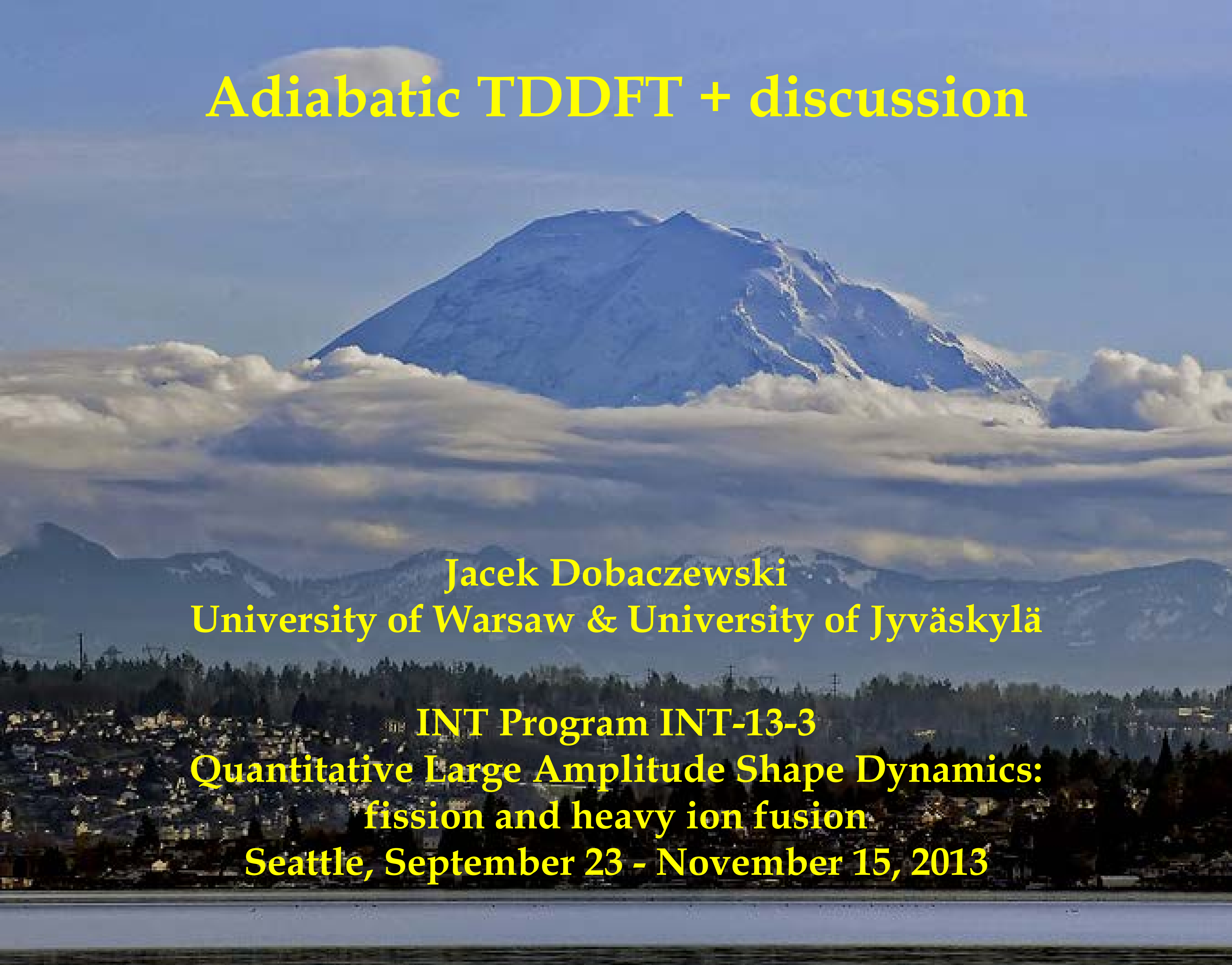

# Adiabatic TDDFT + discussion

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INT Program INT-13-3
Quantitative Large Amplitude Shape Dynamics:
fission and heavy ion fusion
Seattle, September 23 - November 15, 2013

# Outline

1. DFT
2. TDDFT
3. Instantons
   (this is what I will not talk about)
4. Adiabatic expansion
5. Adiabatic equations
6. Adiabatic solutions
7. Discussion



# What is DFT?

**Density Functional Theory:**

**A variational method that uses observables as variational parameters.**

$$\delta\langle\hat{H} - \lambda\hat{Q}\rangle = 0$$
$$\Downarrow$$
$$E = E(Q)$$

for $E(\lambda) \equiv \langle\hat{H}\rangle$ and $Q(\lambda) \equiv \langle\hat{Q}\rangle$

J. D., J. Phys.: Conf. Ser. 312, 092002 (2011)



# Which DFT?

$$\delta\langle\hat{H}-\lambda\hat{Q}\rangle=0 \Longrightarrow E=E(Q)$$

$$\delta\langle\hat{H}-\textstyle\sum_k\lambda_k\hat{Q}_k\rangle=0 \Longrightarrow E=E(Q_k)$$

$$\delta\langle\hat{H}-\int\mathrm{d}q\,\lambda(q)\hat{Q}(q)\rangle=0 \Longrightarrow E=E[Q(q)]$$

$$\delta\langle\hat{H}-\int\mathrm{d}\vec{r}\,\lambda(\vec{r})\hat{\rho}(\vec{r})\rangle=0 \Longrightarrow E=E[\rho(\vec{r})]$$

$$\text{for}\quad \hat{\rho}(\vec{r}) \;=\; \textstyle\sum_{i=1}^{A}\delta(\vec{r}-\vec{r}_i)$$

$$\delta\langle\hat{H}-\int\!\!\int\mathrm{d}\vec{r}\mathrm{d}\vec{r}\,'\,\lambda(\vec{r},\vec{r}\,')\hat{\rho}(\vec{r},\vec{r}\,')\rangle=0 \Longrightarrow E=E[\rho(\vec{r},\vec{r}\,')]$$

J. D., J. Phys.: Conf. Ser. 312, 092002 (2011)



# How the nuclear EDF is built?

**Local energy density is a function of local density**

$$E[\rho(\vec{r})] = \int \mathrm{d}\vec{r}\, \mathcal{H}(\rho(\vec{r}))$$

**LDA**

**Non-local energy density is a function of non-local density**

$$E[\rho(\vec{r},\vec{r}\,')] = \int\int \mathrm{d}\vec{r}\,\mathrm{d}\vec{r}\,'\, \mathcal{H}(\rho(\vec{r},\vec{r}\,'))$$

**Gogny, M3Y,...**

$$\mathcal{H}_P(\rho(\vec{r},\vec{r}\,')) = V(\vec{r}-\vec{r}\,')\Big[\rho(\vec{r})\rho(\vec{r}\,') - \rho(\vec{r},\vec{r}\,')\rho(\vec{r}\,',\vec{r})\Big]$$



# Configuration representation of the DFT and TDDFT

$$E[\rho] \;=\; \int \mathrm{d}\vec{r}\,\frac{\hbar^2}{2m}\tau(\vec{r}) + \mathcal{H}_P(\rho(\vec{r}))$$

$$\rho(\vec{r}) \;=\; \sum_{i=1}^{A} \psi_i(\vec{r})\rho_{ij}\psi_j^*(\vec{r})$$

$$\tau(\vec{r}) \;=\; \sum_{i=1}^{A} \vec{\nabla}\psi_i(\vec{r}) \cdot \rho_{ij}\vec{\nabla}\psi_j^*(\vec{r})$$

$$h_{ij} \;=\; T_{ij} + \Gamma_{ij} = \frac{\partial E[\rho]}{\partial \rho_{ji}}$$

W. Kohn and L.J. Sham
Phys. Rev. 140, A1133 (1965)
cited 23 000 times

**DFT** **TDDFT** for $h(t) = h(\rho(t))$

$$0 \;=\; [h, \rho] \qquad i\hbar\dot{\rho}(t) \;=\; [h(t), \rho(t)]$$



# TDDFT

VOLUME 52, NUMBER 12 PHYSICAL REVIEW LETTERS 19 MARCH 1984

## Density-Functional Theory for Time-Dependent Systems

Erich Runge and E. K. U. Gross
*Institut für Theoretische Physik, Johann Wolfgang Goethe-Universität, D-6000 Frankfurt, Federal Republic of Germany*
(Received 16 August 1983)

A density-functional formalism comparable to the Hohenberg-Kohn-Sham theory of the ground state is developed for arbitrary time-dependent systems. It is proven that the single-particle potential $v(\vec{r}\,t)$ leading to a given $v$-representable density $n(\vec{r}\,t)$ is uniquely determined so that the corresponding map $v \rightarrow n$ is invertible. On the basis of this theorem, three schemes are derived to calculate the density: a set of hydrodynamical equations, a stationary action principle, and an effective single-particle Schrödinger equation.

Cited 2600 times

$$\delta\langle\Psi(t)|\hat{H} - i\hbar\frac{\mathrm{d}}{\mathrm{d}t} - \int \mathrm{d}\vec{r}\,\lambda(\vec{r},t)\hat{\rho}(\vec{r})|\Psi(t)\rangle = 0$$

$$\Downarrow$$

$$E = E[\rho(\vec{r}, t_0 \rightarrow t)]$$

$$E = E[\lambda(\vec{r}, t_0 \rightarrow t)] \quad \text{and} \quad \rho(\vec{r}, t) = \rho[\lambda(\vec{r}, t_0 \rightarrow t)]$$



# Instantons

PHYSICAL REVIEW C **77**, 064610 (2008)

## Nuclear fission with mean-field instantons

Janusz Skalski*
*Sołtan Institute for Nuclear Studies, ul. Hoża 69, PL-00681, Warsaw, Poland*
(Received 3 December 2007; published 30 June 2008)

We present a description of nuclear spontaneous fission, and generally of quantum tunneling, in terms of instantons, that is, periodic imaginary-time solutions to time-dependent mean-field equations. This description allows comparisons to be made with the more familiar generator coordinate (GCM) and adiabatic time-dependent Hartree-Fock (ATDHF) methods. It is shown that the action functional whose value for the instanton is the quasiclassical estimate of the decay exponent fulfills the minimum principle when additional constraints are imposed on trial fission paths. In analogy with mechanics, these are conditions of energy conservation and the velocity-momentum relations. In the adiabatic limit, the instanton method reduces to the time-odd ATDHF equation, with collective mass including the time-odd Thouless-Valatin term, while the GCM mass completely ignores velocity-momentum relations. This implies that GCM inertia generally overestimates the instanton-related decay rate. The very existence of the minimum principle offers hope for a variational search for instantons. After the inclusion of pairing, the instanton equations and the variational principle can be expressed in terms of the imaginary-time-dependent Hartree-Fock-Bogoliubov (TDHFB) theory. The adiabatic limit of this theory reproduces ATDHFB inertia.



We present a description of nuclear spontaneous fission, and generally of quantum tunneling, in terms of instantons, that is, periodic imaginary-time solutions to time-dependent mean-field equations. This description allows comparisons to be made with the more familiar generator coordinate (GCM) and adiabatic time-dependent Hartree-Fock (ATDHF) methods. It is shown that the action functional whose value for the instanton is the quasiclassical estimate of the decay exponent fulfills the minimum principle when additional constraints are imposed on trial fission paths.



# Instantons

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# Instantons

International Journal of Modern Physics E
Vol. 18, No. 4 (2009) 798–807


World Scientific
www.worldscientific.com

## NUCLEAR FISSION WITHIN THE MEAN-FIELD APPROACH

J. SKALSKI

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Received November 24, 2008

We discuss the mean-field instanton method for calculating decay rates of the spontaneous fission. Based on our recent work, we stress the role of the velocity-momentum relations which make the instanton action minimal, and the choice of a time-even density matrix and a time-odd hermitian operator as special variables making the adiabatic limit transparent. We also present an analytic study of a simple model of barrier tunneling, which illustrates the importance of the velocity-momentum constraints.



# Instantons

PHYSICAL REVIEW C VOLUME 36, NUMBER 5 NOVEMBER 1987

## Model for tunneling in many-particle systems

P. Arve and G. F. Bertsch
*Department of Physics, Michigan State University, East Lansing, Michigan 48824*

J. W. Negele and G. Puddu
*Center for Theoretical Physics, Laboratory for Nuclear Science, and Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139*

(Received 29 June 1987)

A model is proposed for studying tunneling in many-particle systems that is simple enough to solve and realistic enough to test various collective models of multidimensional barrier penetration. We find that the imaginary time dependent mean-field theory works very well in its domain of applicability, as does a continuum hopping approximation. The usual cranked mean field approximation is much less reliable.



# TDDFT quantized

TDDFT corresponds to the time-evolution equation of a quantum state moving along the manifold parameterized by $\rho_{mn}(t)$

$$i\hbar\dot{\rho}(t) = [h(t), \rho(t)]$$

Explicit quantum picture is recovered by:

Path integrals → Instantons

Quantization → Collective Hamiltonian



# Collective coordinates

Constraint variation of the functional,

$$E[\rho] \;=\; \int d\vec{r}\,\frac{\hbar^2}{2m}\tau(\vec{r}) + \mathcal{H}_P(\rho(\vec{r})) + \sum_\mu C_\mu\left(\langle\hat{Q}_\mu\rangle - q_\mu\right)^2$$

for $\langle\hat{Q}_\mu\rangle \;=\; \mathrm{Tr}\hat{Q}_\mu\rho = \sum_{ij} Q_{\mu,ij}\rho_{ji}$

gives the potential energy surface (PES): $E(q_\mu)$. Use physics to pick the right constraints! Physics of fission requires (at least?, at most?,...):

$$\begin{aligned}
\hat{Q}_1 &= \hat{Q}_{20} && \text{(axial quadrupole)}\\
\hat{Q}_2 &= \hat{Q}_{22} && \text{(nonaxial quadrupole)}\\
\hat{Q}_3 &= \hat{Q}_{30} && \text{(axial octupole)}\\
\hat{Q}_4 &= \hat{Q}_{40} && \text{(axial hexadecapole)}\\
\hat{Q}_5 &= \hat{N}^2 && \text{(neutron pairing)}\\
\hat{Q}_6 &= \hat{Z}^2 && \text{(proton pairing)}
\end{aligned}$$



# Class 3 PESs

## Continuous regular mesh ?

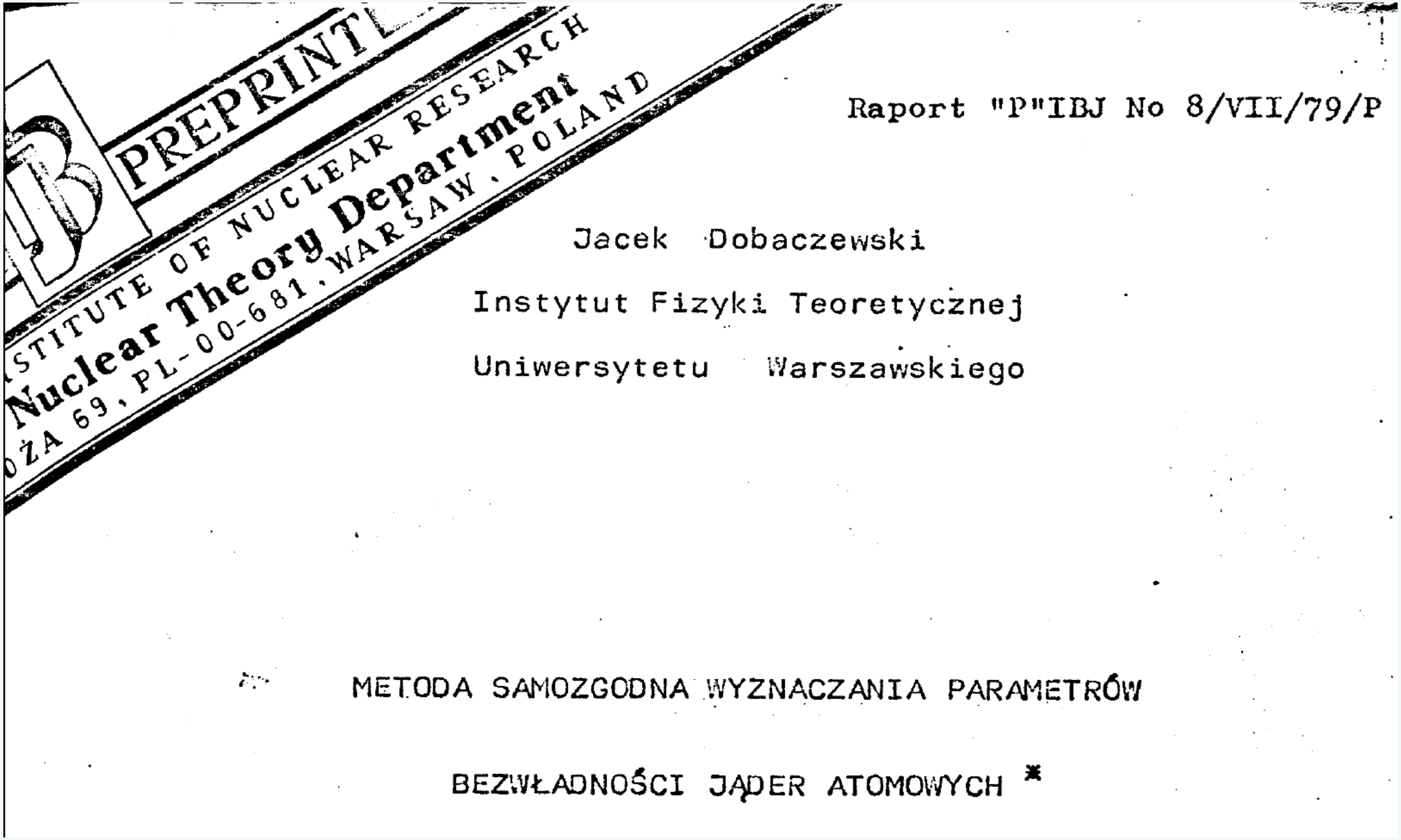
- dimension of a class 3 PES: $N = 1, 2, 3, 4, 5, 6, \ldots$ ?
- regular mesh + hypercube + $N > 2$ = huge number of points $N_p$
- dimension of the TDGCM + GOA hamiltonian matrix: $N_p^2$

## Use a sparse mesh !

- no hypercube, focus on the physics in any dimension
- optimal number of points
- solve the TDGCM + GOA equation with FEM

# How to solve the ATDDFT equation

$$i\hbar[\dot{\rho}_0, \rho_0] = [[h_0, \rho_1] + [h_1, \rho_0], \rho_0]$$



PREPRINT

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METODA SAMOZGODNA WYZNACZANIA PARAMETRÓW

BEZWŁADNOŚCI JĄDER ATOMOWYCH *

J. D. and J. Skalski, Nuc. Phys. A369 (1981) 123



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$$i\hbar[\dot{\rho}_0, \rho_0] = [[h_0, \rho_1] + [h_1, \rho_0], \rho_0]$$

J. D. and J. Skalski, Nuc. Phys. A369 (1981) 123

- 39 -

$$i\,[\dot{\mathcal{R}}_0, \mathcal{R}_0] = \mathcal{M}_0 \mathcal{R}_1 \qquad 3.46$$

lub dla kilku zmiennych kolektywnych (3.33)

$$B_{ij} = \frac{1}{2\dot{\alpha}_i\dot{\alpha}_j}\left(\mathcal{R}_1^i \middle| \mathcal{M}_0 \mathcal{R}_1^j\right) , \qquad 3.47$$

$$i\dot{\alpha}_i\left[\frac{\partial \mathcal{R}_0}{\partial \alpha_i}, \mathcal{R}_0\right] = \mathcal{M}_0 \mathcal{R}_1^i \; . \qquad 3.48$$



# CDC Cyber 170 series

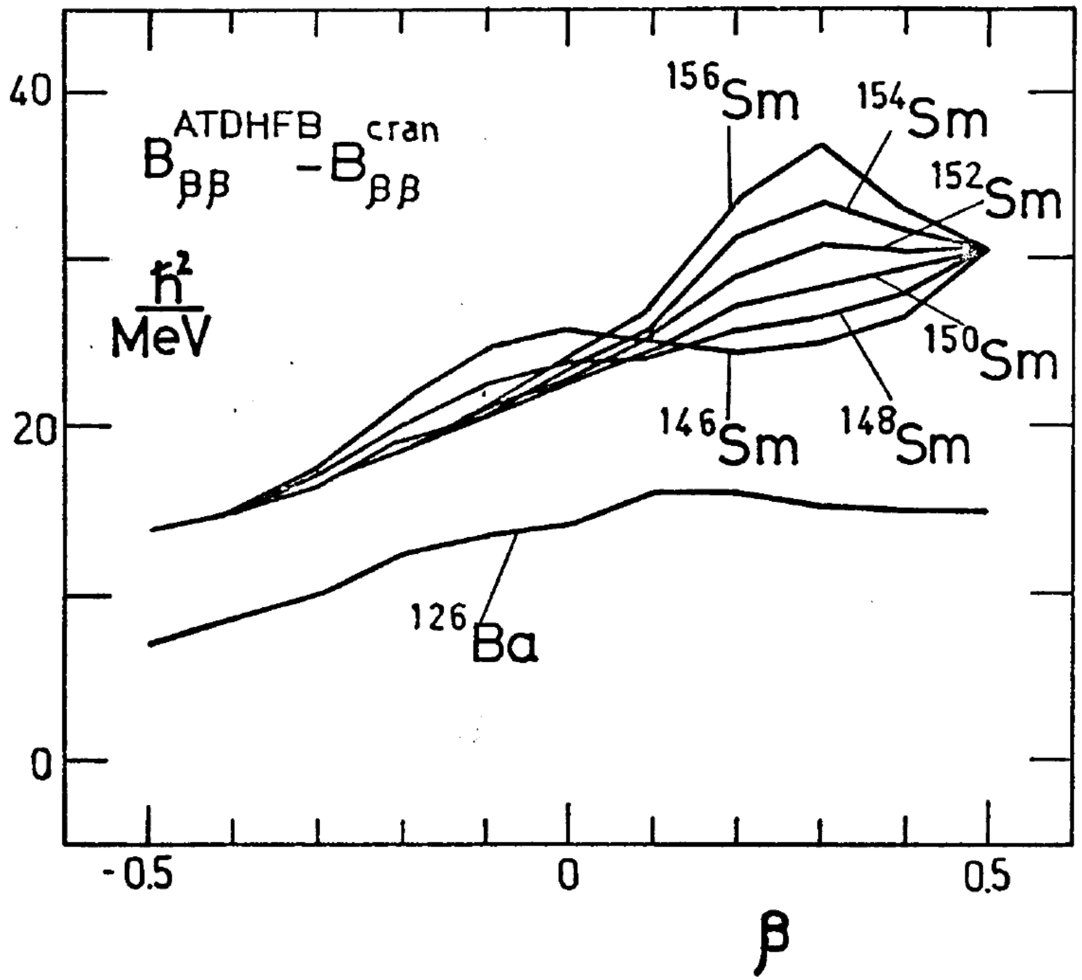
http://en.wikipedia.org/wiki/CDC_Cyber

## 60-bit 40MHz CPU
## 256 kwords memory of 12-bit words



# How to solve the ATDDFT equation

$$i\hbar[\dot{\rho}_0, \rho_0] = [[h_0, \rho_1] + [h_1, \rho_0], \rho_0]$$




Rys. 5.6 . Zależność od deformacji różnicy między parametrem masowym $B_{\beta\beta}$ policzonym w metodzie ATDHFB i w modelu ruchu wymuszonego (siły Skyrme'a SIV)

J. D. and J. Skalski, Nuc. Phys. A369 (1981) 123



# Perturbation expansion of ρ

For a perturbation expansion of the density matrix, $\rho = \rho_0 + \rho_1 + \rho_2 + \ldots$, projectivity of the density matrix, $\rho^2 = \rho$, up to second order gives:

$$(\rho_0 + \rho_1 + \rho_2)^2 = \rho_0 + \rho_1 + \rho_2,$$

which in zero, first, and second order reads:

$$\begin{aligned} \rho_0^2 &= \rho_0 = \rho_0^2, \\ \rho_0\rho_1 + \rho_1\rho_0 &= \rho_1 = [[\rho_1, \rho_0], \rho_0], \\ \rho_0\rho_2 + \rho_1\rho_1 + \rho_2\rho_0 &= \rho_2 = [[\rho_2, \rho_0], \rho_0] + \tfrac{1}{2}[[\rho_1, \rho_0], \rho_1]. \end{aligned}$$

Therefore, in the particle-hole (p-h) basis, the density matrices $\rho_0$, $\rho_1$, and $\rho_2$ have very specific structures:

$$\rho_0 = \begin{pmatrix} 1\ , & 0 \\ 0\ , & 0 \end{pmatrix}, \quad \rho_1 = \begin{pmatrix} 0\ , & \tilde{\rho}_1^+ \\ \tilde{\rho}_1\ , & 0 \end{pmatrix}, \quad \rho_2 = \begin{pmatrix} -\tilde{\rho}_1^+\tilde{\rho}_1\ , & \tilde{\rho}_2^+ \\ \tilde{\rho}_2\ , & \tilde{\rho}_1\tilde{\rho}_1^+ \end{pmatrix},$$

where $\tilde{\rho}_1$ and $\tilde{\rho}_2$ are rectangular matrices having only p-h matrix elements.



# Perturbation expansion of energy

For the perturbation expansion of the energy, one has:

$$E[\rho] = E[\rho_0 + \rho_1 + \rho_2] = E[\rho_0] + \sum_{ij} \frac{\partial E[\rho]}{\partial \rho_{ij}} (\rho_{1,ij} + \rho_{2,ij})$$

$$+ \frac{1}{2} \sum_{iji'j'} \frac{\partial^2 E[\rho]}{\partial \rho_{ij} \partial \rho_{i'j'}} (\rho_{1,ij} + \cancel{\rho_{2,ij}}) (\rho_{1,i'j'} + \cancel{\rho_{2,i'j'}})$$

where all derivatives are taken at $\rho \equiv \rho_0$. We now define the mean fields at zero and first order:

$$h_{0,ji} := \frac{\partial E[\rho]}{\partial \rho_{ij}} \quad , \quad h_{1,ji} := \sum_{i'j'} \frac{\partial^2 E[\rho]}{\partial \rho_{ij} \partial \rho_{i'j'}} \rho_{1,i'j'},$$

which gives the prturbation expansion of the energy,
$E[\rho] = E_0 + E_1 + E_2$, for

$$E_0 = E[\rho_0], \quad E_1 = \mathrm{Tr}\, h_0 \rho_1, \quad E_2 = \mathrm{Tr}\, h_0 \rho_2 + \tfrac{1}{2} \mathrm{Tr}\, h_1 \rho_1.$$



# Perturbation expansion of energy

By using the perturbation expansion of the density matrix, one has:

$$
\begin{aligned}
E_1 &= \mathrm{Tr}\,[h_0,\rho_0][\rho_0,\rho_1], \\
E_2 &= \mathrm{Tr}\,[h_0,\rho_0][\rho_0,\rho_2] + \tfrac{1}{2}\mathrm{Tr}\,\rho_1\,[[h_0,\rho_1]+[h_1,\rho_0],\rho_0].
\end{aligned}
$$

We see that the second term of $E_2$ contains the stability operator $\mathtt{M}_0$ which is a linear operator in the space of first-order corrections $\rho_1$:

$$\mathtt{M}_0\rho_1 := [[h_0,\rho_1]+[h_1,\rho_0],\rho_0]\,.$$

When expanded in the p-h indices $\mathtt{M}_0$ gives the standard RPA matrix. With respect to the scalar product $(\rho_1'|\rho_1) = \mathrm{Tr}\,\rho_1'^{+}\rho_1$, the stability operator is hermitian, $\mathtt{M}_0^\dagger = \mathtt{M}_0$.
At the stationary density $\rho_0$, given by the solution of the self-consistent equation $[h_0,\rho_0]=0$, we have

$$E_1 = 0 \quad , \quad E_2 = \tfrac{1}{2}(\rho_1|\mathtt{M}_0|\rho_1).$$



# Adiabatic expansion of ρ

We look for solutions of the TDDFT equations in the form of series expansion:

$$\rho(t) = \rho_0(t) + \rho_1(t) + \rho_2(t) + \ldots,$$

where every next term is “of the next order in velocities”. Proposals of how to define the "next order in velocities" are plenty. Let us begin with the simplest one:

$$\rho_0 = \rho_0(q_\mu(t)), \quad \text{(preseleced collective path)} \quad ,$$
$$\rho = \exp(i\chi)\rho_0 \exp(-i\chi), \quad \text{(Baranger \& Vénéroni)}$$

for a hermitian and time-even small $\chi$, $\mathrm{Tr}\{\chi^+\chi\} << 1$. For transparency, we drop the time arguments, since $\rho$, $\rho_0$, and $\chi$ all depend on time only through $q_\mu(t)$. Classical trajectories $q_\mu(t)$ are never looked upon – see quantization. We then obtain:

$$\rho = \rho_0 + i[\chi, \rho_0] - \tfrac{1}{2}[\chi, [\chi, \rho_0]] + \ldots,$$

that is, $\rho_1 = i[\chi, \rho_0]$ (time odd) and $\rho_2 = -\frac{1}{2}[\chi, [\chi, \rho_0]]$ (time even).



# Adiabatic TDDFT equation (ATDDFT)

By inserting the adiabatic expansion $\rho = \rho_0 + \rho_1 + \rho_2$ into the TDDFT equation $i\hbar\dot{\rho} = [h(\rho), \rho]$, one obtains:

$$i\hbar\dot{\rho}_0 + i\hbar\dot{\rho}_1 + i\hbar\dot{\rho}_2 = [h(\rho_0 + \rho_1 + \rho_2), \rho_0 + \rho_1 + \rho_2],$$

where the perturbation expansion of $h(\rho)$ gives

$$\begin{aligned} h(\rho) &= h(\rho_0 + \rho_1 + \rho_2) = h_0 + \sum_{ij} \frac{\partial h(\rho)}{\partial \rho_{ij}} (\rho_{1,ij} + \rho_{2,ij}) \\ &+ \tfrac{1}{2}\sum_{iji'j'} \frac{\partial^2 h(\rho)}{\partial \rho_{ij}\partial \rho_{i'j'}} (\rho_{1,ij} + \cancel{\rho_{2,ij}}) (\rho_{1,i'j'} + \cancel{\rho_{2,i'j'}}) \\ &= h_0 + h_1 + h_2. \end{aligned}$$

This allows us to split the TDDFT equation into the time-odd and time-even parts. After droping higher-order terms, we obtain

$$\begin{aligned} i\hbar\dot{\rho}_0 &= [h_0, \rho_1] + [h_1, \rho_0], \\ i\hbar\dot{\rho}_1 &= [h_0, \rho_0] + [h_0, \rho_2] + [h_1, \rho_1] + [h_2, \rho_0]. \end{aligned}$$



# ATDDFT equation

$$i\hbar[\dot{\rho}_0, \rho_0] = [[h_0, \rho_1] + [h_1, \rho_0], \rho_0]$$

$$i\hbar\dot{q}\left[\frac{\partial\rho_0}{\partial q}, \rho_0\right] = \mathtt{M}_0\rho_1$$

$$E_2 = \tfrac{1}{2}(\rho_1|\mathtt{M}_0|\rho_1) = \tfrac{1}{2}B\dot{q}^2 = \frac{p^2}{2B}$$

$$B = \left(\tfrac{\rho_1}{\dot{q}}\middle|\mathtt{M}_0\middle|\tfrac{\rho_1}{\dot{q}}\right) = \tfrac{i\hbar}{\dot{q}}\mathrm{Tr}\,\rho_1\left[\tfrac{\partial\rho_0}{\partial q}, \rho_0\right]$$

## ATDDFT mass parameter



# How to solve the ATDDFT equation

$$i\hbar[\dot{\rho}_0, \rho_0] = [[h_0, \rho_1] + [h_1, \rho_0], \rho_0]$$

1) For several collective variables use the chain rule:

$$\dot{\rho}_0 = \sum_\mu \dot{q}_\mu \frac{\partial \rho_0}{\partial q_\mu}$$

2) Work in the particle-hole basis:

$$i\hbar\dot{\rho}_{0,ph} = (\epsilon_p - \epsilon_h)\rho_{1,ph} + h_{1,ph}$$

3) Use the fixed-point method and iterate:

$$i\hbar\dot{\rho}_{0,ph} = (\epsilon_p - \epsilon_h)\rho^{(n+1)}_{1,ph} + h^{(n)}_{1,ph}$$



# How to solve the ATDDFT equation

$$i\hbar\dot{\rho}_{0,ph} = (\epsilon_p - \epsilon_h)\rho_{1,ph}^{(n+1)} + h_{1,ph}^{(n)}$$

$$\rho_{1,ph}^{(0)} = 0$$

$$\rho_{1,ph}^{(1)} = \frac{i\hbar\dot{\rho}_{0,ph}}{\epsilon_p - \epsilon_h}$$

$$B^{(1)} = 2\hbar^2\sum_{ph}\frac{|(\partial\rho_0/\partial q)_{ph}|^2}{\epsilon_p - \epsilon_h}$$

**Cranking mass parameter**

**(1), (2), (3), infinity = ATDDFT(B) mass parameter**



# How to solve the ATDDFT(B) equation

$$i\hbar\dot{\mathcal{R}}_{0,\alpha\beta} = (E_\alpha - (-E_\beta))\mathcal{R}^{(n+1)}_{1,\alpha\beta} + \mathcal{H}^{(n)}_{1,\alpha\beta}$$

$$\mathcal{R}^{(0)}_{1,\alpha\beta} = 0$$

$$\mathcal{R}^{(1)}_{1,\alpha\beta} = \frac{i\hbar\dot{\mathcal{R}}_{0,\alpha\beta}}{E_\alpha + E_\beta}$$

$$B^{(1)} = 2\hbar^2\sum_{\alpha\beta}\frac{|(\partial\mathcal{R}_0/\partial q)_{\alpha\beta}|^2}{E_\alpha + E_\beta}$$

**Cranking mass parameter**

**(1), (2), (3), infinity = ATDDFT(B) mass parameter**



# Quantization

816 *BORIS PODOLSKY*

momenta, we obtain the expression for $H$ in the quantum-mechanically correct form

$$H=\frac{1}{2\mu}\sum_{r=1}^{r=n}\sum_{s=1}^{s=n} g^{-1/4}p_r g^{1/2} g^{rs} p_s g^{-1/4}+U \tag{19}$$

In the classical case, when the order of factors is immaterial, this reduces to the usual form

$$H=\frac{1}{2\mu}\sum_{r=1}^{r=n}\sum_{s=1}^{s=n} g^{rs}p_r p_s+U \tag{20}$$

B. Podolsky, Phys. Rev. 32, 812 (1928)

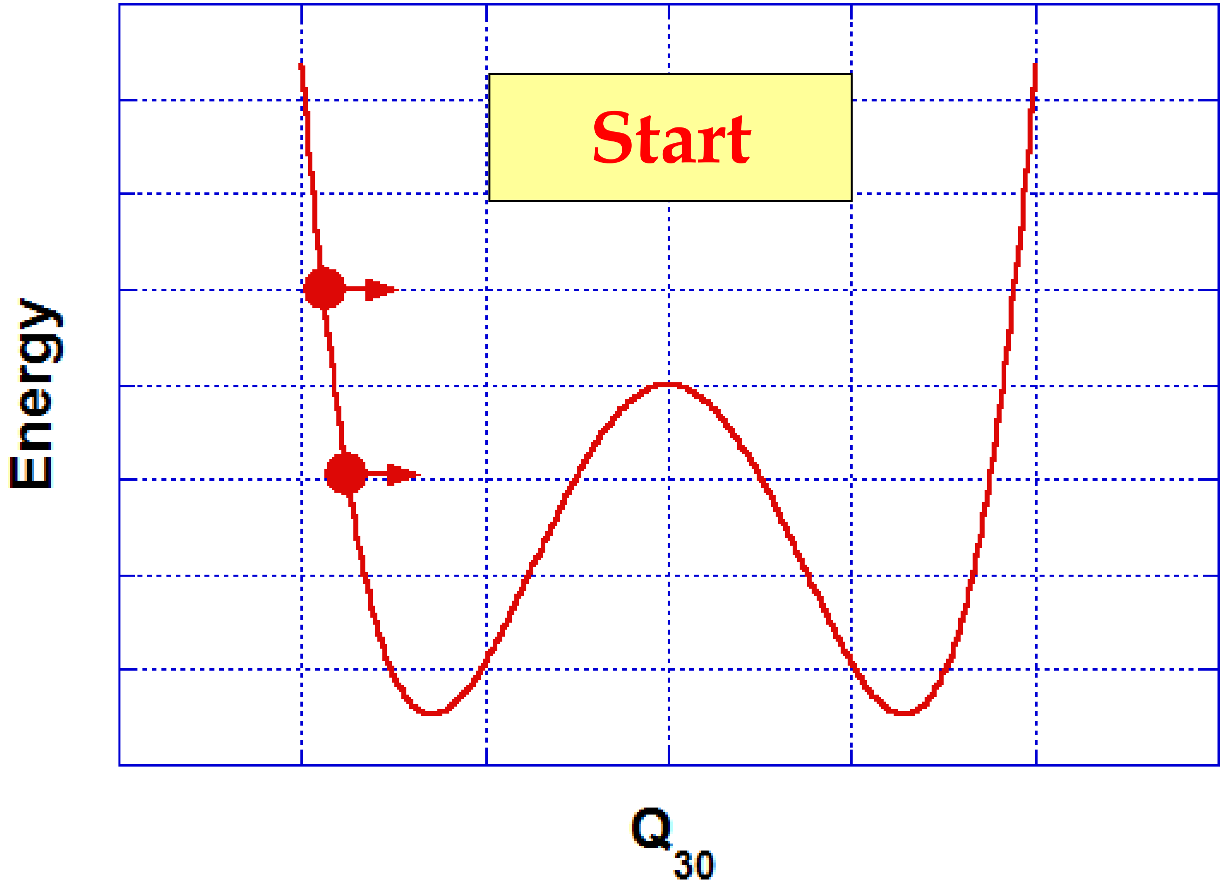
$$\hat{T} = \frac{1}{2}\sum_{\mu\nu} B^{-1/4}\hat{p}_\mu B^{1/2} B^{\mu\nu}\hat{p}_\nu B^{-1/4}$$

for $B = \det(B^{\mu\nu})$



# Can TDDFT penetrate the barrier?

## Homework exercise for the proud owners and users of the TDDFT codes:




$^{224}$Ra

axial

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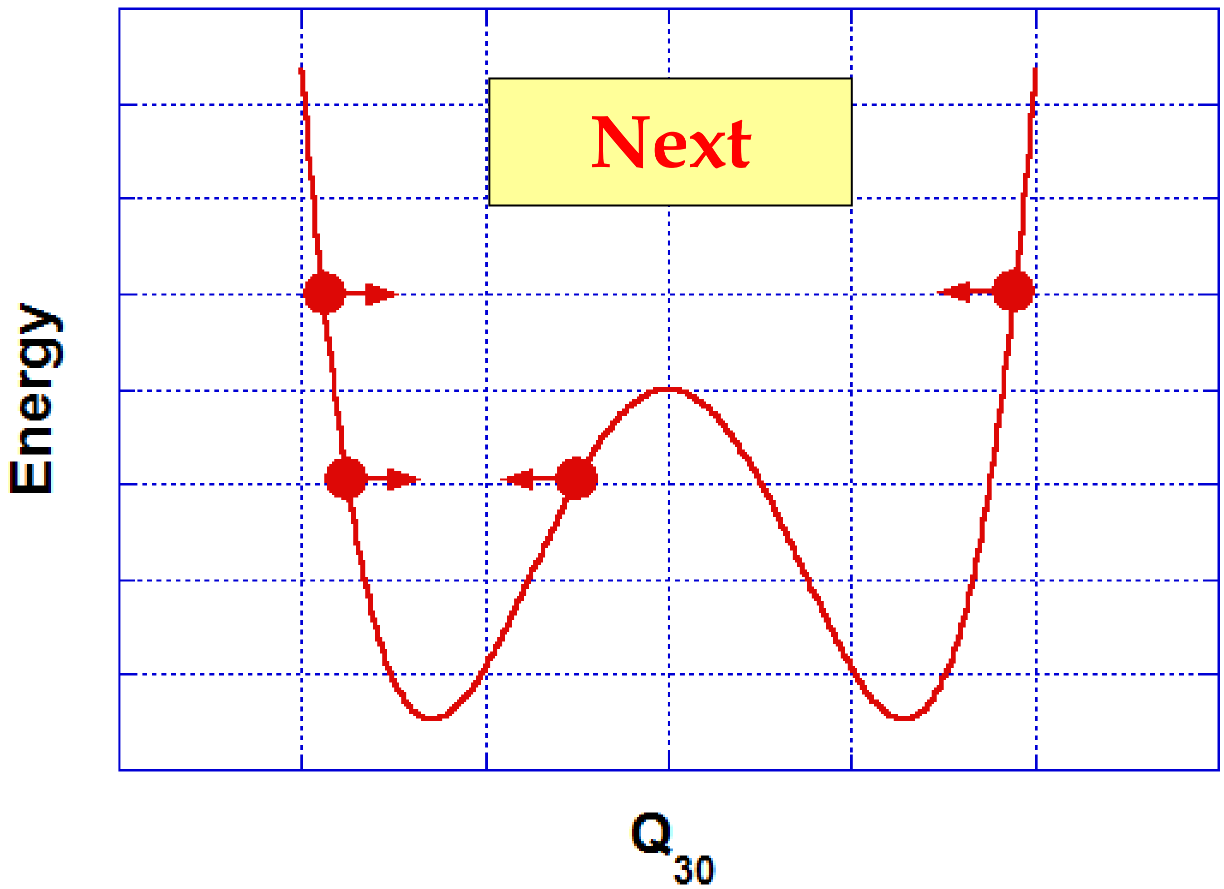


$^{224}Ra$

**axial**

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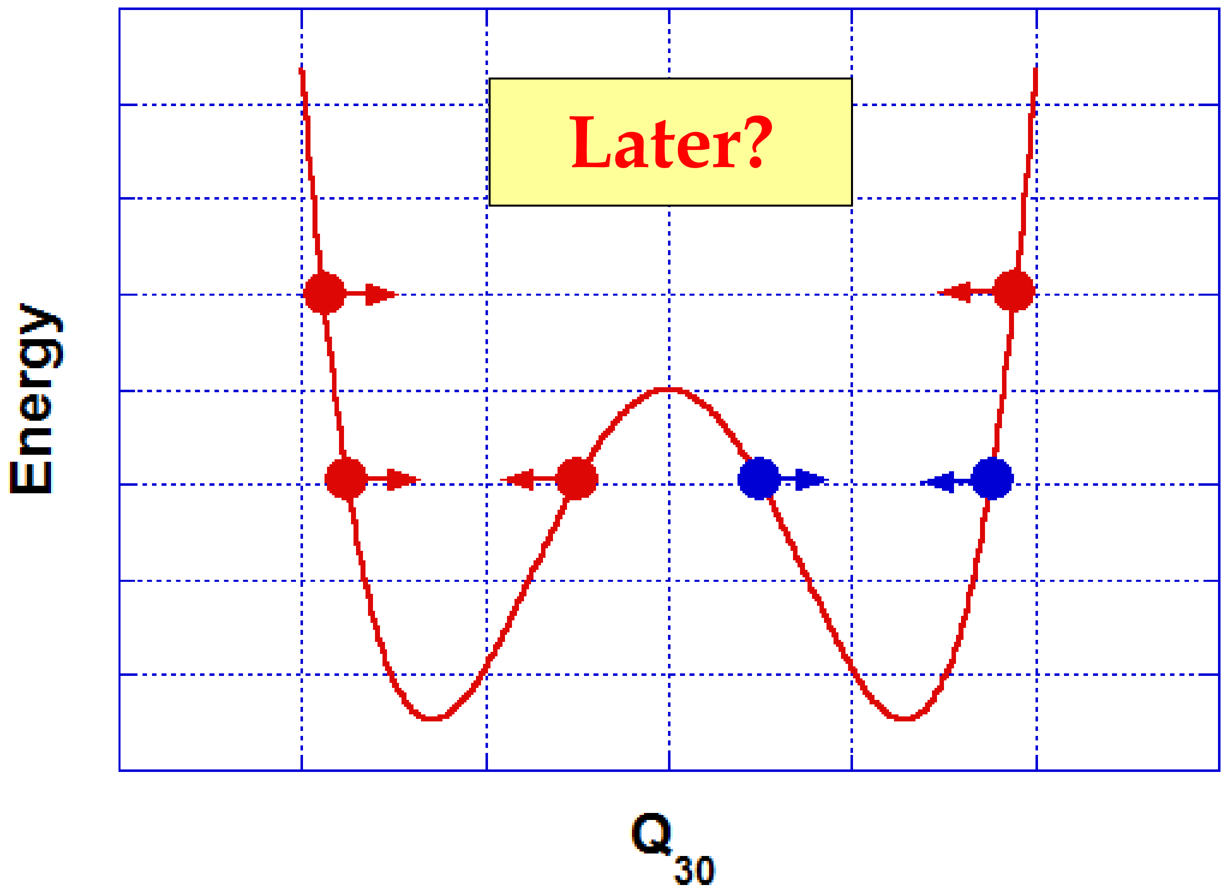


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# Is TDDFT classical or quantal?

Exact many-body time-dependent Schrödinger equation expressed in a complete basis of time-independent many-body states:

$$i\hbar|\dot{\Psi}(t)\rangle = \hat{H}|\Psi(t)\rangle \quad \text{for} \quad |\Psi(t)\rangle = \sum_n a_n(t)|\Psi_n\rangle .$$

This gives the evolution equations for the time-dependent probability amplitudes $a_n(t)$,

$$i\hbar\dot{a}_n(t) = \sum_m H_{nm}a_m(t),$$

where matrix elements $H_{nm} = \langle\Psi_n|\hat{H}|\Psi_m\rangle$ define the classical hamiltonian $\mathcal{H}(a^*,a)$, which governs the classical equations of motion,

$$\mathcal{H}(a^*,a) := \sum_{nm} a_n^* H_{nm} a_m \quad \Longrightarrow \quad i\hbar\dot{a}_n = \frac{\partial\mathcal{H}}{\partial a_n^*}.$$

Classical Hamilton equations of motion are obtained for real and imaginary parts of the probability amplitudes:

$$a_n = \frac{1}{\sqrt{2}}(q_n + ip_n/\hbar) \quad \Longrightarrow \quad \dot{q}_n = \frac{\partial\mathcal{H}}{\partial p_n} \quad , \quad \dot{p}_n = -\frac{\partial\mathcal{H}}{\partial q_n}.$$



# Can TDDFT penetrate the barrier?

Does TDDFT see any barrier at all? What barrier?

- Consider the PES defined in the standard way:

$$E(q) = \min_{\rho} E[\rho] \quad \text{for} \quad \begin{cases} q = \operatorname{Tr} Q\rho, \\ T^{+}\rho T = \rho, \\ \rho = \rho^{2} \end{cases}$$

This pertains to $q := q_{PES} = \operatorname{Tr} Q\rho$

- Consider the value of $q$ during the TDDFT evolution:

$$q = \operatorname{Tr} Q\rho = \operatorname{Tr} Q\rho_{+} \quad \text{for} \quad \begin{cases} T^{+}\rho T \neq \rho = \rho_{+} + \rho_{-}, \\ \rho = \rho^{2} \\ T^{+}\rho_{\pm} T = \pm\rho_{\pm}, \\ \rho_{+} = \rho_{+}^{2} + \rho_{-}^{2} \neq \rho_{+}^{2} \end{cases}$$

This pertains to $q := q_{TDDFT} = \operatorname{Tr} Q\rho_{+}$



# Can TDDFT penetrate the barrier?

**Does TDDFT see any barrier at all? What barrier?**

- $q_{PES}$ has nothing to do with $q_{TDDFT}$.
- $\rho_+$ does not belong to the class of dnsity matrices over which the PES was minimized.
- When TDDFT arrives at the "classical turning point", E=E(q), that is, $q = q_{TDDFT} = q_{PES}$, the TDDFT Slater determinant has nothing to do with the PES Slater determinant.

**Why wouldn't TDDFT go through?**

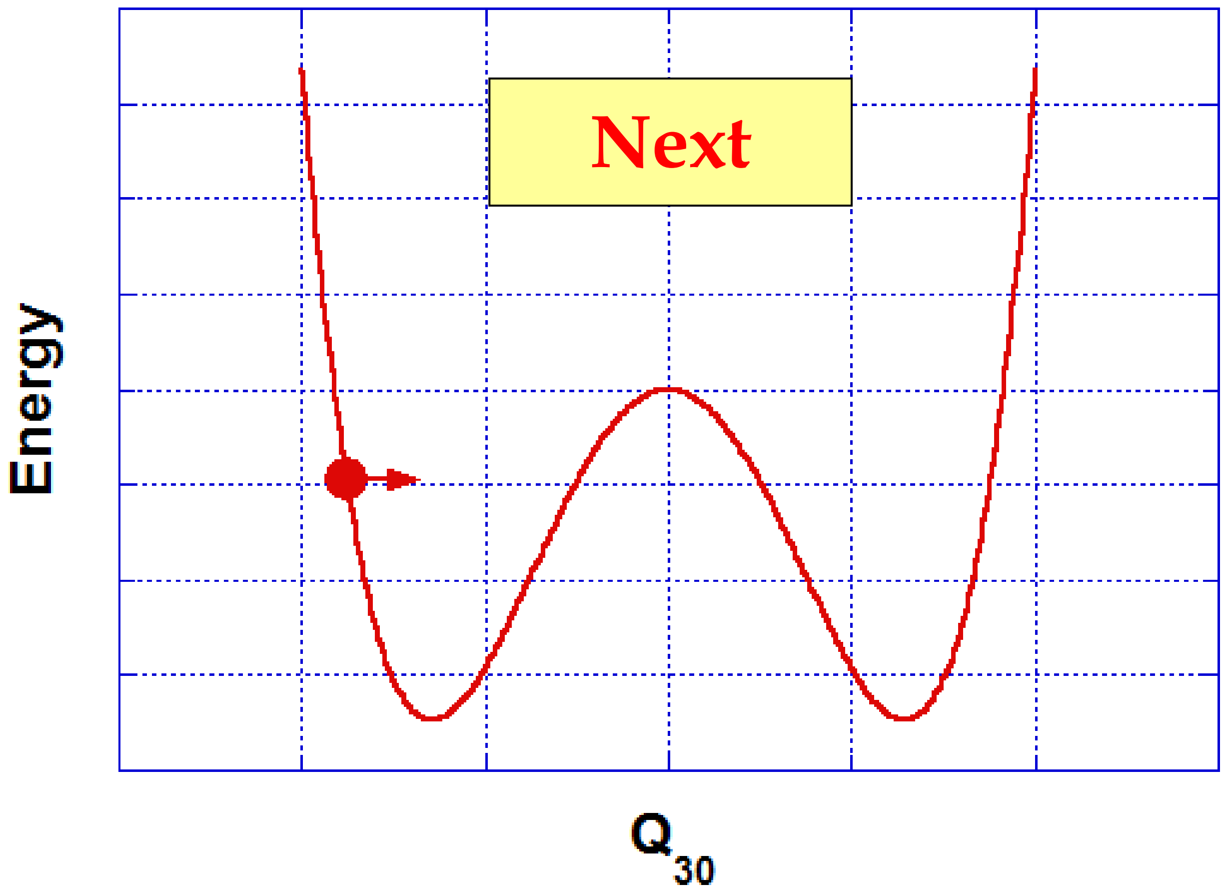


# Thank you



# Can TDDFT penetrate the barrier?

## Homework exercise for the proud owners and users of the TDDFT codes:



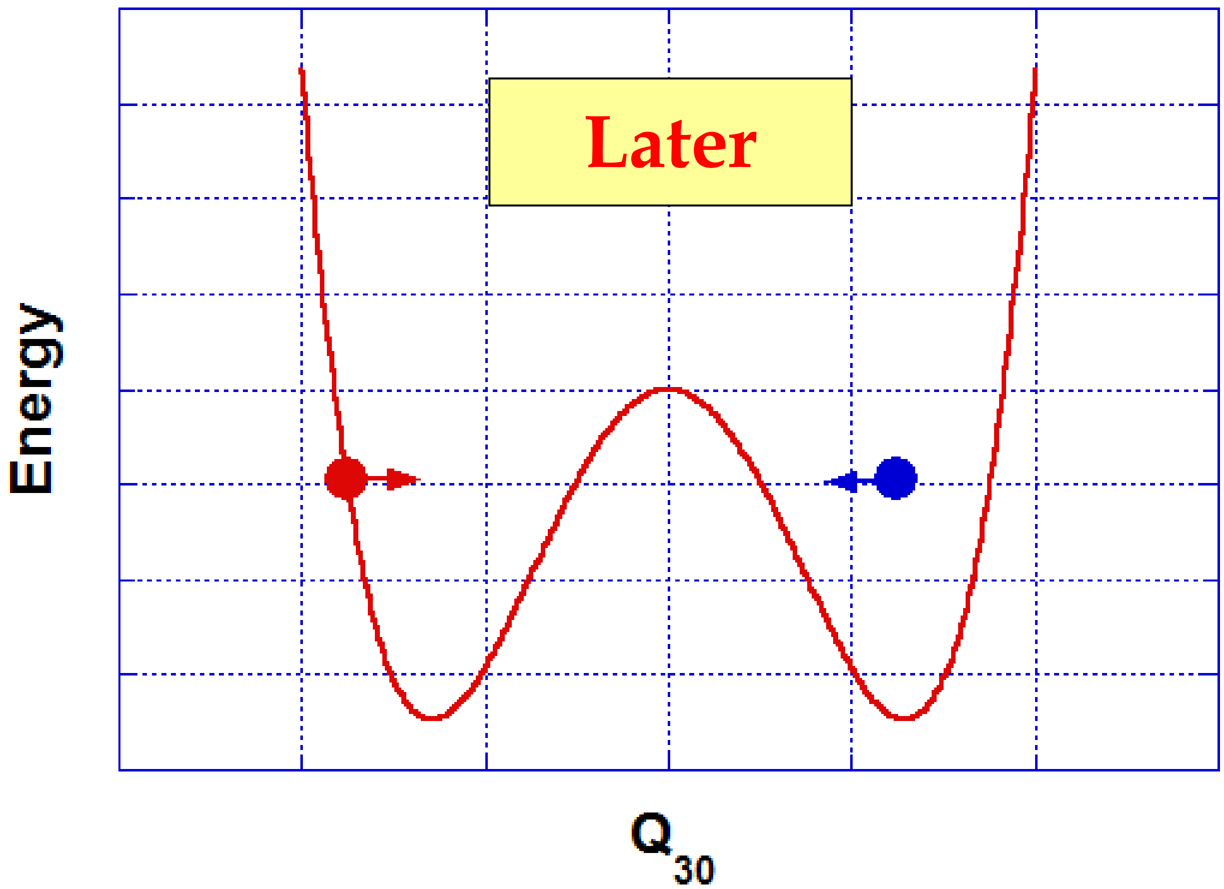

**$^{224}Ra$**

**axial**



# Can TDDFT penetrate the barrier?

## Homework exercise for the proud owners and users of the TDDFT codes:




$^{224}$Ra

axial