CEA, ESNT, Saclay,
Nuclear EDF method: going beyond the minefield
November 22nd, 2023

Multiconfigurational TDDFT model of nuclear dynamics: Some recent developments

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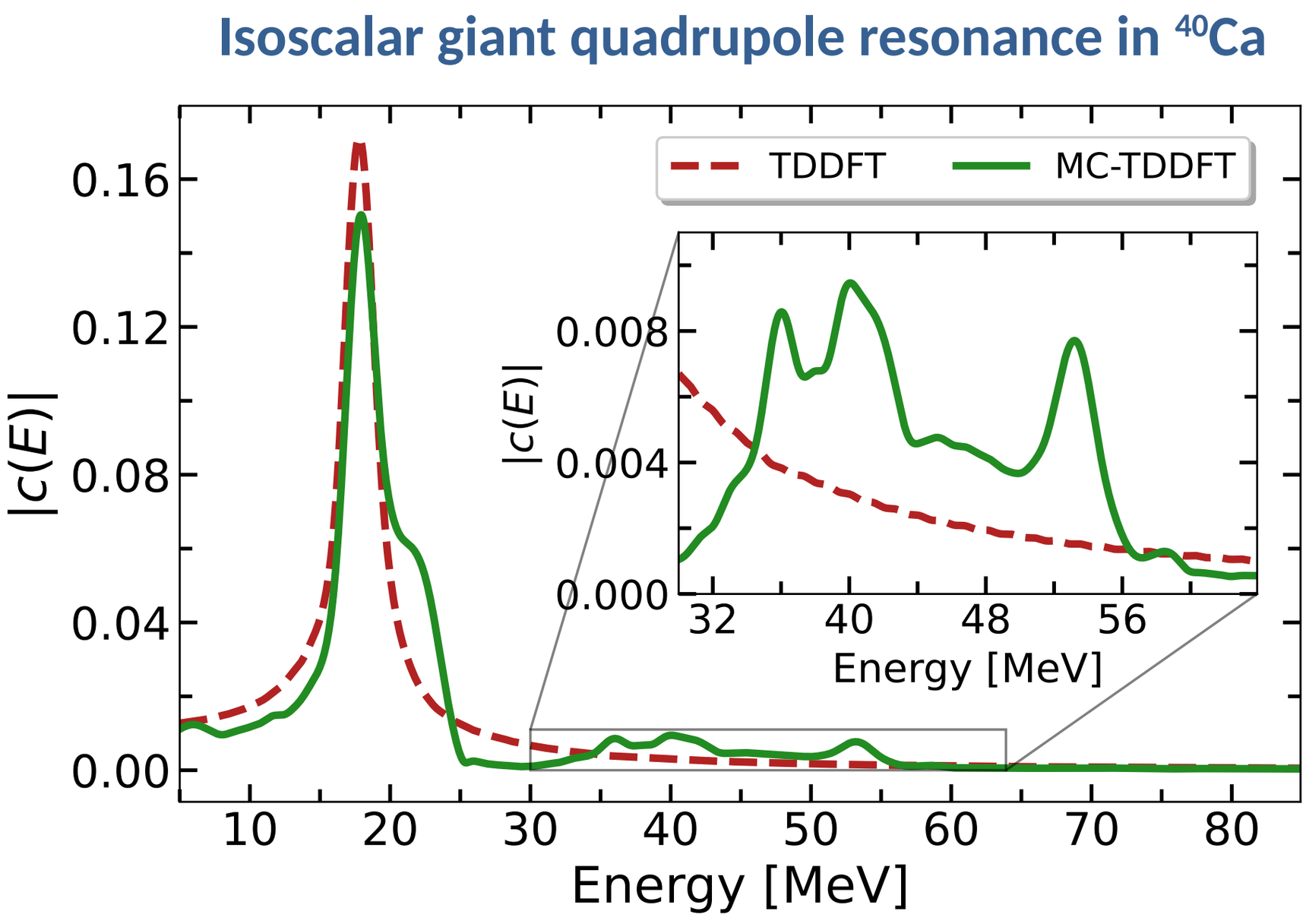
with **David Regnier** (*CEA, DAM*) and **Denis Lacroix** (*IJCLab*)

Peaks at nearly 2x and 3x the energy of the main ISGQR peak!



Outline

1. Introduction: Configuration-mixing EDF models
2. Multiconfigurational TDDFT model
3. First application: ISGQR multiphonons in ^{40}Ca
4. Several technical remarks
5. Conclusion

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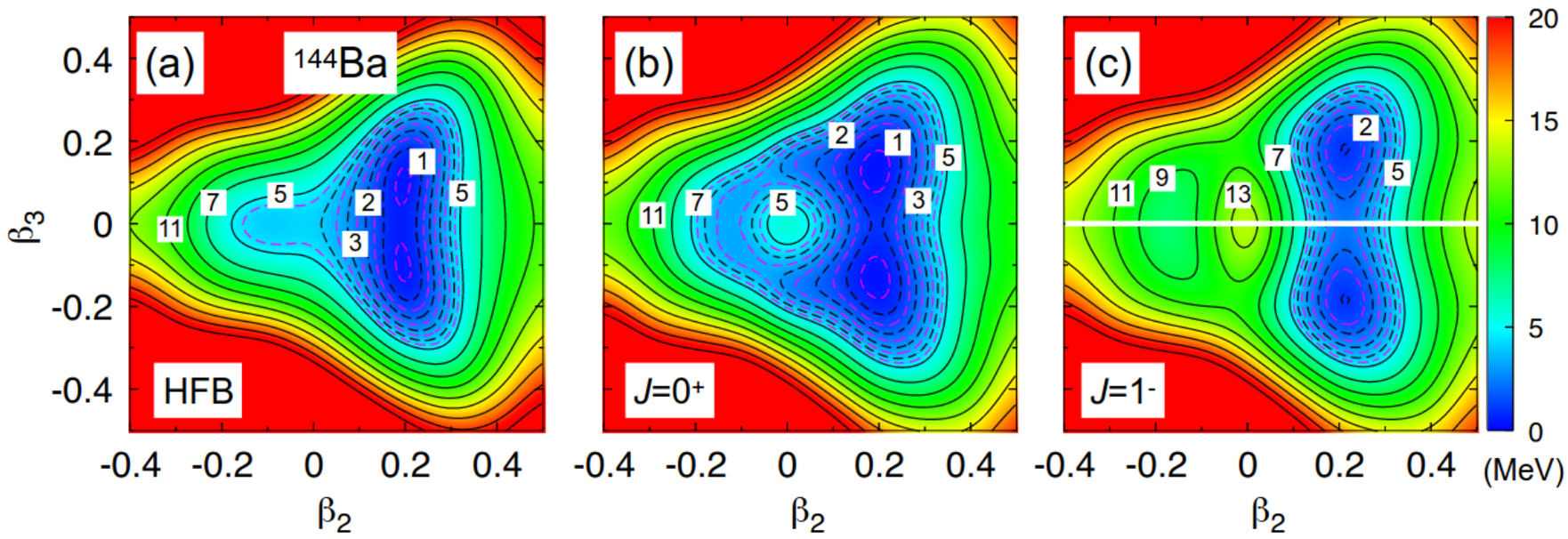
Configuration-mixing EDF models

Generator coordinate method for nuclear structure

Configuration mixing models have been used for decades in nuclear structure studies.

- Usually referred to as the **generator coordinate method** (GCM) or the **multi-reference energy density functional** (MR-EDF) theory
- Based on the **mean-field approximation** and the **Hill-Wheeler-Griffin's theory** from 1950s
  - The idea is to account for quantum fluctuations that are disregarded by the mean-field approach
  - The reference states are typically obtained by constrained HF+BCS or HFB calculations
  - Most applications use Skyrme, Gogny, or relativistic EDFs
  - With or without symmetry restoration prior to the mixing

Symmetry-restored potential energy surfaces of ^{144}Ba obtained with a Gogny EDF



R. Bernard, L. Robledo, T. R. Rodriguez, Phys. Rev. C 93, 061302(R) (2016) .

Configuration-mixing EDF models

Generator coordinate method for nuclear structure

- Starting from a mixed state, one ends up solving the **HWG equation**

$$|\Psi_\sigma^\alpha\rangle = \sum_{\mathbf{q}} f_\sigma^\alpha(\mathbf{q})\,|\Phi^\alpha(\mathbf{q})\rangle \rightarrow \sum_{\mathbf{q}'}\Big(\mathcal{H}^\alpha(\mathbf{q},\mathbf{q}') - E_\sigma^\alpha \mathcal{N}^\alpha(\mathbf{q},\mathbf{q}')\Big) f_\sigma^\alpha(\mathbf{q}') = 0$$

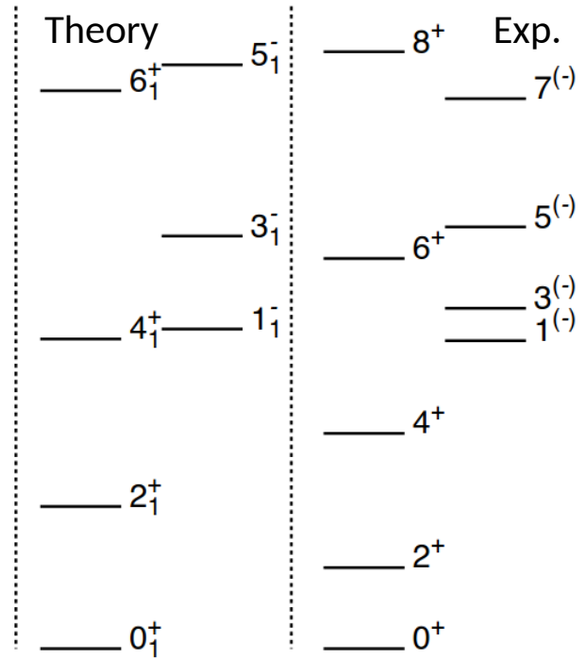
- This provides an access to various spectroscopic properties
  - Excitation spectra for different spins/parities
  - Collective w.f., used to calculate expectation values of observables

- The framework has been extensively applied in numerous studies and thoroughly reviewed in articles and texbooks

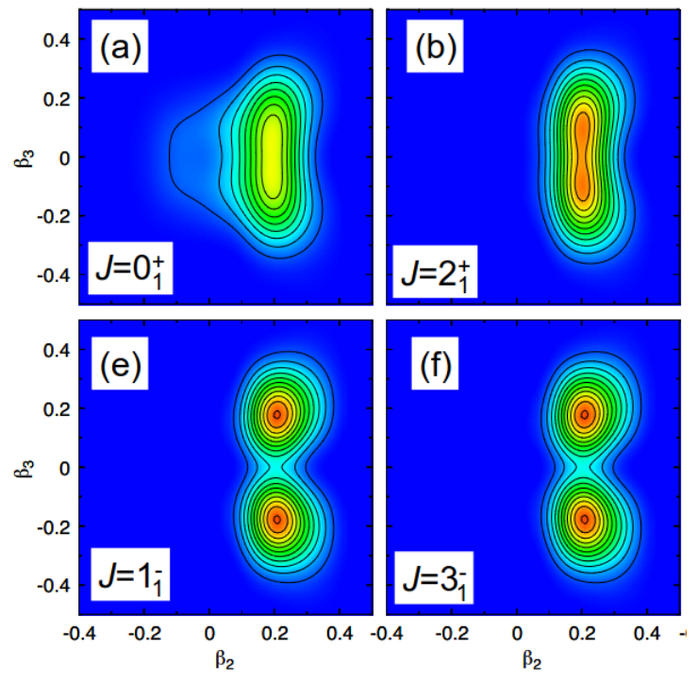
M. Bender, N. Schunck, J.-P. Ebran and Th. Duguet, Chapter 3 in "*Energy Density Functional Methods for Atomic Nuclei*" (2019)

- However, many formal and occasionally practical issues persist (i.e., the minefield)

GCM spectra of ^{144}Ba



Collective w.f. of ^{144}Ba



R. Bernard et al, PRC C 93, 061302(R) (2016) .

Configuration-mixing EDF models

Time-dependent generator coordinate method for nuclear dynamics

A popular extension for nuclear dynamics is the **time-dependent GCM** (TDGCM).

- Based on the same HWG theory, applied to nuclear reactions in the 80s
- Nuclear dynamics is described in terms of several collective coordinates
  - Motivated by the separation in time-scales (single-particle vs collective)
  - Considers a reduced Hilbert space spanned by collective coordinates (deformations, pairing, etc.)
  - Accounts for quantum fluctuations by mixing many **time-independent** basis states

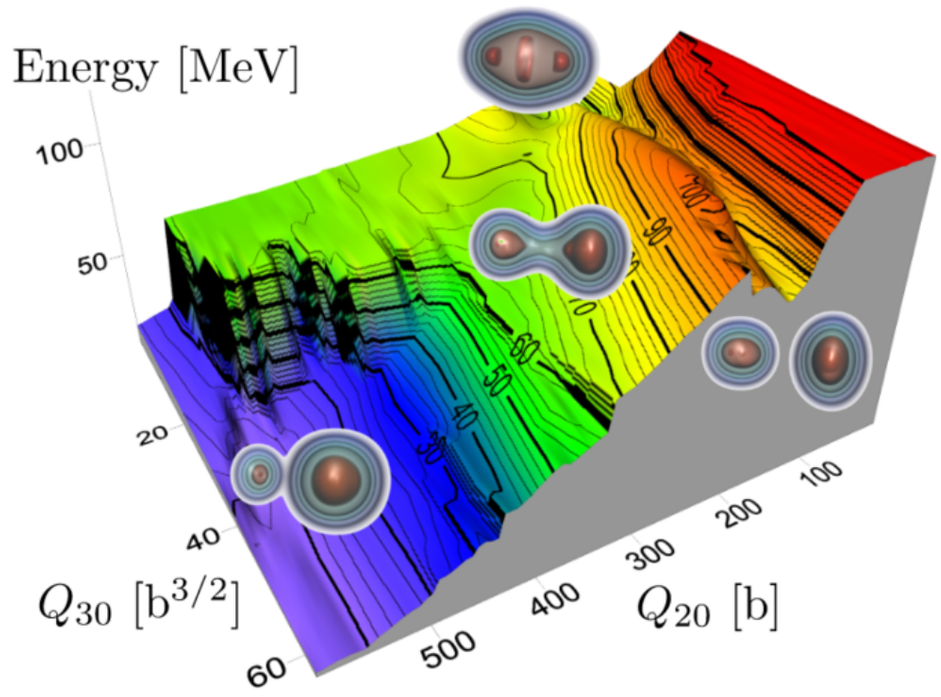
$$|\Psi(t)\rangle = \sum_{\mathbf{q}} f_{\mathbf{q}}(t)\,|\Phi_{\mathbf{q}}\rangle$$

- One ends up solving the time-dependent HWG equation

$$\sum_{\mathbf{q}'}\left(\mathcal{H}_{\mathbf{q}\mathbf{q}'} - i\hbar\mathcal{N}_{\mathbf{q}\mathbf{q}'}\frac{d}{dt}\right) f_{\mathbf{q}'}(t) = 0, \quad \forall\mathbf{q}$$

- Usually combined with the GOA, yielding a Schrödinger-like equation for the collective wave function

2D Potential Energy Surface of ^{240}Pu



N. Schunck and D. Regnier, PPNP **47**, 103963 (2022).

Configuration-mixing EDF models

Time-dependent generator coordinate method for nuclear dynamics

- In the past decade, TDGCM has been widely applied to fission

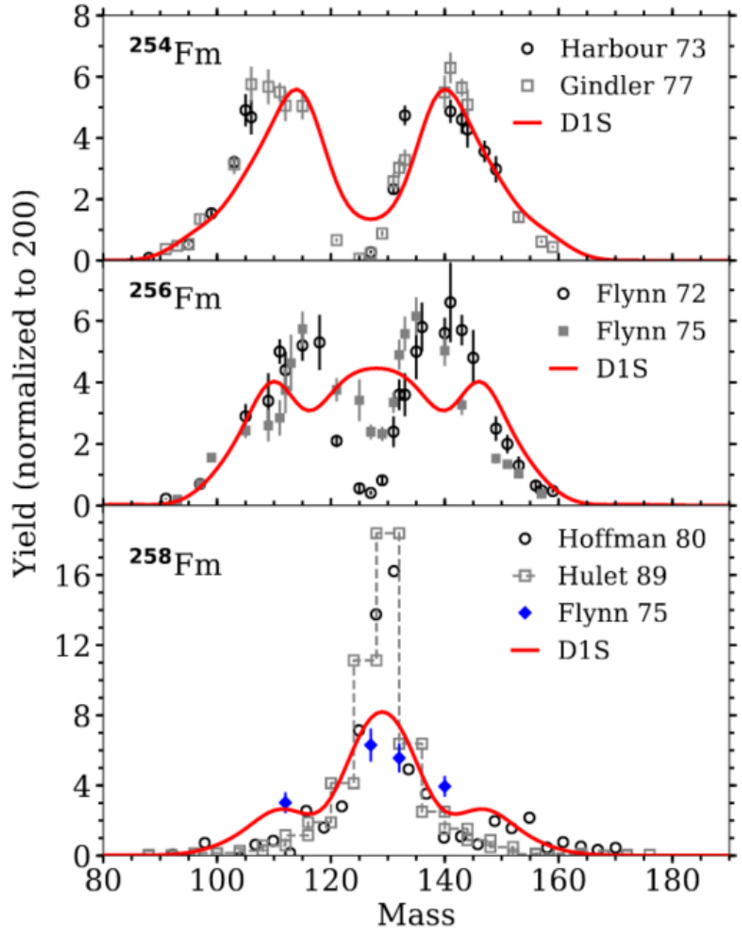
  W. Younes, D. Gogny, and J. F. Berger, "*A Microscopic Theory of Fission Dynamics Based on the Generator Coordinate Method*", Lecture Notes in Physics (2019).

  D. Regnier and M. Verriere, "*The Time-Dependent Generator Coordinate Method in Nuclear Physics*", Front. Phys., Vol. 8 (2020).

- It provides an excellent description of some observables (e.g., preneutron yields)

- However, some major issues remain:
  - ✗ Very large bases of time-independent states are needed
  - ✗ There is no dissipation mechanism like in TDDFT (qp excitations could be added, but not trivial)
  - ✗ For fission, the scission line is not uniquely defined
  - ✗ PES are often discontinuous in the reduced Hilbert space

From asymmetric to symmetric fission in fermium isotopes



D. Regnier *et al.*, PRC **99**, 024661 (2019).

Mixing of time-dependent, non-adiabatic configurations could account for some of these issues.

Configuration-mixing EDF models

Mixing of time-dependent configurations

- More generally, **both the mixing functions and the basis states can be made time-dependent**

$$|\Psi(t)\rangle = \sum_{\mathbf{q}} f_{\mathbf{q}}(t)\,|\Phi_{\mathbf{q}}(t)\rangle$$

In principle encompasses both the **dissipation** and **quantum fluctuation** aspects od dynamics

- Theoretical foundations of such a framework were laid out in the 1980s

P. G. Reinhard, R. Y. Cusson, K. Goeke, Nucl. Phys. A 398, 141 (1983).
P. G. Reinhard and K. Goeke, Rep. Prog. Phys. 50, 1 (1987).

- A few years ago, a couple of toy-model calculations were reported

D. Regnier and D. Lacroix, Phys. Rev. C 99, 064615 (2019).
N. Hasegawa, K. Hagino, Y. Tanimura, Phys. Lett. B 808, 135693 (2020).

- During 2023 a significant progress is made, **first applications in real nuclei**

Apr2023

PM, David Regnier, Denis Lacroix, Phys. Rev. C 108, 014620 (2023).
B. Li, D. Vretenar, T. Nikšić *et al.*, Phys. Rev. C 108, 014321 (2023).

Mixing of TDHF trajectories

Sep2023

B. Li, D. Vretenar, T. Nikšić *et al.*, arXiv:2309.12564 [nucl-th] (2023).

Mixing of TDHF+BCS trajectories

Oct2023

PM, David Regnier, Denis Lacroix, arXiv:2310.20557 [nucl-th] (2023).

Numerical and technical details

Outline

1. Introduction: Configuration-mixing EDF models
2. **Multiconfigurational TDDFT model**
3. First application: ISGQR multiphonons in ^{40}Ca
4. Several technical remarks
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Multiconfigurational TDDFT model

Properties of the many-body ansatz

- Starting point is a linear combination of **time-dependent** many-body states

$$|\Psi(t)\rangle = \int d\mathbf{q}\; f_{\boldsymbol{q}}(t)\,|\Phi_{\boldsymbol{q}}(t)\rangle$$

- Depending on the application, there is a freedom in choosing:
  - **Generating coordinates**
    - Magnitude/phase of the order parameter for symmetry breaking
    - Relative position/momentum for collisions
    - Boost magnitude for oscillations
    - …
  - **Generating states**
    - Many-body level: Slater determinants/U(1)-breaking vacua
    - Single-particle level: from an ansatz or microscopic (schematic interaction or EDF)
  - **Variational procedure**
    - Different variants of the time-dependent variational principle
    - Variational parameters

Multiconfigurational TDDFT model

Time-dependent variational principle

- We apply the time-dependent variational principle **on a set of mixing functions** (and not also on the basis states; consequences are discussed later)

- These mixing functions satisfy the following properties:

  1) They are differentiable in time.
  2) They lead to a many-body state that is normalized in time.
  3) They are associated to non-zero eigenvalues of the norm kernel (to ensure bijection).

- The action to be minimized reads:

$$S(f, f^*, \xi_1, \xi_2) = \int_{t_0}^{t_1} dt \, \langle \Psi(t) | \hat{H} - i\hbar\partial_t | \Psi(t) \rangle \qquad \textit{Dirac-Frenkel variational principle}$$

$$+ \int_{t_0}^{t_1} dt \, \xi_1(t) \Big(\langle \Psi(t) | \Psi(t) \rangle - 1 \Big) \qquad \textit{Condition 2)}$$

$$+ \int_{t_0}^{t_1} dt \, \xi_2(t) || \mathcal{P}^{\mathcal{K}} f ||^2 \qquad \textit{Condition 3)}$$

$$\mathcal{F} = \mathcal{I}(t) \oplus \mathcal{K}(t) \qquad \mathcal{P}^{\mathcal{K}}_{\boldsymbol{q}\boldsymbol{q}'}(t) = \sum_{k \leq d-r} \mathcal{U}_{\boldsymbol{q}k}(t) \mathcal{U}^{\dagger}_{\boldsymbol{q}'k}(t)$$

Multiconfigurational TDDFT model

Equations of motion

- By minimizing the action, we obtain the equation for the collective wave function

$$i\hbar\dot{g}(t) = \left(\mathcal{H}^c(t) - \mathcal{D}^c(t) + i\hbar\dot{\mathcal{N}}^{1/2}(t)\mathcal{N}^{-1/2}(t)\right)g(t) \qquad g = \mathcal{N}^{1/2}f$$

- Various kernels are transformed to the corresponding collective operators

Hamiltonian kernel

$$\mathcal{H}_{\boldsymbol{qq'}}(t) = \langle\Phi_{\boldsymbol{q}}(t)|\hat{H}|\Phi_{\boldsymbol{q'}}(t)\rangle$$

Derivative kernel

$$\mathcal{D}_{\boldsymbol{qq'}}(t) = \langle\Phi_{\boldsymbol{q}}(t)|i\hbar\partial_t|\Phi_{\boldsymbol{q'}}(t)\rangle$$

Collective operators

$$\mathcal{O}^c = \mathcal{N}^{-1/2}\mathcal{O}\mathcal{N}^{-1/2}$$

- The expectation value of any observable is calculated as

$$\langle\hat{O}\rangle(t) = g^\dagger(t)\mathcal{O}^c(t)g(t)$$

- Basis states are propagated **independently**, following the corresponding equations

$$i\hbar\dot{\rho}_{\boldsymbol{q}}(t) = \left[h[\rho_{\boldsymbol{q}}(t)], \rho_{\boldsymbol{q}}(t)\right]$$

  - In the current implementation, it is the nuclear Hartree-Fock equation
  - Simplifies the problem, but neglects the feedback of mixing on individual trajectories

Multiconfigurational TDDFT model

Evaluation of kernels

- Without pairing, the **norm kernel** is a simple determinant of single-particle overlaps
- The **"Hamiltonian" kernel** is evaluated through the GWT + density prescription
- The **inverted norm kernel** is calculated by removing the vanishing norm eigenvalues

$$\mathcal{N}^{-1/2}_{\boldsymbol{q}\boldsymbol{q}'}(t) = \sum_{k>d-r} \mathcal{U}_{\boldsymbol{q}k}(t)\lambda_k^{-1/2}(t)\mathcal{U}^{\dagger}_{\boldsymbol{q}'k}(t)$$

- Kernels with explicit time derivatives are calculated in the **finite differences** scheme

$$\partial_t \left|\Phi_{\boldsymbol{q}}(t)\right\rangle \approx \frac{1}{\Delta t}\left(\left|\Phi_{\boldsymbol{q}}(t)\right\rangle - \left|\Phi_{\boldsymbol{q}}(t_-)\right\rangle \right)$$

- The **time-derivative kernel** then reads

$$\mathcal{D}_{\boldsymbol{q}\boldsymbol{q}'}(t) = \frac{i\hbar}{\Delta t}\left(\left\langle\Phi_{\boldsymbol{q}}(t)|\Phi_{\boldsymbol{q}'}(t)\right\rangle - \left\langle\Phi_{\boldsymbol{q}}(t)|\Phi_{\boldsymbol{q}'}(t_-)\right\rangle \right)$$

- Time derivative of $N^{1/2}$ is the solution of the Sylvester equation

$$\dot{\mathcal{N}}(t) = \dot{\mathcal{N}}^{1/2}(t)\mathcal{N}^{1/2}(t) + \mathcal{N}^{1/2}(t)\dot{\mathcal{N}}^{1/2}(t) \qquad \dot{\mathcal{N}}^{1/2}(t) = \mathrm{vec}^{-1}\left[\mathcal{S}^{-1}(t)\ \mathrm{vec}\left(\dot{\mathcal{N}}(t)\right)\right.$$

$$\dot{\mathcal{N}}_{\boldsymbol{q}\boldsymbol{q}'}(t) = \left\langle\Phi_{\boldsymbol{q}}(t)|\dot{\Phi}_{\boldsymbol{q}'}(t)\right\rangle + \left\langle\dot{\Phi}_{\boldsymbol{q}}(t)|\Phi_{\boldsymbol{q}'}(t)\right\rangle \qquad \mathcal{S}(t) = \mathbb{1}\otimes\mathcal{N}^{1/2}(t) + \left(\mathcal{N}^{1/2}(t)\right)^T\otimes\mathbb{1}$$

Multiconfigurational TDDFT model

Resolving the equations of motion

$$i\hbar\dot{g}(t) = \left(\mathcal{H}^c(t) - \mathcal{D}^c(t) + i\hbar\dot{\mathcal{N}}^{1/2}(t)\mathcal{N}^{-1/2}(t)\right)g(t)$$

$$i\hbar\dot{\rho}_{\boldsymbol{q}}(t) = \left[h[\rho_{\boldsymbol{q}}(t)], \rho_{\boldsymbol{q}}(t)\right]$$

- The equation for g can be solved using a time-propagation method of choice
  - The “direct” method feasible for smaller bases

$$g_{\boldsymbol{q}}(t_0 + \Delta t) = \exp\left(-\frac{i}{\hbar}\mathcal{T}(t_0)\Delta t\right)g_{\boldsymbol{q}}(t_0)$$

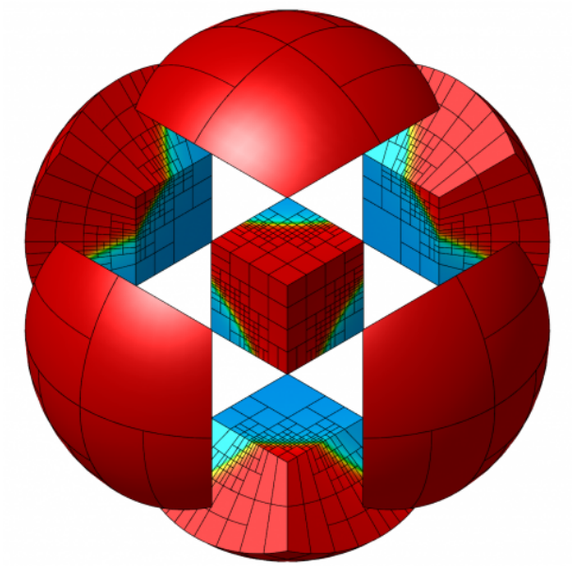
  - Common alternatives: Runge-Kutta (RK), Crank-Nicolson, …
- TDHF equation is resolved using the RK4 scheme

We developed a new TDDFT code and an MC-TDDFT solver on top of it.

Multiconfigurational TDDFT model

The NYMFE computer program

- **N**uclear d**Y**na**M**ics on **F**inite **E**lements (**NYMFE**)
  - The main author is David Regnier (CEA, DAM)
  - Uses modern C++ standard
  - Simulation in a box, Skyrme interactions
  - Space discretization with the finite element method
  - Static and dynamic DFT/EDF solver, MC solver on top
  - Benchmarked against *HFBTHO* and *Sky3D*
  - Will be made publicly available, eventually



MFEM: Scalable Finite Element Discretization Library

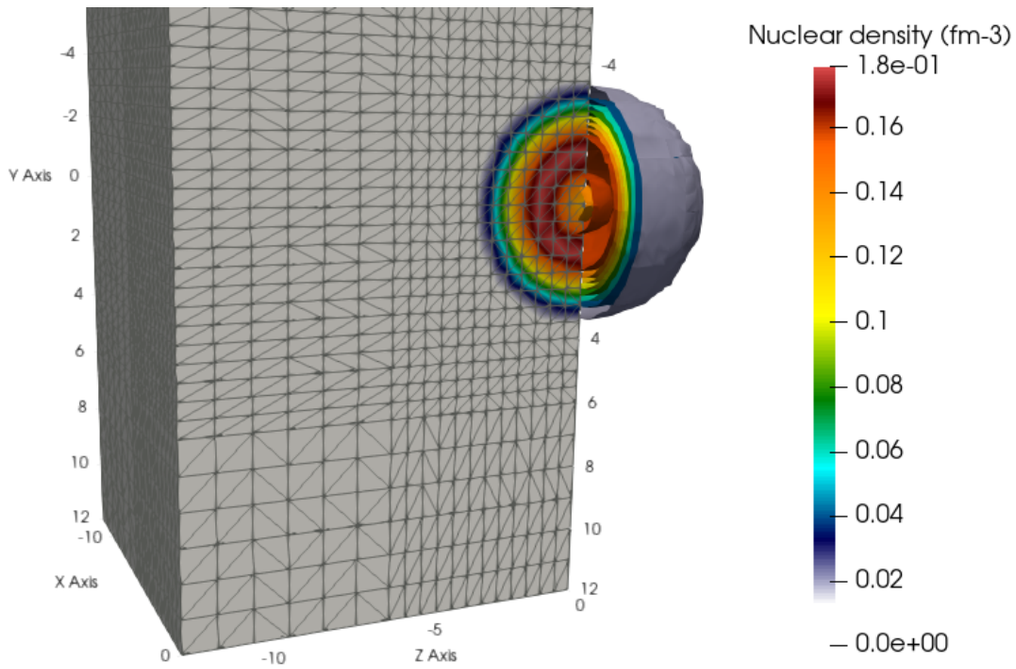


Illustration: Simulation in a finite elements box

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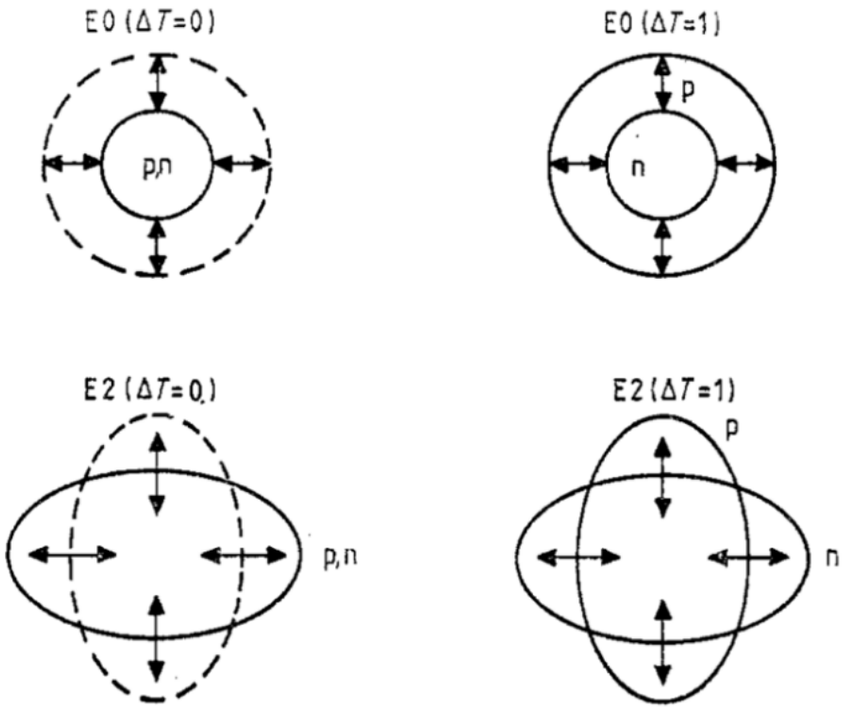
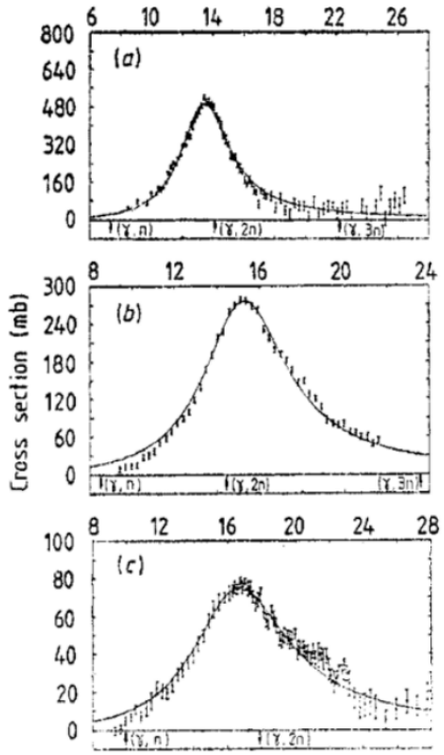
ISGQR multiphonons in ^{40}Ca

Giant resonances as collective nuclear excitations

- Collective response to an external perturbation is a common feature of quantum many-body systems (phonons in solids, plasmons in electronic gases)
- In nuclei, **giant resonances** have been studies for many decades



Textbook illustration of giant resonances



^{208}Pb

^{124}Sn

^{65}Cu

A typical signature of giant resonances

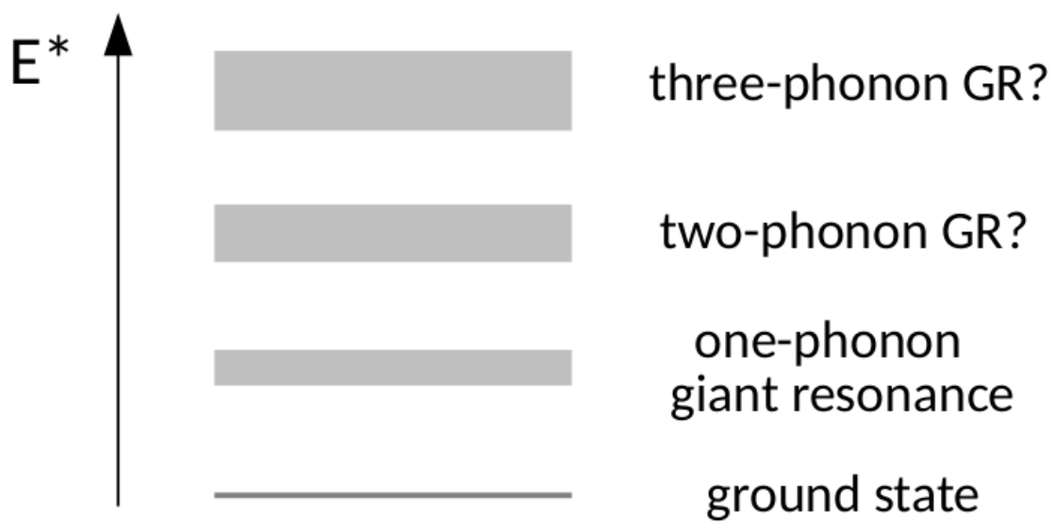
- Giant resonances correspond to small-amplitude, (nearly-) harmonic oscillations

The question is - **are there higher excitation quanta (multiphonons)**?

ISGQR multiphonons in ^{40}Ca

Multiphonons in nuclei

- Multiphonons – higher quanta of the main GR excitation



Do collective multiphonon excitations exist in nuclei?

If yes, are they harmonic?

There are decades of excellent research on the topic!

A scheme of a (hypothetical) multiphonon spectrum

Experiment:

- 2nd phonon observed in multiple nuclei
- 3nd phonon possibly observed in two cases
  Schmidt *et al.*, IVGDR in ^{136}Xe (1993) (?)
  Fallot *et al.*, ISGQR in ^{40}Ca (2006)

Theory:

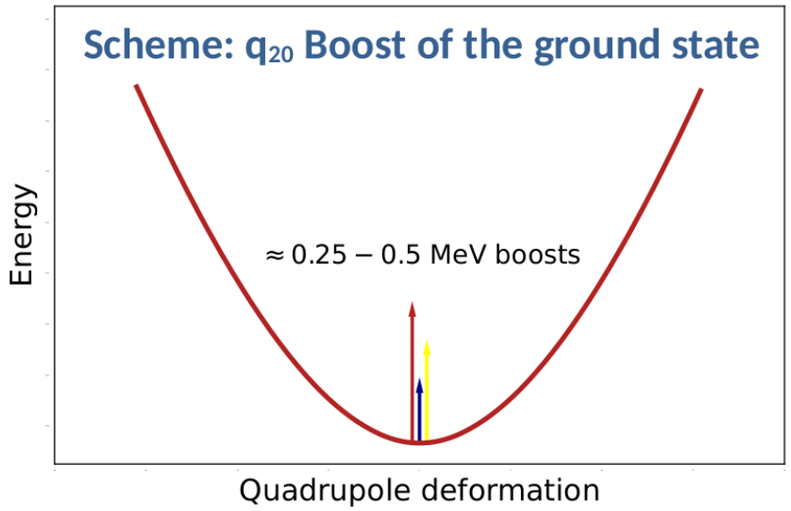
- In principle, a requantized theory is needed
- Many models on the market, usually an *ad hoc* introduction of phonon d.o.f.

  Very recently: Ab initio calculations (A. Porro)

ISGQR multiphonons in ^{40}Ca

Calculation parameters

- Some calculation parameters:
  - L = 24 fm box, regular mesh of 14 cells, FE basis od 3rd ord. poly.
  - SLy4d Skyrme EDF (common choice for dynamical studies)
  - Basis states correspond to the HF g.s. with different q_{20} boosts
  - $\Delta t = 5 \times 10^{-4}$ zs step for time propagation



- We consider a simple case of quantum configuration mixing:

At *t* = 0

TDDFT $$|\Psi(0)\rangle = \exp(i\lambda\hat{Q})\,|\Phi_{\mathrm{GS}}\rangle$$

MC-TDDFT $$|\Psi(0)\rangle = \exp(i\lambda\hat{Q})\Big(f_1(0)\,|\Phi_{\mathrm{GS}}\rangle + f_2(0)\,|\Phi_2(0)\rangle + f_3(0)\,|\Phi_3(0)\rangle\Big)$$

At later *t*

TDDFT $$|\Psi(t)\rangle = |\Phi_1(t)\rangle$$

MC-TDDFT $$|\Psi(t)\rangle = f_1(t)\,|\Phi_1(t)\rangle + f_2(t)\,|\Phi_2(t)\rangle + f_3(t)\,|\Phi_3(t)\rangle$$

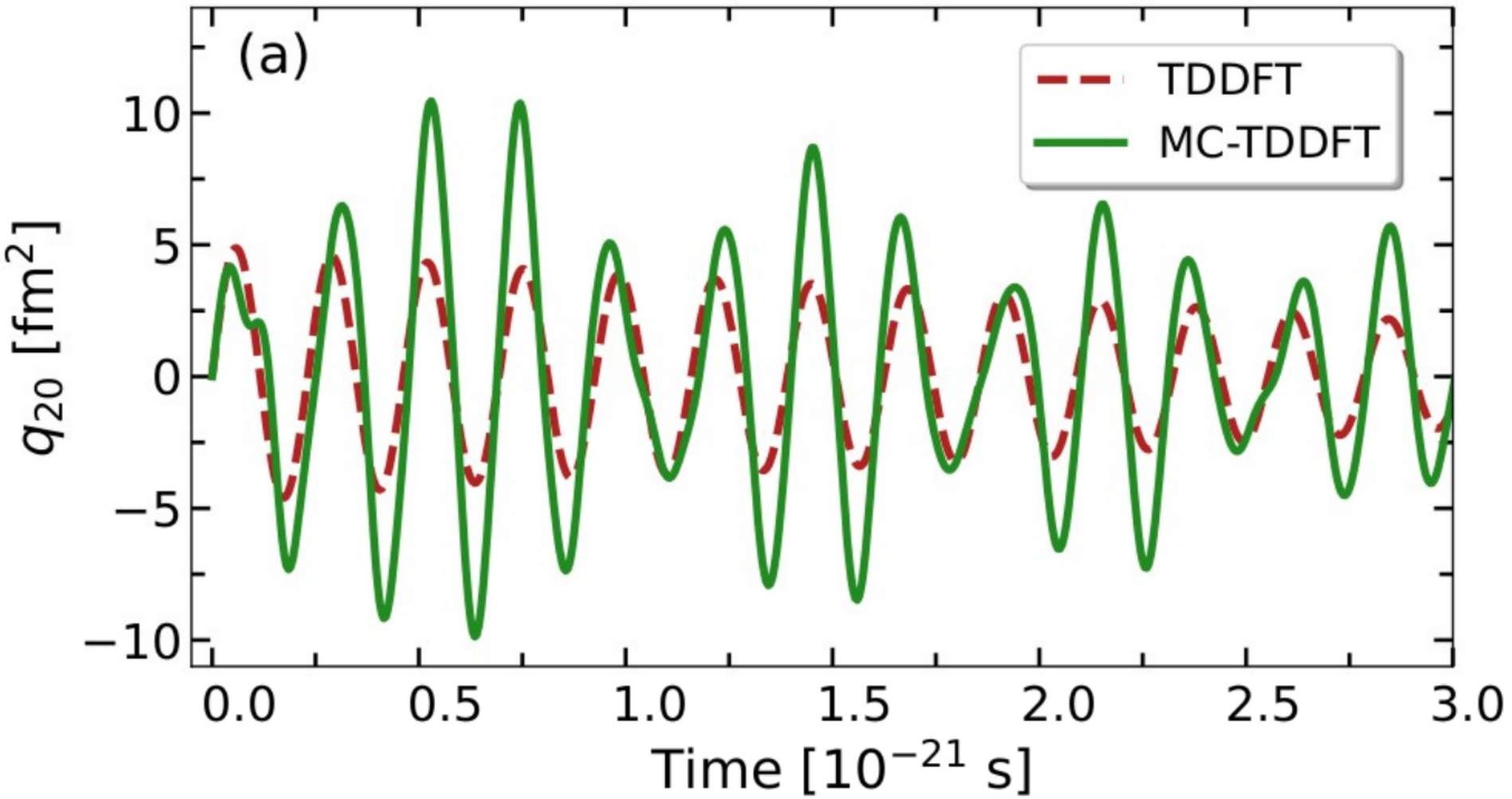
$$f_1(0) = 1 \qquad f_2(0) = f_3(0) = 0$$

We can compare predictions of the **quasi-classical TDDFT** and the **quantum MC-TDDFT**.

ISGQR multiphonons in ^{40}Ca

Nuclear response to quadrupole perturbation

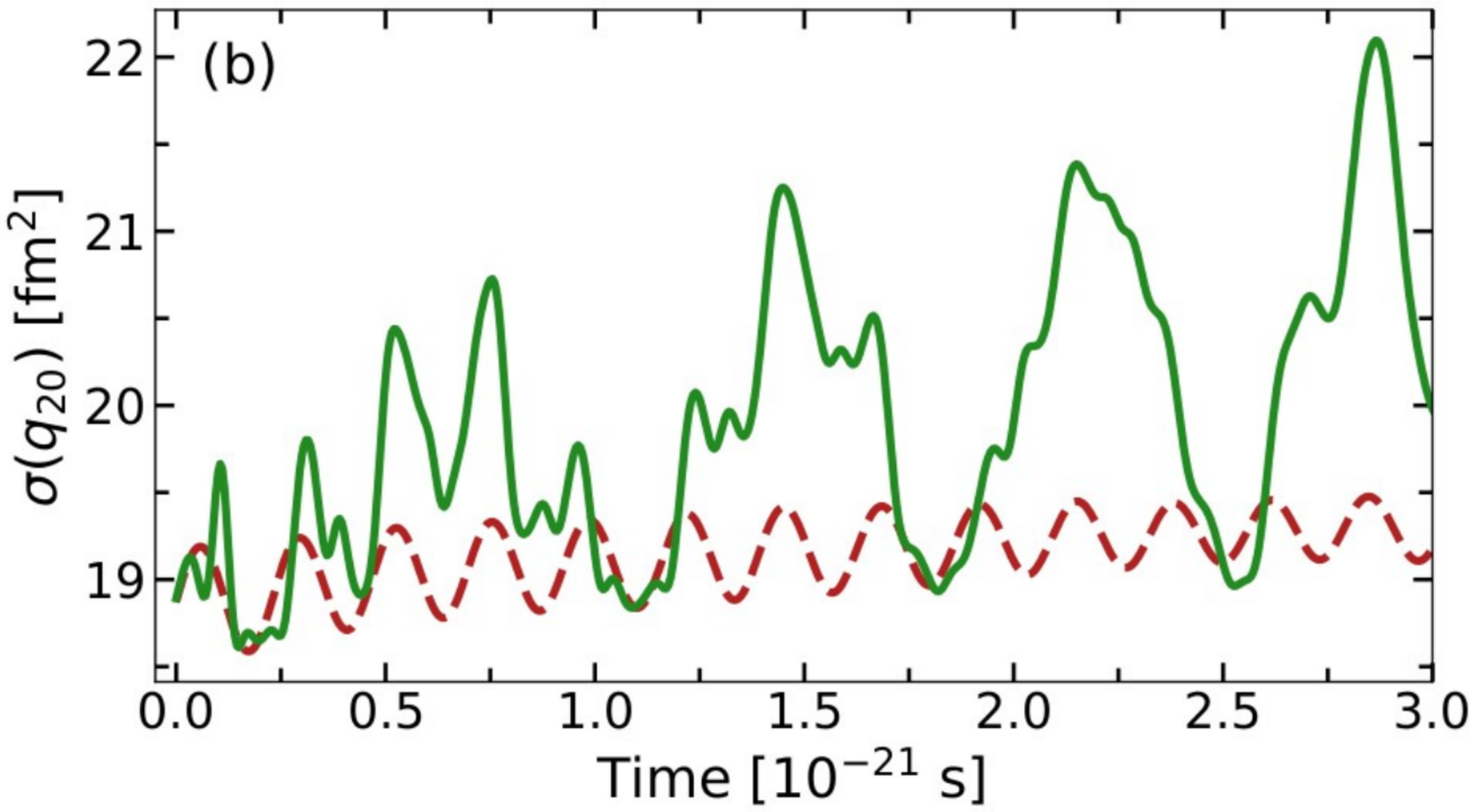
- Quadrupole response for MC-TDDFT is **more complex** and exhibits **multiple frequencies**



ISGQR multiphonons in ^{40}Ca

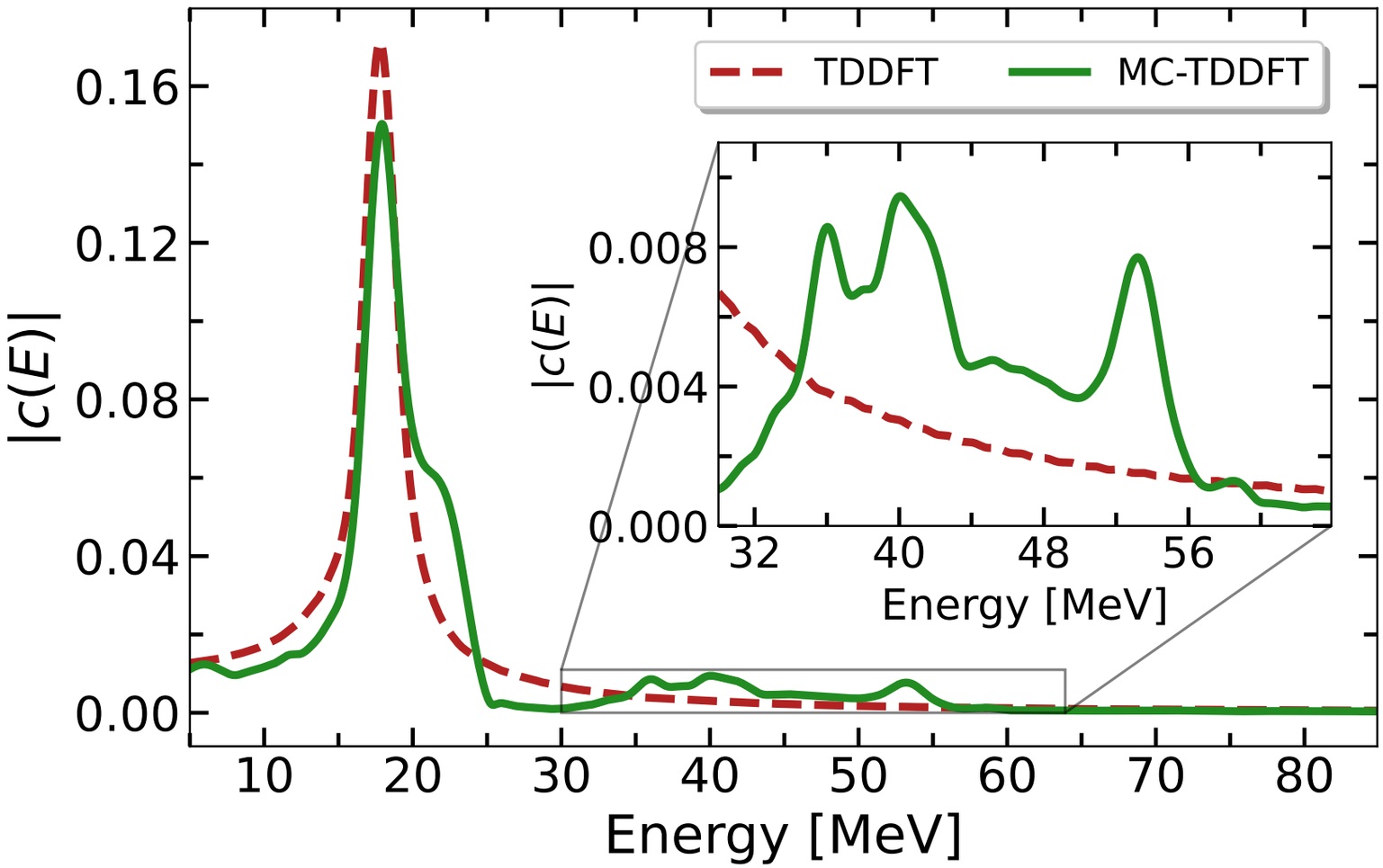
Nuclear response to quadrupole perturbation

- Quadrupole response for MC-TDDFT is **more complex** and exhibits **multiple frequencies**
- Quadrupole fluctuations are **larger** and also exhibit multiple frequencies
- Frequency (energy) spectrum can be extracted through Fourier analysis



ISGQR multiphonons in ^{40}Ca

Energy spectrum



- TDDFT yields a **single peak** (all trajectories are equivalent)
- MC-TDDFT yields multiple **peaks**
  - The 1st peak is split (but we do not aim to describe all the fine details of fragmentation)
  - The 2nd and 3rd peak appear at 2x and 3x the energy and reflect this splitting
  - There is no 4th peak

ISGQR multiphonons in ^{40}Ca

Robustness of results

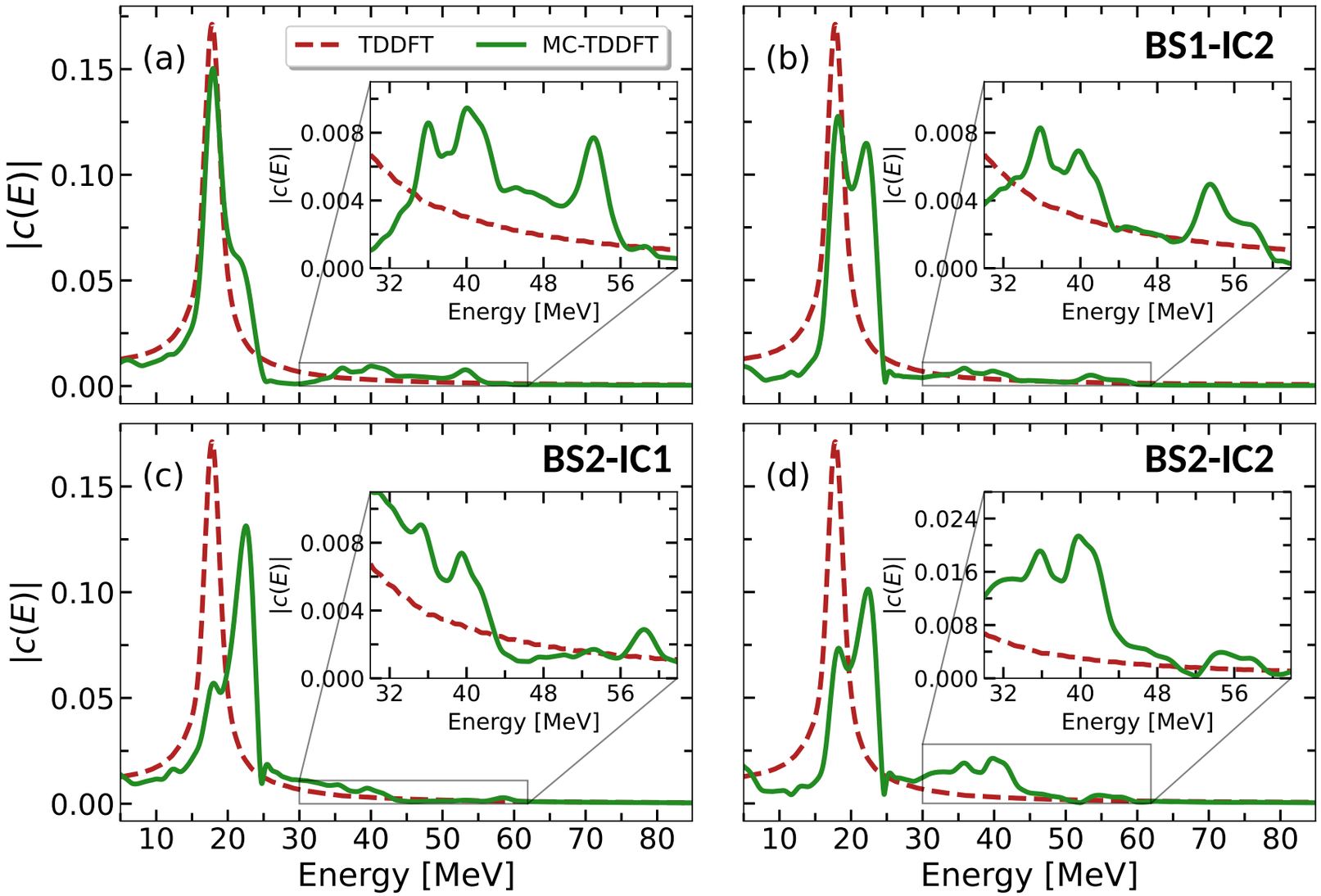
- We consider two sets of basis states (BS) and two sets of initial conditions (IC)

BS1 – Different boost magnitudes

BS2 – The same boost, different points along the trajectory

IC1 – $f_1(0) = 1$, $f_2(0) = f_3(0) = 0$

IC2 – Diagonalization of the initial collective Hamiltonian

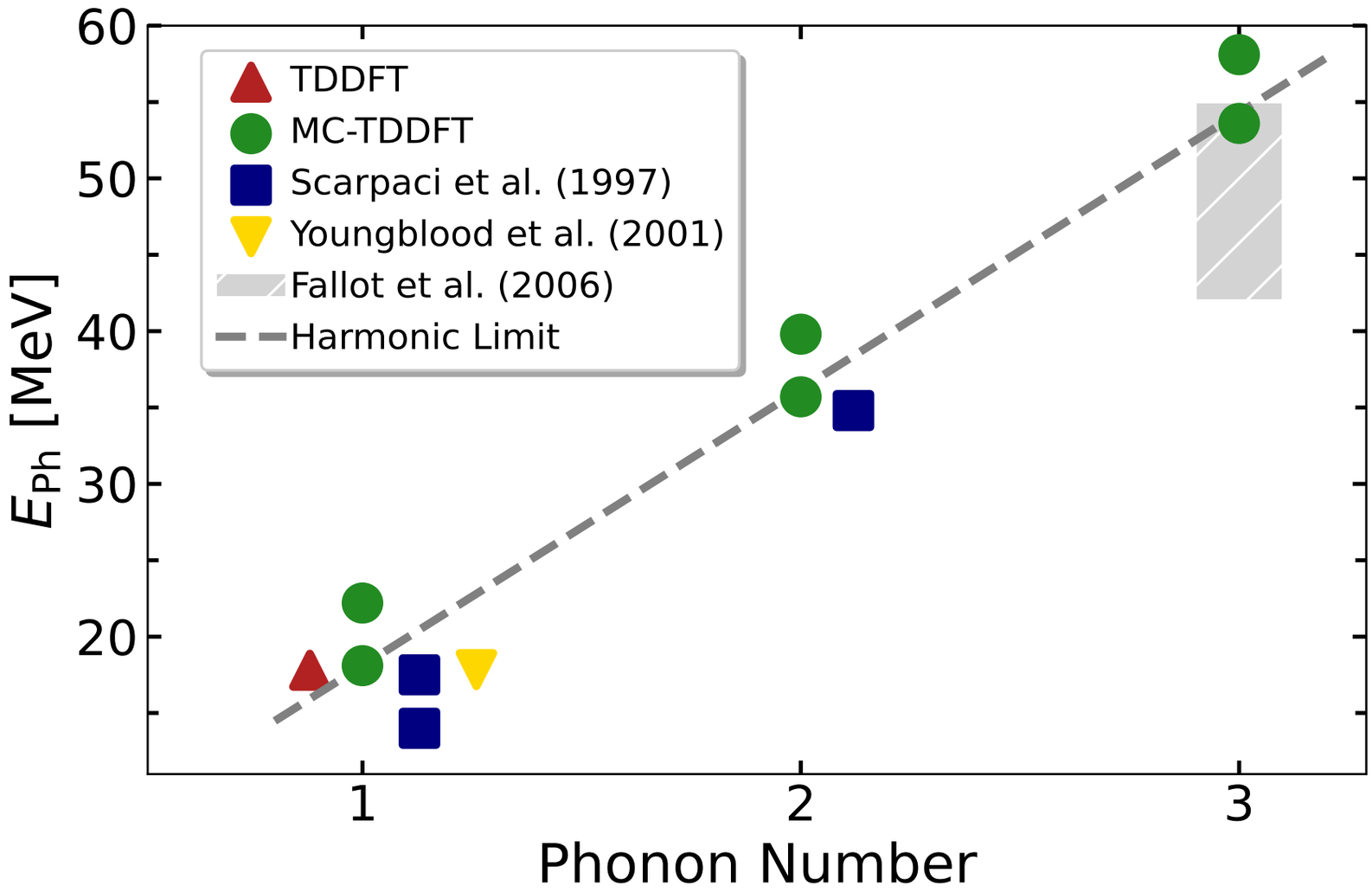


The appearance of peaks and their energies are **robust** w.r.t. this choice (but amplitudes vary).

ISGQR multiphonons in ^{40}Ca

Comparison with experiment

Energy spectrum is in **excellent agreement** with experiment and **nearly harmonic** (~2%).



Excitations of the main peak:

E_{1ph} = 18.1 (0.2) MeV

E_{2ph} = 35.7 (0.5) MeV

E_{3ph} = 53.6 (0.7) MeV

(not full theoretical uncertainties)

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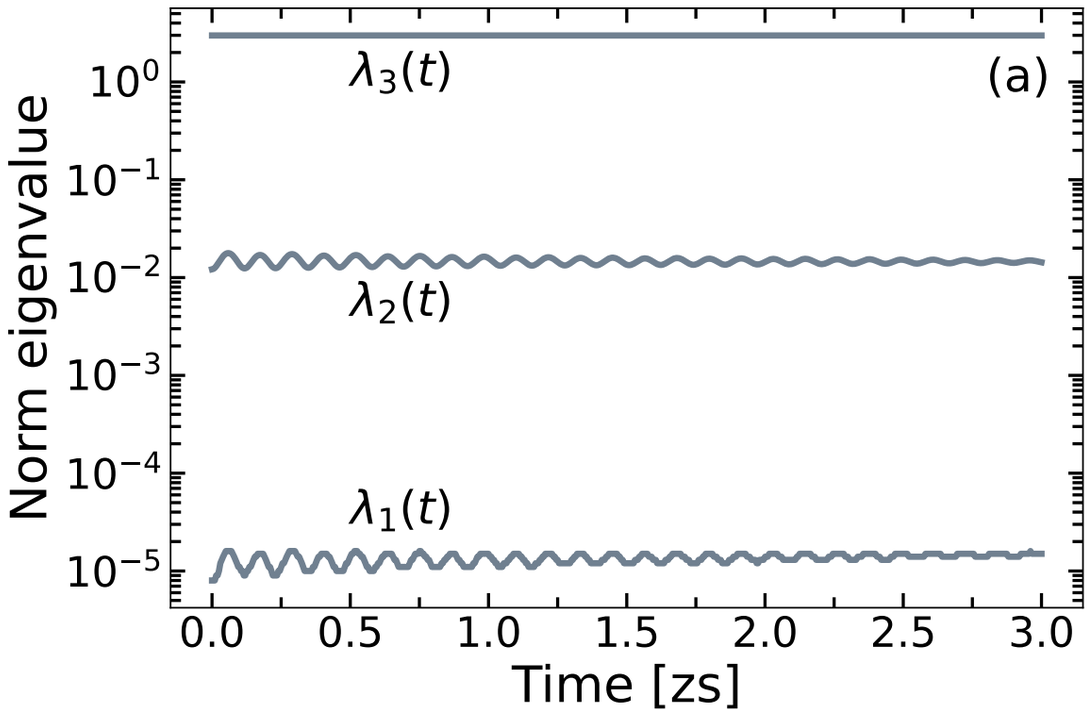
Several technical remarks

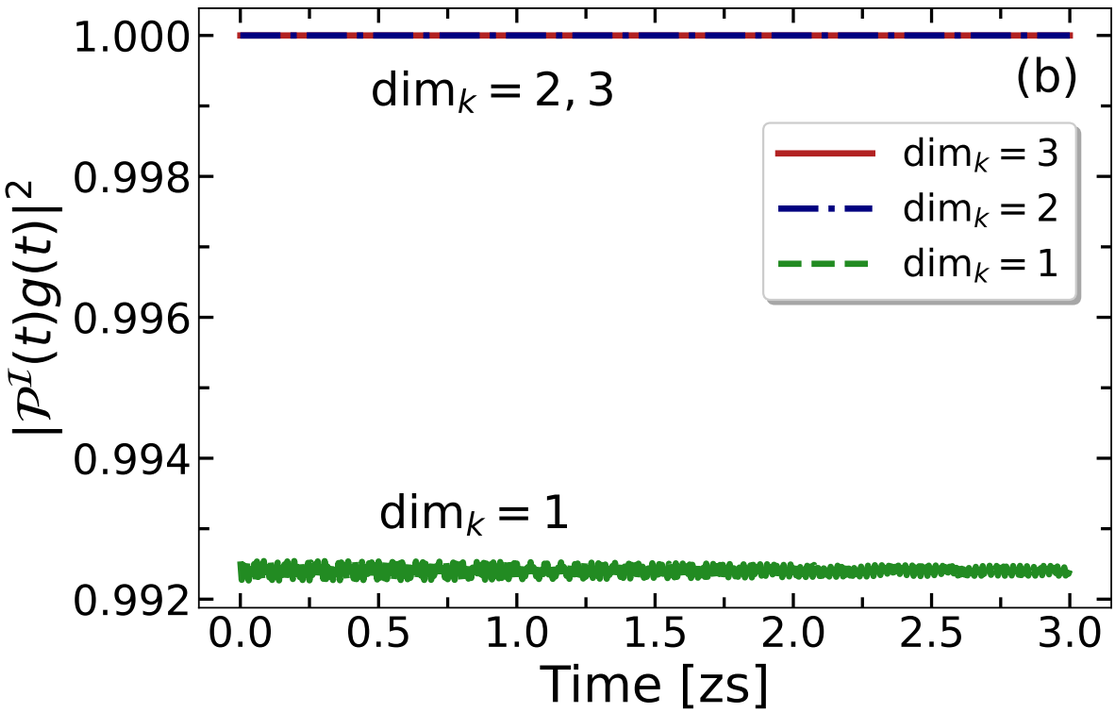
Treatment of linear dependencies in the basis

- In a recent publication, we addressed numerical and technical details of the model

PM, David Regnier, Denis Lacroix, "*Multiconfigurational time-dependent density functional theory for atomic nuclei: Technical and numerical aspects*", arXiv:2310.20557 [nucl-th] (2023).

- Like in static GCM, there is an issue of linear dependencies in the basis set





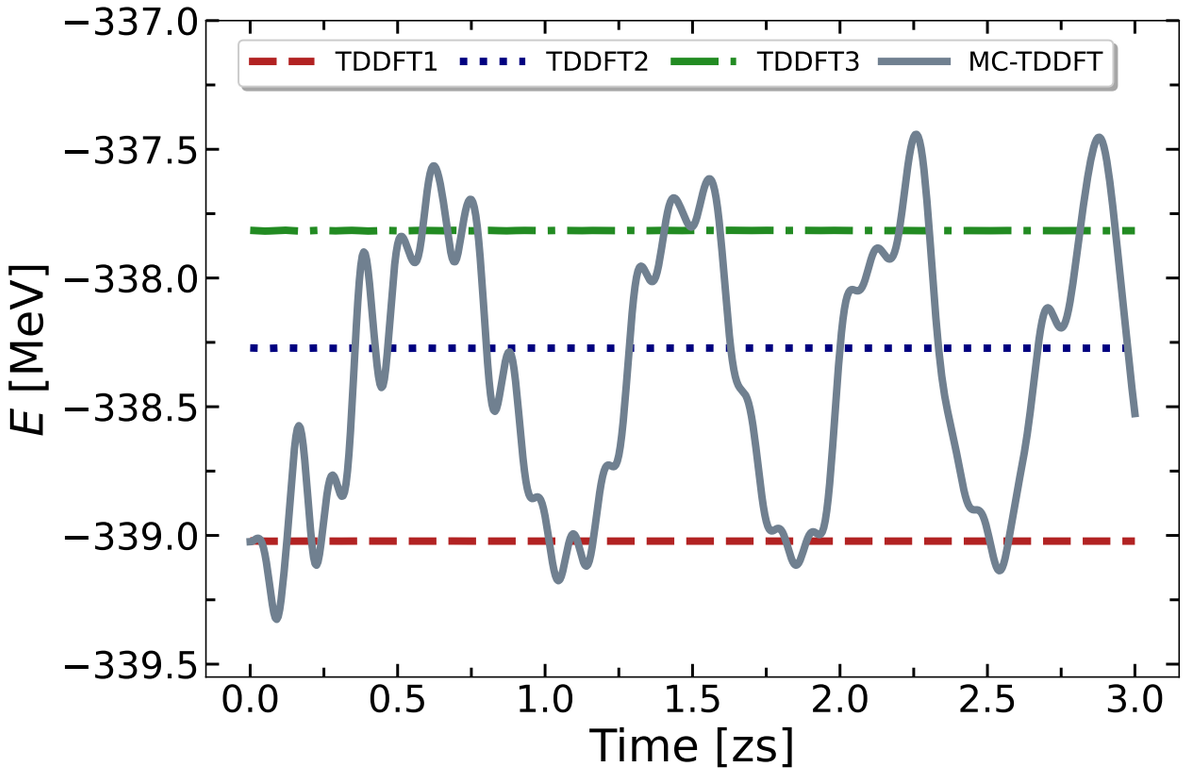
- Small norm eigenvalues carry little physical content but induce numerical noise through $N^{-1/2}$
- Consequently, they are removed from the basis, analogously to the static case
- Caution: removing too large eigenvalues removes a part of physical information

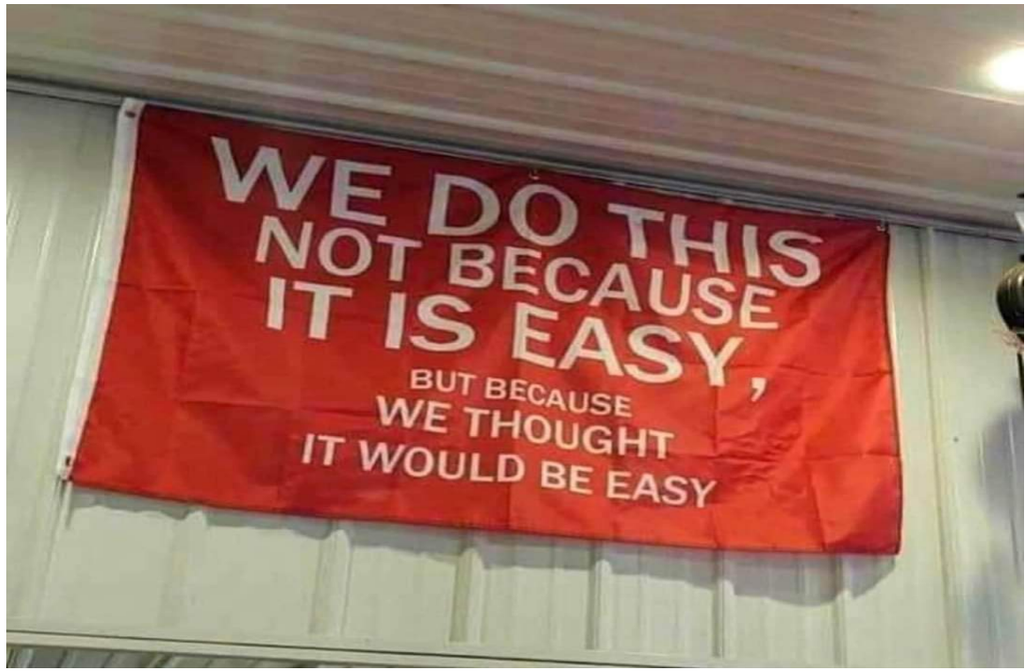
Several technical remarks

A somewhat unexpected mine: Energy conservation

- In TDDFT, the energy is guaranteed to be conserved throughout time evolution
- However, the energy is **not conserved** on the multiconfigurational level

Energy of the MC-TDDFT state (left) and motivation revised (right)





- Variations will depend on the particular application (here ~1.5 MeV)
- This appears to be a **consequence of not being fully variational**
- Including the effect of configuration mixing on individual trajectories may be necessary

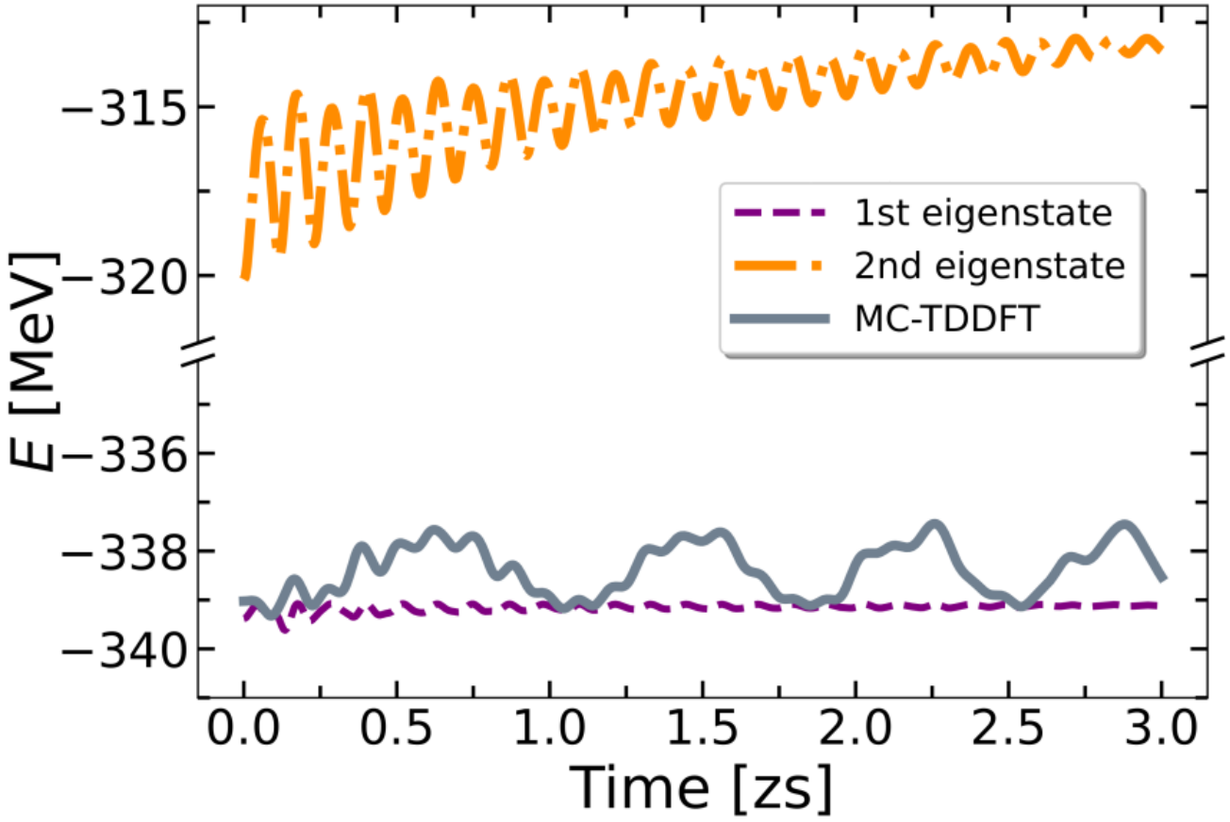
Several technical remarks

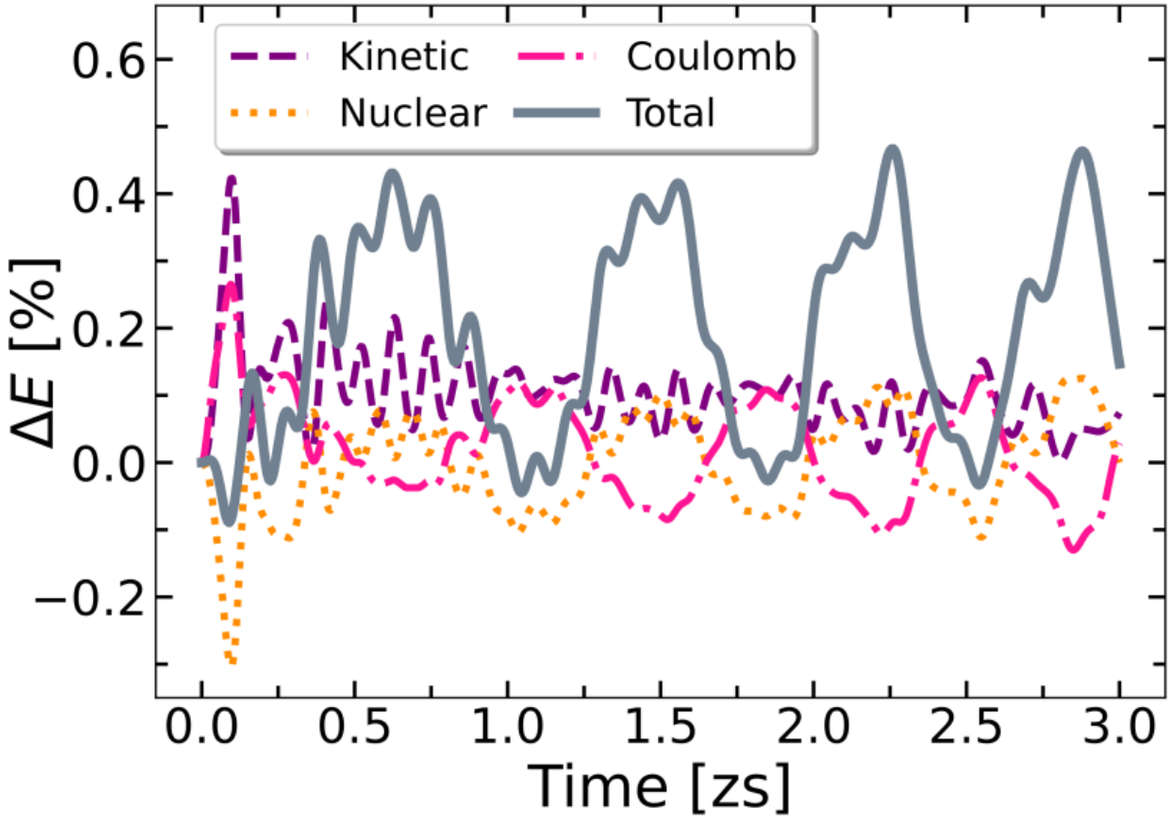
A somewhat unexpected mine: Energy conservation

- In fact, the MC-TDDFT energy is bound by the eigenvalues of the collective Hamiltonian

$$E_{\mathrm{MC-TDDFT}}(t) = \tilde{g}^{\dagger}(t)\Lambda_H(t)\tilde{g}(t) \qquad \tilde{g}(t) = U_H^{\dagger}(t)g(t)U_H(t)$$

- All energy components contribute to the total variation





Several technical remarks

Stepping into the minefield: Density-dependent prescription

- Certain components of an EDF are density dependent
  - A Skyrme component proportional to ρ^{α} ($\alpha = 1/6$ for SLy4d)
  - Coulomb exchange ($\alpha = 4/3$)
- These terms are not uniquely defined on a multi-configurational level
- We can use the **average density** prescription… but which one?

$$\rho_D^{\alpha}(\boldsymbol{r};t) = \left[\frac{1}{2}\left(\rho_{\boldsymbol{qq}}(\boldsymbol{r};t) + \rho_{\boldsymbol{q'q'}}(\boldsymbol{r};t)\right)\right]^{\alpha}$$ *Average Density 1*

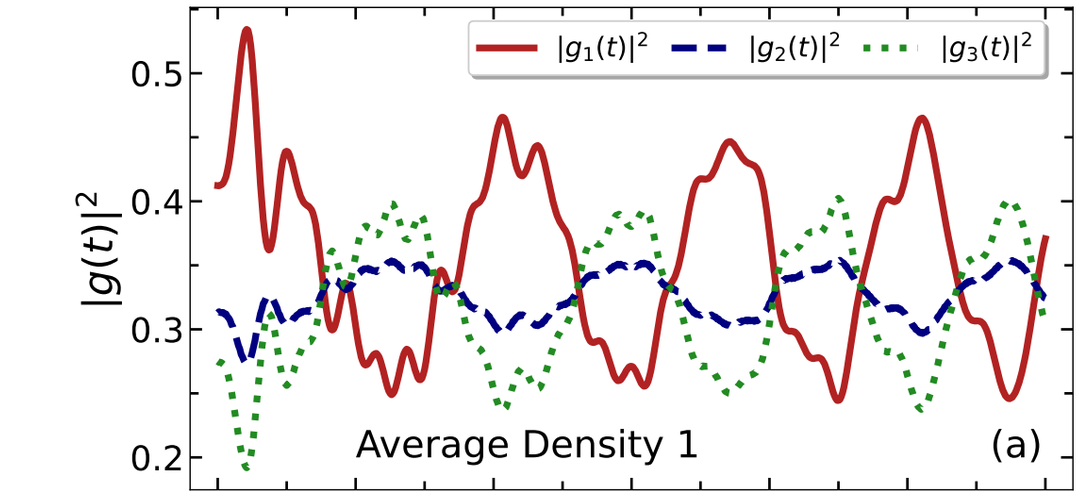
$$\rho_D^{\alpha}(\boldsymbol{r};t) = \frac{1}{2}\left(\rho_{\boldsymbol{qq}}^{\alpha}(\boldsymbol{r};t) + \rho_{\boldsymbol{q'q'}}^{\alpha}(\boldsymbol{r};t)\right)$$ *Average Density 2*

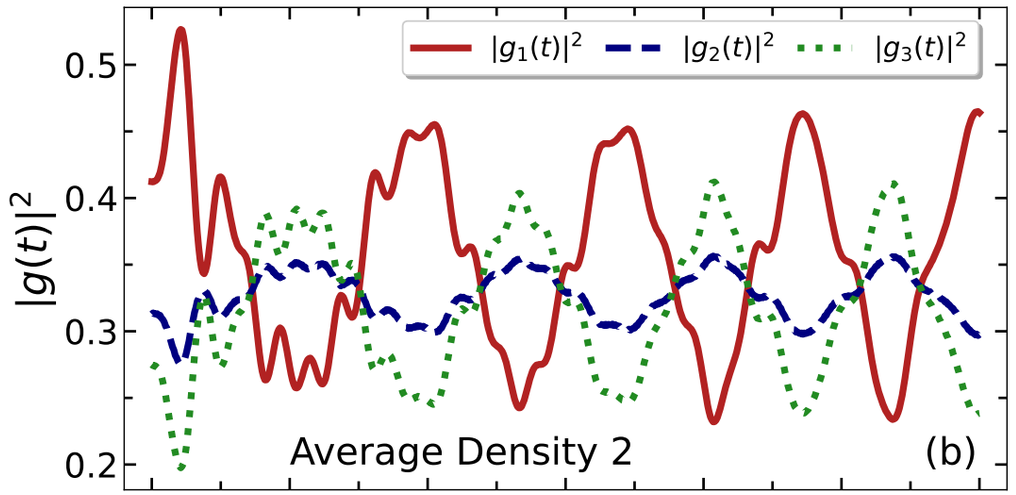
The two prescriptions are **real** and **good in the *q* = *q'* limit**, but **not equivalent**.

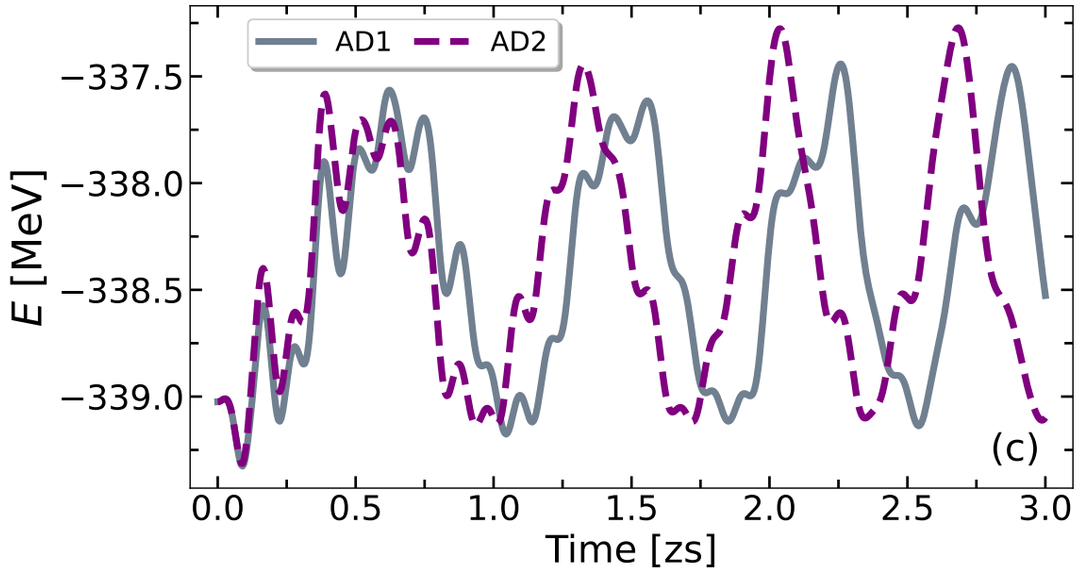
We can, at best, try to assess an impact of this choice on predictions of the model.

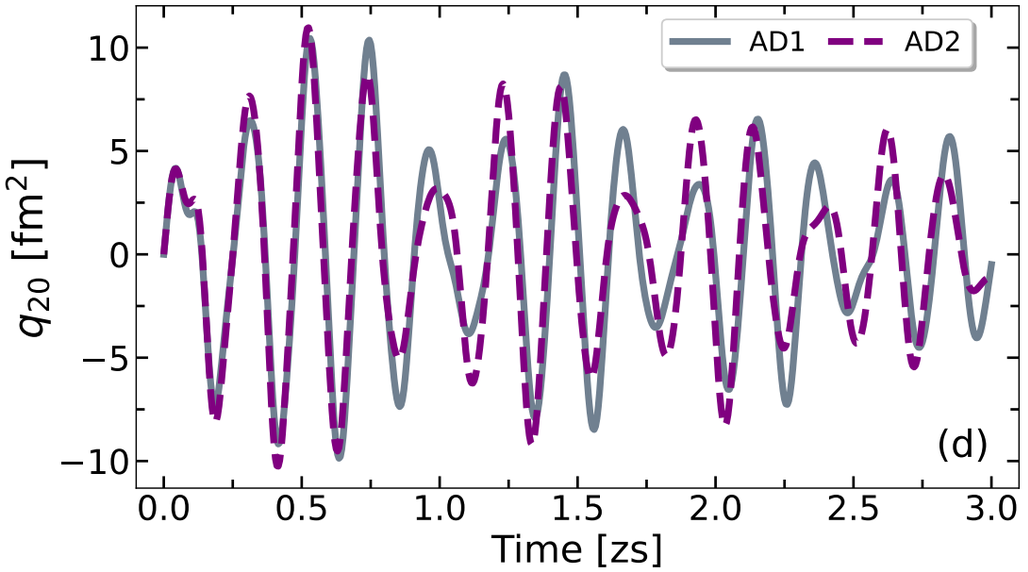
Several technical remarks

Stepping into the minefield: Density-dependent prescription









- Overall dynamics ($|g|^2$) is largely unaffected, except for a moderate shift in phase for large t
- Energy variation is comparable, with a similar shift in phase
- Quadrupole moment is identical up to t = 0.75zs, afterwards moderate variations
- **Energy spectrum is robust**: all peaks are shifted by less than 0.1 MeV

Outline

1. Introduction: Configuration-mixing EDF models
2. Multiconfigurational TDDFT model
3. First application: ISGQR multiphonons in ^{40}Ca
4. Several technical remarks
5. Conclusion

Conclusion

1. MC-TDDFT is an advanced and versatile model of nuclear dynamics.

- Time dependence is embedded in both the basis states and the mixing function
- Includes both the dissipation and quantum fluctuation aspects
- In principle, applicable to nuclear vibrations, collisions, fission, and more

2. This year, the model was for the first time applied in nuclei, using EDFs.

- We considered a simple quantum mixing model for quadrupole vibrations in ^{40}Ca
- The model predicts multiphonon ISGQR excitations without any *a priori* assumptions
- Excitation spectrum is in an excellent agreement with experimental data

3. The model and its extensions will soon be applicable to other systems and phenomena.

- Inclusion of pairing and code optimization/parallelization
- Fully variational approach by treating basis states as variational parameters

"*Quantum fluctuations induce collective multiphonons in finite Fermi liquids*"
PM, David Regnier, Denis Lacroix, Phys. Rev. C 108, 014620 (2023).

"*MC-TDDFT theory for atomic nuclei: Technical and numerical aspects*",
PM, David Regnier, Denis Lacroix, arXiv:2310.20557 [nucl-th] (2023).