Theory of Fission Product Yields

FIESTA Summer School

September, 30th 2017

Nicolas Schunck

LLNL-PRES-737743

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

Lawrence Livermore National Laboratory

Introduction

- Historical Remarks
- Some Definitions
- The Big Picture

LLNL-PRES-737743

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

Lawrence Livermore National Laboratory

Experimental Discovery of Nuclear Fission

Hahn Strassman Meitner

Hahn's Worktable

- 1939: experimental discovery of neutron-induced fission

that Hahn and Strassmann were forced to conclude that *isotopes of barium* ($Z = 56$) *are formed as a consequence of the bombardment of uranium* ($Z = 92$) *with neutrons.* L. Meitner, Nature **3615**, 239 (1939).

On the basis, however, of present ideas about the behaviour of heavy nuclei[6], an entirely different and essentially classical picture of these new disintegration processes suggests itself. On account of their close packing and strong energy exchange, the particles in a heavy nucleus would be expected to move in a collective way which has some resemblance to the movement of a liquid drop. If the movement is made sufficiently violent by adding energy, such a drop may divide itself into two smaller drops.

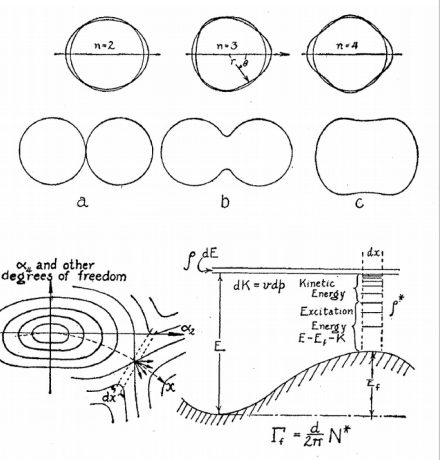
- 1940: experimental discovery of spontaneous fission by K.A. Petrzhak and G.N. Flerov

Fission: A Large Amplitude Collective Motion

- Induced fission as a two-step process: formation of a compound nucleus followed by its decay
- Semi-classical process that can be described by a set of collective variables (=deformations)
- Separate collective motion and intrinsic excitations

$$\hat{H} = \hat{H}_{\text{int.}} + \hat{H}_{\text{col.}} + \hat{H}_{\text{coup.}}$$

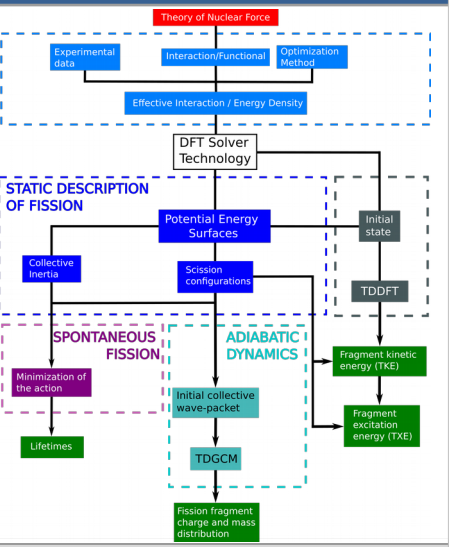
- Adiabatic approximation: the coupling between intrinsic and collective can be neglected



- Experimental observations are compatible with the picture of the nucleus as a small liquid drop that splits into two smaller drops [1].
- Keep in mind some of the approximations in this historical picture of fission (neutron-induced)
  - Two-step process: formation of an excited compound nucleus with a "long" lifetimes, followed by fission;
  - Driven by a few collective variables, which are often some deformations characterizing the nuclear shape;
  - No coupling between intrinsic and collective degrees of freedom, the system remains in equilibrium at all times.
- Already in Bohr & Wheeler paper, there were arguments about the possibility of spontaneous fission via quantum tunneling through some potential energy surface [2].

Theorist's TODO List

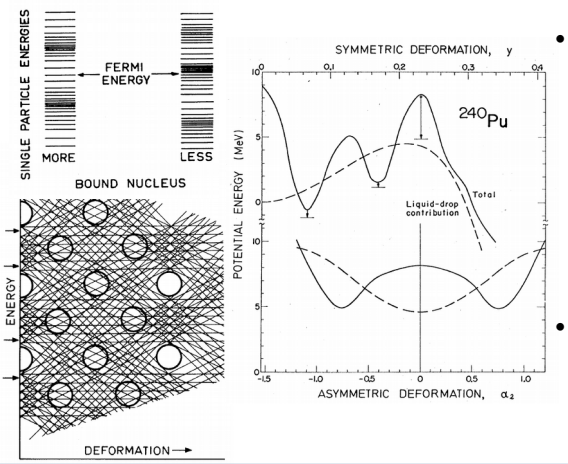
- Choose collective variables
- Compute energy as a function of collective variables
- Compute dynamical evolution through that space
  - Quantum tunneling for spontaneous fission
  - Quantum collective flow for fission fragment distributions
  - Real-time dynamics for single fission events
  - Alternative: statistical model



- Nuclear fission is one of the most complex scientific problems you can imagine
  - ✔ It requires quantum mechanics in full glory, including concepts such as delocalization, entanglement, fluctuations, etc.;
  - ✔ It is a many-body problem with no analytical solution: One of the main difficulties is to explain how various types of correlations induced by the nuclear Hamiltonian are responsible for the "emergence" of collective effects;
  - ✔ It involves nucleons in interaction: nuclear forces should in principle be mathematically derived from quantum chromodynamics, or at least be constrained by it. This is a research area of its own;
  - ✔ Fission is naturally a time-dependent process, which significantly adds to the computational difficulty;
  - ✔ It is also an open system: a fissioning nucleus and/or its fragments can emit particles and radiation (gamma), hence it it coupled to its environment. Furthermore, some of the observables that are important for energy applications are precisely these neutrons and gammas.

- Computing the properties of the fissioning nucleus as a function of the collective variables is the first, most important task in almost any theory of fission
  - This seems pretty obvious for adiabatic approaches, since it is assumed there that everything else does not matter that much;
  - Less obviously, we will see that it is also important for non-adiabatic approaches based on real-time dynamics, as it helps preparing a suitable initial state.
- Most of the work on fission product yields (FPY) done over the years point to the fact that most of the properties of these FPY can be directly inferred from the properties of the potential energy surface (PES)
- The devil is in the details, though…

Macroscopic-microscopic Models (1/4)
Introduction



- Take into account nucleon degrees of freedom
  - Shell correction coming from the distribution of single-particle levels
  - Pairing correction to mock up the effect of residual interactions
- Extensions to finite angular momentum or temperature are also available

In the macroscopic-microscopic approach, the basic degrees of freedom are the single-particle states and the nuclear deformations, and the equation of motion is the Schrödinger equation

- The liquid drop picture of the nucleus was very successful but could not explain the existence of shape isomers: long-lived states characterized by a large quadrupole moment (or, equivalently, large electric quadrupole gamma transitions) [3–5].
- In 1950, Goeppert-Mayer and Jensen were awarded the Nobel prize for their discovery of the nuclear shell structure: how could we reconcile this with the liquid drop picture?
  - In the independent particle model (which was called shell model at the time but has little to do with what we call shell model today), a nucleus is a bunch of nucleons that occupy single-particle (s.p.) orbitals;
  - Gaps in the distribution of s.p. orbitals bring extra binding energy, hence the special role of doubly closed shell nuclei;
  - The shell structure changes with deformation, hence we can observe nuclei that are more bound in a deformed configuration than in a spherical one (deformed ground-state).
- The macroscopic-microscopic approach incorporates information about the shell structure (microscopic) into the liquid drop picture of the nucleus (macroscopic) [6,7].

Macroscopic-microscopic Models (2/4)
Components of the Total Energy

- Total energy is written

$$E(\boldsymbol{q}) = E_{\mathrm{mac}}(\boldsymbol{q}) + \delta R_{\mathrm{shell}}(\boldsymbol{q}) + \delta R_{\mathrm{pair}}(\boldsymbol{q})$$

- Macroscopic liquid drop energy

$$E_{\mathrm{mac}}(\boldsymbol{q}) = E_{\mathrm{vol}} + E_{\mathrm{surf}}(\boldsymbol{q}) + E_{\mathrm{asym}}(\boldsymbol{q}) + E_{\mathrm{Coul.}}(\boldsymbol{q})$$

- Shell correction

$$\delta R_{\mathrm{shell}}(\boldsymbol{q}) = \sum_n e_n - \left\langle \sum_n e_n \right\rangle$$

- Pairing correction

$$\delta R_{\mathrm{pair}} = E_{\mathrm{pair}} - \tilde{E}_{\mathrm{pair}}$$

- Shell and pairing corrections require a set of single-particle energies e_n: need to solve the Schrödinger equation

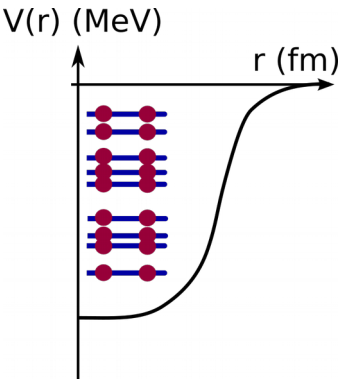
- The liquid drop contribution gives more than 99% of the mass of the nucleus; the shell correction gives the remaining 1% (the contribution of the pairing correction is of the order of 0.1%) [3–7].
  - Everything is relative: 1% of a nucleus with 1,000 MeV still gives about 10 MeV effect;
  - Overall quality of mass prediction in macro-micro method is of the order of 0.5 MeV!
- Shell and pairing effects are essential to understand the origin of, and quantitatively explain
  - Deformation effects: shape isomers, super- and hyper-deformed configurations;
  - Qualitative differences between the spectra of even-even and odd-even/even-odd/odd-odd nuclei.
- The liquid drop (and its more advanced formulation, the droplet model) has connection with nuclear matter properties: a nuclear liquid drop is nothing but a little chunk of nuclear matter enclosed in a finite volume [3-5].

Macroscopic-microscopic Models (3/4)
Single-Particle Degrees of Freedom

- (One-body) Schrödinger equation

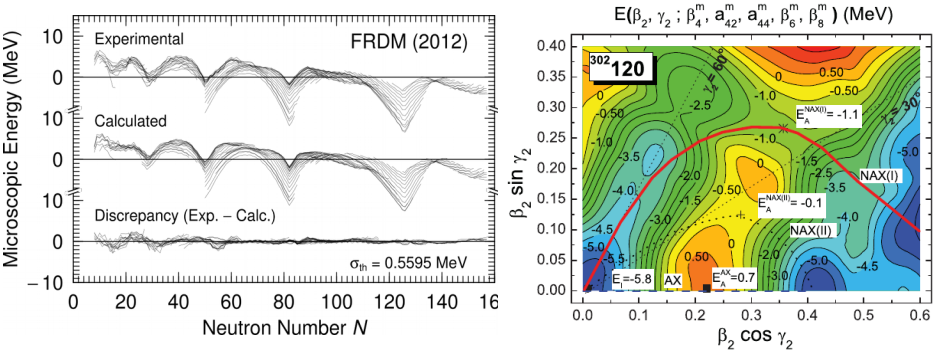
$$\left[-\frac{\hbar^2}{2m}\boldsymbol{\nabla}^2 + V(\boldsymbol{q})\right]\varphi_n(\boldsymbol{r}) = e_n\varphi_n(\boldsymbol{r})$$

- Mean-field potential can be Nilsson, Woods-Saxon, Folded-yukawa, etc.
- Solve BCS equation (for example) to compute occupation of s.p. states and extract pairing energy
- Collective variables are deformations that define the shape of the potential



- Solving the one-body Schrödinger equation is the most time-consuming part of the macroscopic-microscopic approach
- The macroscopic-microscopic approach defines a *nuclear phenomenology* based on *single-particle orbitals and collective deformations* as basic degrees of freedom. It is an extremely powerful tool
  - The distribution of s.p. levels is an excellent predictor of the stability of a nucleus (shell gaps), whether these gaps appear as a function of deformation or rotational frequency (high-spin physics);
  - Specific excited configurations can be constructed by multiple particle-hole excitations;
  - In some cases (odd nuclei in particular), the quantum numbers of s.p. states can be directly related to quantum numbers of the nucleus.
- There exist extensions of the macroscopic-microscopic approach to account for collective rotation (cranking model) and high-energy excitation (finite-temperature)[8–10].

Macroscopic-microscopic Models (4/4)
Examples



- The macroscopic-microscopic model can be applied to ground-state properties, e.g., masses
- Given a set of collective variables, we can calculate the potential energy surface, that is, the function *E(**q**)*

- The macroscopic-microscopic approach remains a very powerful tool to analyze properties of nuclei [11–13].
- The root of its success is in… the Pauli principle, which justifies seeing a nucleus as a bunch of independent particles in some average potential.
- Current state-of-the-art: liquid drop + shell correction + pairing correction (with/without particle number projection) for g.s. properties, QRPA with residual interaction for low-lying excited states; Langevin equations for collective dynamics.
- There is a lack of consistency between the various components of the model. For instance, one may think that the parameters of the liquid drop (related to nuclear matter) should be somewhat connected to the shell and pairing structure.
- Going beyond the current accuracy requires either more parameters (less predictive since they need to be fit) or … a real two-body Hamiltonian, which defeats the purpose of the method.

Nuclear Density Functional Theory (1/4)

Introduction

- Describe fission as emerging from nuclear forces and quantum many-body effects
- We cannot use modern nuclear forces as given by chiral effective field theory because direct solution of the full many-body problem is not possible for heavy nuclei
- Use effective methods:
  - Enforce a simple form of the many-body wave function for the system
  - Design an effective Hamiltonian such that

$$\langle \Psi | \hat{H} | \Psi \rangle \approx \langle \Phi | \hat{H}_{\text{eff.}} | \Phi \rangle$$

  - Minimize energy with respect to unknown parameters of $|\Phi\rangle$
- Examples of reference states $|\Phi\rangle$
  - Slater determinant (Hartree-Fock, HF)⇔System described by density matrix
  - Quasiparticle vacuum (Hartree-Fock-Bogoliubov, HFB)⇔System described by generalized density

- Density functional theory (DFT) in nuclear physics is different from DFT for atomic or molecular structure. In this presentation, I will not discuss any of these differences.
- Note that the term DFT has been popularized in the nuclear physics community after 1998. Coincidence: In 1998, Walter Kohn was awarded the Nobel Prize (in Chemistry) for the development of DFT for electrons…
- Other names frequently used for the set of methods that I am going to present: self-consistent mean-field theory (and beyond mean-field), energy density functional methods [14].
- Bottom line of nuclear DFT: turn the original nuclear many-body problem into something that is tractable.
- Key elements:
  - ✔ Choose a "simple" form for the many-body wave function describing the nucleus. In nuclear DFT, the most important of these "simple" forms is called a HFB vacuum, see next slide;
  - ✔ Determine the parameters of this simple form by solving an equation obtained by the requirement that the energy should be minimal with respect to these parameters. This will be the HFB equation.

Nuclear Density Functional Theory (2/4)

Hartree-Fock-Bogoliubov Theory

- Introduce Bogoliubov transformation (second quantization)

$$\begin{pmatrix} \beta \\ \beta^\dagger \end{pmatrix} = \begin{pmatrix} U^\dagger & V^\dagger \\ V^T & U^T \end{pmatrix} \begin{pmatrix} c \\ c^\dagger \end{pmatrix}$$

- What is the meaning of these quasiparticles?
  - Creating one quasiparticle is equivalent to moving a bunch of particles from some states to some other states (with some probability): Quasiparticles represent excitations of the system
  - HFB gives the ground-state of a nucleus and its excitations
- Unknown parameters of $|\Phi\rangle$: the matrices U and V
- We can express the energy as a function of only two objects, the density matrix ρ and pairing tensor κ, which are both expressed as function of U and V

- The HFB theory is the cornerstone of modern DFT. It was designed to describe pairing on the same footing as s.p. levels.
- It reduces the number of "unknowns" to just the density and the pairing tensor. Once these 2 objects are known (by solving the HFB equation), you can in principle calculate any observable.
- The key point is to find a way to determine U and V. This is equivalent to determining ρ and $\varkappa$; this is also equivalent to knowing the quasiparticle operators, hence the many-body wave function

$$|\Phi\rangle = \prod_\mu \beta_\mu |0\rangle$$

  which obeys the fundamental property $\beta_\nu |\Phi\rangle = 0, \forall \nu$
- The density matrix and pairing tensors are defined by

$$\rho_{ij} = \langle \Phi | c_j^\dagger c_i | \Phi \rangle, \qquad \kappa_{ij} = \langle \Phi | c_j c_i | \Phi \rangle,$$

- In other words, look at the overlap between the wavefunction obtained by moving a particle from state i from to state j with the original wave function: it is the element (i,j) of the density matrix.
- We can show that in the HFB theory they are actually given by

$$\rho_{ij} = \left(V^* V^T\right)_{ij}, \qquad \kappa_{ij} = \left(V^* U^T\right)_{ij}$$

Nuclear Density Functional Theory (3/4)

Single- and Multi-Reference Energy Density Functional Theory

- Single-reference energy density functional approach (SR-EDF) is built on top of the HFB theory
  - Choose an effective Hamiltonian or, equivalently, a form for the energy dependence E[ρ,κ] (Skyrme, Gogny)
  - Solve the HFB equation in order to determine ρ and κ
- Why the name? Because it only depends on one reference state…
- What does it have to do with fission?
  - Solve HFB equation with constraints on the expectation value of multipole moments operators: potential energy surfaces, fission barriers
  - Same philosophy as macro-micro, only the energy is obtained differently
- Multi-reference EDF
  - Take two different reference states (say, 2 different deformations)
  - Energy is now functional of transition densities

- Once we have chosen a nuclear Hamiltonian, we can express its expectation value on the HFB ground-state. Suppose we take only a two-body Hamiltonian, we find

$$E = \sum_{ij} t_{ij}\rho_{ji} + \frac{1}{2}\sum_{ij}\Gamma_{ij}\rho_{ji} + \frac{1}{2}\sum_{ij}\Delta_{ij}\kappa^*_{ij}$$

- With the mean-field potential and pairing field given by

$$h_{ij} = t_{ij} + \Gamma_{ij}, \quad \Gamma_{ij} = 2\frac{\partial E}{\partial \rho_{ji}}, \quad \Delta_{ij} = 2\frac{\partial E}{\partial \kappa^*_{ij}}$$

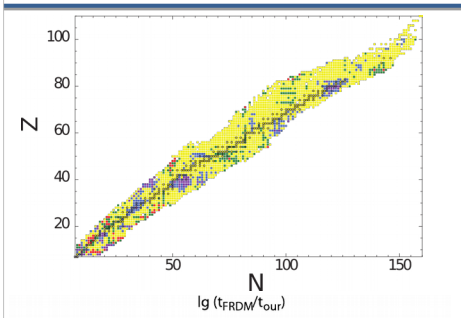
- Imposing that the energy be a minimum with respect to variations of ρ and ϰ (remember: these are the only "parameters") leads to the HFB equation, which can be put into the form of a pseudo-eigenvalue problem (here written with some constraints Q_n)

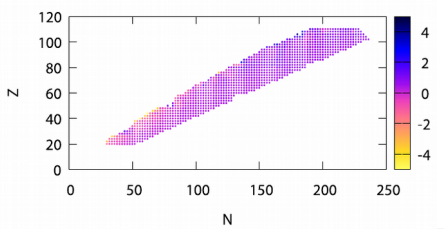
$$\begin{pmatrix} h - \sum_n \lambda_n Q_n & \Delta \\ -\Delta^* & -h^* + \sum_n \lambda_n Q_n \end{pmatrix} \begin{pmatrix} U_\mu \\ V_\mu \end{pmatrix} = E_\mu \begin{pmatrix} U_\mu \\ V_\mu \end{pmatrix}$$

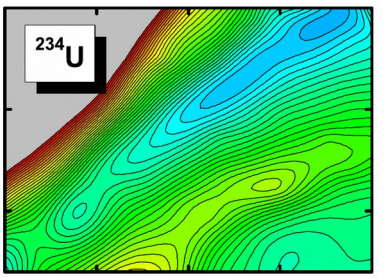
- Note that both mean-field and pairing field depends on the eigenvectors through ρ and ϰ: this is an example of a non-linear eigenvalue problem.

Nuclear Density Functional Theory (4/4)

Examples

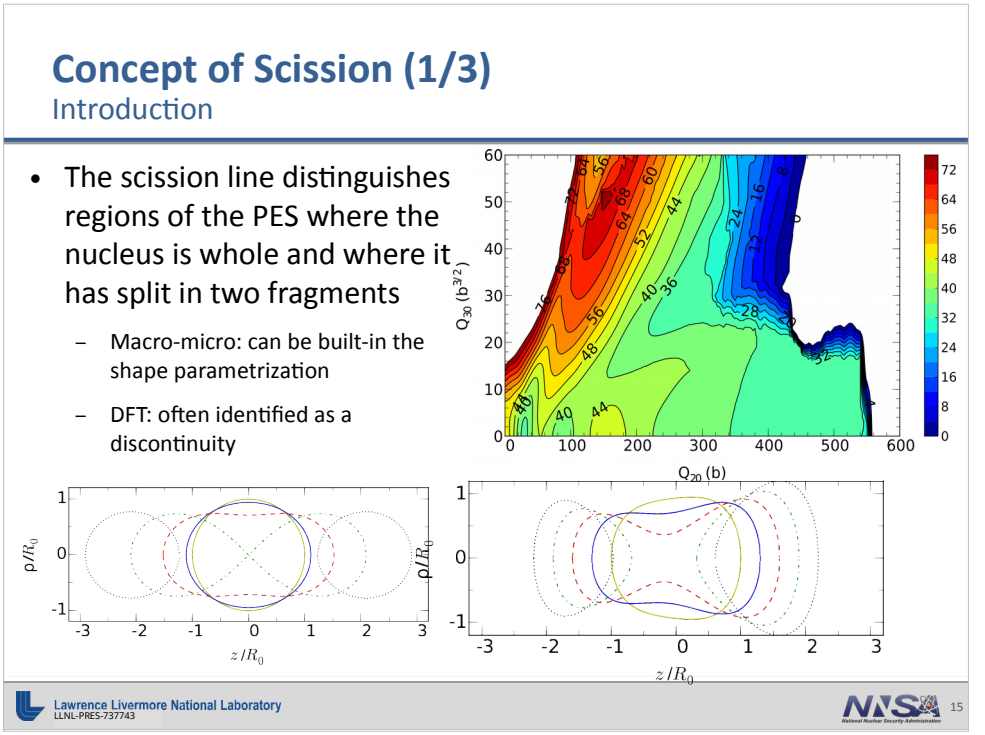






- DFT can now be applied to g.s. properties (masses), decays (beta-decay)
- Potential energy surfaces can be computed easily

- There are probably about a dozen computer programs on the market that can solve the HFB equation under various approximations/conditions [15–22]
  - Solve them as presented before, that is, "in a basis" (typically, the harmonic oscillator basis for energy functionals that can be Skyrme- or Gogny-like;
  - Solve them by direct integration in coordinate space (beyond the scope of this lecture);
  - Assume, or not, that specific symmetries are present in the system. For example, that the densities are axially symmetric.
- Solving the HFB equation for one nucleus gives its ground-state energy, hence its mass; repeating it for all known nuclei gives you a full mass table. In the same run, you can calculate, e.g., the r.m.s. radius of the system, or its (intrinsic) quadrupole moment, or (a not so great approximation of) its first excited state, etc.
- The HFB solution is also the starting point for several other theories capable of describing beta-decay (random phase approximation), shape coexistence (generator coordinate method), etc.

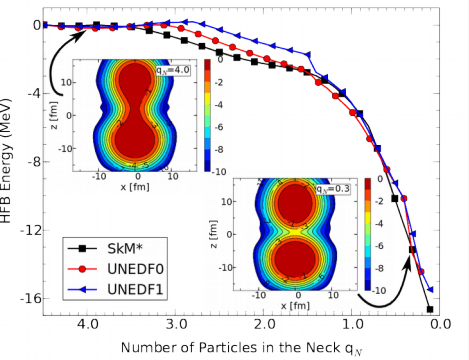


- The potential energy surface (PES) of the fissioning nucleus (whether computed in the macro-micro or DFT approach) encapsulates how the energy changes with the deformation(s) of the system.
- The scission line (in a 2D PES, the scission (N-1)d-surface in a Nd-PES) defines the frontier between regions of the PES where the nucleus is whole and regions where it has split [23].
- In macroscopic-microscopic models, the nuclear shape has to be parametrized. All parametrizations have a finite number of parameters, and some of them allow going continuously from a single compact object to 2 fragments.
- In DFT, the nuclear shape is automatically determined by the requirement that some constraints on multipole moments are satisfied and that the energy be minimal with respect to every other multipole moment: a DFT PES is a projection of an infinitely-dimensional "surface" into the finite-dimensional collective space [24].

Concept of Scission (2/3)

Continuous scission

- Sharp, geometric scission unrealistic
  - Coulomb forces take over nuclear forces before neck vanish
  - DFT: scission often viewed as discontinuity but only because finite number of collective variables
- More realistic descriptions imply scission (radius, density, etc.) becomes a parameter of the calculation



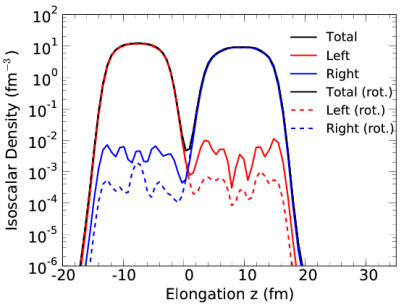
$$\langle \hat{Q}_N \rangle = \int d^3\mathbf{r}\ \rho(\mathbf{r}) e^{-\left(\frac{z-z_N}{a}\right)^2}$$

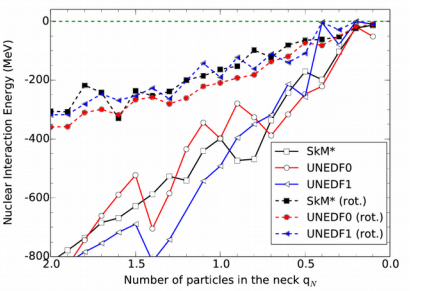
- In DFT, the concept of scission poses serious problems [25–28]:
  - Describing all possible 3D shapes require an infinity of parameters (think, multipole expansion) and there is no evidence that the shape of the system should be allowed to change in a discontinuous way – contrary to what happens in practice;
  - By virtue of the variational principle, fission fragments come out in their ground-state as soon as they are far enough from one another that the nuclear interaction between them becomes negligible.
- The concept of scission is, therefore, an artifact of using a *static*, *finite-dimensional* representation of the nuclear shape: if we add more collective variables, these discontinuities will be removed: where will scission be, then?
- This problem is specific to DFT, but the macro-micro has its own set of difficulties:
  - Sharp nuclear surfaces are incompatible with experimental evidence that the density has a diffuseness;
  - Coulomb forces may render the nucleus unstable against fission before we reach the point where the two fragments are separated: scission must occur before what geometry tells us [29].

Concept of Scission (3/3)

Scission and quantum entanglement

- Simple, user-defined criteria for scission often ignore
  - Quantum nature of fission fragments and neck region: antisymmetry
  - Finite-range of nuclear and Coulomb forces
- Adiabatic theory not adapted to describe dynamical, non-equilibrium process such as scission





- Wait, there is more: the nucleus is a quantum-mechanical system, with a density of particles that decays exponentially outside the nucleus – but never goes, mathematically speaking, to 0.
- Therefore, the density of particles in the left fragment extends into the right fragment and vice-versa: the two are *entangled* – in a quantum mechanical sense.
- Fission fragment observables (energy, deformation, spin, etc.) depend crucially on the characteristics of this entanglement.
- In fact, mathematically, you can perform a unitary transformation of the whole fissioning system that leaves it invariant but completely changes the properties of the fragments: fission fragments are not uniquely defined in the adiabatic theory!
- This fascinating aspect of fission is not visible in macroscopic-microscopic approaches, but its practical consequences are the same: every prediction of fission fragment yields very sensitively depend on a somewhat arbitrary definition of scission configurations [26-28].

Fission Dynamics

- Classical Dynamics (2)
- Quantum Dynamics – TDGCM (3)
- Quantum Dynamics – TDDFT (3)

LLNL-PRES-737743

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

Lawrence Livermore National Laboratory

The Scission Point Model

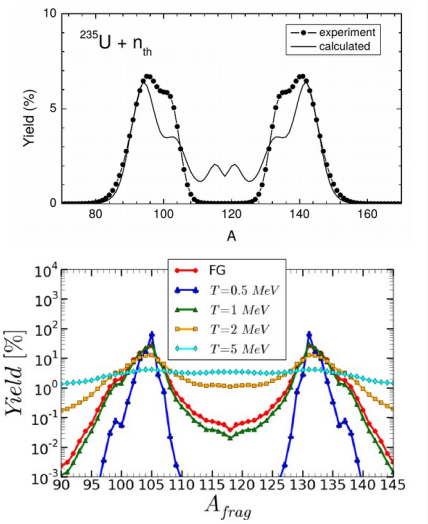
Statistical Approximation to Fission Dynamic

- Static picture exclusively based on the structure of the potential energy surface at scission (including the fragment characteristics)
- Probability of fission is simply related to (Wilkins)

$$\propto \int d\boldsymbol{q}_1 \int d\boldsymbol{q}_2 e^{-V(\boldsymbol{q}_1,\boldsymbol{q}_2;\alpha)/T}$$

or the level densities of the two fragments (SPY)

$$\propto \int d\boldsymbol{q}_1 \int d\boldsymbol{q}_2 \rho_1(\boldsymbol{q}_1;\alpha)\rho_2(\boldsymbol{q}_2;\alpha)$$



- The scission point was originally invented by Wilkins in 1976 and relied on calculations of potential energy surfaces within the macroscopic-microscopic picture and the canonical ensemble for the statistical phase space (Boltzman factor)[30].
- The CEA recently proposed a modification of the scission point model based on the explicit calculation of level densities for the fragments (=microcanonical ensemble)[31].
- The main advantages of scission point models are
  - To skip entirely the dynamical phase of the calculation of FPY;
  - To give a average-to-good reproduction of total kinetic energy.
- The downsides are
  - An overall poor-to-average reproduction of experimental data – not as good as explicitly time-dependent approaches;
  - A strong dependence on scission configurations, including the position of the scission (acknowledged by scission point practitioners) but also the entanglement of fission fragments (not necessarily realized).

Classical Dynamics (1/2)

Langevin equations

- How to extract fission product yields from the knowledge of the potential energy surface?
  - Analogy with classical theory of diffusion
  - Collective variable = generalized coordinate
  - Define related momentum
- Langevin equations

Fluctuation-dissipation theorem

$$\sum_k \Theta_{ik}\Theta_{kj} = \Gamma_{ij}T$$

$$\dot{q}_\alpha = \sum_\beta B_{\alpha\beta}p_\beta,$$

Friction tensor

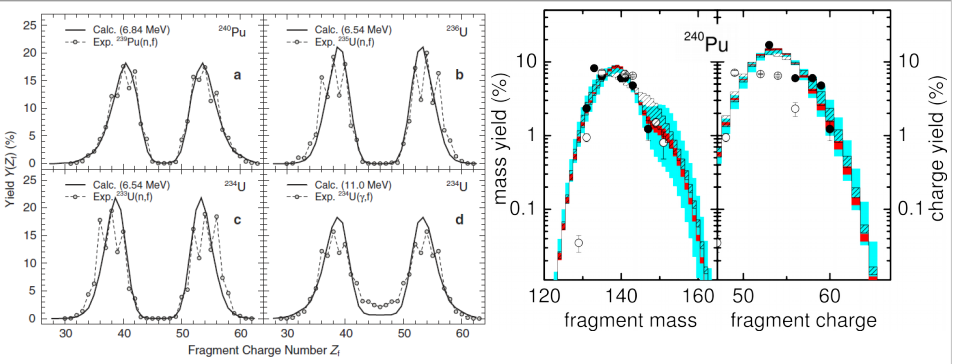
Random force

$$\dot{p}_\alpha = -\sum_{\beta\gamma}\Gamma_{\alpha\beta}B_{\beta\gamma}p_\gamma + \sum_\beta \Theta_{\alpha\beta}\xi_\beta(t) - \frac{1}{2}\sum_{\beta\gamma}\frac{\partial B_{\beta\gamma}}{\partial q_\alpha}p_\beta p_\gamma - \frac{\partial V}{\partial q_\alpha}$$

- The Langevin equation is a classical approach to nuclear dynamics
  - Nucleus is assumed point-like;
  - The PES determines the entire phase space.
- The presence of the random force allows to simulate quantum tunneling effects: if this term is absent, the particle can only go downhill; with this term, it can "climb" and go over barriers.
- Solving the Langevin equation requires the knowledge of the collective mass tensor $B_{\alpha\beta}$ and of the friction tensor $\Gamma_{\alpha\beta}$; there are theories available to compute the mass tensor [23,32,33], but the friction tensor is often more phenomenological.
- The inputs to the Langevin equations can be computed with any model capable of producing at each point of the collective space, the energy, the collective mass tensor and the friction tensor.
- There are variants of the Langevin equation in the literature where the mass tensor is set to 0 (strongly-damped evolution)[34–36].
- The Langevin equations are non-deterministic and give trajectories in phase space; they can be related to a deterministic Kramers equation, which gives the evolution of the probability distribution function of being at any point in phase space at time t [37].

Classical Dynamics (2/2)

Practical examples



- Start beyond the saddle point (or close enough)
- Build trajectories through the collective space by generating at each step the needed random variable
- Enough trajectories (in the thousands) allow reconstructing FPY

- Solving the Langevin equation is relatively cheap compared to the calculation of the PES itself (especially in more than 2 dimensions).
- Reconstructing fission fragment distributions, however, requires large samples of Langevin trajectories (at least several thousands, if not millions) to get good statistics.
- Note that a given initial point on the PES generates an infinity of possible trajectories thanks to the presence of the random force. However, the tails of the distribution (very asymmetric fission with a very light and a very heavy fragment) requires different initial conditions from the peaks of the same distribution.
- Note that the Langevin equation is a great tool to obtain distributions for spontaneous fission: the initial point is chosen not on the saddle (the top of the second barrier), but at the outer turning point of the WKB theory [38].

Quantum Dynamics - TDGCM (1/3)

Computing the flow of probability in the collective space

- Ansatz for the time-dependent many-body wave function

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

- Minimization of the time-dependent quantum mechanical action + ansatz + Gaussian overlap approximation

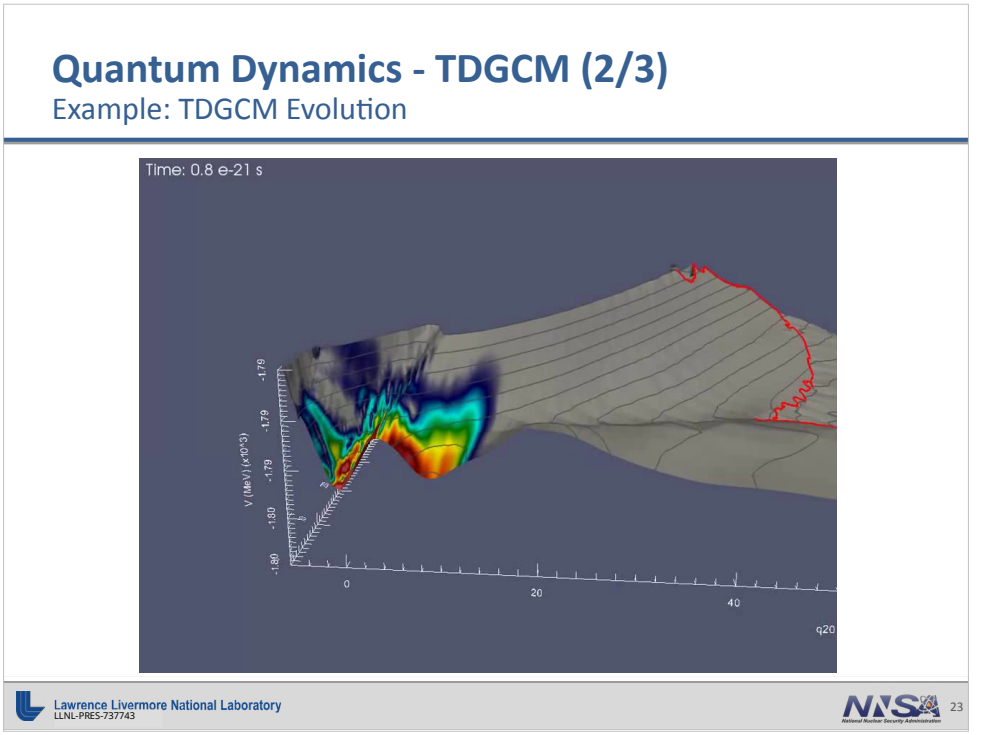
$$i\hbar\frac{\partial}{\partial t}g(\mathbf{q},t) = \left[-\frac{\hbar^2}{2}\sum_{kl}\frac{\partial}{\partial q_k}B_{kl}\frac{\partial}{\partial q_l} + V(\mathbf{q})\right]g(\mathbf{q},t)$$

- Interpretation
  - $g(\mathbf{q},t)$ is probability amplitude to be at point **q** at time t
  - Related probability current
  - Flux of probability current through scission line gives yields

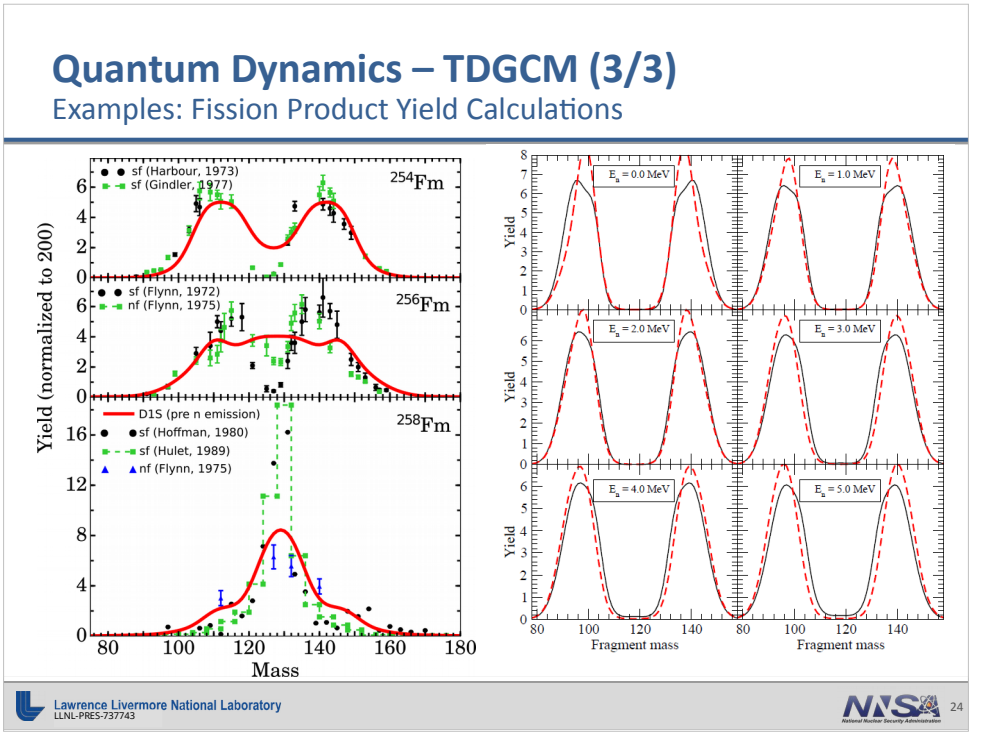
- The TDGCM is a fully quantum-mechanical approach to treat nuclear dynamics which is an extension of the multi-reference EDF very briefly mentioned earlier [39–43].
- It provides a fully-consistent description of static and dynamics properties: the same energy functional is used to determine the potential energy surface and the collective inertia tensor, which are the only inputs to the dynamics.
- The TDGCM equation has not been solved exactly (yet) to produce fission product yields. Instead, people have used the Gaussian Overlap Approximation (GOA), which assumes that

$$\langle\Phi(\boldsymbol{q})|\Phi(\boldsymbol{q}')\rangle \approx e^{-(\boldsymbol{q}-\boldsymbol{q}')^2}$$

It is the GOA approximation that allows to recast the whole time-dependent evolution into a simple, collective Schrödinger equation [23].



The movie shown here is an example of time-evolution of the collective wave-packet, $g(\mathbf{q},t)$, that is, of the solution of the collective Schrödinger equation, see slide 22. Calculations were done for the neutron-induced fission of ^{239}Pu, that is, the collective wave-packet represents the ^{240}Pu compound nucleus. The collective space is two-dimensional and characterized by the axial quadrupole and axial octupole moments. The red line represents the scission line, defined here as by the criterion that the number of points in the neck region is lower than 4. For more details on the calculation, see [43] and references therein.



The figures represent state-of-the-art TDGCM calculations of fission fragment yields for neutron-induced fission. The figure on the left (unpublished) shows the transition between symmetric and asymmetric fission in Fermium isotopes. It was obtained with the code FELIX developed jointly by CEA and LLNL (D. Regnier, lead developer). The figure on the right shows the evolution of fission fragment yields in ^{239}Pu(n,f) as a function of the energy of the initial wave-packet; see [42] for additional details.

Quantum Dynamics – TDDFT (1/2)

Brief Introduction

- Main limitation of Langevin and TDGCM: adiabaticity is built-in
  - Need to precompute potential energy surfaces (costly)
  - Invoke arbitrary criteria for scission
  - Does not (easily) include dissipation = exchange between intrinsic (=single-particle) and collective degrees of freedom
- Solution: Generalize DFT to time-dependent processes
- Start from many-body Schrödinger equation

$$i\hbar\frac{\partial|\Psi(t)\rangle}{\partial t} = \hat{H}|\Psi(t)\rangle$$

- Insert approximation that many-body state is q.p. vacuum at all time

$$i\hbar\frac{\partial R}{\partial t} = \left[\mathcal{H},\mathcal{R}\right]$$

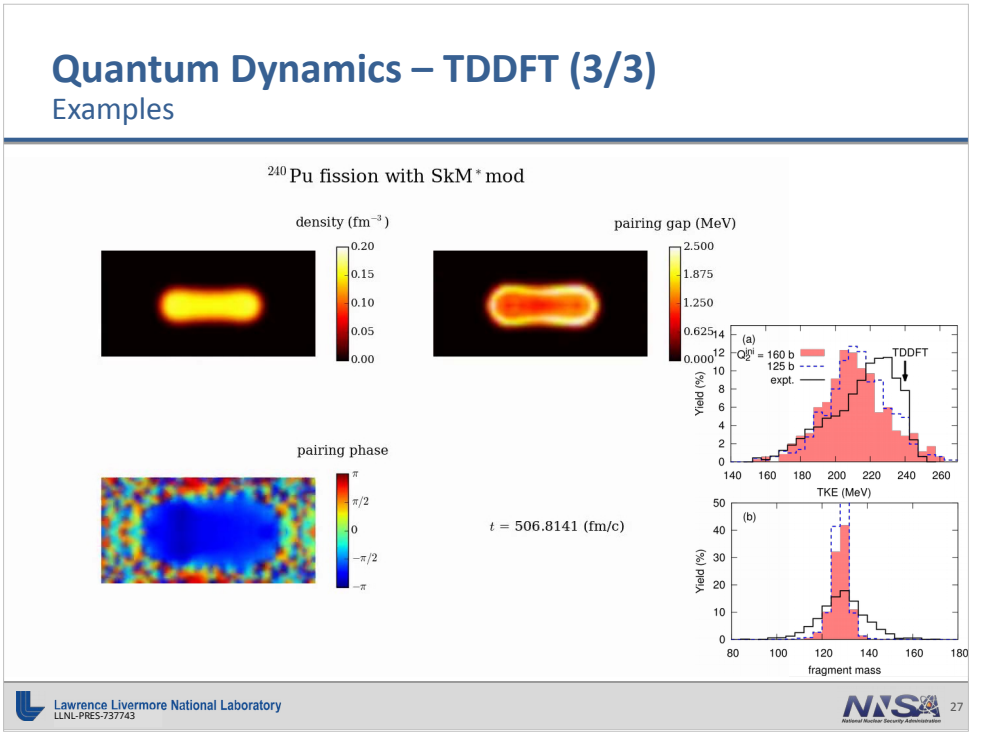
- Applications of time-dependent DFT to fission are still in their infancy – although the first application of time-dependent Hartree-Fock was done back in 1978.
- TDDFT removes many of the limitations of adiabatic approaches, most notably the need for the adiabatic approximation itself, and provides a real-time description of the fissioning nucleus.
- It can be viewed as the quantum-mechanical analogue of the Langevin equation: solving the TDDFT equation for one initial condition gives a "trajectory" in some phase space.
- Most work until now has been based on time-dependent Hartree-Fock (TDHF); very recently (past 2 years), there have been the first applications of TDDFT with pairing correlations built-in (TDHF+BCS, TDHFB) or with some stochastic fluctuations included [44–47].

Quantum Dynamics – TDDFT (2/3)

Advantages and Limitations

- Advantages
  - TDDFT does not require adiabaticity, total energy is conserved: diabatic excitation of s.p./q.p. states
  - Dynamic shape evolution: normal and pairing vibrations, giant resonances
  - Produces 'naturally' excited fission fragments
- Limitations
  - Computational cost is enormous (especially for TDHFB)
  - Nucleus cannot tunnel through (semi-classical): not adapted to SF
  - Need HFB solver in coordinate space
- Computing FPY from TDDFT by sampling trajectories is in principle possible but would require computational resources at or beyond exascale (100x what we have now)

- By providing an explicit mechanism to describe dissipation, TDDFT promises to give much more accurate estimates of fragment kinetic energies.
- TDDFT does not really need a scission point – or rather, it is much less sensitive to the definition of scission. This is because the total energy is conserved, hence the fragments will remain excited even after they have split. In the adiabatic picture based on the HFB theory (or the macroscopic-microscopic approach), the fragments dive in their ground-state as soon as they are far from one another.
- Three reasons why the computational cost is enormous for TDHFB
  - We need precise solution of the TDHFB equation even when the two fragments have separated. This requires covering of the order of 25 fm range;
  - Some HFB quasiparticles (with energy $E_k > |\lambda|$) are delocalized, meaning they extend over all space. Once again, this means that one must be able to describe accurately what is happening "far" from the center of the fissioning nucleus;
  - Quasiparticle energies evolve as a function of deformation, hence as a function of time. This means that one must keep track of a very large number of q.p. orbitals.



The movie on the left (unpublished) shows the real-time nuclear dynamics of ^{240}Pu computed from time-dependent density functional theory, including a full treatment of pairing correlations. The theoretical and computational frameworks are the same as in [46]. The figure on the right shows the fission fragment distributions for Fermium isotopes obtained with a particular variant of TDDFT called stochastic time-dependent Hartree-Fock; see [47] for details.

Conclusions

- Navigating the zoo of methods
- Perspectives

LLNL-PRES-737743

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

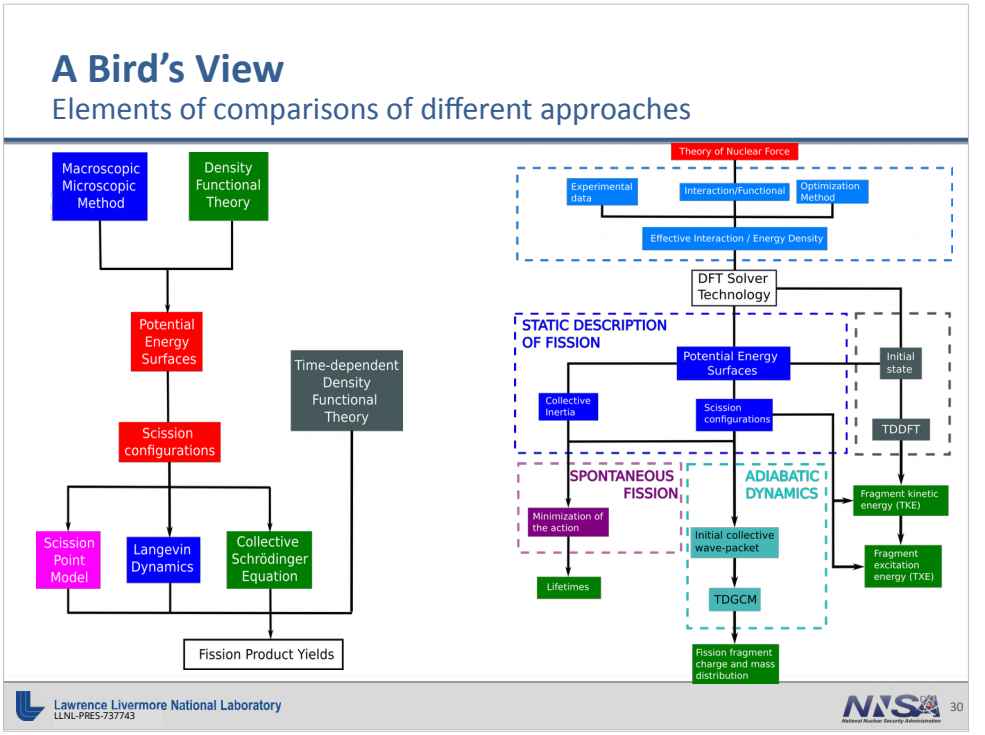
Lawrence Livermore National Laboratory

A Bird's View

Elements of comparisons of different approaches

	Quantum	Description	Adiabaticity	Observable	Computational cost
Scission point model	Half	Static	Yes	Fission yields	Low
Macro-micro + Langevin	Half	Static + dynamic	Yes	Fission events	Low
DFT + TDGCM	Full	Static + dynamic	Yes	Fission yields	Moderate-high
TDDFT	Full	Dynamic	No	Fission events	Very high

- The cardinal rule of nuclear theory: there is no perfect theory, no miracle recipe, no "there is only one way"...
- Each approach has its own merits, its own strengths and its own weaknesses. Practitioners of a given approach are experts at both selling its strengths and downplaying its weaknesses.
- Factors to take in consideration
  - Can the model deliver the *observables* that I want?
  - Can it deliver them with the *accuracy* and *precision* that I need for my applications (note for non-english speakers: these are two distinct concepts)?
  - What is its *demonstrated* predictive power and its *potential* predictive power?
  - What are the *computational* resources needed?
  - What is the *time-to-solution*?
- Note that there are connections between these various approaches: the macro-micro approach provides the PES for the scission point model; the Langevin equations can be solved on top of a DFT-generated PES, etc.



- Historically, the scission point model was the first attempt at *predicting* fission product yields based on a model of the nucleus (in contrast to fitting data).
- Until the beginning of the 21st century, the macroscopic-microscopic method coupled with the Langevin equation was the only theoretical approach that was doable in practice.
- Over the past 2 decades, microscopic theories based on time-dependent extensions of DFT (TDGCM and more recently TDDFT) have become very competitive – even though they still require substantial computational resources.
- Ballpark estimate of accuracy on FPY predictions for current models: of the order of 30% for actinides
- Some of the challenges to go below 5% accuracy
  - Know where you stand: *Quantify and propagate uncertainties* of nuclear models. To which of their components are the models most sensitive?
  - Adiabatic approaches: everything depends on scission. What are the static nuclear properties at scission (which is really non-static by nature)?
  - Non-adiabatic approaches: how to include information on collective degrees of freedom into TDDFT?

Bibliography

[1] L. Meitner and O. R. Frisch, Nature **143**, 239 (1939).

[2] N. Bohr and J. A. Wheeler, Phys. Rev. **56**, 426 (1939).

[3] W. D. Myers and W. J. Swiatecki, Nucl. Phys. **81**, 1 (1966).

[4] W. D. Myers and W. J. Swiatecki, Ann. Phys. **55**, 395 (1969).

[5] W. D. Myers and W. J. Swiatecki, Ann. Phys. **84**, 186 (1974).

[6] M. Brack, J. Damgaard, A. S. Jensen, H. C. Pauli, V. M. Strutinsky, and C. Y. Wong, Rev. Mod. Phys. **44**, 320 (1972).

[7] M. Bolsterli, E. O. Fiset, J. R. Nix, and J. L. Norton, Phys. Rev. C **5**, 1050 (1972).

[8] M. J. A. de Voigt, J. Dudek, and Z. Szymański, Rev. Mod. Phys. **55**, 949 (1983).

[9] T. R. Werner and J. Dudek, At. Data Nucl. Data Tables **59**, 1 (1995).

[10] J. Dudek, B. Herskind, W. Nazarewicz, Z. Szymanski, and T. R. Werner, Phys. Rev. C **38**, 940 (1988).

[11] P. Möller, A. J. Sierk, T. Ichikawa, and H. Sagawa, At. Data Nucl. Data Tables **109**, 1 (2016).

[12] P. Möller, R. Bengtsson, B. G. Carlsson, P. Olivius, and T. Ichikawa, Phys. Rev. Lett. **97**, 162502 (2006).

[13] P. Möller, J. R. Nix, and K. L. Kratz, At. Data Nucl. Data Tables **66**, 131 (1997).

[14] M. Bender, P.-H. Heenen, and P.-G. Reinhard, Rev. Mod. Phys. **75**, 121 (2003).

[15] M. V. Stoitsov, N. Schunck, M. Kortelainen, N. Michel, H. Nam, E. Olsen, J. Sarich, and S. Wild, Comput. Phys. Commun. **184**, 1592 (2013).

[16] N. Schunck, J. Dobaczewski, W. Satuła, P. Bączyk, J. Dudek, Y. Gao, M. Konieczka, K. Sato, Y. Shi, X. B. Wang, and T. R. Werner, Comput. Phys. Commun. **216**, 145 (2017).

[17] T. Nikšic, N. Paar, D. Vretenar, and P. Ring, Comput. Phys. Commun. **185**, 1808 (2014).

[18] W. Ryssens, V. Hellemans, M. Bender, and P.-H. Heenen, Comput. Phys. Commun. **187**, 175 (2015).

[19] J. Pei, M. Stoitsov, G. Fann, W. Nazarewicz, N. Schunck, and F. Xu, Phys. Rev. C **78**, 064306 (2008).

[20] S. Jin, A. Bulgac, K. Roche, and G. Wlazłowski, Phys. Rev. C **95**, 044302 (2017).

[21] B. G. Carlsson, J. Dobaczewski, J. Toivanen, and P. Veselý, Comput. Phys. Commun. **181**, 1641 (2010).

[22] K. Bennaceur and J. Dobaczewski, Comput. Phys. Commun. **168**, 96 (2005).

[23] N. Schunck and L. M. Robledo, Rep. Prog. Phys. **79**, 116301 (2016).

[24] N. Dubray and D. Regnier, Comput. Phys. Commun. **183**, 2035 (2012).

[25] L. Bonneau, P. Quentin, and I. N. Mikhailov, Phys. Rev. C **75**, 064313 (2007).

[26] W. Younes and D. Gogny, Phys. Rev. Lett. **107**, 132501 (2011).

[27] N. Schunck, D. Duke, H. Carr, and A. Knoll, Phys. Rev. C **90**, 054305 (2014).

[28] N. Schunck, D. Duke, and H. Carr, Phys. Rev. C **91**, 034327 (2015).

[29] K. T. R. Davies, R. A. Managan, J. R. Nix, and A. J. Sierk, Phys. Rev. C **16**, 1890 (1977).

[30] B. D. Wilkins, E. P. Steinberg, and R. R. Chasman, Phys. Rev. C **14**, 1832 (1976).

[31] J.-F. Lemaître, S. Panebianco, J.-L. Sida, S. Hilaire, and S. Heinrich, Phys. Rev. C 92, 034617 (2015).

[32] M. Baranger and M. Veneroni, Ann. Phys. **114**, 123 (1978).

[33] P.-G. Reinhard and K. Goeke, Rep. Prog. Phys. **50**, 1 (1987).

[34] J. Randrup and P. Möller, Phys. Rev. Lett. **106**, 132503 (2011).

[35] J. Randrup, P. Möller, and A. J. Sierk, Phys. Rev. C **84**, 034613 (2011).

[36] J. Randrup and P. Möller, Phys. Rev. C **88**, 064606 (2013).

[37] Y. Abe, S. Ayik, P.-G. Reinhard, and E. Suraud, Phys. Rep. **275**, 49 (1996).

[38] J. Sadhukhan, W. Nazarewicz, and N. Schunck, Phys. Rev. C **93**, 011304 (2016).

[39] J. F. Berger, in Dyn. Wave Pack. Mol. Nucl. Phys., edited by J. Broeckhove, L. Lathouwers, and P. van Leuven (Springer Berlin Heidelberg, 1986), p. 21.

[40] J. F. Berger, M. Girod, and D. Gogny, Comput. Phys. Commun. **63**, 365 (1991).

[41] H. Goutte, J.-F. Berger, P. Casoli, and D. Gogny, Phys. Rev. C **71**, 024316 (2005).

[42] W. Younes and D. Gogny, Fragment Yields Calculated in a Time-Dependent Microscopic Theory of Fission (Lawrence Livermore National Laboratory (LLNL), Livermore, CA, 2012).

[43] D. Regnier, N. Dubray, N. Schunck, and M. Verrière, Phys. Rev. C **93**, 054611 (2016).

[44] C. Simenel and A. S. Umar, Phys. Rev. C **89**, 031601(R) (2014).

[45] G. Scamps, C. Simenel, and D. Lacroix, Phys. Rev. C **92**, 011602 (2015).

[46] A. Bulgac, P. Magierski, K. J. Roche, and I. Stetcu, Phys. Rev. Lett. **116**, 122504 (2016).

[47] Y. Tanimura, D. Lacroix, and S. Ayik, Phys. Rev. Lett. **118**, 152501 (2017).


Lawrence Livermore
National Laboratory