{\rm n} \propto \sqrt{\epsilon_{\rm n}} d\epsilon_{\rm n}$) so that

$$\begin{aligned} \frac{d^3\nu}{d^3\mathbf{p}_{\rm n}} d^3\mathbf{p}_{\rm n} &\propto \sqrt{\epsilon_{\rm n}}\, {\rm e}^{-\epsilon_{\rm n}/T_f^{\rm max}} \sqrt{\epsilon_{\rm n}} d\epsilon_{\rm n}\, d\Omega \\ &= \epsilon_{\rm n}\, {\rm e}^{-\epsilon_{\rm n}/T_f^{\rm max}} d\epsilon_{\rm n}\, d\Omega \ , \end{aligned}$$

Resulting excitation energy and mass of daughter nucleus

$$E_f^* = Q_{\rm n} - \epsilon_{\rm n} \quad M_f^* = M_f^{\rm gs} + E_f^*$$

Emission continues until no further emssion energetically possible, $E_f^* < S_{\rm n}$ where $S_{\rm n}$ is neutron separation energy of daughter,

$$S_{\rm n} = M(^AZ) - M(^{A-1}Z) - m_{\rm n}$$

Evaluation requires improved fission modeling at all energies, not just thermal

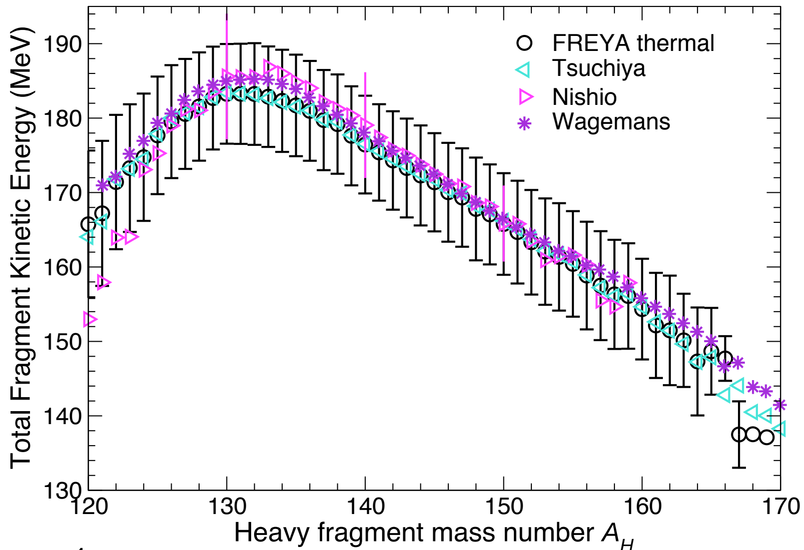
Extract shape of TKE(A_H) from thermal data and assume energy independence, moves TKE up and down

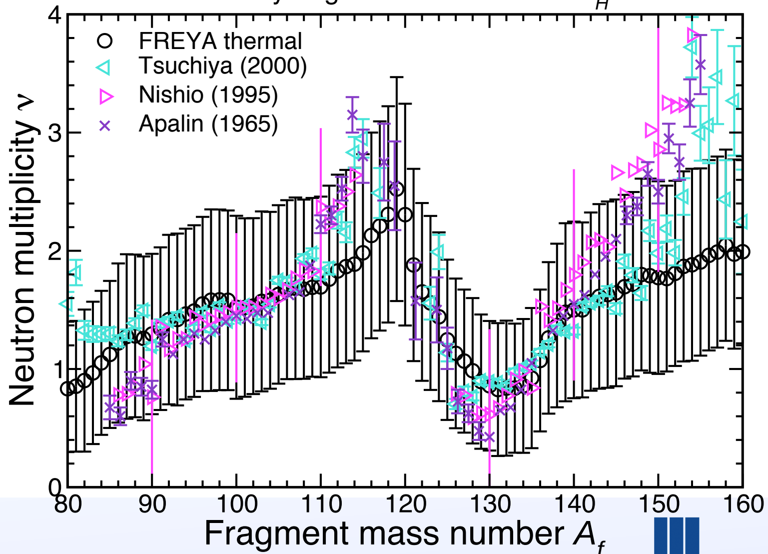
Sawtooth shape of ν falls out from TKE shape
And energy balance

Seems to work well but more data, e.g. from fission TPC,
May help fill in gaps if taken over a range of incident
Energies

Poor quality of spectral data means we must rely on extrapolations and inclusive quantities such as average total multiplicity

Improved spectral evaluations require better modeling of all aspects of the physics of fission process and/or new data over entire energy range of evaluation similar to that shown here





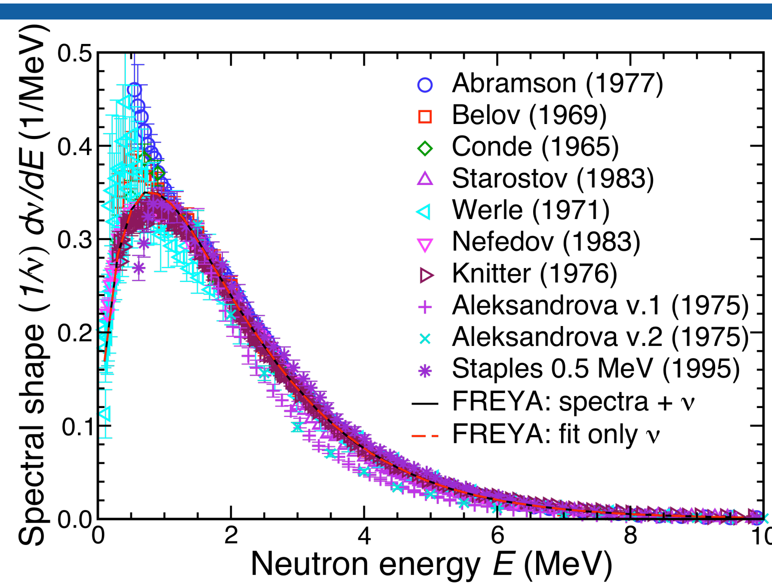
Fitting to data isn't all it's cracked up to be

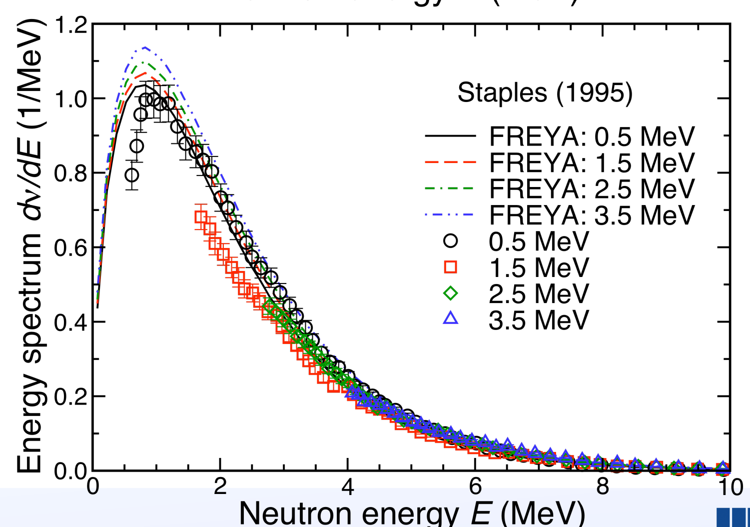
An improved spectral evaluation involves understanding more than just the spectra

- There are big 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 the spectra.
- Both the average neutron multiplicity, $\overline{\nu}$, and the spectra, $d\overline{\nu}/dE$, depend on the physics of the fission process. The two are intimately linked and can't really be treated separately, N.B. $\int dE(d\overline{\nu}/dE) = \overline{\nu}$.
- Improvements in the spectral evaluation will come with improved modeling of fission.

Data are often insufficient for comprehensive understanding of fission process

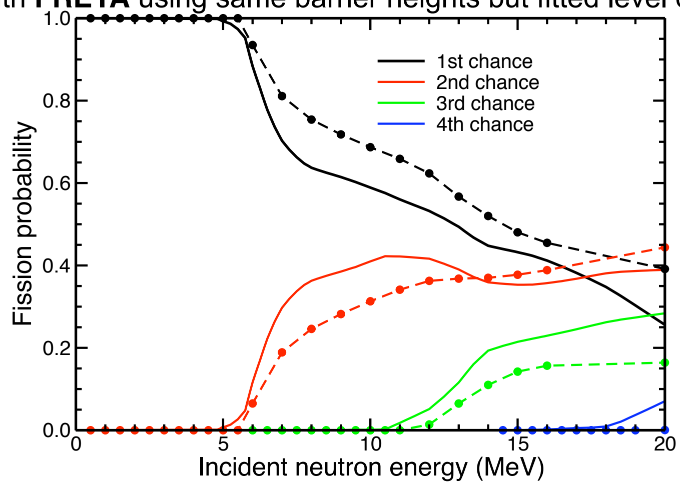
- Spectral data for thermal neutrons are inconsistent with each other and have large uncertainties in important regions, much larger than the constraints on $\overline{\nu}$ itself
- Published spectral data do not extend into the low energy region, only extend to incident neutron energies of a few MeV
- Measurements of other quantities such as total fragment kinetic energy and neutron multiplicity as a function of fragment mass only exist for low incident energies
- Modeling of complete fission events helps fill the gaps in data



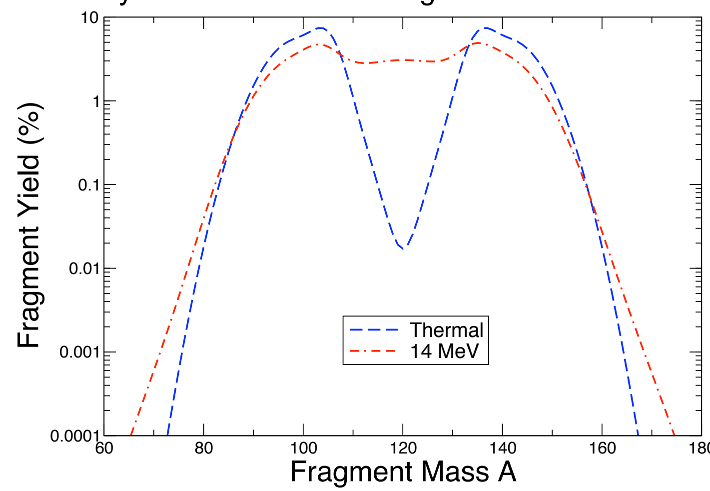


Three important contributions at high incident neutron energy

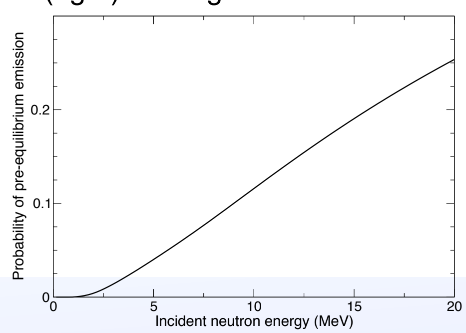
- **Multi-chance fission**, emission of one or more neutrons from excited compound nucleus pre-fission, turns on when incident neutron energy is above neutron separation energy for compound nucleus (^{240}Pu, 1st chance; ^{239}Pu, 2nd chance; ^{238}Pu, 3rd chance)
- Comparison to ENDF ^{239}Pu evaluation (**GNASH** calculation, not measured) with **FREYA** using same barrier heights but fitted level density parameter

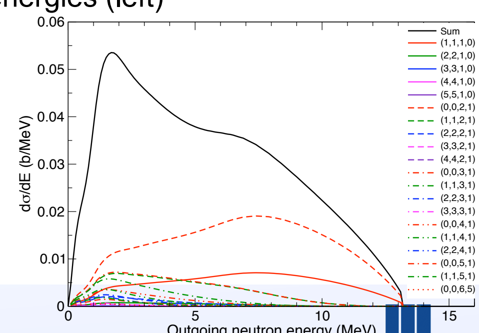


- **Transition from asymmetric to symmetric fission:** more symmetric with increasing incident neutron energy



- **Pre-equilibrium emission:** captured neutron fails to equilibrate and is re-emitted; biggest effect for smallest number of intermediate excited states (right) and high incident neutron energies (left)





- **Missing ingredients to improve analysis at all incident neutron energies:**
  - **more comparison data e.g. Y(A,TKE)**
  - **more differential data like $n(A_f)$**
  - **better modeling of yields, kinetic energies with 'hot' fission (dynamics)**

Obtaining the most likely set of fit parameters

- Assume our model parameters $\{\alpha_k\} = \{s, e_0, x\}$ are uniformly distributed in parameter space
- For each specified set of parameters, $\{\alpha_k^{(m)}\}$, generate a large set of fission events with FREYA (about 1M for each m) and extract observables $\{\mathcal{C}_i\}$ which are compared to experimental values, $\{\mathcal{E}_i\}$
- We then calculate the χ^2 deviation of the observables from their measured values,

$$\chi_m^2 \equiv \chi^2\{\alpha_k^{(m)}\} \equiv \sum_i \frac{(\mathcal{C}_i\{\alpha_k^{(m)}\} - \mathcal{E}_i)^2}{\sigma_i^2}$$

and obtain relative weights that give likelihood for calculations with the given set of parameters to give the "correct" result,

$$w_m \equiv w\{\alpha_k^{(m)}\} \propto \mathrm{e}^{-\frac{1}{2}\chi^2\{\alpha_k^{(m)}\}}$$

- Obtain probability density in model parameter space, $P\{\alpha_k\} \equiv w\{\alpha_k\}/W$, where $W \equiv \sum_m w_m$, used to obtain best estimate for the model parameter values, the likelihood-weighted average

$$\tilde{\alpha}_k \equiv \prec \alpha_k \succ \equiv \frac{1}{W}\sum_m w_m \alpha_k^{(m)} \approx \alpha_k^0$$

where the best estimate is that with the largest likelihood

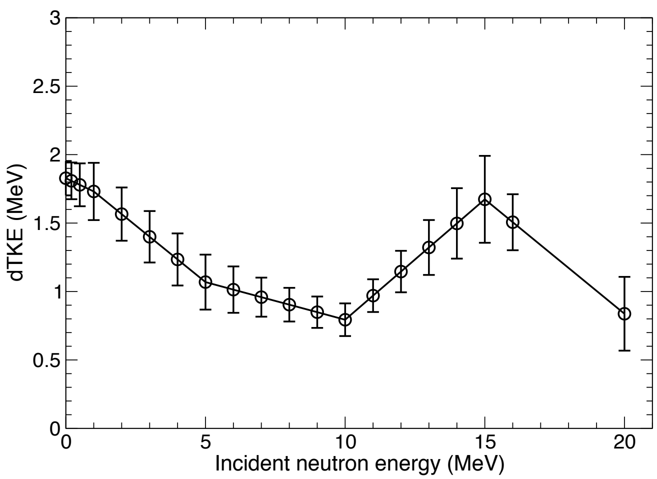
Statistical methods used to match average FREYA neutron multiplicity to the ENDF-B/VII evaluated result, including covariance

Three FREYA parameters 'tuned' to average neutron multiplicity, $\overline{\nu}$:

- shift of TKE(A_H), dTKE, from average of data at thermal energies – the shape is assumed to remain the same as a function of energy, not necessarily the case if system is excited enough for shell effects to be negligible
- the asymptotic level density parameter, e_0, which sets the fragment 'temperature' for neutron evaporation;
- the relative excitation of the light and heavy fragments, x where $x = 1$ is the equal temperature situation with the same number of neutrons emitted from both fragments while $x > 1$ gives more neutrons evaporated from the light fragment than the heavy fragment.

Randomized set of parameters dTKE, e_0 and x chosen over a reasonable range to obtain spectra and $\overline{\nu}$ for each set of parameters, χ^2 minimized to obtain optimal parameter set. We assume that e_0 and x are independent of energy and that dTKE varies linearly between fixed points at incident energies of 10^{-11}, 1, 5, 10, 15 and 20 MeV to ensure a good fit with a reasonable number of realizations (1000 on a Latin Hypercube parameter grid).

Result for dTKE

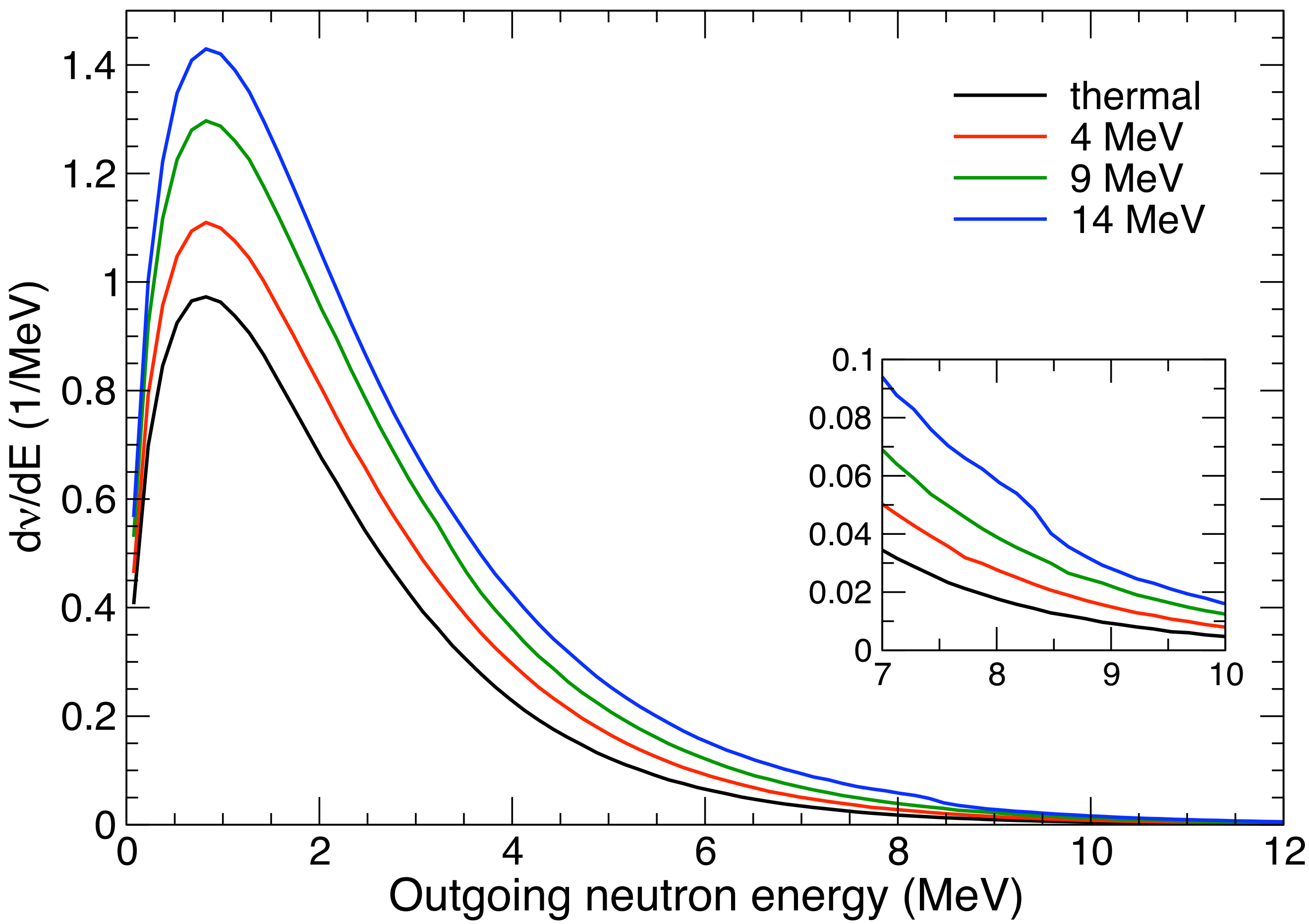


Energy-independent parameters:

e_0 = 8.542 ± 0.5449 MeV^{-1}

x = 1.139 ± 0.0616

Spectral results including multi-chance fission and pre-equilibrium neutron emission



Correlations between input parameters reveals characteristics of fit

We can calculate the covariances among pairs of input parameters in the set α_k,

$$\tilde{\sigma}_{kk'} \equiv \prec (\alpha_k - \tilde{\alpha}_k)(\alpha_{k'} - \tilde{\alpha}_{k'}) \succ \quad .$$

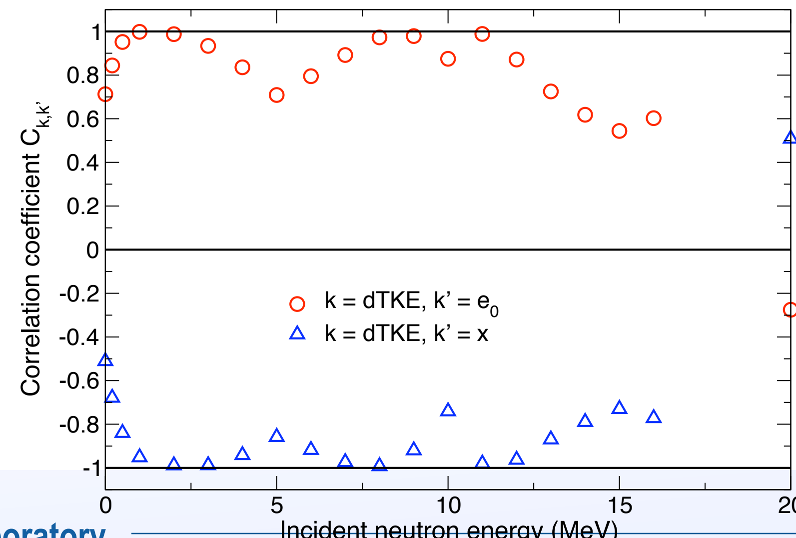
The diagonal elements, $\tilde{\sigma}_{kk} = \tilde{\sigma}_k^2$, are the variances and $\tilde{\sigma}_k$'s are the standard deviations of the parameter values (squares of the individual model parameter uncertainties). The off-diagonal elements give the covariances between two parameters.

We can also calculate the sometimes more transparent associated *correlation coefficients*,

$$C_{kk'} \equiv \frac{\tilde{\sigma}_{kk'}}{\tilde{\sigma}_k \tilde{\sigma}_{k'}} \ .$$

If the parameters are uncorrelated, $C_{kk'} = 0$. Correlated parameters lead to nonzero correlation coefficients. If $C_{kk'} > 0$, α_k increases as $\alpha_{k'}$ increases. On the other hand, if $C_{kk'} < 0$, α_k increases as $\alpha_{k'}$ decreases.

Changes in correlation coefficient occur near fixed points of the fits

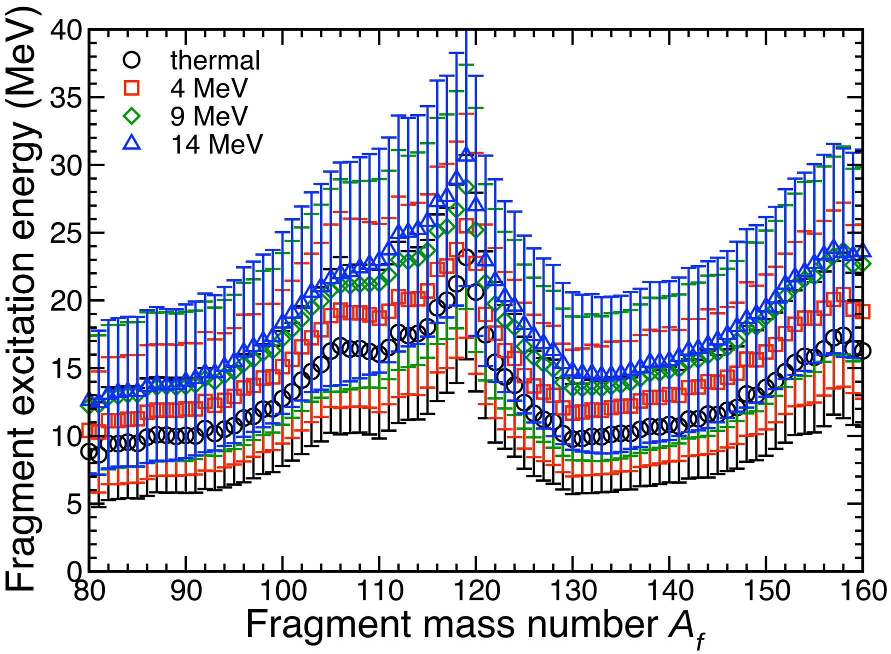


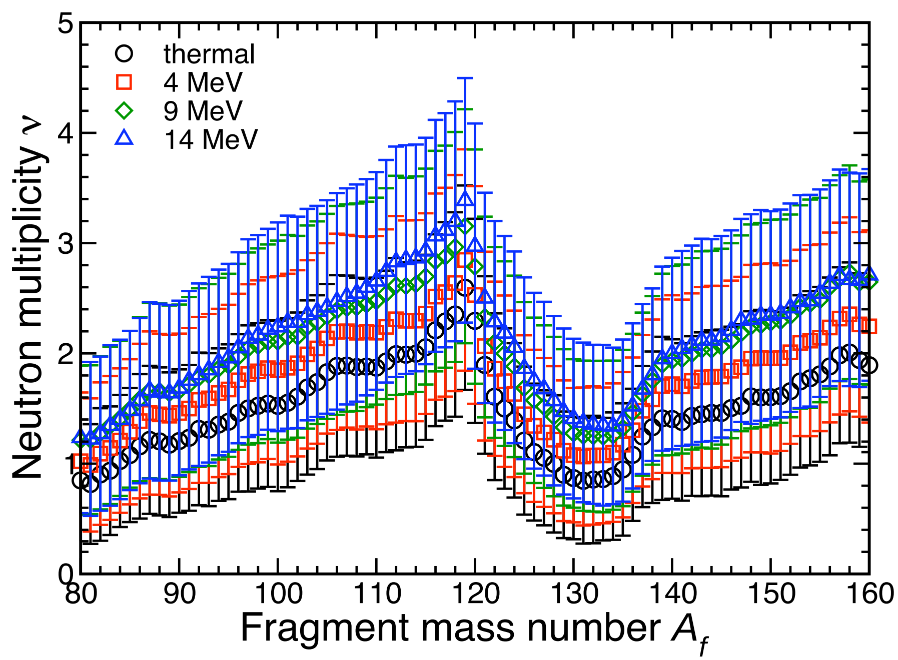
$C_{e0,x}$ = -0.946, both e_0 and x are energy independent

Neutron multiplicity correlated with excitation energy

(Left) Mean fragment excitation energy and the associated dispersion

(Right) Corresponding mean neutron multiplicity as a function of fragment mass; agrees well with sawtooth shape of data

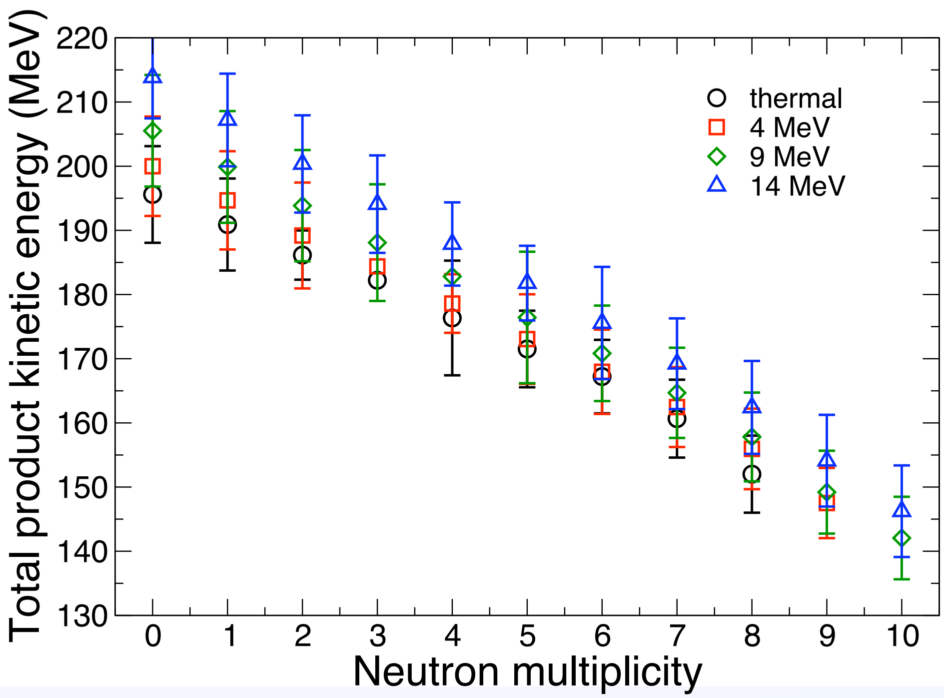


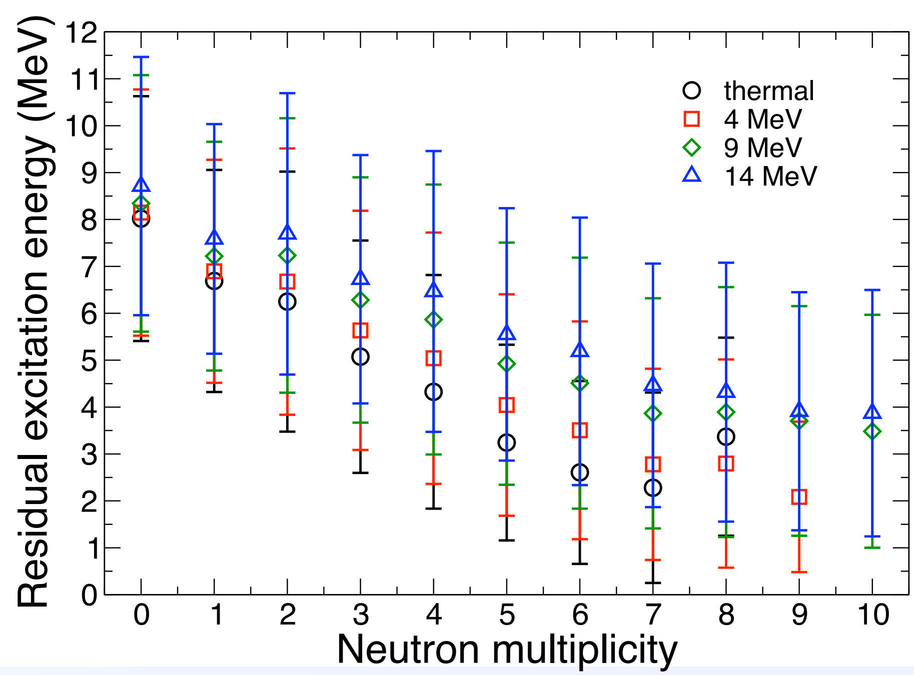


Dependence of fragment kinetic and excitation energies on neutron multiplicity correlated with gamma emission

(Left) Mean total kinetic energy of fission products (after evaporation) – larger multiplicities come from more excited fragments and thus lower kinetic energies

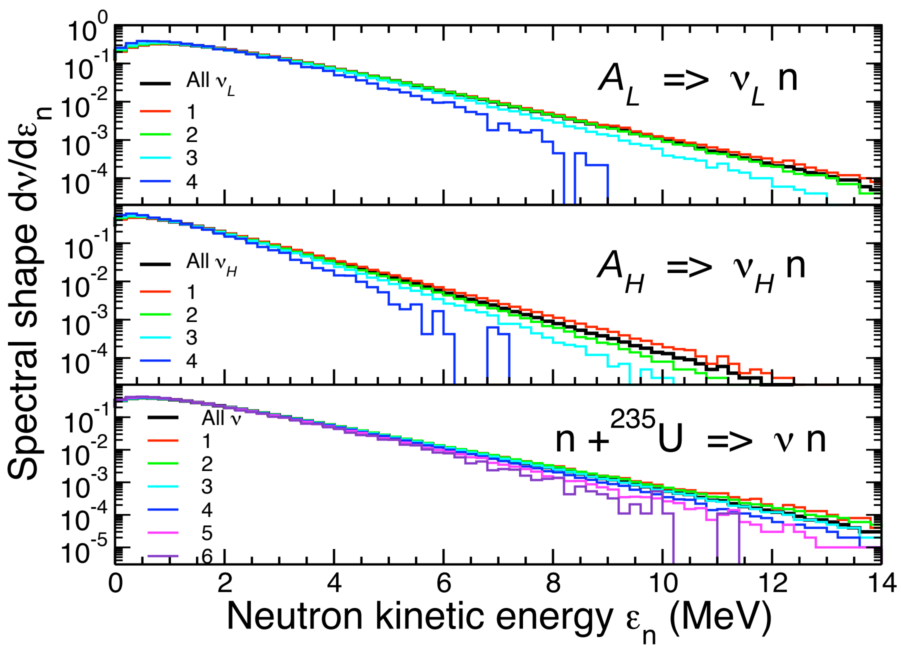
(Right) Mean total excitation energy of the fission products left after evaporation – provides a qualitative indication of neutron-photon correlations: the more neutrons, the less excitation left for photon emission, the two multiplicities (neutron and photon) are anti-correlated

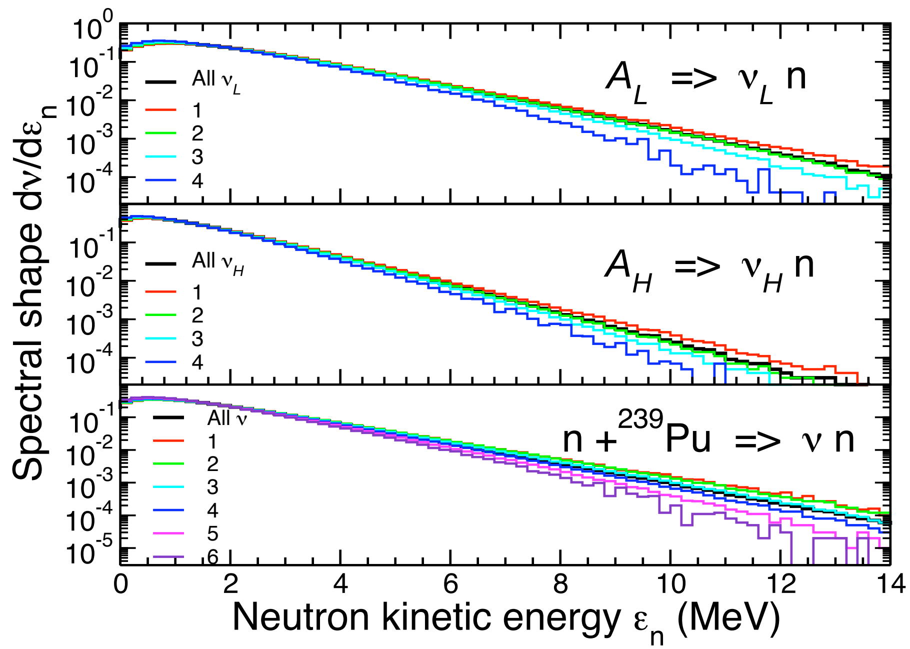




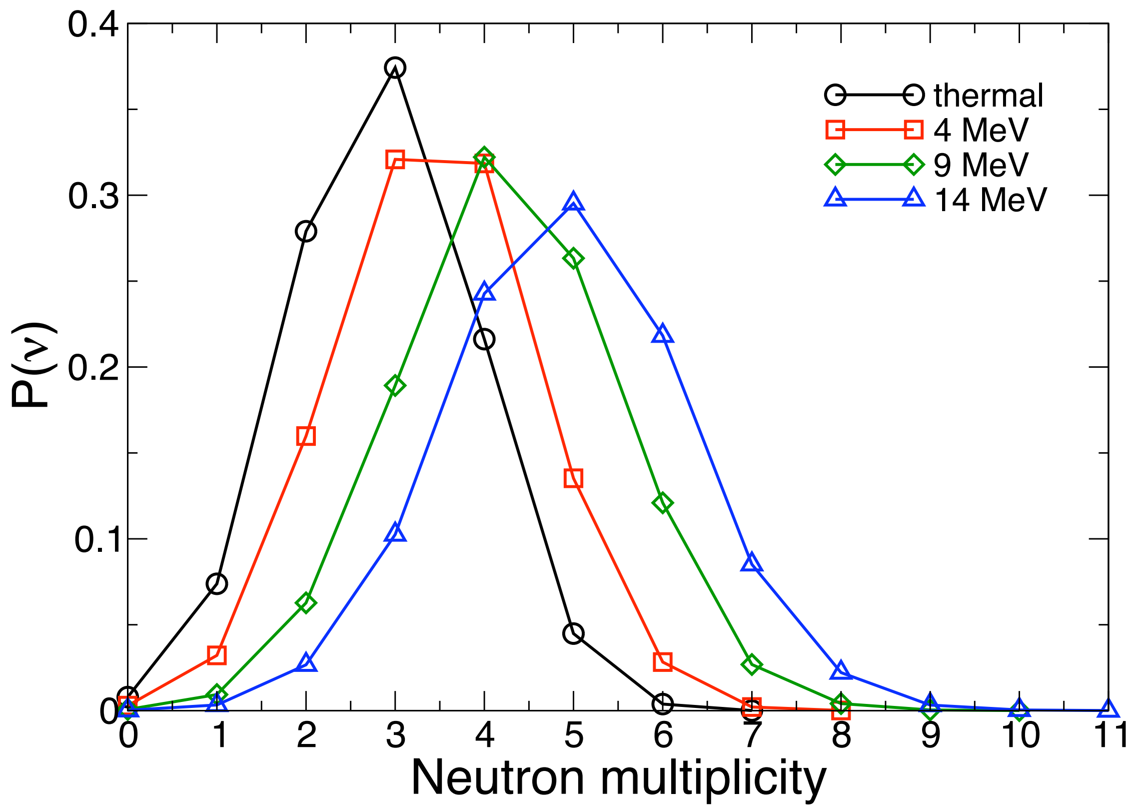
Neutron number-energy correlations

Preliminary **FREYA** results show softening of spectra for higher multiplicities





Probability for prompt neutron emission



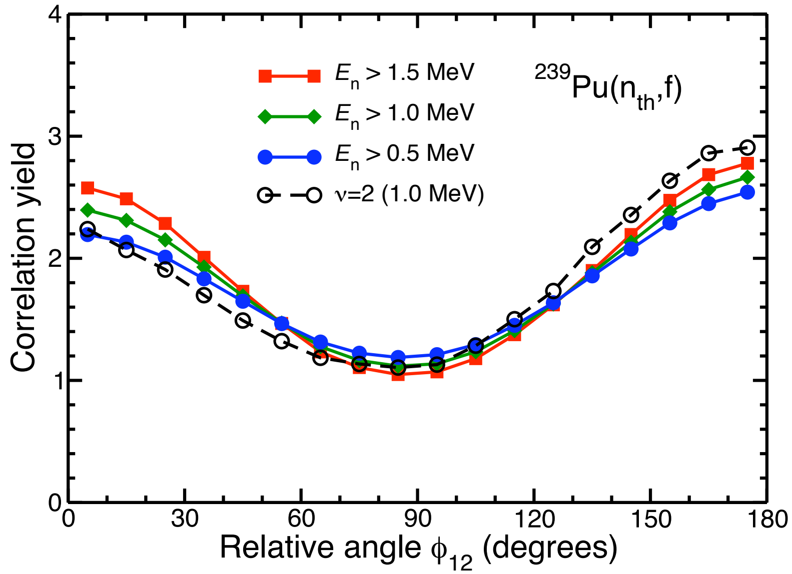
Examples of neutron angular correlations in FREYA

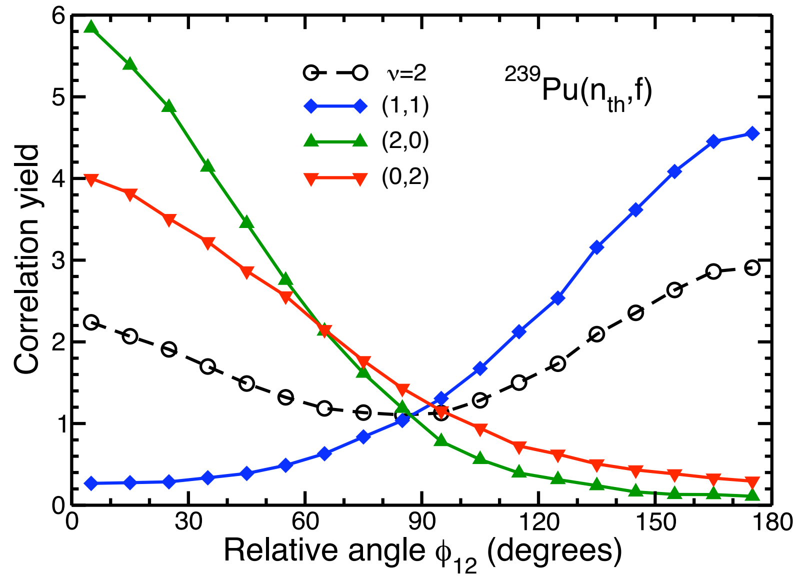
Correlations of neutrons with energies above a specified threshold energy

Yield forward and backward is more symmetric for higher energy neutrons

Correlations between neutrons when exactly 2 neutrons with $E_n > 1$ MeV emitted

One from each fragment (blue) back to back; both from single fragment emitted in same direction, tighter correlation when both from light fragment (green) than from heavy (red); open circles show sum of all possibilities





Correlations between outputs reveals physics features

Covariances between different observables, $\mathcal{C}_i$, are calculated as

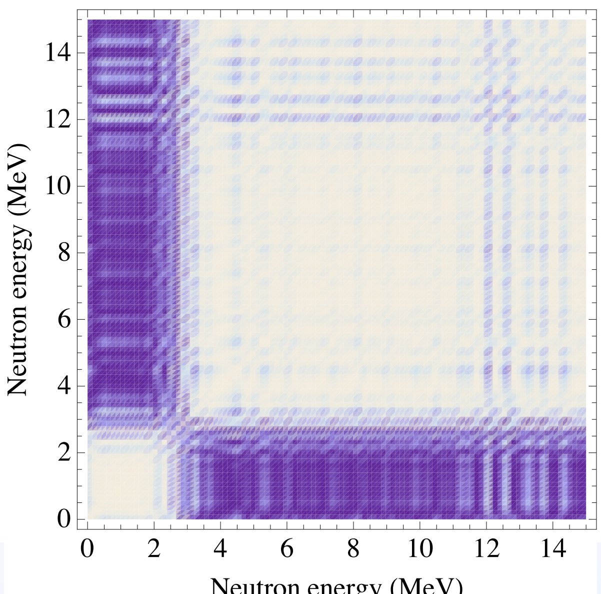
$$\tilde{\sigma}_{ij} \equiv \prec (\mathcal{C}_i - \tilde{\mathcal{C}}_i)(\mathcal{C}_j - \tilde{\mathcal{C}}_j) \succ \ .$$

The diagonal elements are the squares of the standard deviations, $\tilde{\sigma}_i$, of the calculated values $\mathcal{C}_i$ resulting from uncertainties in the model parameter values. Here the correlation coefficients between the observables $\mathcal{C}_i$ and $\mathcal{C}_j$ are

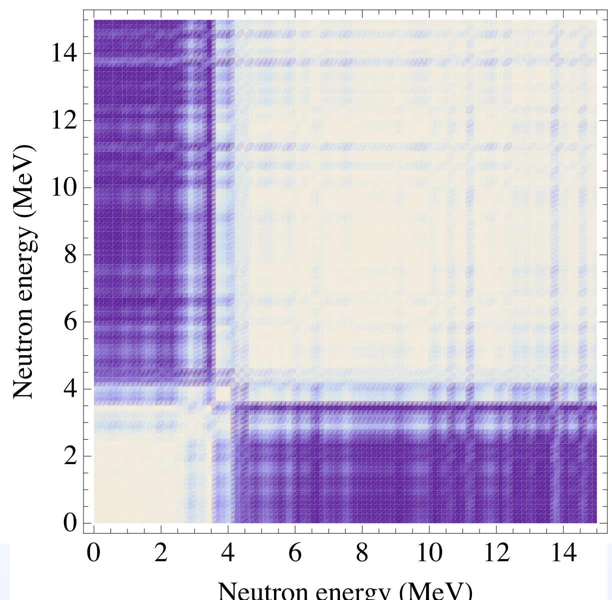
$$C_{ij} \equiv \frac{\tilde{\sigma}_{ij}}{\tilde{\sigma}_i \tilde{\sigma}_j} \ .$$

Spectral correlations between outgoing neutron energies

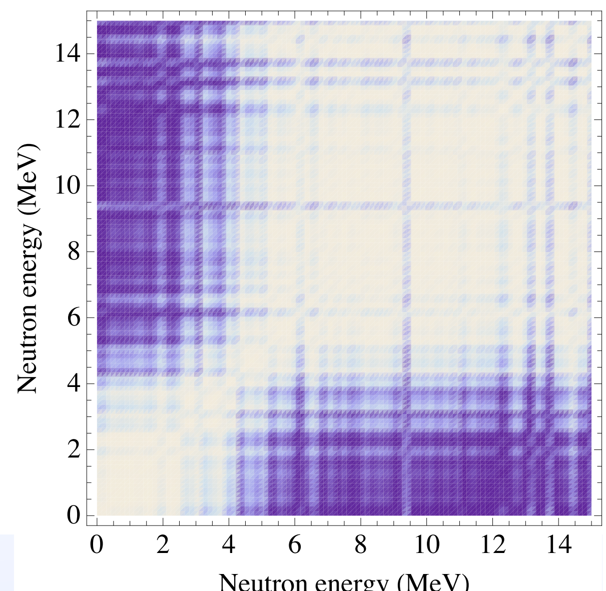
E_n = 0.5 MeV



E_n = 6 MeV

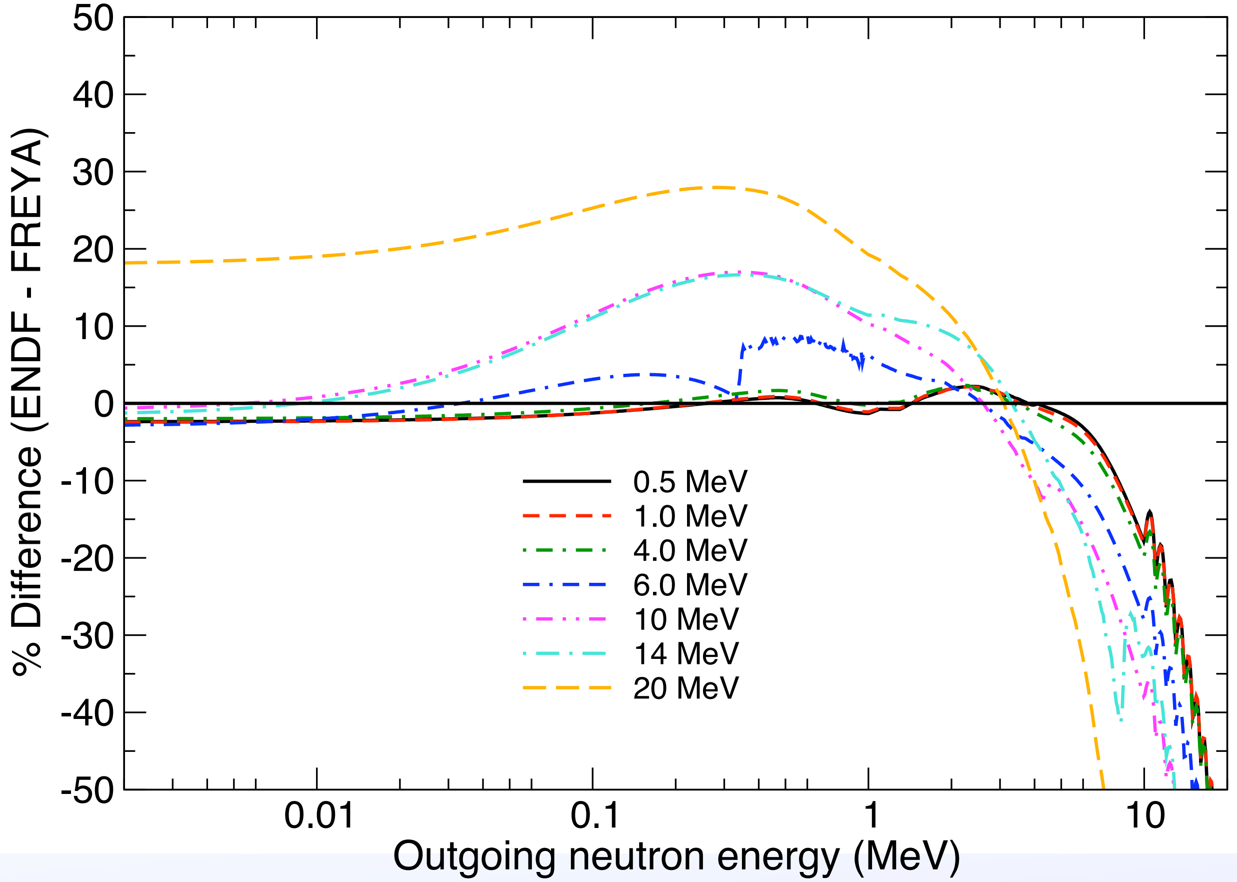


E_n = 14 MeV



FREYA evaluation can be compared to existing ENDF evaluation

- Evaluation prepared with FREYA fits
  - Extrapolation to 10^{-5} MeV and up to 20 MeV done by fitting two different Watt spectra
- Differences in spectral shapes at both low and high energies – FREYA higher for E < 1 MeV



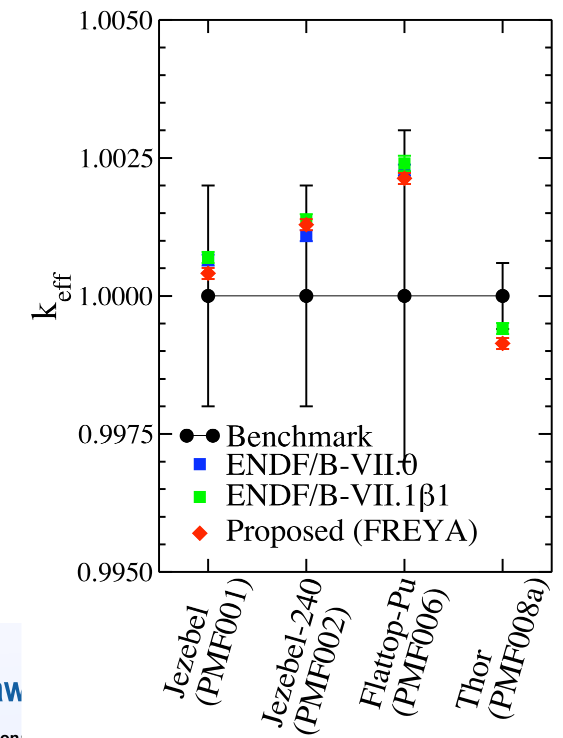
New evaluation passes key integral tests

Critical assemblies (k_{eff}) designed to determine critical mass

k_{eff} is eigenvalue of neutron transport equation, measures relative neutron gain and loss in material

Benchmark for PFNS evaluations, Jezebel is classic criticality test

FREYA agrees well with Pu assemblies

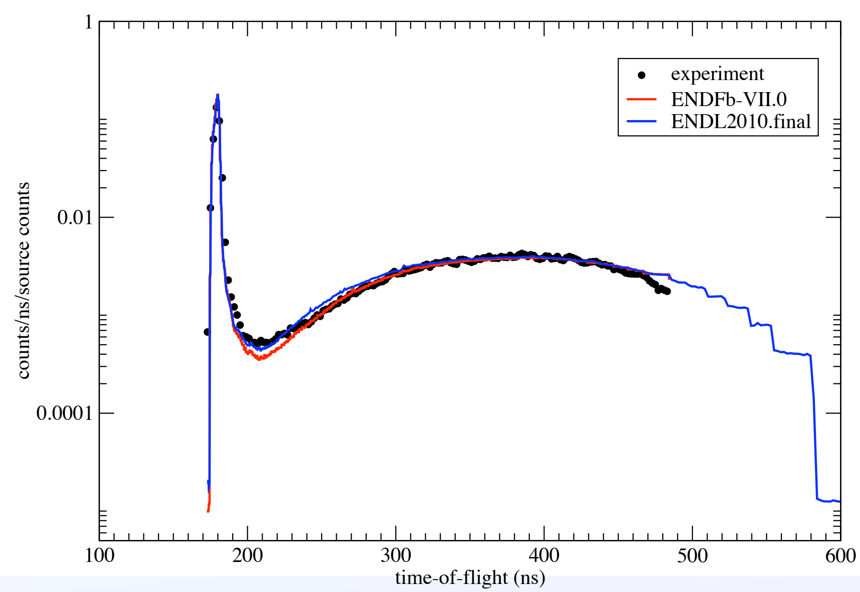


Pulsed spheres have 14 MeV neutron source at center of material to be tested

Description of neutron time of flight immediately after pulse tests PFNS in material

FREYA result improves pulsed sphere description in important region, ~ 200 ns

(Disagreement at late times due to imprecise modeling of objects in room outside sphere)



FREYA capabilities for AFCI

- **FREYA** is becoming a mature fission modeling tool
- Evaluation produces neutron-neutron covariances as a by-product of fitting procedure
- Applied so far to plutonium; spectral evaluations of ^{235}U and ^{238}U to follow this year
- New funding for **FREYA** obtained to make detailed studies of correlations for DOE NA22 and to make **FREYA** generally available
  - For more actinides
  - In codes where it can be called for single events such as **MCNP**, **GEANT**, etc.