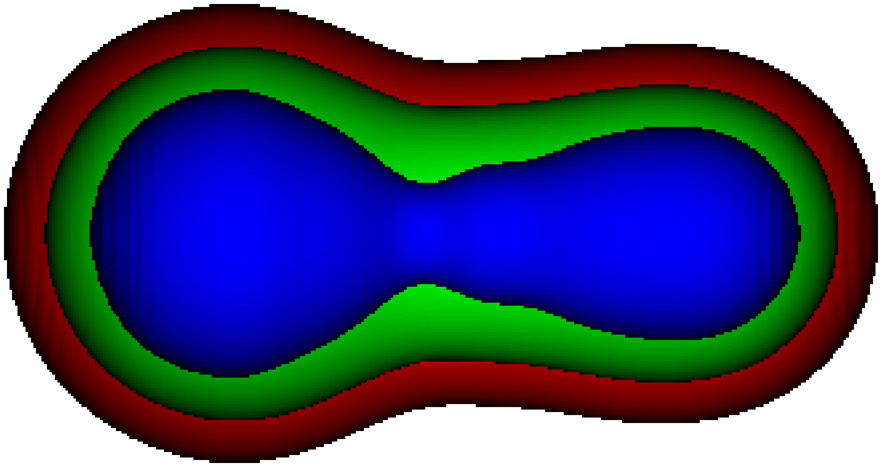
Towards a Microscopic Theory of Nuclear Fission
With New Nuclear Density Functionals



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Stewardship Science Graduate Fellowship Conference 2012
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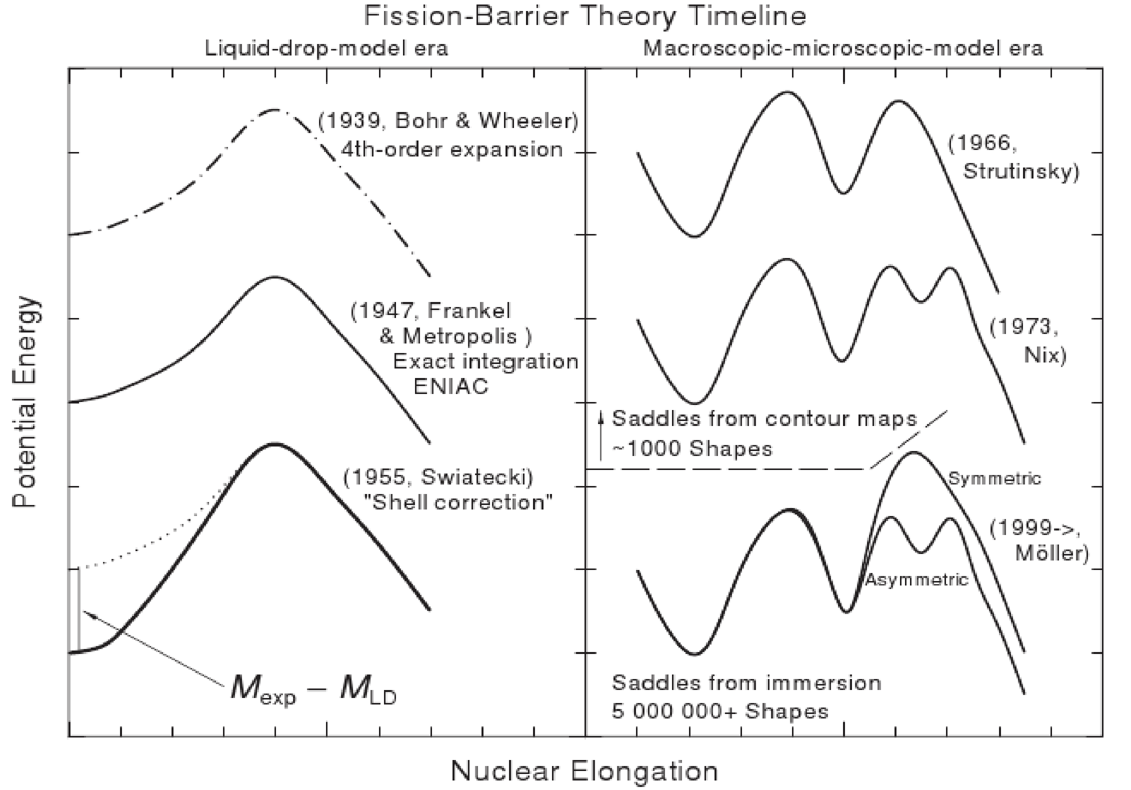
State of Fission Theory

-Fission has been thoroughly studied experimentally and phenomenologically since 1939.
Original Paper:
-O. Hahn and F. Strassmann, Naturwissenschaften 27, 11 (1939).
Review articles:
-J.R. Nix, Nucl. Phys. A 502, 609c (1989).
-S. Bjørnholm and J.E. Lynn, Rev. Mod. Phys. 52, 725 (1980).

-Macroscopic-microscopic methods reproduce fission quantities well – but are their predictions reliable?

-Great strides have been made with density functional theory (DFT) in describing nuclear structure...

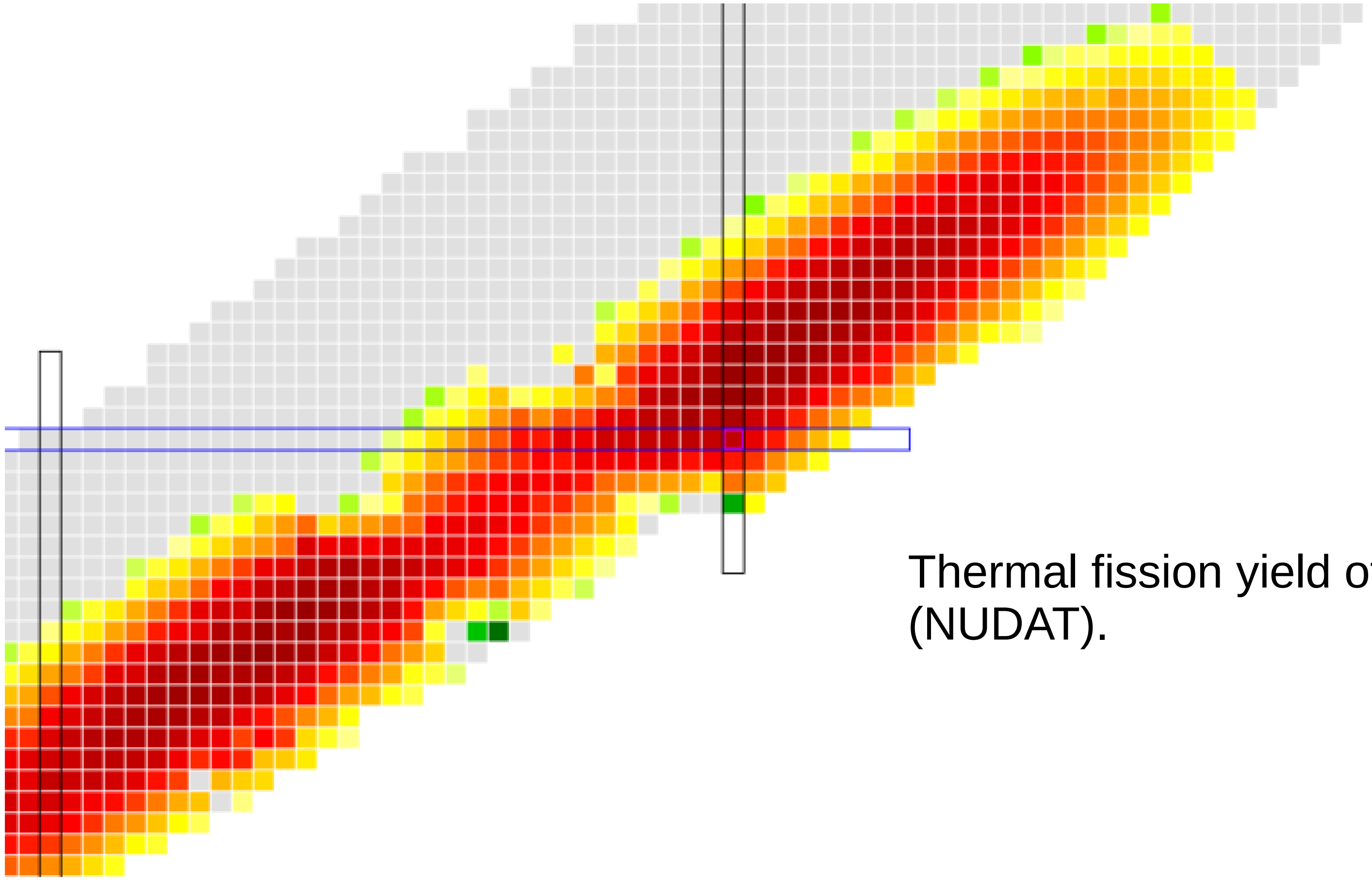


Möller *et al*, Phys. Rev. C **79**, 064304 (2009)

How well does a fully microscopic theory of fission, based on DFT, perform where data are known? Can we be confident in that theory's predictions for new situations?

Outline

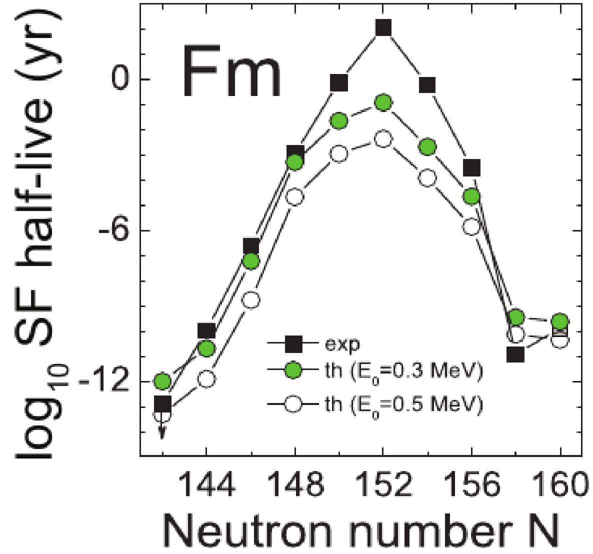
- Review of DFT for Fission
- Third Minima in Thorium and Uranium Isotopes
- The Fission of Mercury Isotopes
- Survey of Spontaneous Fission in the Actinides

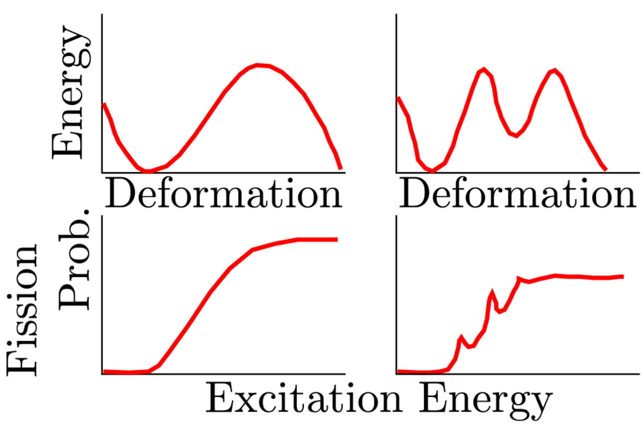


Thermal fission yield of 239Pu (NUDAT).

Motivation

- Why work towards a microscopic theory of fission?
  - Predictive power needed for:
    - Superheavy Elements
    - r-process Elements
  - Defense and energy applications
    - Understanding nuclei in extreme conditions
- A Universal Nuclear Energy Density Functional (UNEDF)
- Spectroscopy of Hyperdeformed Isomers in Actinides

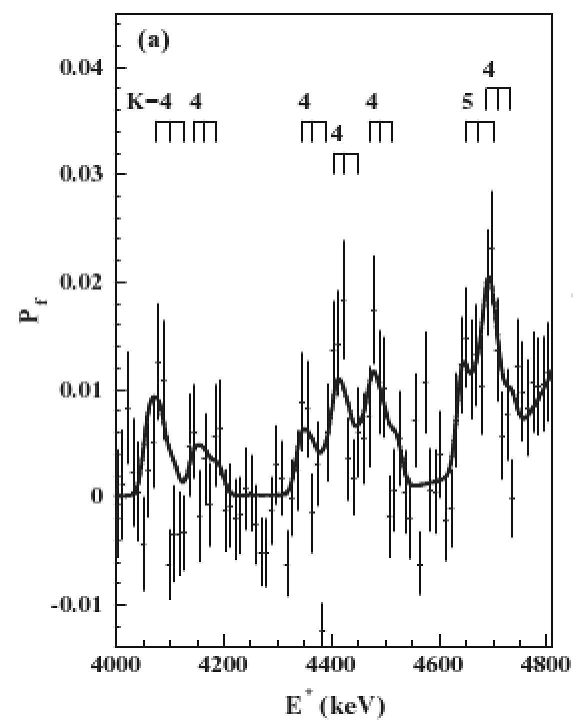




(Top) Predicted half-lives for fermium isotopes
(from Staszczak (2009)).

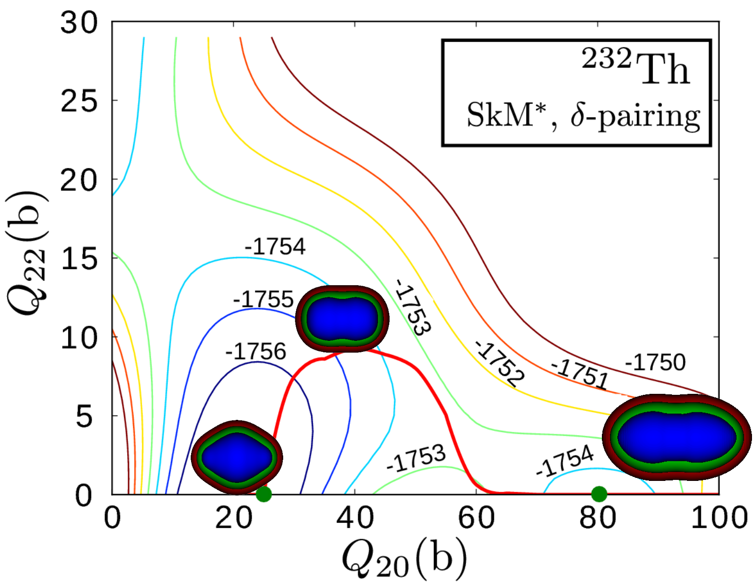
(Left) Fission probabilities yield clues about nuclear structure (from Bjornholm and Lynn (1980)).

(Right) Fission probability for uranium-232, from which Csige *et al* (2009)
infer a third potential well.



Review of DFT for Fission

For spontaneous fission, we explore a potential energy surface (potential energy as a function of nuclear configuration) to map out the barrier through which a nucleus tunnels to fission.



Elongation:

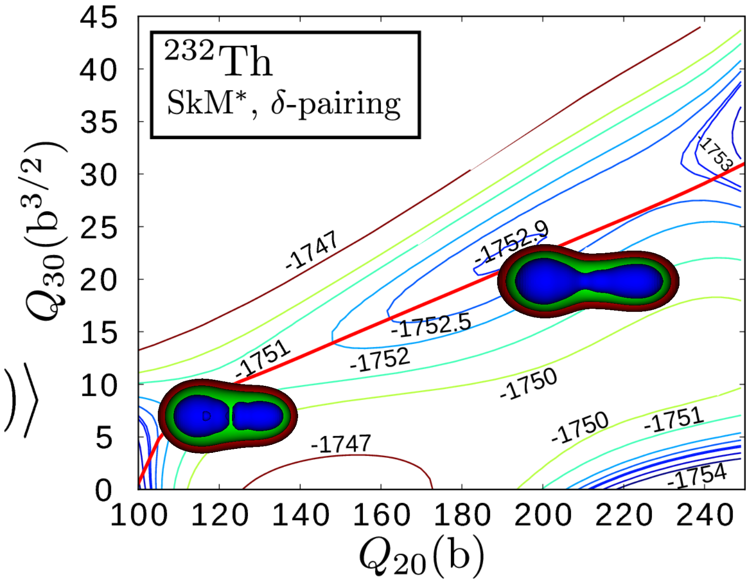
$$Q_{20} \propto \langle r^2 Y_{20} \rangle$$

Triaxiality:

$$Q_{22} \propto \langle r^2 (Y_{22} + Y_{2-2}) \rangle$$

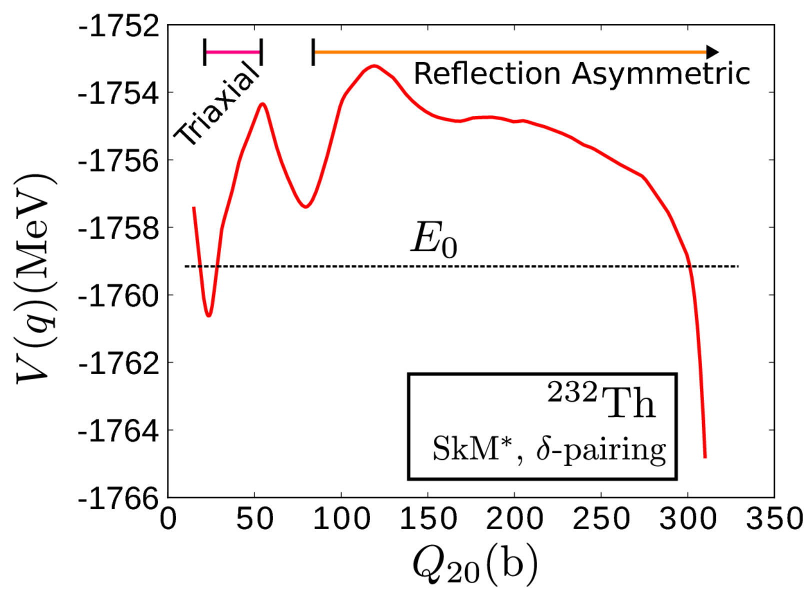
Refl. Asym.:

$$Q_{30} \propto \langle r^3 Y_{30} \rangle$$



•A typical actinide nucleus tends to break axial symmetry near the first saddle point, and it tends to break reflection symmetry between the second isomer and scission. The exploration of many degrees of freedom is crucial for an accurate calculation of action (important for half-life estimates).

$$\text{Action} = \int_{(s)} \left\{ 2[V(q) - E_0] \sum_{ij} \mathcal{M}_{ij}(q) \frac{dq_i}{ds} \frac{dq_j}{ds} \right\}^{1/2} ds$$



Finite-temperature DFT for Excited Nuclei

- Finite-Temperature Hartree-Fock-Bogoliubov (FT-HFB) theory allows us to consider compound nuclei formed with some excitation energy
  - A self-consistent mean field equation with pairing correlations
  - Fission is believed to be an isentropic process. Curves of free energy (F=E-TS) at constant temperature correspond exactly to curves of internal energy at constant entropy...

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

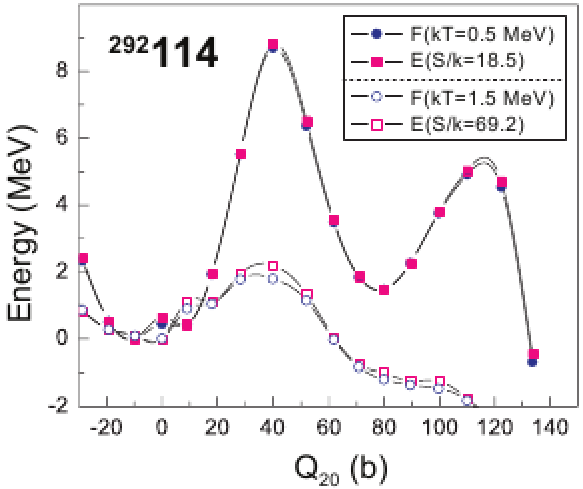
$$h = \epsilon + \Gamma$$

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

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

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

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



Numerical verification of correspondence between isentropic internal energy curves and isothermal free energy curves. From Pei *et al* (2009).

Computational Methods

HFODD – symmetry-unrestricted, iterative-diagonalization solution of the Hartree-Fock-Bogoliubov equations

* N. Schunck *et al*. *Comp. Phys. Comm*. **183**, 166-192 (2012).

A. Nuclear Energy Density Functionals
   1. Skyrme(-like) functionals in particle-hole channel: SkM* and UNEDF1 focused on here
   2. HFB and Lipkin-Nogami for pairing; Density-dependent delta pairing

B. One-center, Cartesian harmonic oscillator basis
   1. 31 major shells with a deformed basis, to cover full range of shapes
   2. Simplex symmetry imposed, but calculations break axial and reflection symmetry.

C. Massively parallel computing platform
   1. MPI distributes potential energy surface calculations
   2. Each configuration calculated with OpenMP, threaded LAPACK

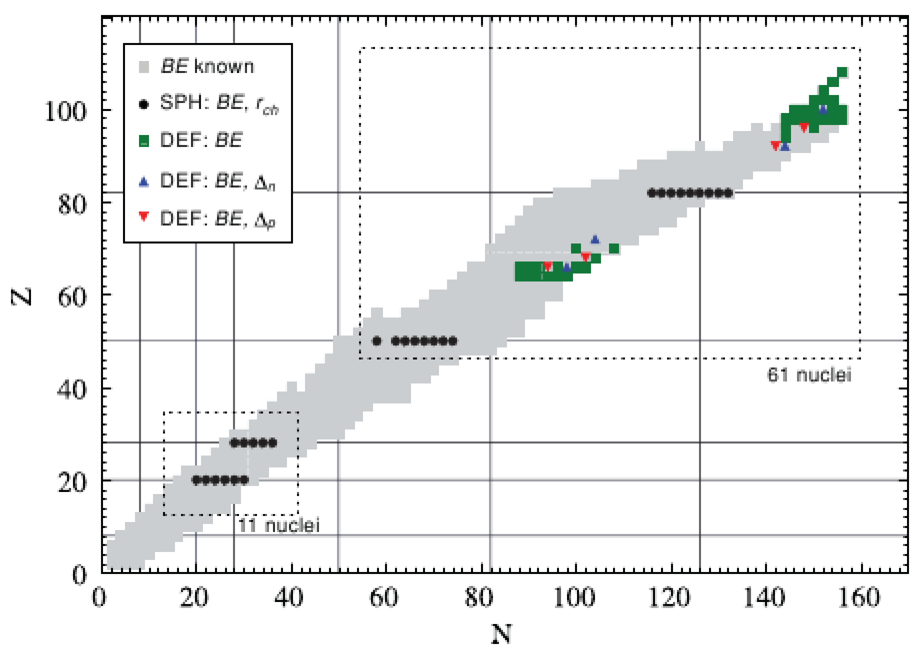
Advances in DFT: UNEDF

- A Skyrme-like energy density functional optimized for studies of fission for actinide nuclei (M. Kortelainen *et al*, PRC 85, 024304 (2012))

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

$$\mathcal{E}_{\text{pair}}(r) = \sum_{q=n,p} \frac{V_0^q}{2}\left\{1 - \frac{\rho_0(r)}{2\rho_c}\right\}\tilde{\rho}^2(r)$$

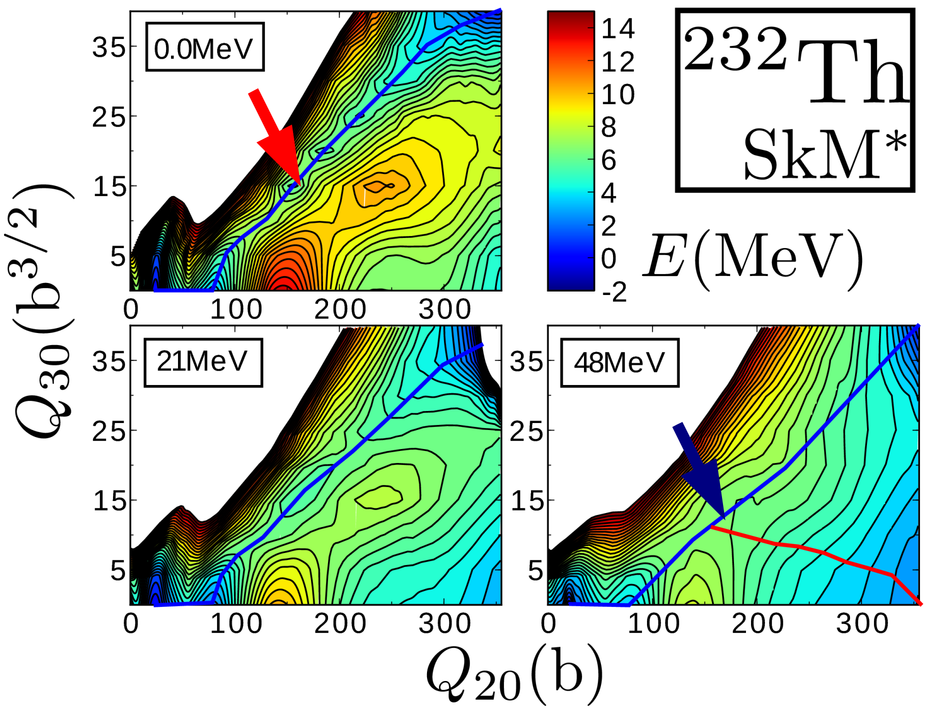
  - Coupling constants optimized to observables (binding energies, radii...) for 72 nuclei (UNEDF0)
  - Additional optimization to data for super-deformed isomers of 236U, 238U, 240Pu, 242Cm (UNEDF1)

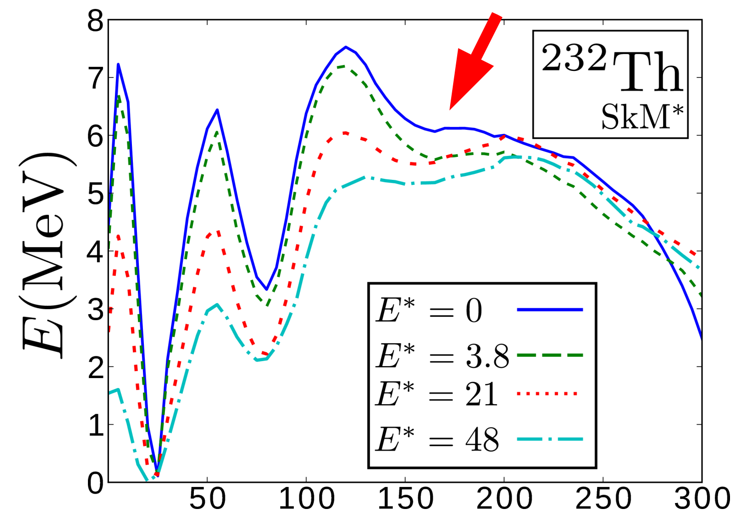


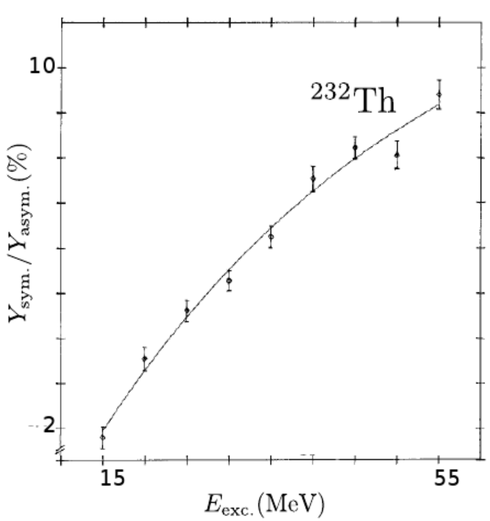
M. Kortelainen *et al*, PRC 82, 024313 (2010).

Third Minima in Thorium and Uranium Isotopes

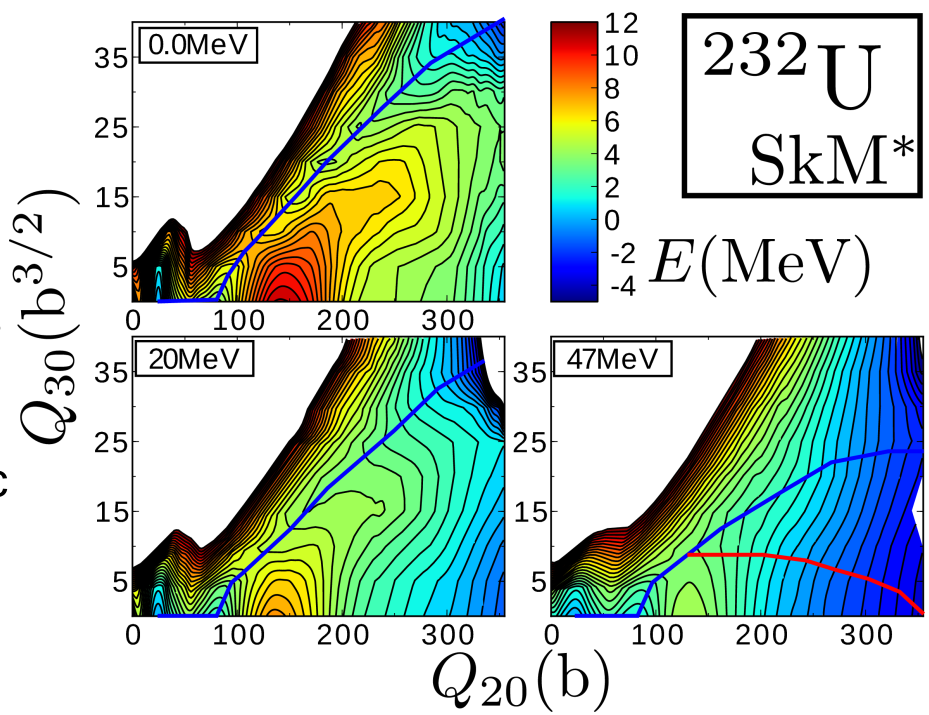
- Do the major actinides possess third minima? Macroscopic-microscopic methods predict a deep third minimum, while DFT predicts a shallower minimum. What physical conditions are favorable for a deep third minimum?





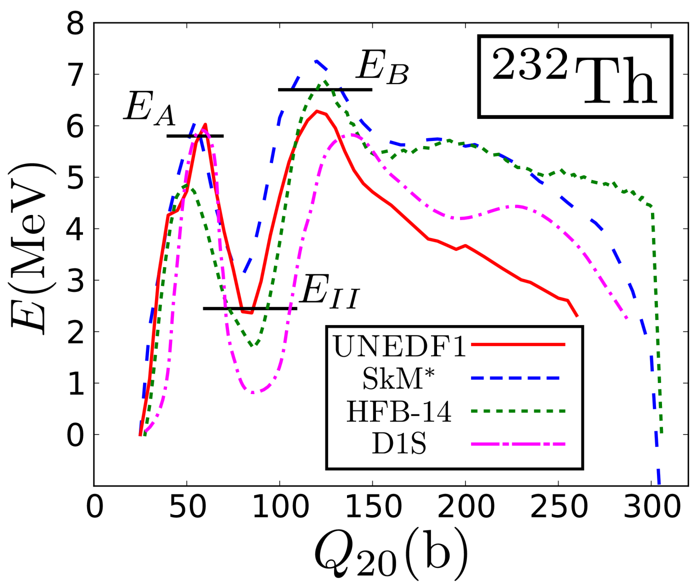


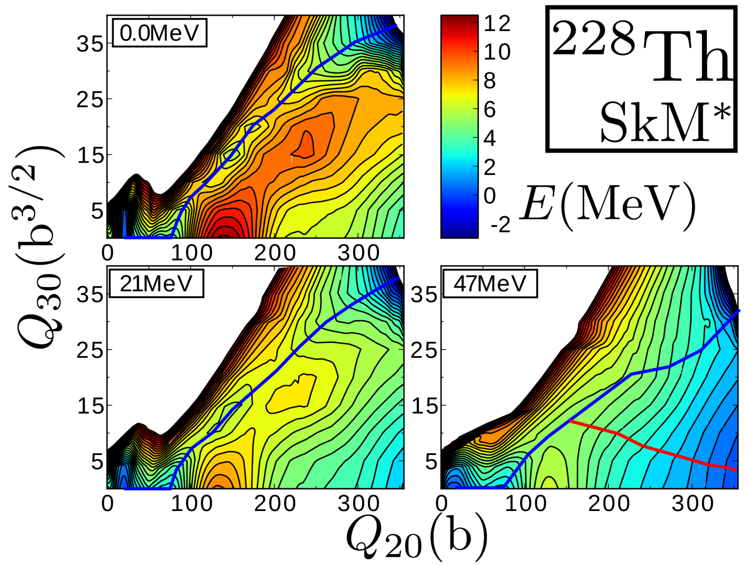
Above and right: Isentropic fission paths for different values of excitation energy. Left: Ratio of symmetric to asymmetric fission mass yield vs. increasing excitation energy (from Gunther (1980)).

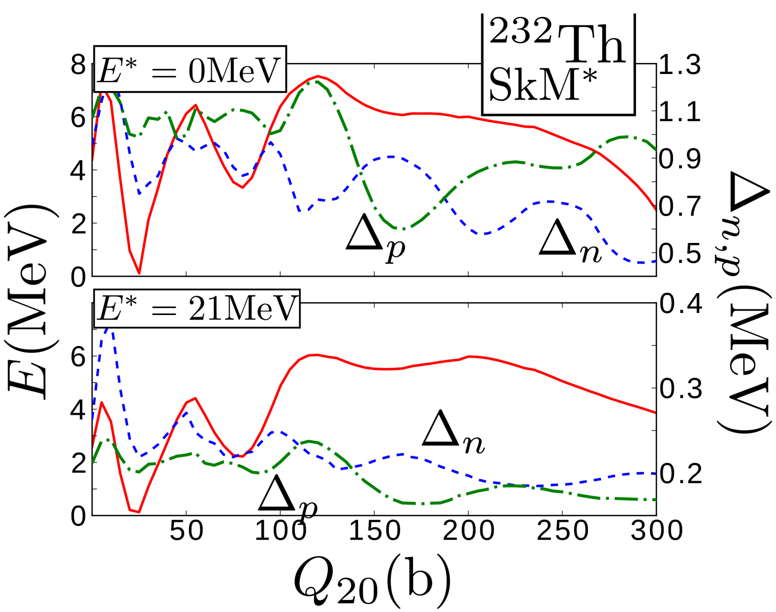


Third Minima in Thorium and Uranium Isotopes

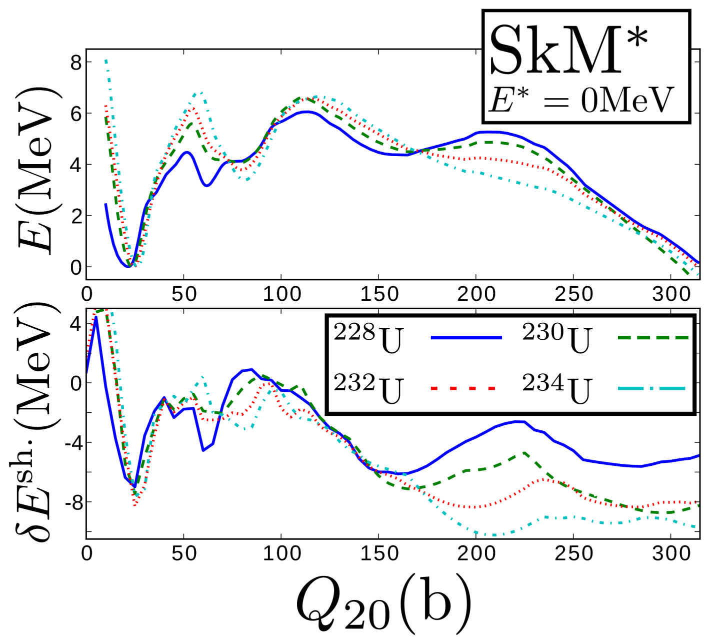
DFT theories consistently predict a shallow third minimum for 232Th and 232U. Lighter isotopes, such as 228Th, are predicted to possess a deeper third minimum.





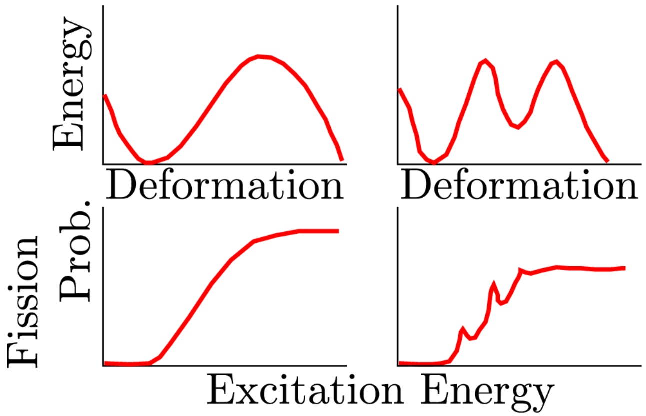


The evolution of the third minimum with excitation energy and neutron number indicates a delicate balance between pairing and shell effects.



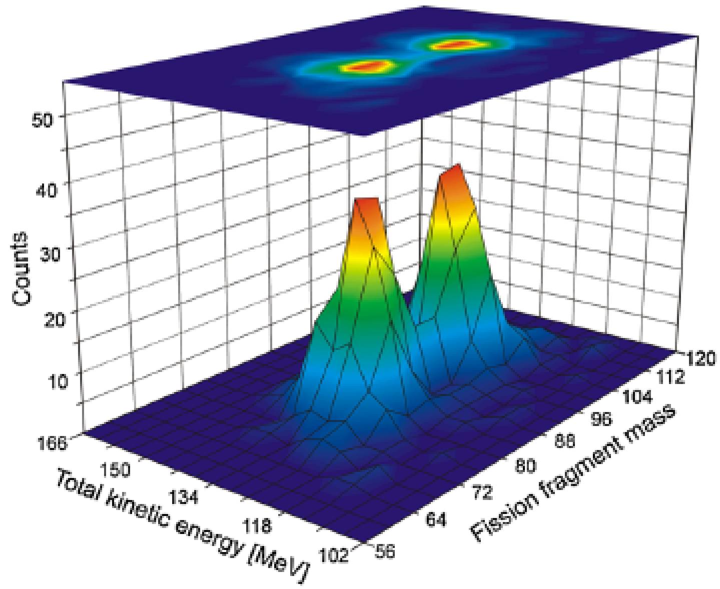
Third Minima in Thorium and Uranium Isotopes

- DFT consistently predicts a shallow third minimum in 232Th and 232U – does that mean DFT is hopeless for fission studies?
  - In fact, several model-dependent assumptions enter the experimental analyses that infer third minima. The analysis of the experimental resonance structures is complicated... (Mirea *et al* (2007), Blokhin and Soldatov (2009)).
  - If an experiment confirmed 100% that the third minima are deep, it would provide a constraint on the optimization of future energy density functionals. This work's analysis of the conditions that favor deeper third minima should prove beneficial.

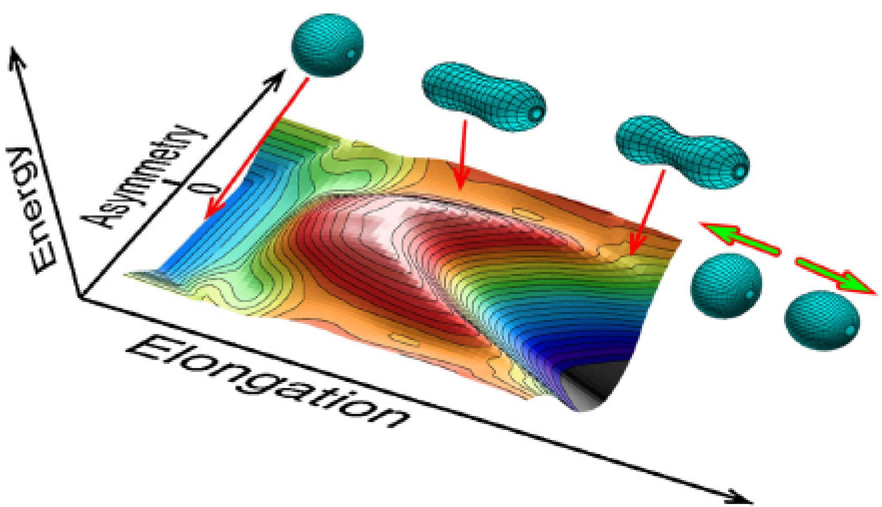


From Bjornholm and Lynn (1980).

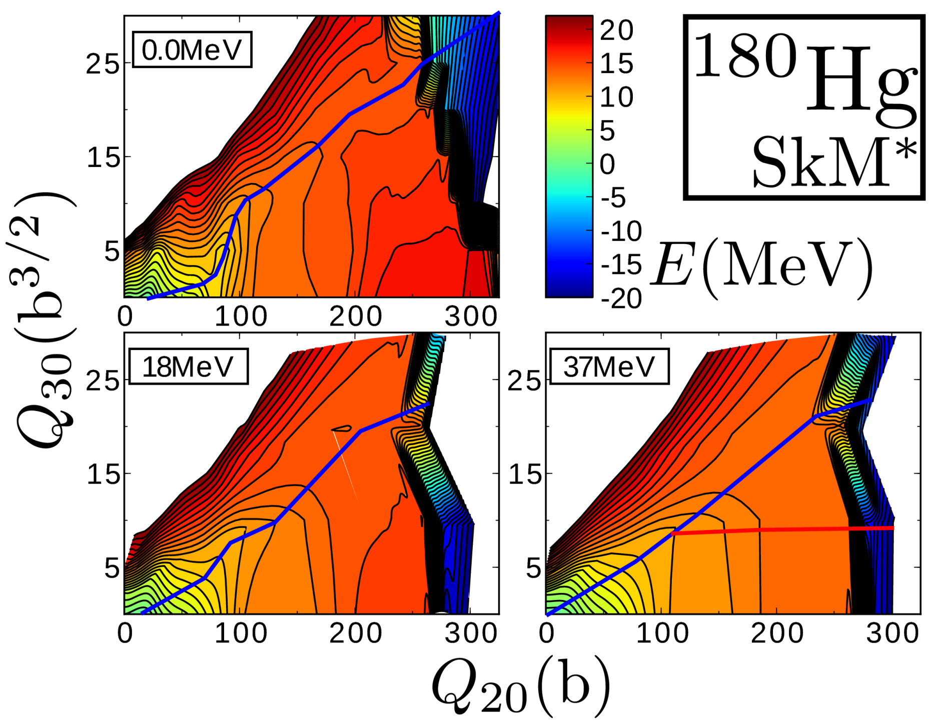
Asymmetric Fission for Mercury Isotopes



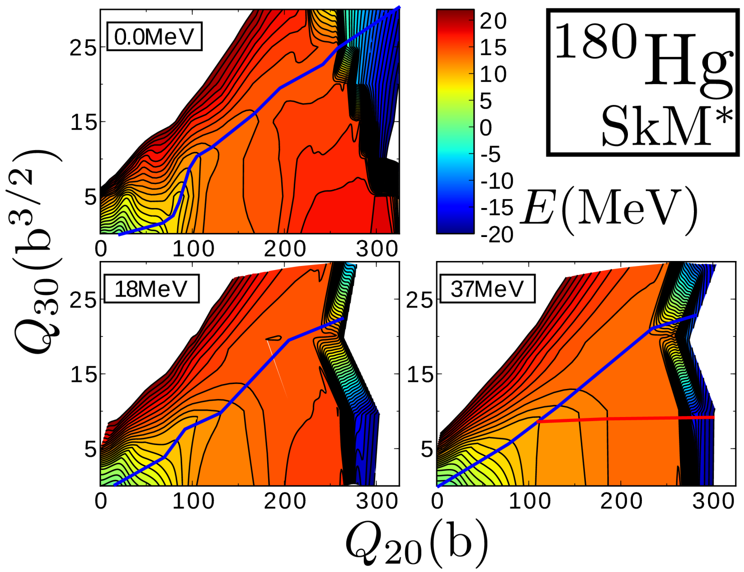
A recent experiment by Andreyev *et al* (2010) obtained previously unseen asymmetric mass fragments from the fission of proton-rich 180Hg. Reasoning purely by the shell structure of the fragments leads one to expect symmetric fission into semi-magic 90Zr. Zero-temperature calculations reveal the mass asymmetry found in the Andreyev experiment.



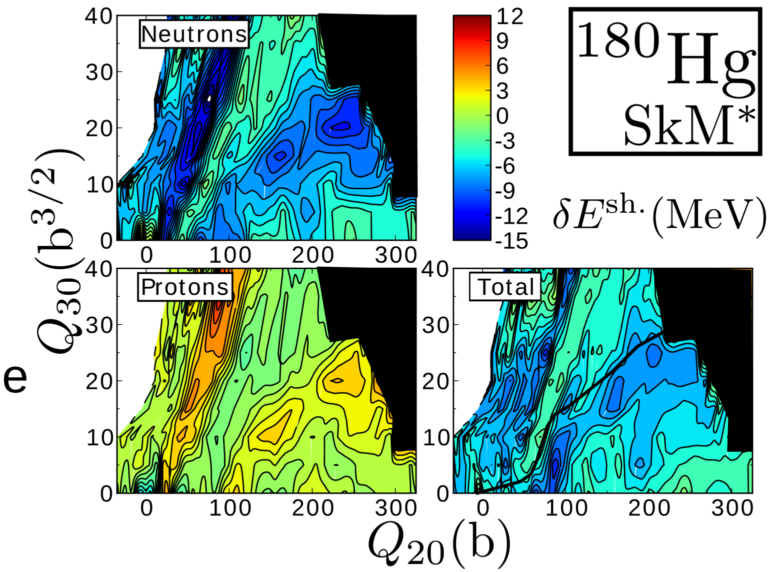
The above plot was generated by Moller with FRLDM for 180Hg; the plot to the right is our result with SkM*. Note the strong preference for asymmetry at zero temperature.

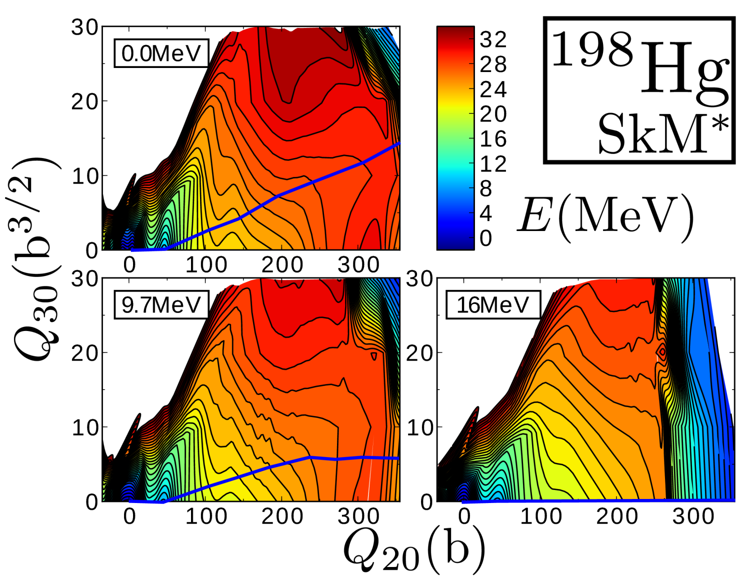


Asymmetric Fission for Mercury Isotopes

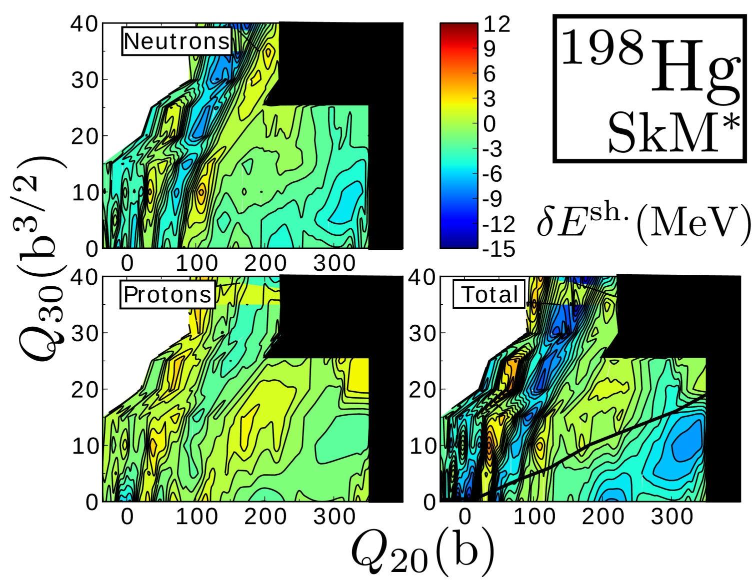


The asymmetric fission of 180Hg persists to rather high excitation energy. It can be seen that shell corrections drive the asymmetric fission mode.





The fission of 198Hg, however, does not prefer asymmetry as strongly. This also is reflected in the shell correction energies.



Asymmetric Fission in the Mercury Isotopes

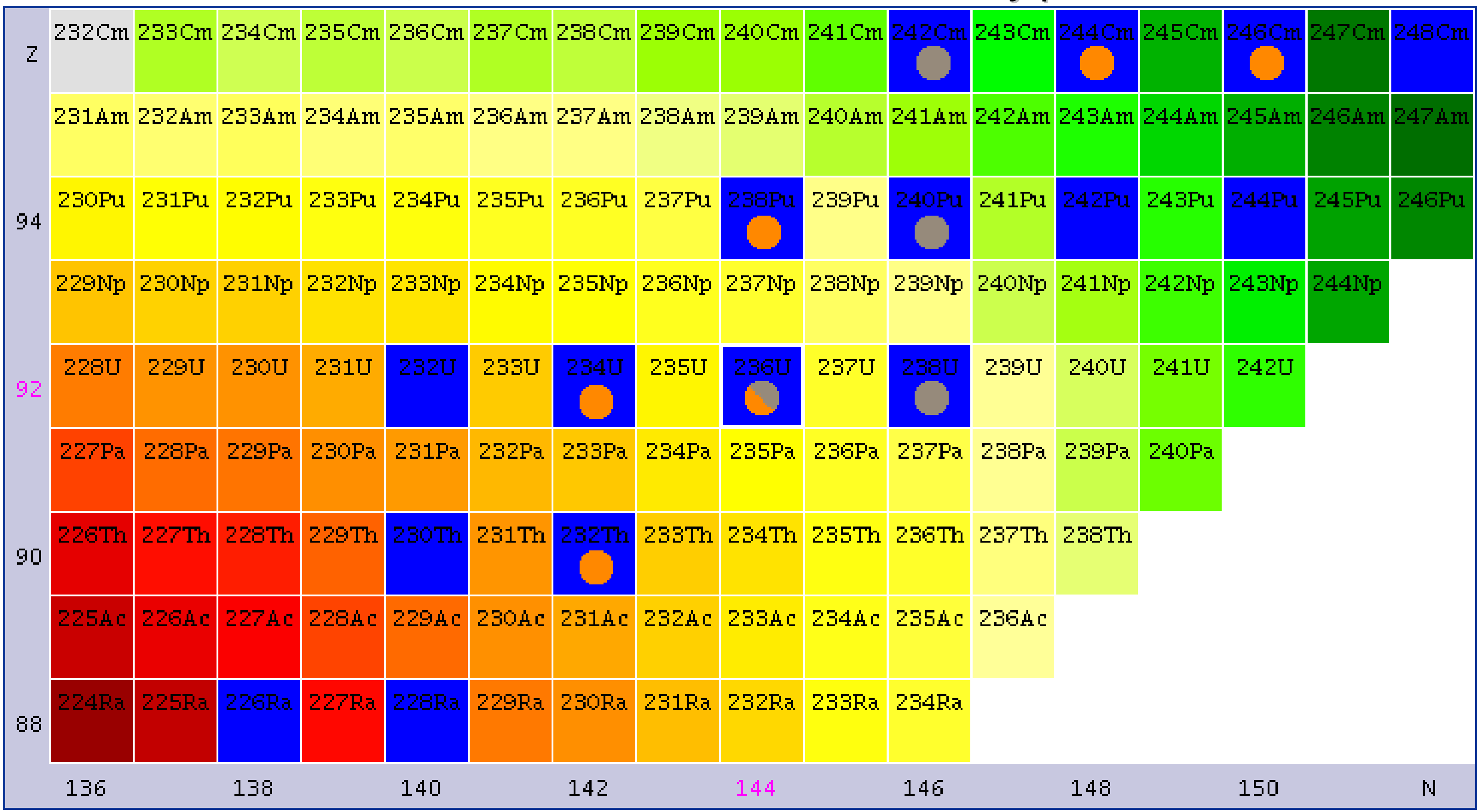
- This important study of an “unusual” case of fission helps to affirm the predictive power of the DFT approach to fission.

- With the DFT approach, we have the tools to study the microscopic physics that drive 180Hg to asymmetric fission and 198Hg to symmetric fission.

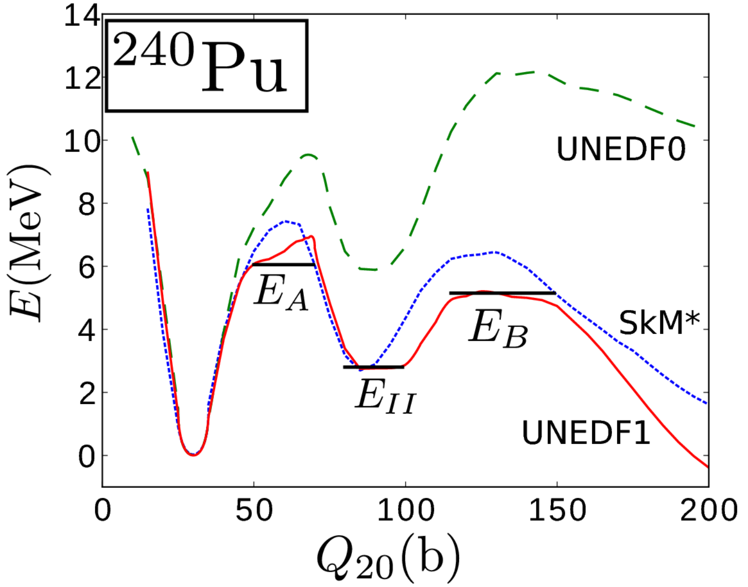
Survey of Spontaneous Fission in the Actinides

- Having studied a few specific cases in depth, we turn to evaluate the performance of nuclear DFT for predicting spontaneous fission observables for several even-even major and minor actinides.

- The UNEDF optimization procedure includes observables such as binding energies, RMS radiii, and pairing gaps (odd-even mass differences). **No** fission data are fit, so that these calculations are truly predictive.

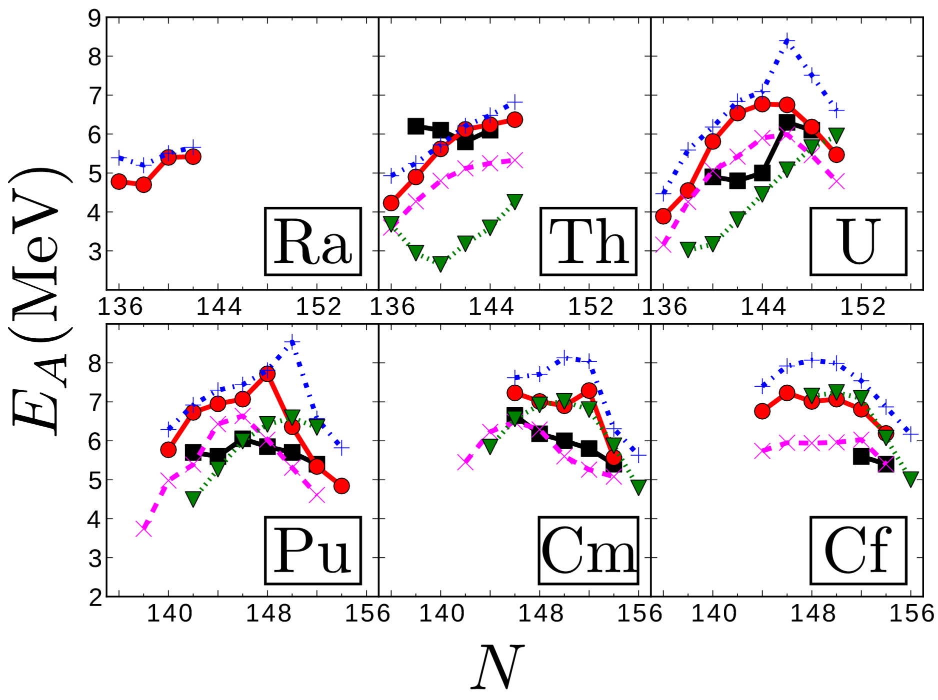


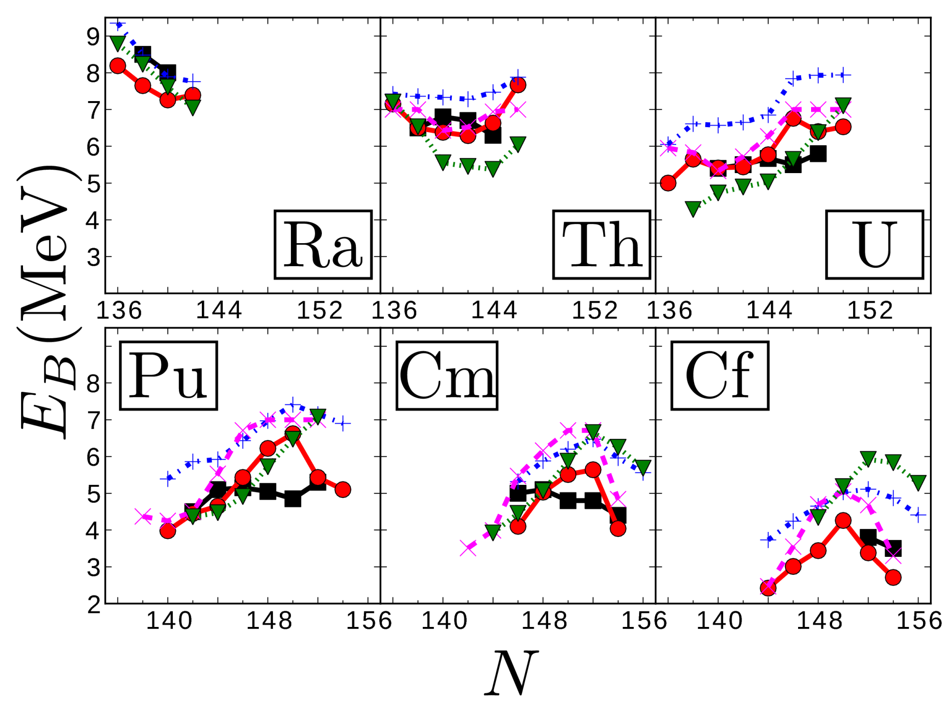
Survey of Spontaneous Fission in the Actinides



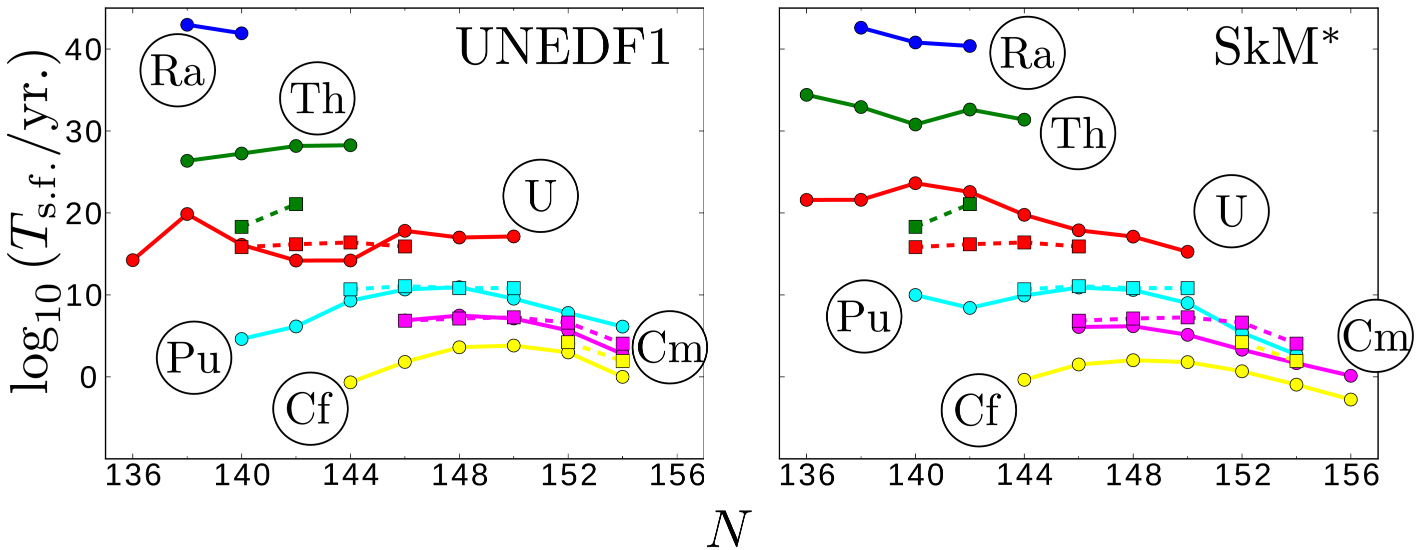
Compared to empirical fission barrier properties, the new UNEDF1 functional performs competitively with the best approaches to fission.

Below, the heights of the first and second fission barriers are plotted for a subset of actinides. UNEDF1 (red) compares well with the RIPL evaluated barrier heights (black), as well as the FRLDM (green) model of the Los Alamos group. Also plotted: SkM* (blue) and D1S (magenta).



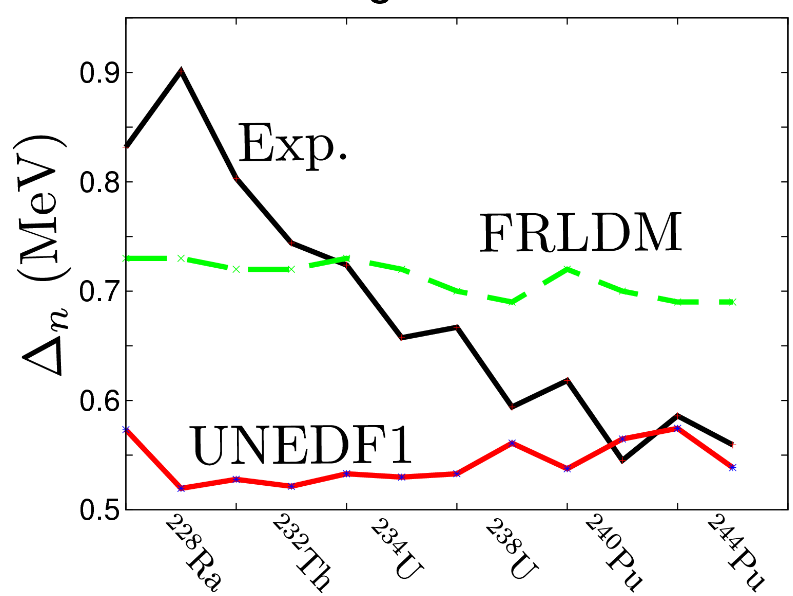


Guiding the Optimization of the Functional



With a simple correction for collective inertias, the spontaneous fission half-lives are reproduced remarkably well across the actinides studied here. The adiabatic time-dependent HFB theory captures fission dynamics well, and is a powerful tool to be coupled with next-generation functionals.

It has been found that the pairing gaps calculated by UNEDF1 are too low, which affects collective inertias and therefore SF half-lives. For the next iteration of UNEDF fits, pairing data for 232Th will be included. The agreement with experimental quantities is already excellent, and we expect an improvement in pairing properties to improve this functional's predictive power overall.



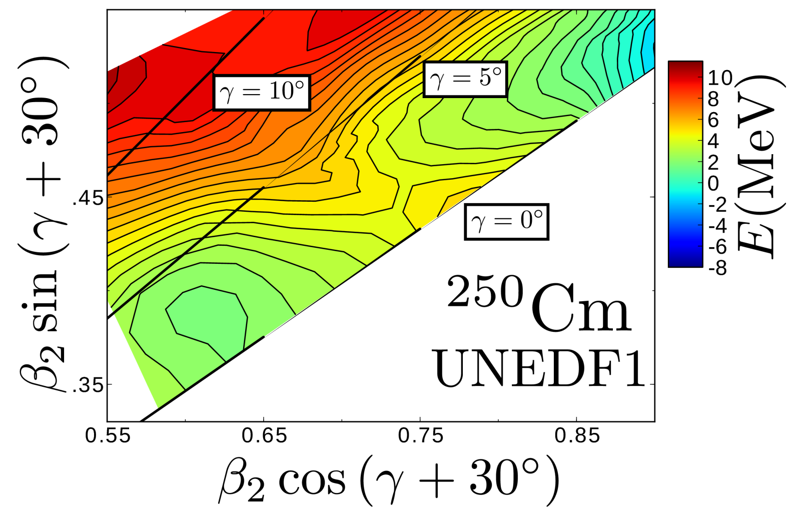
Conclusion

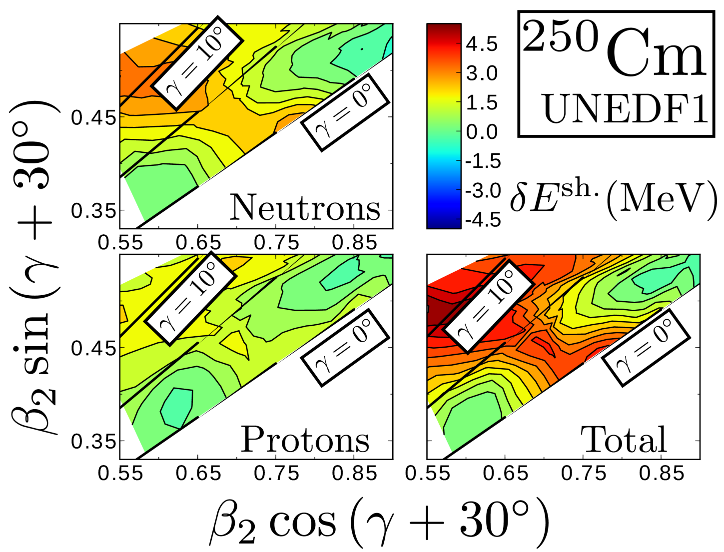
- Ongoing work towards a predictive theory of fission via DFT is motivated by the agreement of current predictions for actinides with known experimental data
  - Why does DFT predict such shallow third minima for 232Th and 232U? The depth of the third minimum is very sensitive to the balance between pairing and shell effects.
  - DFT predicts an asymmetric mass yield for 180Hg for substantial excitation energies; shell effects drive 180Hg to asymmetric fission, but they favor symmetric fission for 198Hg.
  - Trends in fission barrier heights and spontaneous fission half-lives are reproduced remarkably by the new UNEDF1
- This microscopic model yields results of a fidelity competitive with the best macroscopic-microscopic methods. Since the microscopic model of the nuclear force is rooted in the properties of nuclear matter itself, we have reason to be confident in the predictions of this model beyond known data.

- Thank you:
  - This work has been funded by the DOE/NNSA Stewardship Science Graduate Fellowship through the Krell Institute.
  - W. Nazarewicz, C. Bingham, R. Grzywacz, R. Harrison, T. Papenbrock
  - A. Baran, H. Nam, N. Schunck, J.A. Sheikh, A. Staszczak
  - Useful discussions with N. Dubray, D. Gogny, E. Ormand, and W. Younes

Exploring Nuclear Shapes: Triaxiality at *Second* Barrier?

We have emphasized that a typical actinide breaks axial symmetry near the inner fission barrier, and it breaks reflection symmetry along the rest of the fission pathway. Can axial symmetry also be broken near the second fission barrier? Lu *et al* (2012) assert that breaking axial symmetry does lower the second fission barrier.

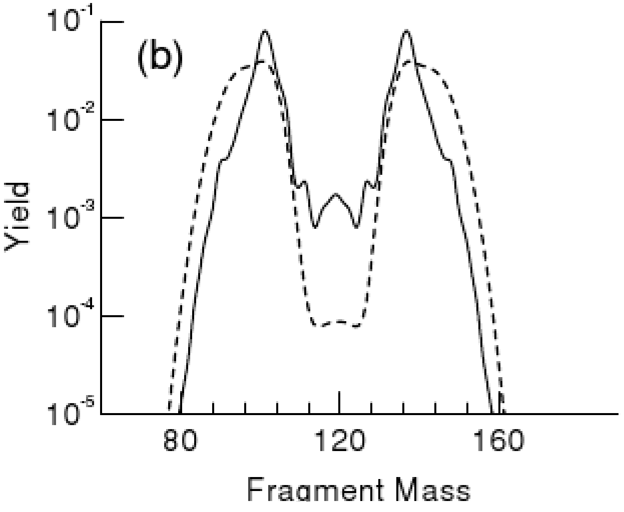


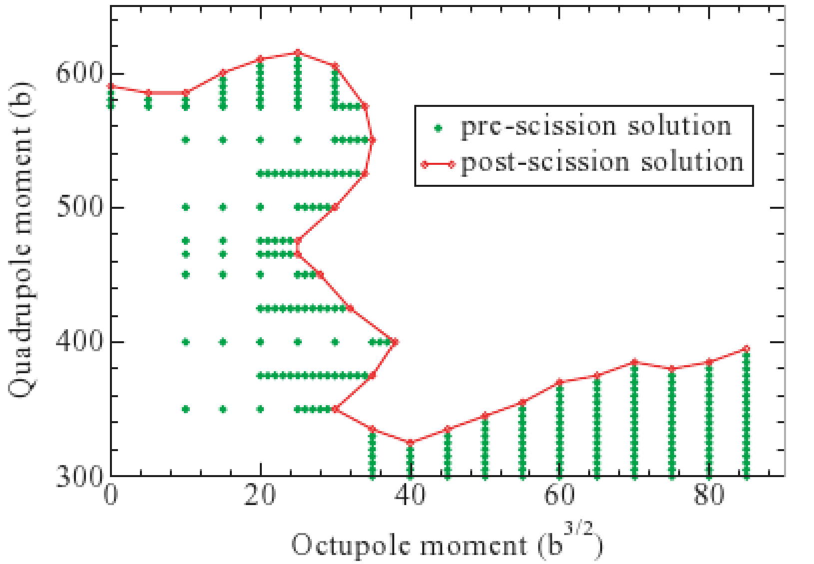


In our survey of even-even major and minor actinides, we found broken axial symmetry to have a negligible effect on the second barrier in most cases. The most dramatic effect was seen for 250Cm, where a real change in configuration appears to drive the system to triaxiality near the second fission barrier.

Perspectives

- Ingredients for Fission Observables
  - For half-lives and mass yields: collective inertias
    - Generator Coordinate Method (GCM) with ATDHFB masses
  - Nuclear configurations at scission
    - How does the deformation at scission depend on temperature?
    - Configuration at scission determines distribution of mass, excitation energy, and kinetic energy between fragments.





Above: Dynamic GCM (solid) vs. evaluated (dashed) mass distribution in fission of U238 (from Goutte (2005)).
Below: Location of scission configurations in Pu240 (from Younes (2009)).

Specific Contributions

- Computational Methods
  - – Helped prepare the MPI functionality of the code HFODD for use with fission
  - – Implemented ScaLAPACK diagonalization routines in HFODD
  - – Compared the performance of ScaLAPACK diagonalization and threaded LAPACK
  - – Visualization for nuclear potential energy surfaces
  - – Visualization for one-proton quasiparticle excitations
- Calculations for Fission
  - – Large-scale computations of potential energy surfaces for major actinides, minor actinides, and mercury isotopes with the Jaguar Cray XT5 and the Kraken XT5
  - – Calculations of spontaneous fission half-lives for major and minor actinides
  - – Expanded scope of the study of third minima for 232 Th and 232 U to include lighter isotopes of both elements

Recent Publications and Presentations

– M. Kortelainen, J. McDonnell, W. Nazarewicz, P.-G. Reinhard, J. Sarich, N. Schunck, M.V. Stoitsov, S.M. Wild. “Nuclear energy density optimization. III. UNEDF2” (in preparation).

– J.D. McDonnell *et al*. “Systematic Study of Spontaneous Fission in the Actinides” (in preparation).

– J.D. McDonnell *et al*. “Microscopic Study of the Fission of Mercury Isotopes at Finite Temperature” (in preparation).

– J.D.McDonnell *et al*. “Microscopic Study of Third Minima in Thorium and Uranium Isotopes” (in preparation).

– M. Kortelainen, J. McDonnell, W. Nazarewicz, P.-G. Reinhard, J. Sarich, N. Schunck, M.V. Stoitsov, S.M. Wild. “Nuclear energy density optimization. II. Large deformations”, Phys. Rev. C85, 024304 (2012).

– N. Schunck, J. Dobaczewski, J. McDonnell, W. Satula, J.A. Sheikh, A. Staczszak, M. Stoitsov, P. Toivanen. “Solution of the Skyrme-Hartree-Fock-Bogolyubov equations in the Cartesian deformed harmonic-oscillator

basis. (VII) hfodd (v2.49t): a new version of the program”, Comp. Phys. Comm. 183, 166 (2012).

– W. Nazarewicz and J. McDonnell. “Towards Predictive Theory of Fission”, Stewardship Science Academic Alliances Annual 2011, DOE/NA-0016, p.18 (2011).

– “Nuclear Fission: From Microscopic Forces to Experimental Observables”, poster, J. McDonnell, W. Nazarewicz, M. Kortelainen, N. Schunck, J.A. Sheikh, M.V. Stoitsov, Stewardship Science Academic Alliance Symposium, Washington, D.C., February 22-23, 2012. Recognized by an “Outstanding Poster Award”.

– “Fission of Actinide Nuclei”, J. McDonnell, M. Kortelainen, W. Nazarewicz, J.A. Sheikh, M.V. Stoitsov, N. Schunck, FUSTIPEN Topical Meeting on ’Theory of Nuclear Fission’ at GANIL, Caen, France, January 4-6, 2012.

– “Fission of Actinide Nuclei”, J. McDonnell, M. Kortelainen, W. Nazarewicz, J.A. Sheikh, M.V. Stoitsov, N. Schunck, Fall Meeting of the American Physical Society Division of Nuclear Physics, East Lansing, MI, October 26-29, 2011.

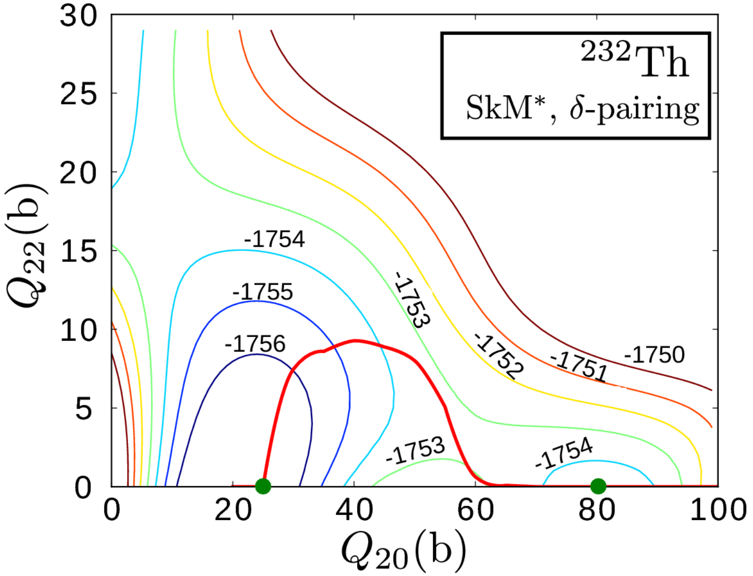
– “Fission Barriers in Actinide Nuclei”, J. McDonnell, M. Kortelainen, W. Nazarewicz, J.A. Sheikh, M.V. Stoitsov, 5th LACM-EFES-JUSTIPEN Workshop, Oak Ridge, TN, March 15-17, 2011.

– “Understanding Nuclear Reactions”, poster, J. McDonnell, W. Nazarewicz, Stewardship Science Graduate Fellowship Annual Meeting, Washington, D.C., July 20 - 22, 2011.

– “Microscopic Description of Fission Process”, W. Nazarewicz, J. McDonnell, Stewardship Science Academic Alliance Symposium, Washington, D.C., February 14-17, 2011.

Fission Theory Review

- Triaxiality lowers the inner fission barrier, and reflection asymmetry lowers the outer barriers. The exploration of many degrees of freedom is crucial for an accurate calculation of half-lives and mass yields.



Elongation:

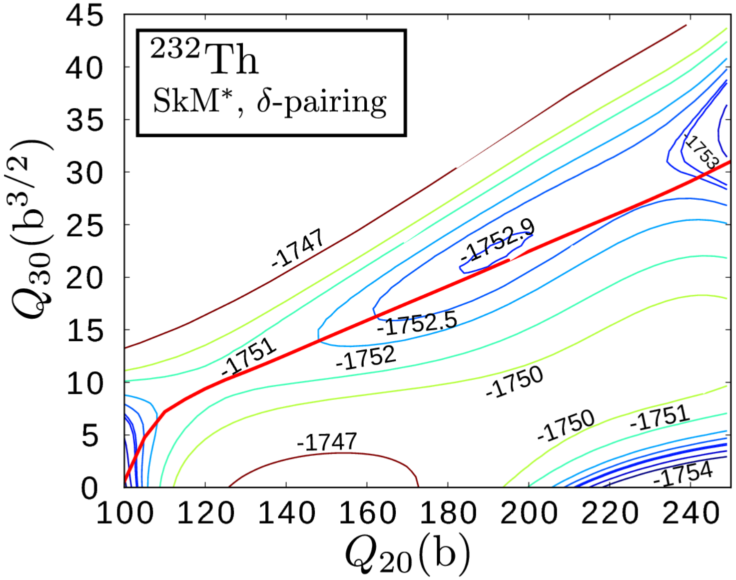
$$Q_{20} \propto \langle r^2 Y_{20} \rangle$$

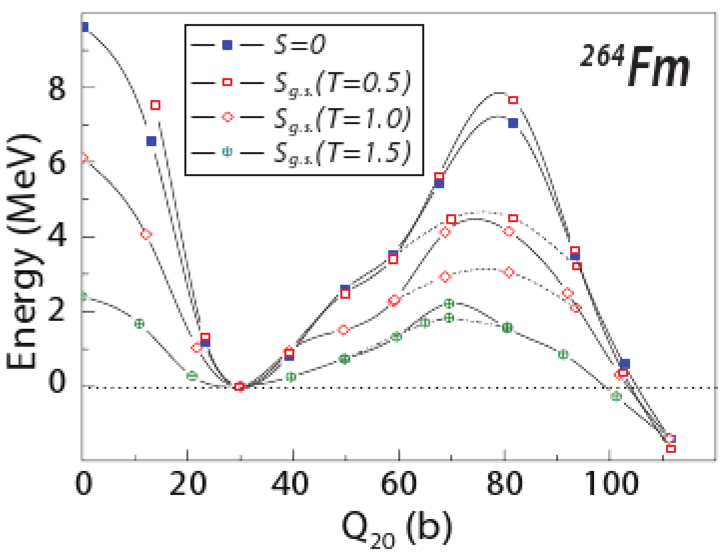
Triaxiality:

$$Q_{22} \propto \langle r^2 (Y_{22} + Y_{2-2}) \rangle$$

Refl. Asym.:

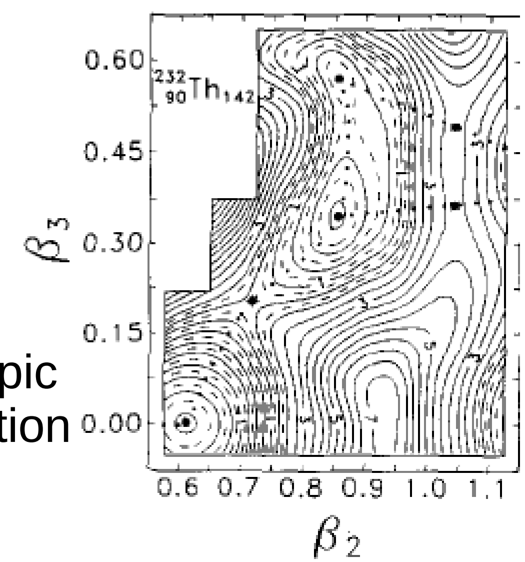
$$Q_{30} \propto \langle r^3 Y_{30} \rangle$$





Left: Effects of triaxiality on inner barrier (from Pei *et al* (2009)).

Right: Reflection asymmetric pathways via macroscopic-microscopic Methods with Strutinsky shell correction find a third minimum (from Cwiok *et al* (1994)).



Conclusion

- Ongoing work towards a predictive theory of fission is motivated by the qualitative agreement of current results for actinides with known experimental data, especially the evolution of symmetric mass distributions with increasing temperature.
- Progress is being made with increasingly accurate nuclear density functionals, with UNEDF1 reproducing empirical fission barriers.
- Our study of fission dynamics and ATDHFB will continue, with half-lives and mass distributions calculated with the new functionals.

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