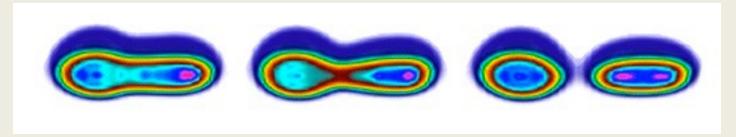
## **Nuclear Fission within a Real-Time Density Functional Framework**



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#### Slides: <u>UW Bulgac's web page</u>

Nuclear fission is unquestionably one of the most challenging quantum many-body problems in theoretical physics.

Superconductivity needed less than 50 years to reach a microscopic understanding, from 1911 to 1957. This is just one example out of many: superfluidity, QHE, FQHE, topological phases of matter, magnetism, essentially any CM topic, etc.

Nuclear fission is more than 80 years old now!

**Still waiting!** 

What are the essential ingredients necessary to describe nuclear fission?

In order of relevance:

- ✓ Surface tension/surface energy and Coulomb energy Bethe, Weizsäcker
- ✓ Pairing interaction Bohr, Mottelson, Pines
- ✓ Spin-orbit interaction Goeppert-Mayer, Jensen
- ✓ Symmetry energy Bethe, Weizsacker
- ✓ Saturation energy and saturation density Bethe, Weizsäcker

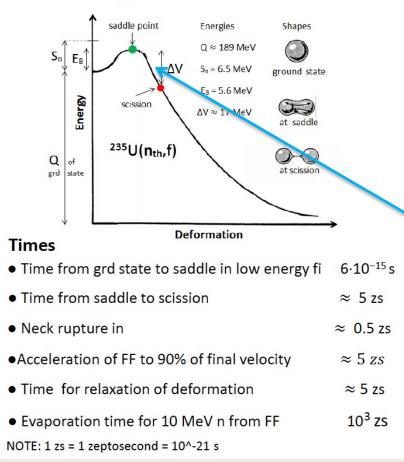
It is reasonable to expect that if these 6 (since Coulomb energy is known a priori) characteristics are accurate and <u>iff the theoretical framework is sound</u> then one should obtain quite accurate predictions for fission (at the percent level), and for masses, charge radii, ...

All these nuclear properties are quite well known for 7 decades now.

Why did it take it so long to have a genuine microscopic quantum description of nuclear fission?

Well, for one reason, nuclear fission is a highly non-equilibrium process.

#### Potential energy versus deformation



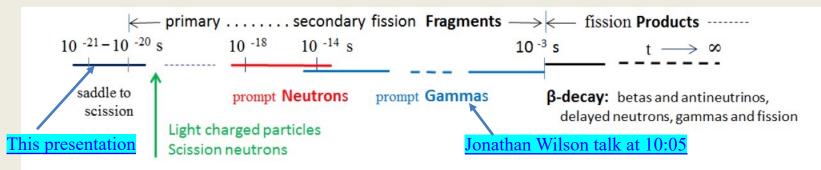
The shape of the PES is governed mostly by the competition between the surface and the Coulomb energies.

#### Lise Meitner and Otto Frisch (1939)

LM was deprived of 2 Nobel prizes: Meitner-Auger effect (she predicted it a year before Auger) nuclear fission (LM coined the term nuclear fission).

From the outer saddle to the scission the dynamics is relatively fast, likely non-adiabatic, and in this region the fission fragments are formed and their properties are (mostly) defined.

 $1 \text{ zs} = 10^{-21} \text{ sec.} = 300 \text{ fm/c}$ 



Notice the extremely wide range of time scales!

From Lectures given by Gönnenwein at LANL Fiesta School, 2014

Several recent developments have radically changed our prospects of attaining a microscopic description of nuclear fission.

• In THEORY:

Formulation of a local extension of the Density Functional Theory (DFT), in the spirit of the Local Density Approximation (LDA) formulation of DFT due to Kohn and Sham, to superfluid time-dependent phenomena, the Superfluid Local Density Approximation (SLDA).

Validation and verification of (TD)SLDA against a large set of theoretical and experimental data for systems of strongly interacting fermions.

Unlike phenomenological models, a sensible non-equilibrium quantum many-body framework should have general validity.

• In HIGH PERFOMANCE COMPUTING:

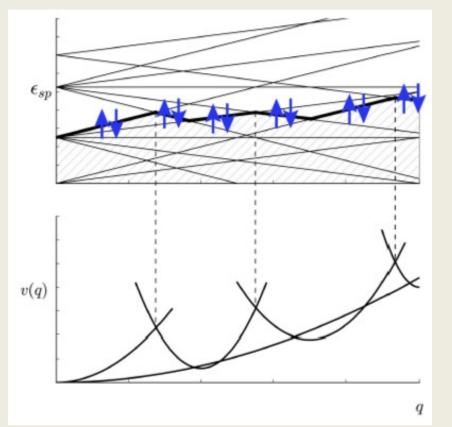
Emergence of very powerful computational resources, advanced capabilities of leadership class computers, in particular tens of thousands of GPUs.

• NUMERICAL IMPLEMENTATION on leading edge supercomputers since 2007: Jaguar, Titan, Summit (all at ORNL), Sierra, Lassen (both at LLNL), Piz Daint (CSCS, Lugano), ...

SLDA and TDSLDA are problems of extreme computational complexity, requiring the solution of up to 1,000,000s coupled complex non-linear time-dependent 3D partial differential equations.

Potential energy surface is a bit more complicated than a liquid drop model would suggest.

Nuclei are quantum compact objects and single-particle motion is quantized!



How nuclei change their shape at a microscopic level?

• While a nucleus elongates its Fermi surface becomes oblate and its sphericity must be restored

Hill and Wheeler, PRC, 89, 1102 (1953)

Bertsch, PLB, 95, 157 (1980)

• Each single-particle level is double degenerate (Kramers' degeneracy) and at each level crossing two nucleons must jump simultaneously!

(m,-m) => (m',-m') "Cooper pair" => "Cooper pair"

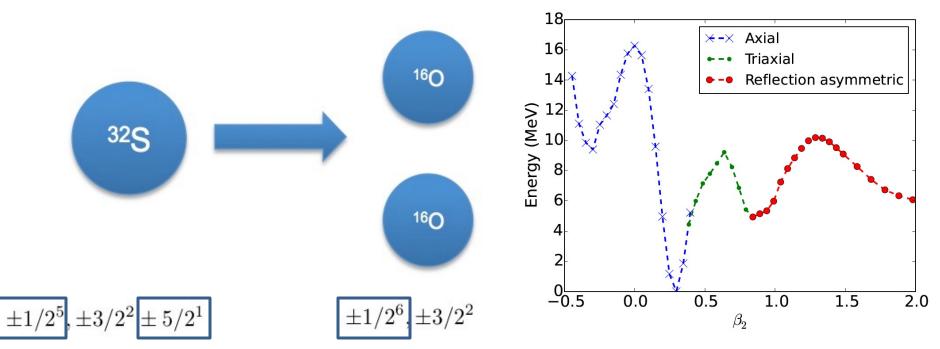
• <u>Pairing interaction/superfluidity is the most</u> <u>effective mechanism at performing shape changes.</u> <u>The transitions are significantly enhanced due to the</u> <u>presence of the Bose condensate of Cooper pairs.</u>

Barranco, Bertsch, Broglia, and Vigezzi Nucl. Phys. A512, 253 (1990)

### During fission the nucleus undergoes a dramatic number of <u>quantum</u> phase transitions!

#### A pedagogical gedanken experiment:

The fictious fission of <sup>32</sup>S was considered by Negele et al in 1980's, by artificially increasing the charge of the proton.



Occupied sp orbitals m-quantum numbers in initial and final configurations

Potential energy curve for <sup>240</sup>Pu with SLy4 Ryssens, et al., Phys. Rev C 92, 064318 (2015)

One more problem! Initial nucleus: 20 positive + 12 negative parity sp orbitals Final nuclei: 16 positive + 16 negative parity sp orbitals Most of the modeling of fission since 1939 is still performed mostly phenomenologically, which is distinct from <u>genuine microscopic approaches</u>!

"Microscopic" approaches (TDGCM, ATDHF) assume <u>the decoupling of collective</u> <u>and intrinsic motion (adiabaticity</u>), making thus the introduction of a collective Hamiltonian legitimate.

Schunck and Robledo, Microscopic theory of nuclear fission, Rep. Prog. Phys. 79, 116301 (2016) Krappe and Pomorski, Theory of Nuclear Fission, Springer, 2012.

We challenged and disproved this never checked assumption!!!

What microscopic conclusions have been firmly established so far?

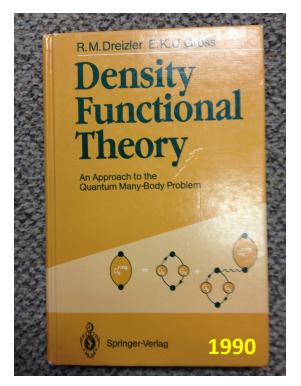
• The revolutionary idea introduced by Meitner and Frisch: fission is controlled by the competition between Coulomb and surface energies. *Meitner and Frisch (1939)* 

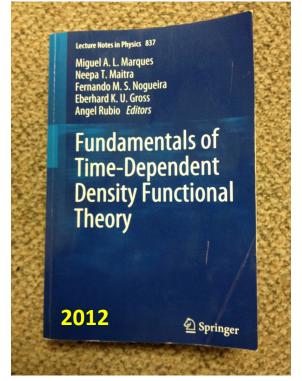
• The formation of a compound nucleus and a very slow evolution of the nuclear shape towards the outer barrier. *Bohr (1936) and Bohr and Wheeler* (1939)

• The crucial role of shell effects at large deformations and of the pairing correlations while the nuclear shape evolves. *Strutinsky, 1967, Bertsch, 1980* 

• The decay of the fission fragments can be described in a statistical approach. Weisskopf (1937), Hauser and Feshbach (1952)

# **The Main Theoretical Tool**





DFT has been developed and used mainly to describe normal (non-superfluid) electron systems – <u>57 years old theory</u>, Kohn and Hohenberg, 1964.

Kohn and Hohenberg proved <u>mathematically</u> that the number density and the energy are identically reproduced within the Density Functional Theory and with the Schrödinger many-body equation.

A new local extension of DFT to superfluid systems (not everyone is normal) and time-dependent phenomena was developed Reviews: A. Bulgac, Ann. Rev. Nucl. Part. Sci. 63, 97 (2013) and Physica Status Solidi B 256, 1800592 (2019) Let us consider the Schrödinger equation for example:

$$H = \sum_{i=1}^{N} T(i) + \sum_{i < j}^{N} U(ij) + \sum_{i < j < k}^{N} U(ijk) + \dots + \sum_{i=1}^{N} V_{ext}(i)$$
$$H\Psi_{0}(1, 2, \dots N) = E_{0}\Psi_{0}(1, 2, \dots N)$$

We know this is <u>the correct framework to describe quantum phenomena</u>, even though we have only an approximate idea about interactions.

In nuclear physics we do not know the exact NN and NNN potentials and use phenomenology.

$$\Psi_{0}(1,2,...N) \equiv \Psi_{0}(1,2,...N;[n(\vec{r})]) \Leftrightarrow V_{ext}(\vec{r}) \Leftrightarrow n(\vec{r})$$
$$E[n(\vec{r})] = \langle \Psi_{0}[n(\vec{r})]|H|\Psi_{0}[n(\vec{r})] \rangle$$

Kohn and Hohenberg (1964)

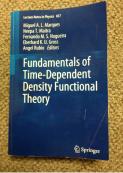
We now also know that <u>DFT is mathematically equivalent to the Schrödinger equation</u>, even though we cannot always in practice show that, and, as a rule, we do not know the exact functional either and use use phenomenology too.

$$E_{0} = \min_{n(\vec{r})} \int d^{3}r \left\{ \frac{\hbar^{2}}{2m^{*}(\vec{r})} \tau(\vec{r}) + \varepsilon \left[ n(\vec{r}) \right] + V_{ext}(\vec{r}) n(\vec{r}) \right\}$$
$$n(\vec{r}) = \sum_{i}^{N} \left| \varphi_{i}(\vec{r}) \right|^{2}, \qquad \tau(\vec{r}) = \sum_{i}^{N} \left| \vec{\nabla} \varphi_{i}(\vec{r}) \right|^{2}$$

Schrödinger equation and DFT are both exact quantum frameworks!

## **Extending the formalism to Time-Dependent Phenomena and Superfluidity**

"The time-dependent density functional theory is viewed in general as a reformulation of the exact quantum mechanical time evolution of a many-body system when only one-body properties are considered."



**Time-Dependent Superfluid Local Density Approximation (TDSLDA)** 

$$E(t) = \int d^{3}r \left[ \mathcal{E}(n(\vec{r},t),\tau(\vec{r},t),\nu(\vec{r},t),\vec{j}(\vec{r},t)) + V_{ext}(\vec{r},t)n(\vec{r},t) + ... \right] \\ \left\{ \begin{array}{l} i\hbar \frac{\partial u_{i}(\vec{r},t)}{\partial t} = [h(\vec{r},t) + V_{ext}(\vec{r},t) - \mu] u_{i}(\vec{r},t) + [\Delta(\vec{r},t) + \Delta_{ext}(\vec{r},t)] v_{i}(\vec{r},t) \\ i\hbar \frac{\partial v_{i}(\vec{r},t)}{\partial t} = [\Delta^{*}(\vec{r},t) + \Delta^{*}_{ext}(\vec{r},t)] u_{i}(\vec{r},t) - [h(\vec{r},t) + V_{ext}(\vec{r},t) - \mu] v_{i}(\vec{r},t) \end{array} \right.$$

#### Galilean invariance determines the functional dependence on currents.

Spin degrees of freedom not shown.

# **The Main Theoretical Tool: DFT**

**TDSLDA** - This is an extension to Superfluids and Time-Dependent Phenomena of DFT, based on Verification and Validation for a variety of strongly interacting fermions systems (cold atoms, neutron star crust, nuclei).

• Since DFT/SLDA is not an approximation, but an exact theoretical framework (unlike HF, HFB, CC, etc.), one has to convincingly prove that its specific realization is equivalent to the Schrödinger equation!

The DFT and the Schrödinger descriptions of one-body observables should be identical.

• One expects that DFT also describes correctly Nature!

In nuclear physics both DFT and Schrödinger equation are at disadvantage, neither the energy density functional nor the NN, NNN, ... interactions are know with sufficient accuracy.

• And, of course, make sure that the numerical implementation faithfully reproduces the theory.

$$i\hbar\frac{\partial}{\partial t} \begin{pmatrix} \mathbf{u}_{n\uparrow}(\vec{r},t) \\ \mathbf{u}_{n\downarrow}(\vec{r},t) \\ \mathbf{v}_{n\uparrow}(\vec{r},t) \\ \mathbf{v}_{n\uparrow}(\vec{r},t) \end{pmatrix} = \begin{pmatrix} \hat{\mathbf{h}}_{\uparrow\uparrow}(\vec{r},t) - \mu & \hat{\mathbf{h}}_{\uparrow\downarrow}(\vec{r},t) & 0 & \Delta(\vec{r},t) \\ \hat{\mathbf{h}}_{\downarrow\uparrow}(\vec{r},t) & \hat{\mathbf{h}}_{\downarrow\downarrow}(\vec{r},t) - \mu & -\Delta(\vec{r},t) & 0 \\ 0 & -\Delta^{*}(\vec{r},t) & -\hat{\mathbf{h}}_{\uparrow\uparrow}^{*}(\vec{r},t) + \mu & -\hat{\mathbf{h}}_{\uparrow\downarrow}^{*}(\vec{r},t) \\ \Delta^{*}(\vec{r},t) & 0 & -\hat{\mathbf{h}}_{\downarrow\uparrow}^{*}(\vec{r},t) & -\hat{\mathbf{h}}_{\downarrow\downarrow}^{*}(\vec{r},t) + \mu \end{pmatrix} \begin{pmatrix} \mathbf{u}_{n\uparrow}(\vec{r},t) \\ \mathbf{u}_{n\downarrow}(\vec{r},t) \\ \mathbf{v}_{n\uparrow}(\vec{r},t) \\ \mathbf{v}_{n\downarrow}(\vec{r},t) \end{pmatrix}$$

•Number of PDEs is of the order of the number of spatial lattice points  $2 \times 2 \times 4 \times N_x N_y N_z$  – from 10,000s to 1-2,000,000

# The Main Computational Tool(s)





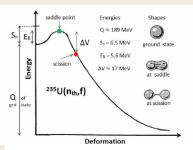




Cray XK7 (Titan), ranked at peak  $\approx 27$  Petaflops (Peta –  $10^{15}$ ) On Titan there are <u>18,688 GPUs</u> which provide <u>24.48 Petaflops !!</u> and <u>299,008 CPUs</u> which provide <u>only 2.94 Petaflops</u>.

A single GPU using a CUDA code on Titan (now retired) performed the same amount of FLOPs as approximately 150 CPUs using a C code.

### Piz Daint is about 3x faster, and Summit is 1.4x faster than Piz Daint

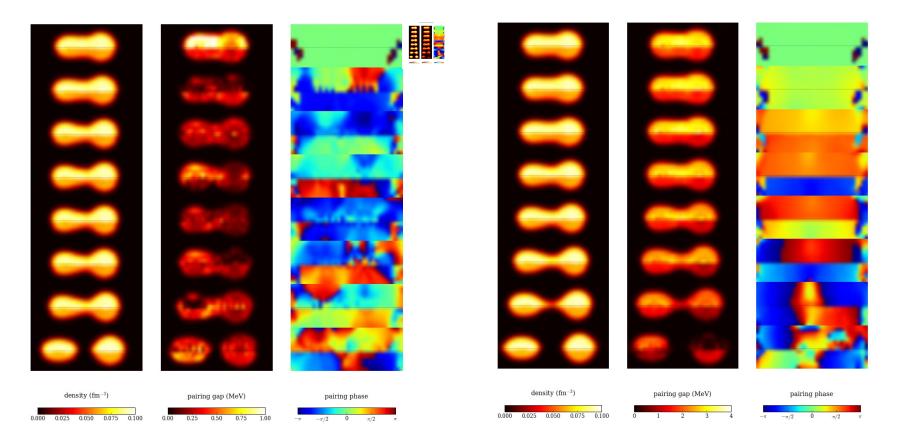


# How important is pairing in fission?

#### Without pairing nuclei will typically will not fission!!!

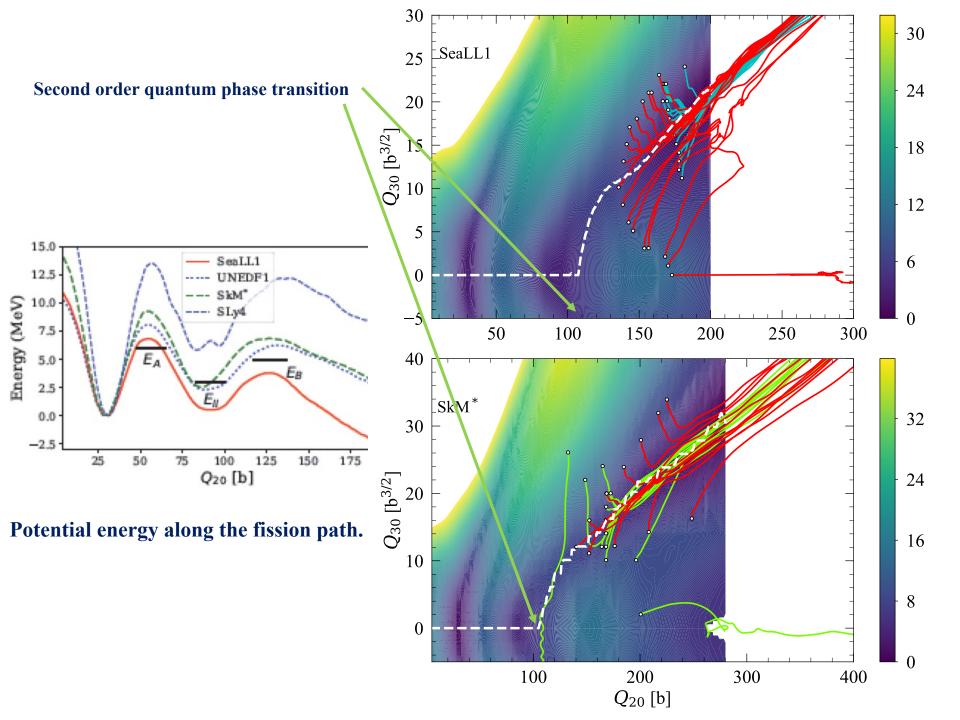
 $^{\rm 240}{\rm Pu}$  fission in the normal pairing gap

<sup>240</sup>Pu fission in a larger pairing gap



### Normal pairing strength Saddle-to-scission 14,000 fm/c

## Enhanced pairing strength Saddle-to-scission 1,400 fm/c !!!

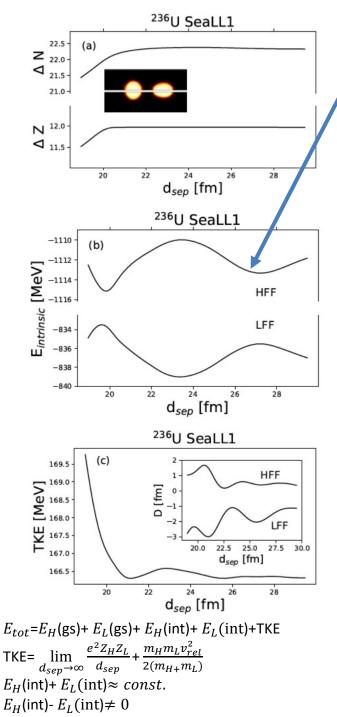


NEDF	$E_{\rm ini}^*$	TKE	N <sub>H</sub>			ZL					$\tau_{s \to s} (\text{fm/c})$
SeaLL1-1asy	7.9(1.7)	177.8(3.1)	83.4(0.4)	53.2(0.4)	62.9(0.5)	41.1(0.4)	17.1(3.0)	20.3(2.0)	37.4(3.1)	215.2(2.5)	2317(781)
SeaLL1-2asy	2.6(1.8)	178.0(2.3)	82.9(0.4)	52.9(0.2)	63.3(0.5)	41.5(0.3)	19.5(3.8)	14.0(1.9)	33.5(5.1)	211.5(3.3)	1460(176)
SeaLL1-sy	9.2	147.1	77.5	48.9	68.8	45.4	45.2	29.0	74.2	221.3	10103
SkM*-1asy	8.2(3.0)	174.5(2.5)	84.1(0.9)	53.0(0.5)	61.8(0.9)	40.9(0.5)	16.6(3.1)	14.9(2.3)	31.5(3.8)	206.0(2.4)	1214(448)
SkM*-1sy	9.6	149.0	73.4	47.2	72.6	46.7	29.4	28.5	57.9	206.9	3673
SkM*-2asy	8.1(0.2)	182.8(4.4)	82.6(1.0)	52.4(0.6)	63.6(1.0)	41.7(0.5)	14.3(3.9)	13.0(3.0)	27.3(3.4)	210.1(1.8)	1349(309)

Table I. The NEDF, the initial excitation energy  $E_{ini}^*$ , TKE, neutron, proton number, and excitation energies of the heavy and light fragments, total excitation energy of fragments TXE, and the sum of TKE and TXE, and the average saddle-to-scission times and their corresponding variances in parentheses. All energies are in MeV and S\*\*\*sy, S\*\*\*asy stand for symmetric and antisymmetric channels. Using Wahl's charge systematics [90] and data from Ref. [91] one obtains for neutrons  $N_L^{syst} \approx 61$  and  $N_H^{syst} \approx 85$  and for protons  $Z_L^{syst} \approx 40$  and  $Z_H^{syst} \approx 54$ , and TKE<sup>syst</sup> = 177 ... 178 MeV from Ref. [92].

NEDF	T <sub>L</sub> [MeV]	$T_{\rm H}$ [MeV]	T <sub>L</sub> [MeV]	T <sub>H</sub> [MeV]	$Q_{20}^{\rm L}$ [b]	$Q_{20}^{\rm H}$ [b]	$Q_{30}^{\rm L}$ [b <sup>3/2</sup> ]	$Q_{30}^{\rm H}$ [b <sup>3/2</sup> ]	$(c/a)_H$	$(c/a)_L$	$\tau_{s \to s}  [\text{fm/c}]$
SeaLL1-1	1.40(0.07)	1.11(0.08)	1.28(0.07)	1.16(0.07)	15.7(0.9)	2.6(0.5)	0.08(0.17)	-0.20(0.06)	1.06(0.01)	1.59(0.03)	2392(800)
SeaLL1-2	1.15(0.08)	1.19(0.12)	1.00(0.08)	1.21(0.08)	17.1(1.1)	2.6(0.6)	0.23(0.08)	-0.19(0.06)	1.06(0.01)	1.63(0.03)	1460(176)
SeaLL1-sy	1.54	1.99			27.4	27.0	0.9	-1.1	1.87	1.73	10103
SkM*-1asy	1.20(0.09)	1.10(0.10)			11.3(1.3)	3.5(0.9)	0.1(0.1)	-0.4(0.1)	1.08(0.02)	1.42(0.04)	1214(448)
SkM*-1sy	1.56	1.55			24.2	25.6	0.9	-1.0	1.72	1.75	3673
SkM*-2asy	1.11(0.14)	1.02(0.14)			14.5(1.7)	2.3(0.7)	0.09(0.08)	-0.3(0.1)	1.05(0.02)	1.53(0.06)	1349(309)

Table II. Internal temperatures for the light  $T_L$  and heavy  $T_H$  fragments computed according to the simple estimate (columns 2 and 3) or finite-temperature HFB calculations (columns 4 and 5). The axial quadrupole and octupole moments of the fragments, the ratios of the long to the short semi-axes, as well as the average scission times are also listed



FFs are well separated and exchange energy, but not nucleons!

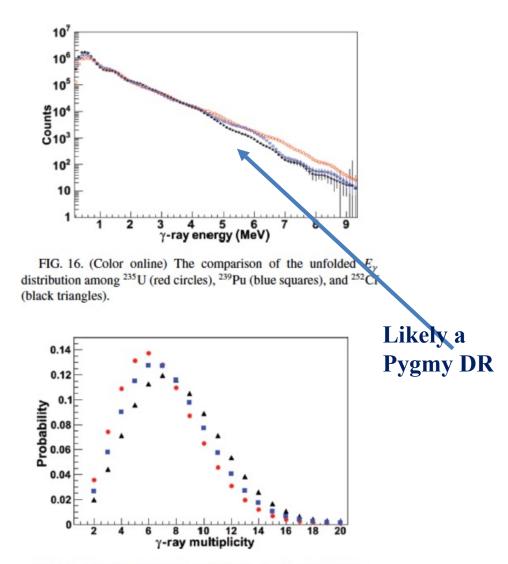


FIG. 17. (Color online) The comparison of the unfolded  $M_{\gamma}$  distribution among <sup>235</sup>U (red circles) and <sup>239</sup>Pu (blue squares), and <sup>252</sup>Cf (black triangles).

#### Chyz et al, Phys. Rev. C 87, 034620 (2013)

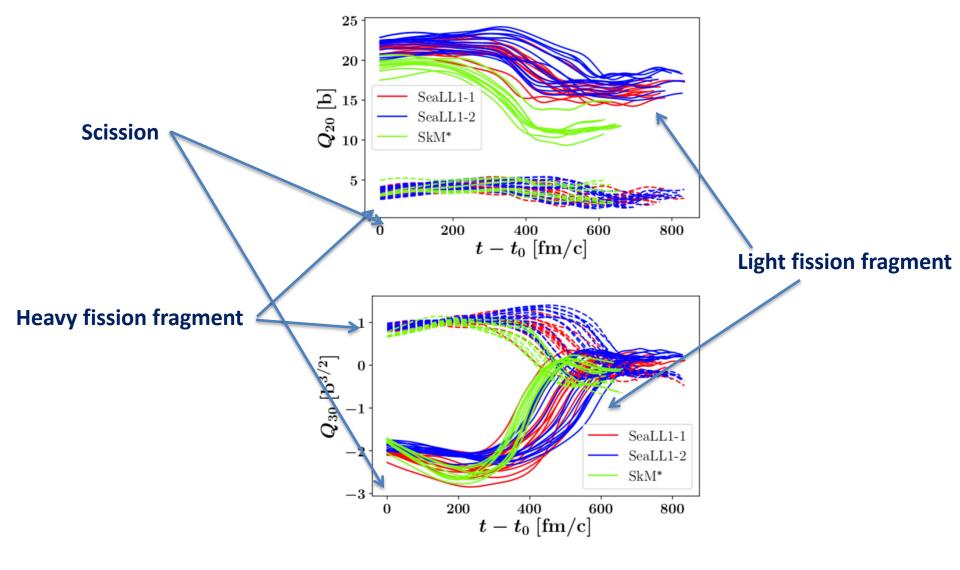
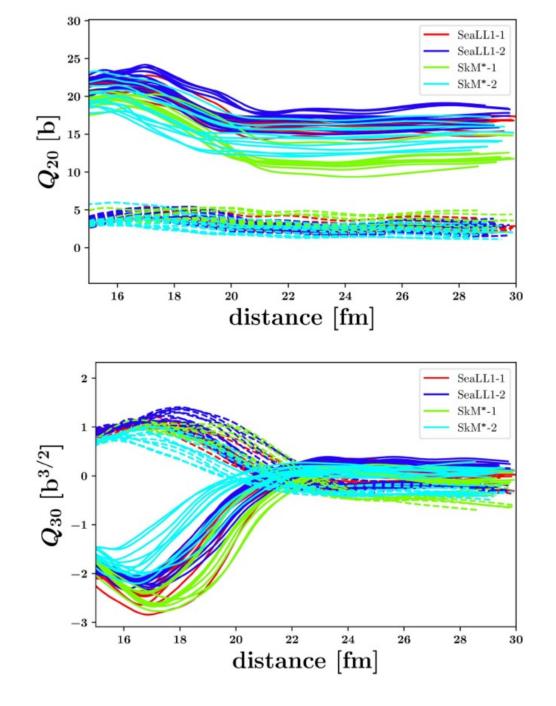
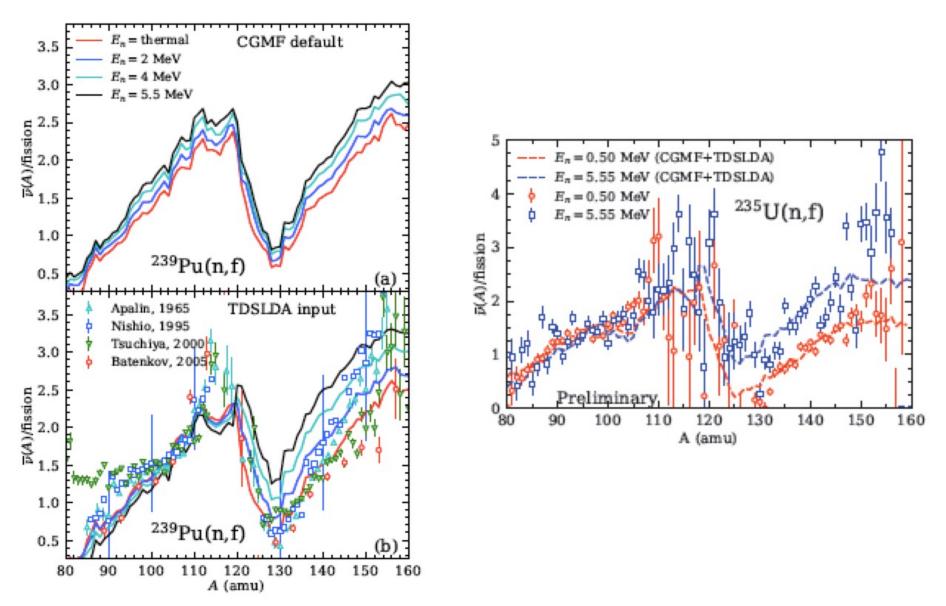


Figure 5. (Color online) The evolution of the quadrupole  $Q_{20}$  and octupole  $Q_{30}$  moments of the light (solid lines) and heavy (dashed lines) FFs after scission. The color codes are the same as in Fig. 3

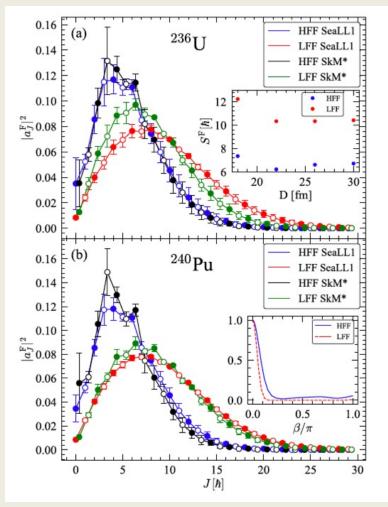
# The light fission fragment emerges at scission (t<sub>0</sub>) very elongated, but it relaxes relatively quickly.





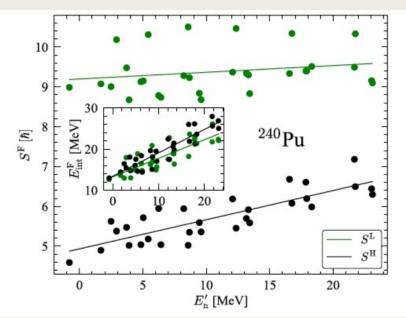
TDSLDA, unlike other approaches, predicts the excitation energy sharing between fission fragments, which are used in a Hauser-Feshbach code.

#### The first microscopic evaluation of the fully separated fission fragment intrinsic spins.



Intrinsic spins distributions of the well separated heavy (HFF) and light (LFF) fission fragments (FF), evaluated with different nuclear energy density functionals.

# NB These are intrinsic spins prior to neutron and $\gamma$ -rays emission.



Evolution of the fission FF intrinsic spins after full separation as a function of the equivalent incident neutron energy in reaction <sup>239</sup>Pu(n,f). Inset: FF excitation energies.

TABLE II. The averages (standard deviations) of  $\langle \Phi | J_{\alpha}^{L} J_{\alpha}^{H} | \Phi \rangle$ , with  $\alpha = x$ , *y*, *z*. The nondiagonal elements of this tensor are negligible and all  $\langle \Phi | J_{\alpha}^{F} | \Phi \rangle = 0$ .

Nucleus	NEDF	$\langle \Phi   J^L_x J^H_x   \Phi  angle$	$\langle \Phi   J^L_y J^H_y   \Phi  angle$	$\langle \Phi   J^L_z J^H_z   \Phi  angle$
<sup>236</sup> U	SeaLL1	-1.16(0.63)	-1.16(0.63)	-2.63(0.47)
236U	SkM*	-0.48(0.71)	-0.48(0.71)	-1.62(0.30)
<sup>240</sup> Pu	SeaLL1	-0.72(0.65)	-0.72(0.65)	-4.43(0.92)
<sup>240</sup> Pu	SkM*	-0.90(0.57)	-0.90(0.57)	-1.80(0.52)
		•		<b></b>

#### Characterization of the FF twisting and bending modes.

## Large Amplitude Collective Motion is strongly dissipative. It is overdamped! It is slower than adiabatic motion!!!

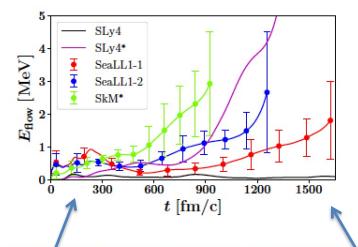


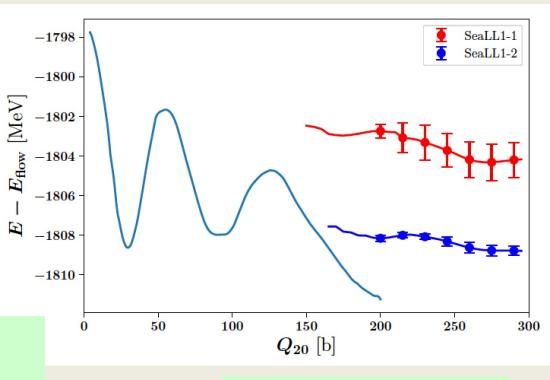
Figure 3 (Color online) The collective flow energy evaluated for NEDFs with realistic pairing SLy4 [41], enhanced pairing SLy4\*, and for SkM\*, SeaLL1-1 and SeaLL1-2 sets. The error bars illustrate the size of the variations due to different initial conditions.

$$E_{flow} = \int d^3 r \frac{\vec{j}^2(\vec{r},t)}{2mn(\vec{r},t)}$$

$$\vec{j}(\vec{r},t) = \frac{i\hbar}{2} \sum_{k} \mathbf{v}_{k}^{*}(\vec{r},t) \vec{\nabla} \mathbf{v}_{k}(\vec{r},t) - \mathbf{v}_{k}(\vec{r},t) \vec{\nabla} \mathbf{v}_{k}^{*}(\vec{r},t)$$

$$n(\vec{r},t) = \sum_{k} \left| \mathbf{v}_{k}(\vec{r},t) \right|^{2}$$

$$E_{total} = E_{flow} + E_{int} \approx E_{int}(q,T) \approx const.$$



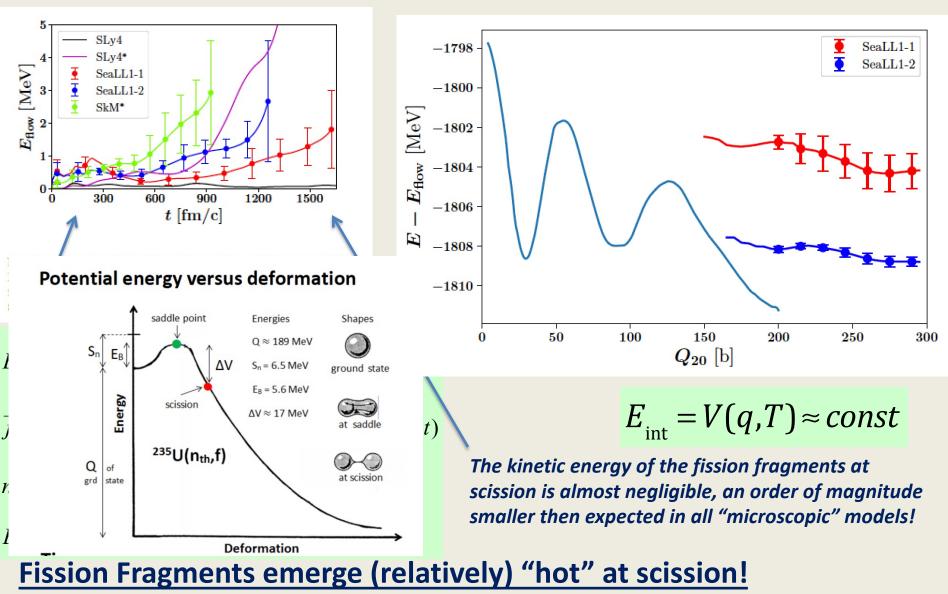
 $E_{\rm int} = V(q,T) \approx const$ 

The kinetic energy of the fission fragments at scission is almost negligible, an order of magnitude smaller then expected in all "microscopic" models!

## Fission Fragments emerge (relatively) "hot" at scission!

And they will get a bit hotter after their shape relaxes, particularly the light fragments.

## Large Amplitude Collective Motion is strongly dissipative. It is overdamped! <u>It is slower than adiabatic motion!!!</u>



And they will get a bit hotter after their shape relaxes, particularly the light fragments.

What consequences do our results have on phenomenological approaches and on GCM or ATDHF based microscopic approaches?

In GCM inspired approaches the total wave function is represented as

and  $\Phi(x|q)$  is chosen roughly from such a picture of the nucleus dynamics. This implies that during the evolution the nucleus follows the lowest adiabatic potential energy surface

#### and

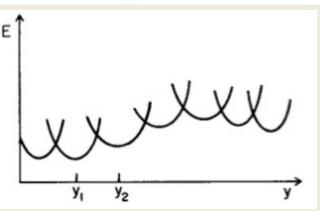
not any other potential surface

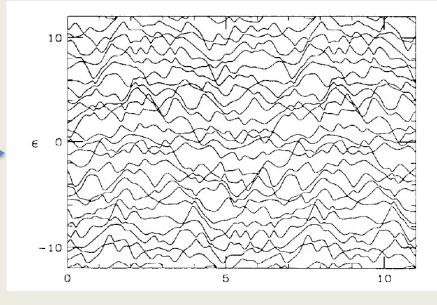
#### and

neither performs any jumps between various potential energy surfaces, as in this more accurate representation of such surfaces.

This is however a typical situation in chemistry. See works of John C. Tully (Yale, Chemistry) starting from 1990s' in J. Chem. Phys. and even earlier.

$$\Psi(\mathbf{x},t) = \int d\mathbf{q} f(\mathbf{q},t) \Phi(\mathbf{x}|\mathbf{q}),$$





## **Summary**

- Large Amplitude Collective Motion is strongly dissipative, it is overdamped, the role of the collective inertia is negligible! The introduction of a collective Hamiltonian is illegitimate. Fluctuations or two-body collisions do not modify this conclusion.
- While pairing is not the engine driving the fission dynamics, <u>(dynamical) pairing provides the essential</u> <u>lubricant</u>, without which the evolution may arrive quickly to a screeching halt.
- TDDFT will offer insights into nuclear processes and quantities which are either not easy or impossible to obtain in the laboratory: fission fragments excitation energies and angular momenta distributions prior to neutron and γ emission, element formation in astrophysical environments, and other nuclear reactions in a parameter free approach ...
- So far we have been able to extract information, which is impossible to extract directly experimentally
- The quality of the agreement with experimental observations is surprisingly good, especially taking into account the fact that we made no effort to reproduce any fission measured data. No fitting of parameters!
- It has been now firmly established microscopically that large amplitude collective motion is strongly dissipative and overdamped and phenomenological models would have to be altered accordingly.
- The fissioning nucleus behaves <u>superficially</u> as a very viscous system.
- The "temperatures" of the fission fragments are not equal.

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