A Microscopic Theory for Nuclear Fission: Excitation Energy Dependence and Uncertainty Quantification

INT Program, Quantitative Large Amplitude Shape Dynamics

9 October 2013

J. D. McDonnell

Lawrence Livermore National Laboratory

LLNL-PRES-644900

This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344. Lawrence Livermore National Security, LLC

Outline

- Density Functional Theory for Nuclear Fission
  - Brief Review of Finite Temperature Formalism
- Results for Thorium and Uranium Isotopes
- Results for the Mercury Region
- Early Work with Uncertainty Quantification
- Conclusion

The FT-DFT Formalism for Nuclear Fission

- Self-consistent, constrained HFB with Skyrme-like EDF
- We use the symmetry-unrestricted DFT solver HFODD.

J. Dobaczewski and J. Dudek, Comput. Phys. Comm. 102, 183 (1997).
N. Schunck *et al*, Comput. Phys. Comm. 183, 166 (2012).

$$\mathcal{E}_{\text{int.}}(r) = \sum_{t=0,1} \left\{ C_t^{\rho\rho}\rho_t^2 + C_t^{\rho\tau}\rho_t\tau_t + C_t^{J^2}\mathbf{J}_t^2 + C_t^{\rho\Delta\rho}\rho_t\Delta\rho_t + C_t^{\rho\nabla J}\rho_t\nabla\cdot\mathbf{J}_t \right\}$$

$$\begin{pmatrix} h & \Delta \\ -\Delta^* & -h^* \end{pmatrix} \begin{pmatrix} U_k \\ V_k \end{pmatrix} = \begin{pmatrix} U_k \\ V_k \end{pmatrix} E_k$$

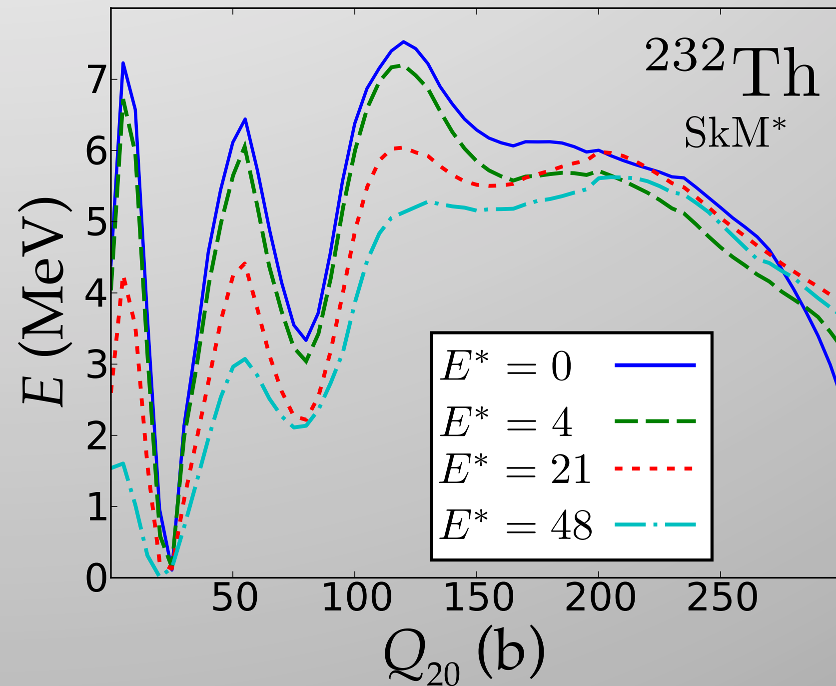
$$h = \epsilon + \Gamma$$

$$\Gamma_{\mu\nu} = \sum_{\alpha\beta} v_{\mu\beta\nu\alpha}\rho_{\alpha\beta} \qquad \rho = UfU^\dagger + V^*(1-f)V^T$$

$$\kappa = UfV^\dagger + V^*(1-f)U^T$$

$$\Delta_{\mu\nu} = \frac{1}{2}\sum_{\alpha\beta} v_{\mu\nu\alpha\beta}\kappa_{\alpha\beta} \qquad f_i = \frac{1}{1+e^{\beta E_i}}$$

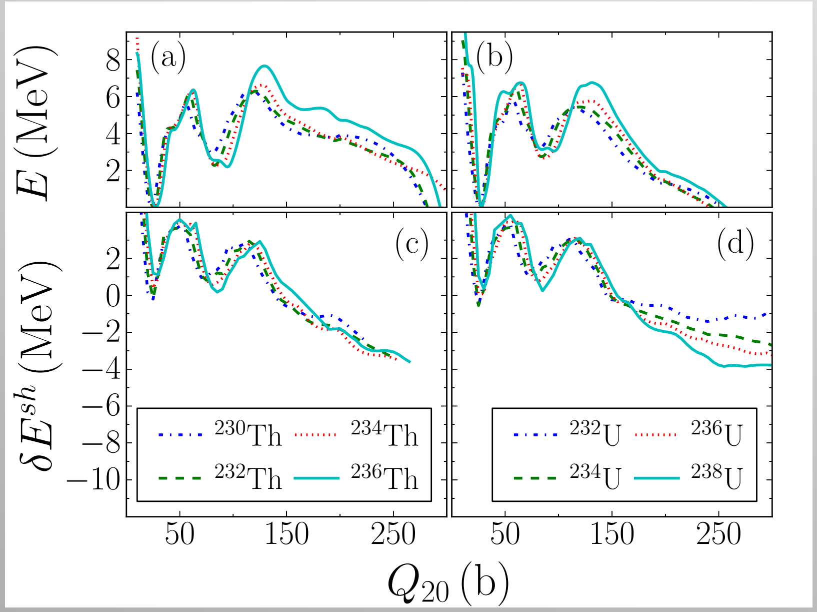
$$\langle S\rangle = -kTr(D\ln D) = \sum_i [(1-f_i)\ln(1-f_i) + f_i\ln f_i]$$



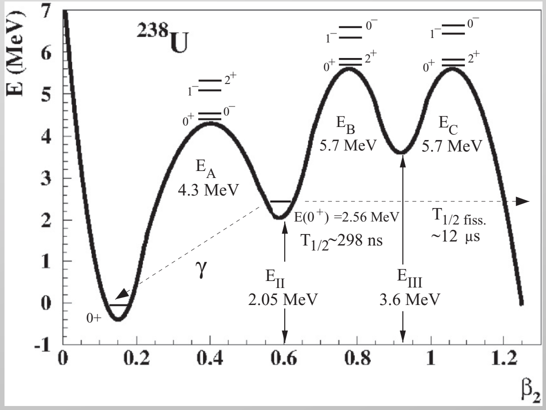
Results – Thorium and Uranium

- Csige *et al* reported experimental evidence for third minima in $^{232,238}U$ – but DFT studies consistently do not predict deep third minima. Why?

UNEDF1 Predicted PESs



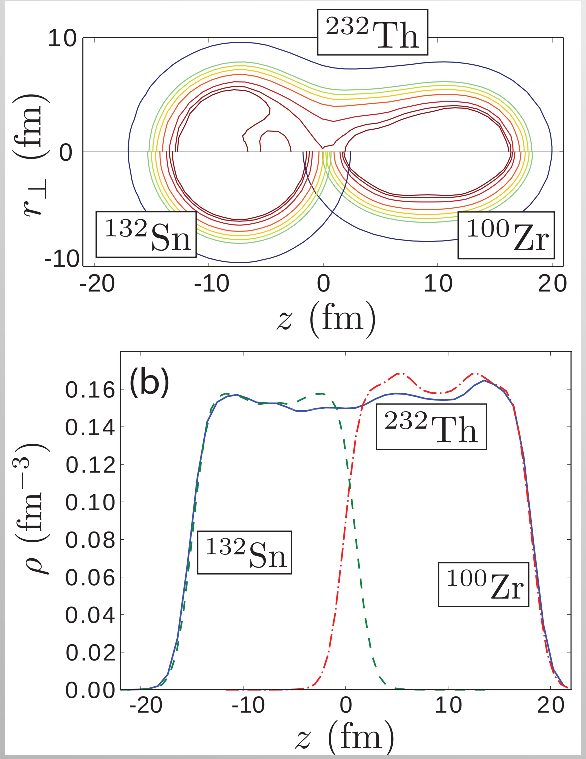
Experimentally Inferred PES, ^{238}U



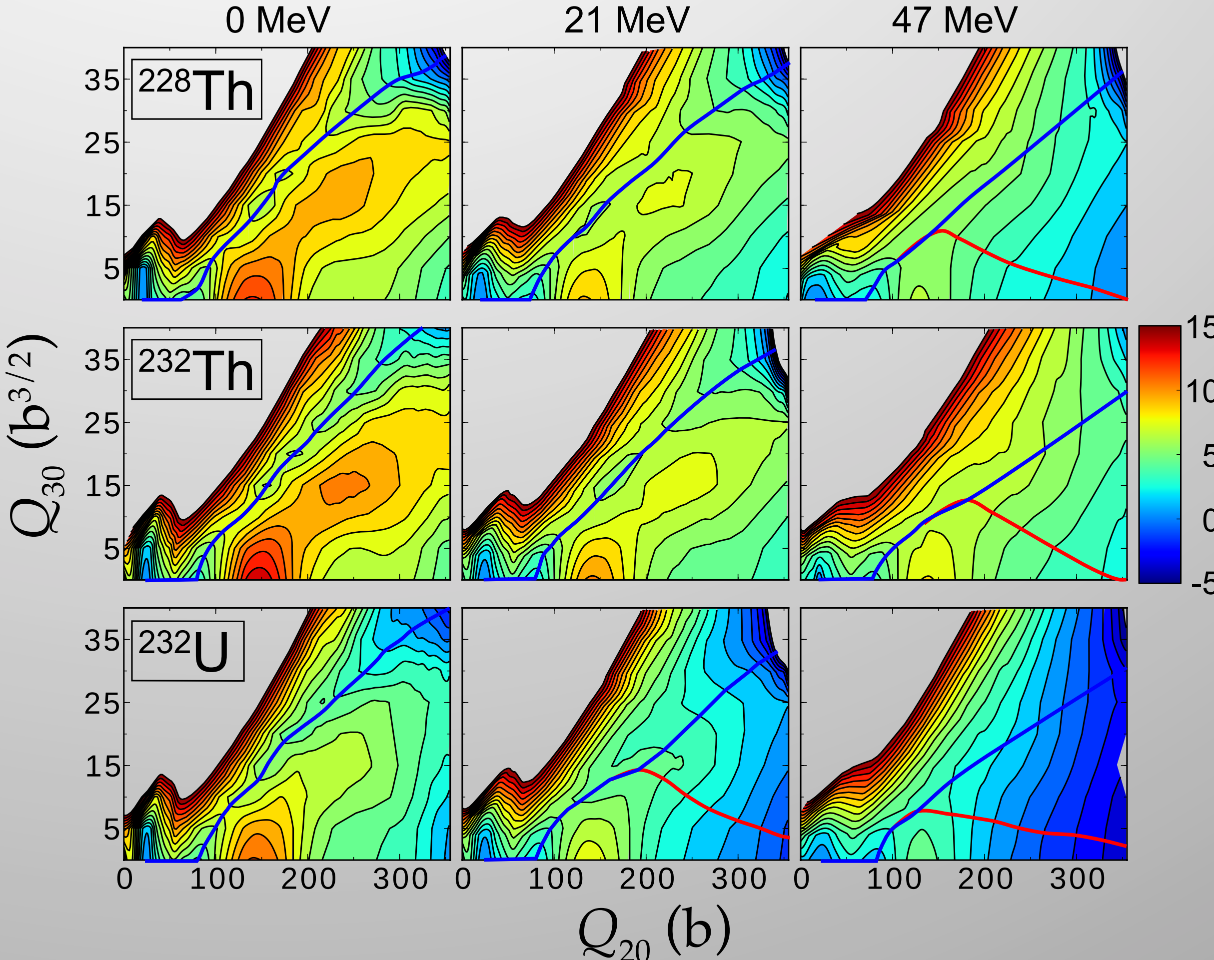
Csige *et al*, *Phys. Rev. C* **87**, 044321 (2013).

Results - Thorium and Uranium

- Despite the lack of a third hump, we do see the formation of well-developed pre-fragments.



At Q_{20} = 165 b.

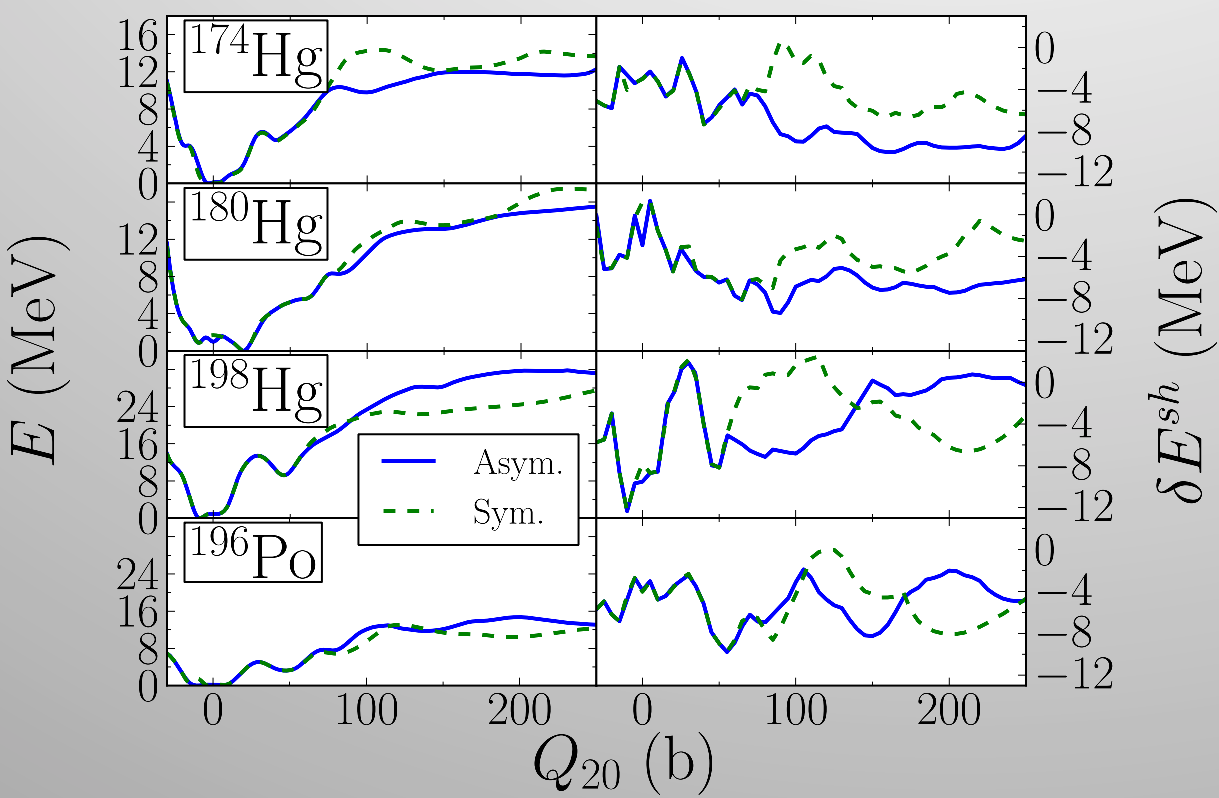


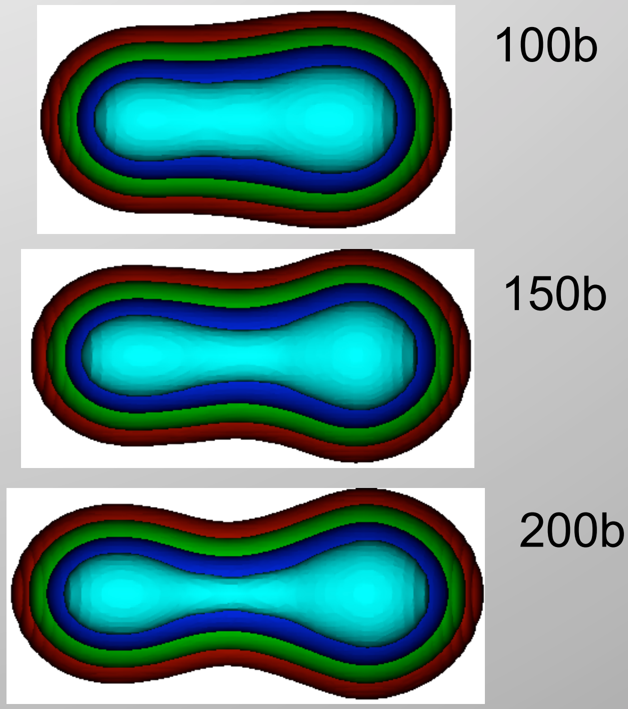
Third Minima – Further Exploration

- Resonances Caused by Richer Level Structure, Rather than Three Humps
  - The di-molecular pictures suggests some stabilizing effect.
  - Explore with Generator Coordinate Method
- A non-phenomenological (non-Skyrme, non-Gogny) EDF Might Yield Three Humps

Results – Mercury Region

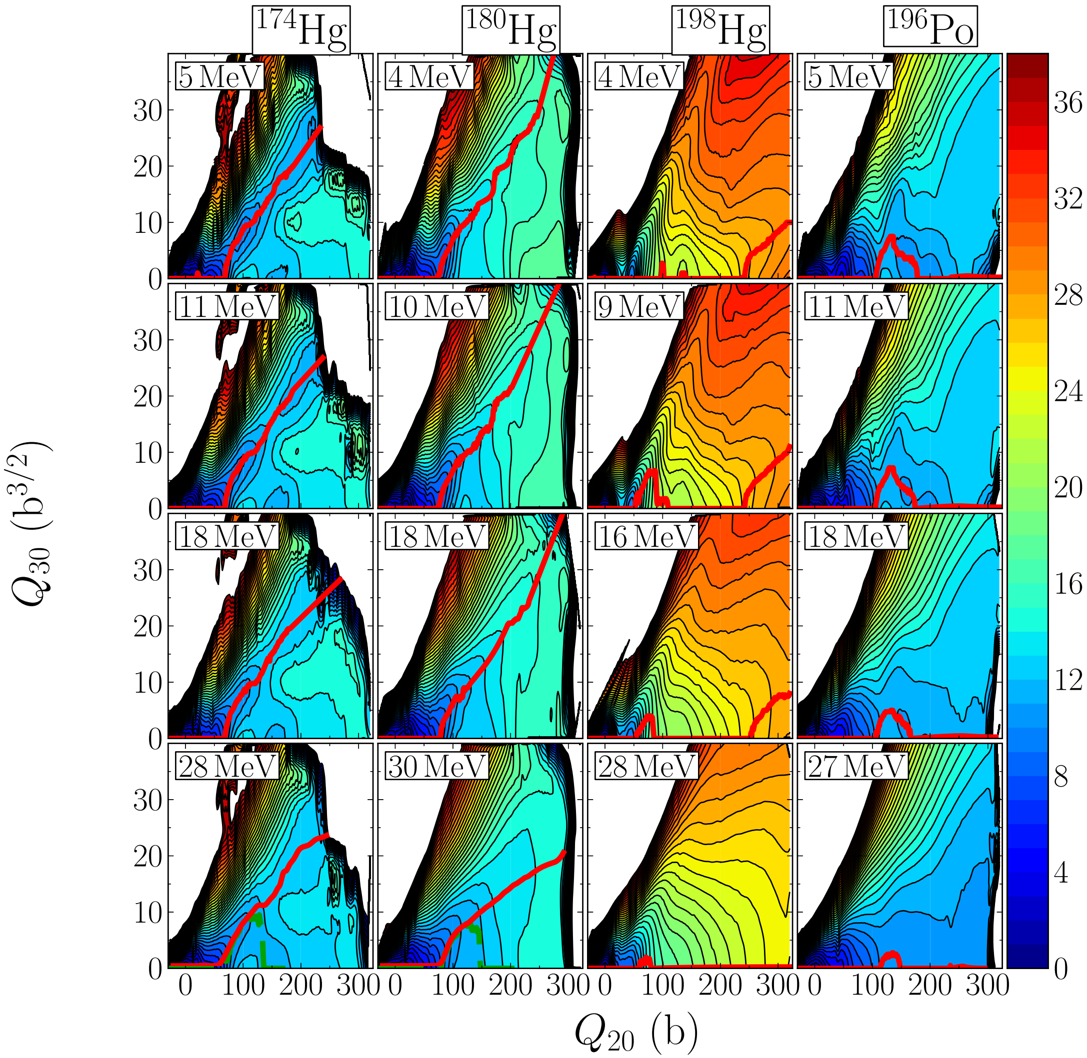
- The asymmetric fission of ^{180}Hg (pictured on right) reveals that "simple" arguments based on magic numbers do not determine fragmentation.





Results - Mercury Region

- Experimentalists discovered asymmetric fission of 180Hg at low energy – what happens at higher energies?
- For ^{174,180}Hg, we see an asymmetry that is robust through E^{*}=0-30MeV.
- For the heavier isotopes, the favored mass distribution is more symmetric.

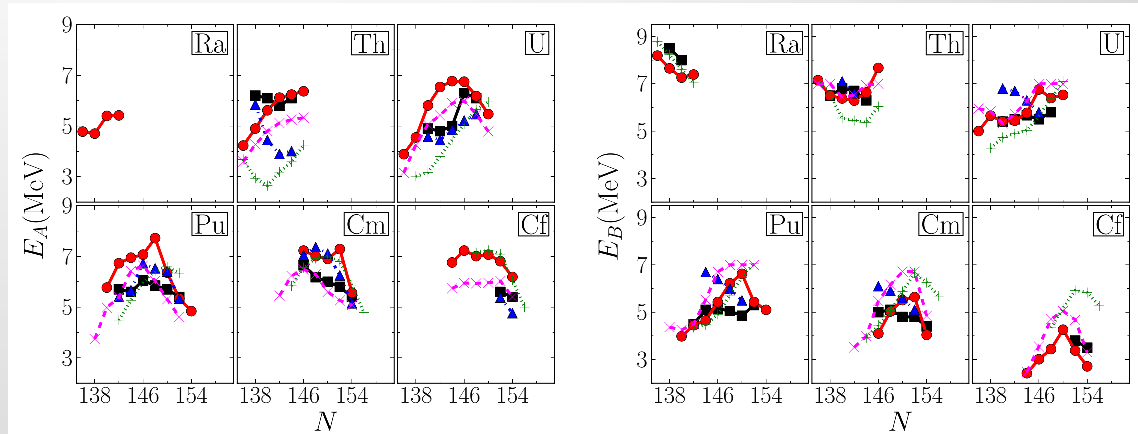


Results – Mercury Region

- This important study of an “unusual” case of fission presents a key test of the predictive power of the DFT approach to fission.
- We turn now to a discussion of uncertainty quantification, to gauge the reliability of our predictions beyond known data.

Uncertainty Quantification – Actinide Fission Barriers

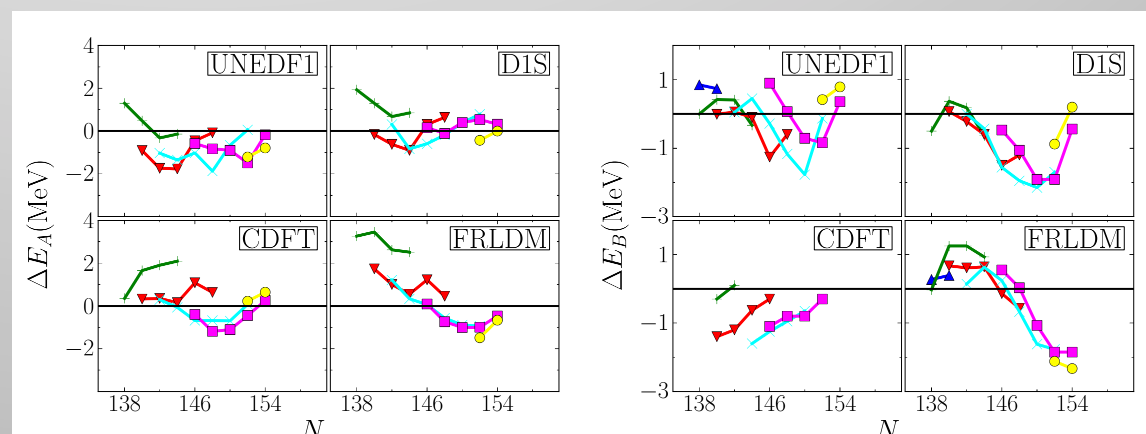
- Many methods in similar ballpark – can we rely on extrapolation?



Red: UNEDF1. Blue: CDFT. Magenta: D1S. Green: FRLDM. Black: Experiment.

RMS Table:

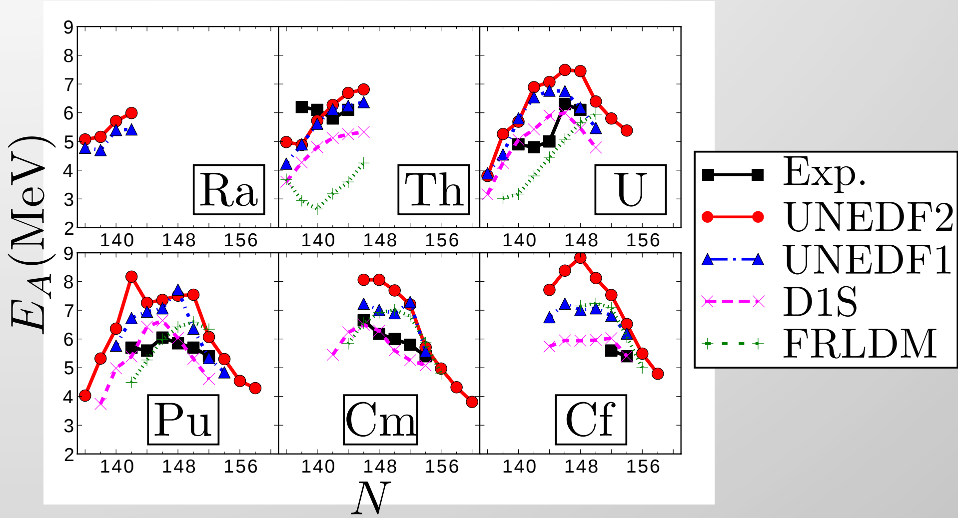
	UNEDF1	CDFT	FRLDM	SkM*	D1S
E_A [MeV]	1.03	0.896	1.52	1.61	0.709
E_{II} [MeV]	0.357	0.977	0.675	0.351	0.339
E_B [MeV]	0.690	0.926	1.13	1.39	1.14



Experiment: RIPL2
FRLDM: Moller et al, 2009
D1S: Delaroche et al, 2006
CDFT: A.V. Afanasjev (private)

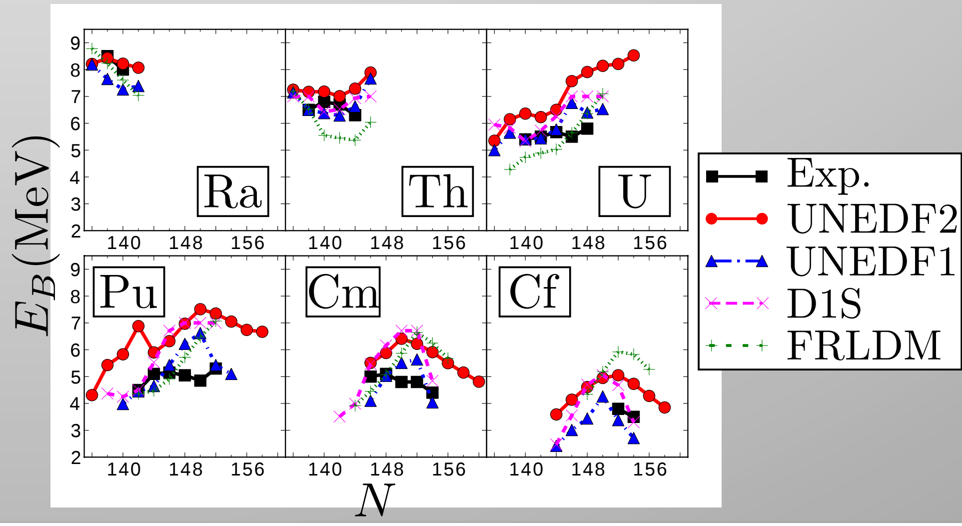
Uncertainty Quantification – Actinide Fission Barriers

- Can we rely on extrapolation? What happens in the UNEDF family?



RMS Table:

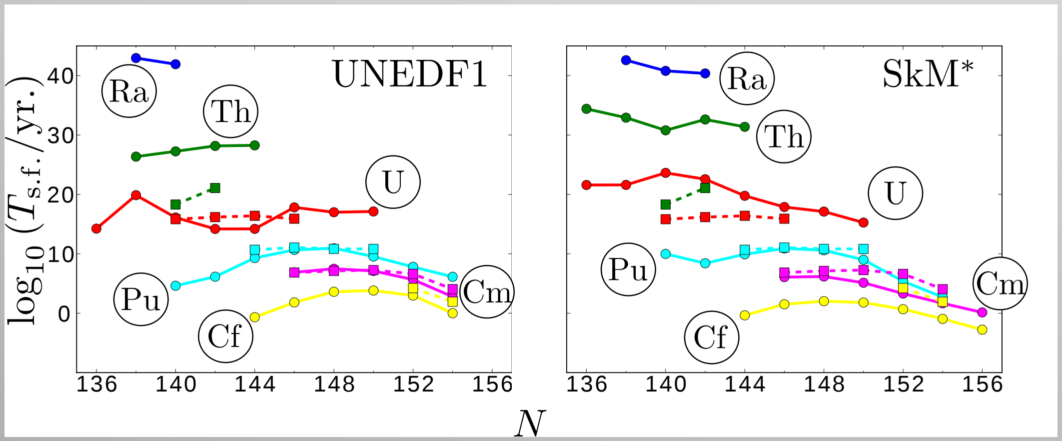
	UNEDF2	UNEDF1	FRLDM	SkM*	D1S
E_A	1.47	1.03	1.52	1.61	0.709
E_{II}	0.515	0.357	0.675	0.351	0.339
E_B	1.39	0.690	1.13	1.39	1.14



Experiment: RIPL2
FRLDM: Moller et al, 2009
D1S: Delaroche et al, 2006

Uncertainty Quantification – Actinide Fission Barriers

- Challenges to Address:
  - For UNEDF1, collective inertia **must** be adjusted to obtain realistic half-lives (this adjustment is not necessary for SkM*)
  - Early Indication: UNEDF2 observables do not agree with data as well as UNEDF1

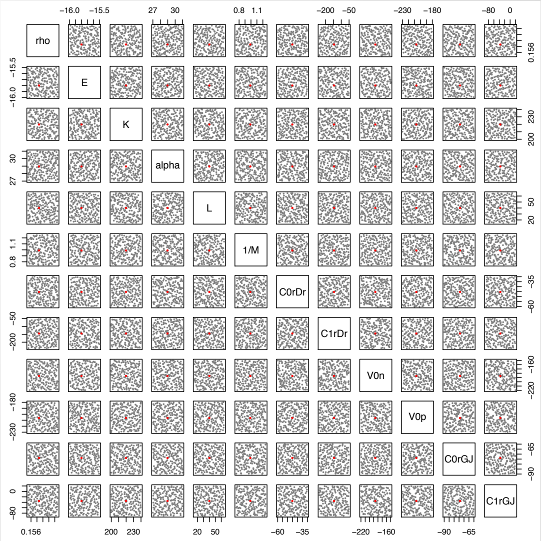


Squares: Experiment (Holden, N. and D. Hoffman, 2000, Pure Appl. Chem. 72, 1525–1562.)
Circles: Theory

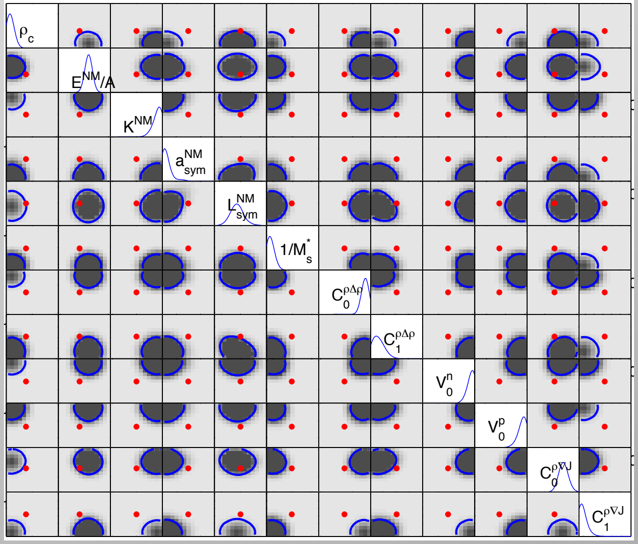
Uncertainty Quantification – Early Results

- Pilot Study Applied to UNEDF1
  - Response surface (based only on mass data) guides us toward new parameter sets to consider
  - Preliminary error bars for our predictions for masses in a newly measured region
- Massively Parallel Approach
  - 200 Test Parameter Sets
  - 130 Nuclei (including deformed nuclei)

Input Parameters

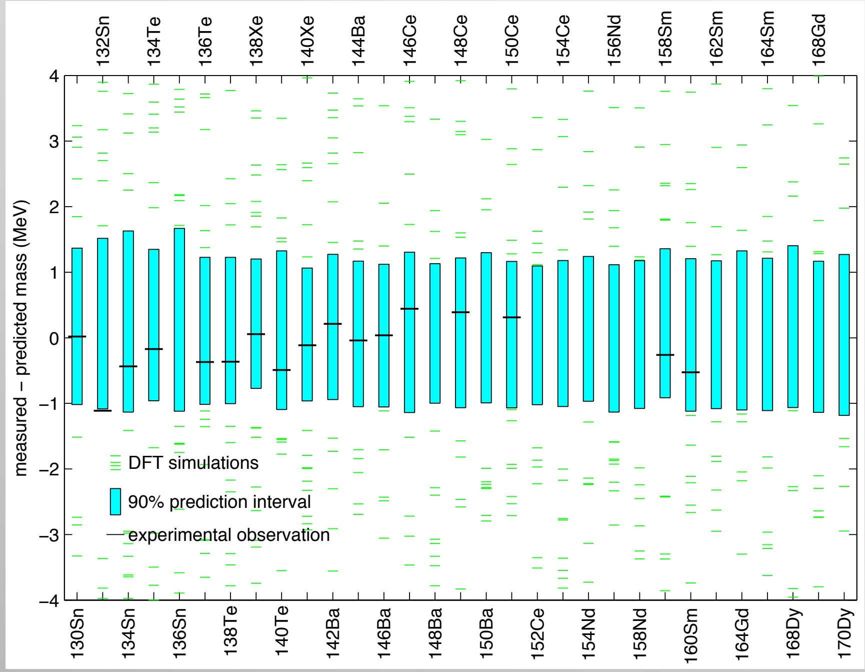


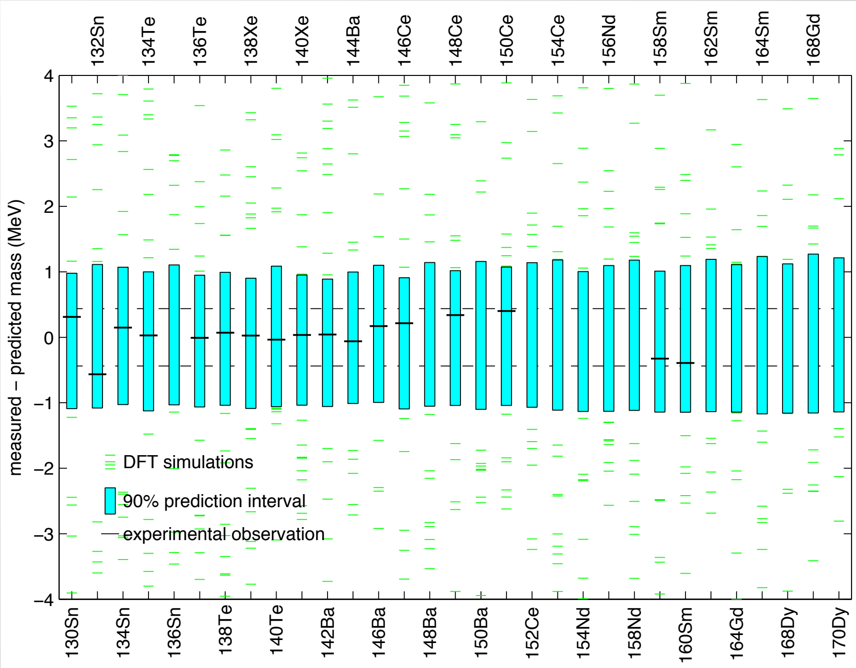
Output Parameter Estimation



Uncertainty Quantification – Early Results

- In the 132Sn region, we see a (small) reduction in uncertainty bars due to mass measurements.





We will study what measurements will have the greatest impact on reducing theoretical error bars for other quantities, such as fission barrier heights.

Conclusion

- We have showcased the capabilities of nuclear DFT for nuclear fission
  - We see a di-molecular configuration in Th and U isotopes, stabilized by something other than a third hump.
  - We predict that symmetry vs. asymmetry of mass yields in the mercury region has a weak dependence on excitation energy.
- We have demonstrated a viable approach for uncertainty quantification for EDFs

Collaborators

- LLNL: N. Schunck
- UTK: W. Nazarewicz, J.A. Sheikh
- LANL: D. Higdon
- Lublin: A. Baran, A. Staszczak, M. Warda
- ANL: J. Sarich, S. Wild