Quantum control of interacting particles: an MCTDHF–approach

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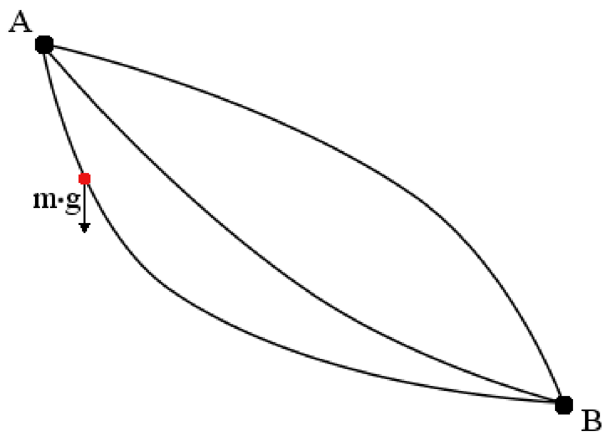
Chemical Physics
Weizmann Institute of Science

KITP, Santa Barbara 2009

Outline

1. Quantum control: goals, examples, and techniques

2. Basic ideas of optimal control theory (OCT)



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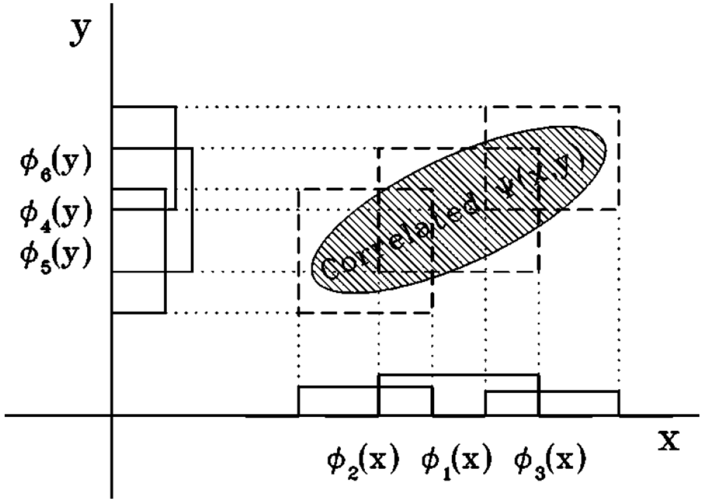
3. The multi-configuration time-dependent Hartree-Fock (MCTDHF) method



taken from J. Caillat, J. Zanghellini, M. Kitzler, O. Koch, W. Kreuzer, and A. Scrinzi, Phys. Rev. A **71**, 012712 (2005).

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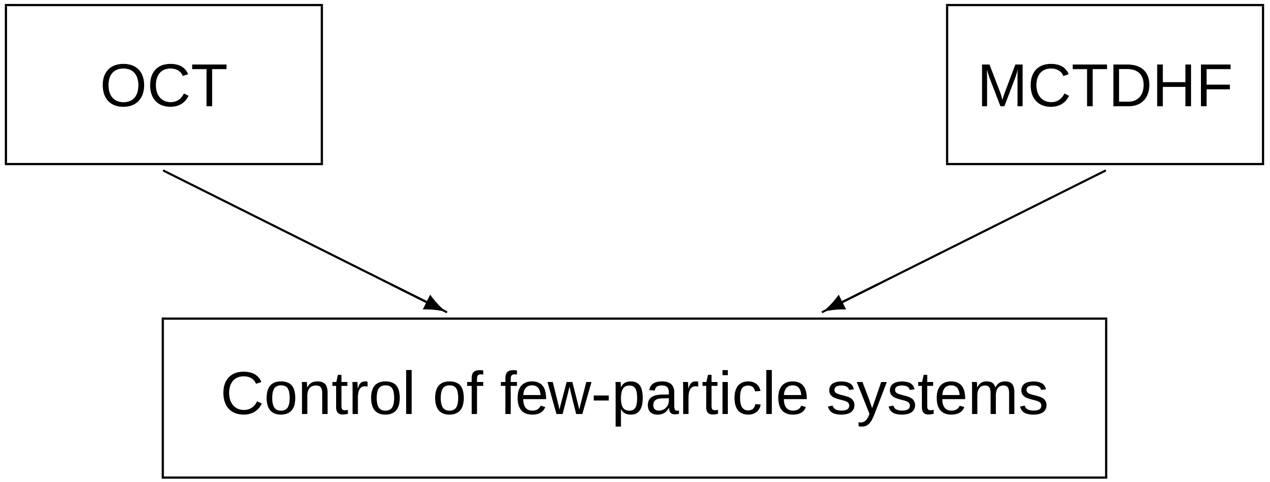
4. Combining OCT and MCTDHF



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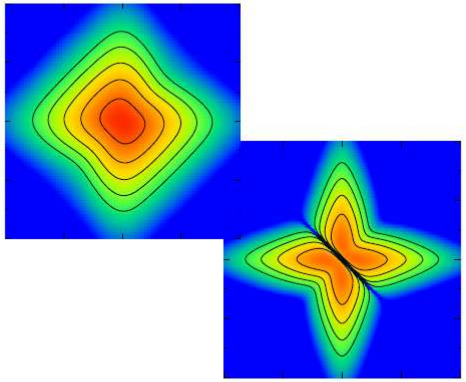
5. Applications: 1-dim. He atom and transport of cold atoms

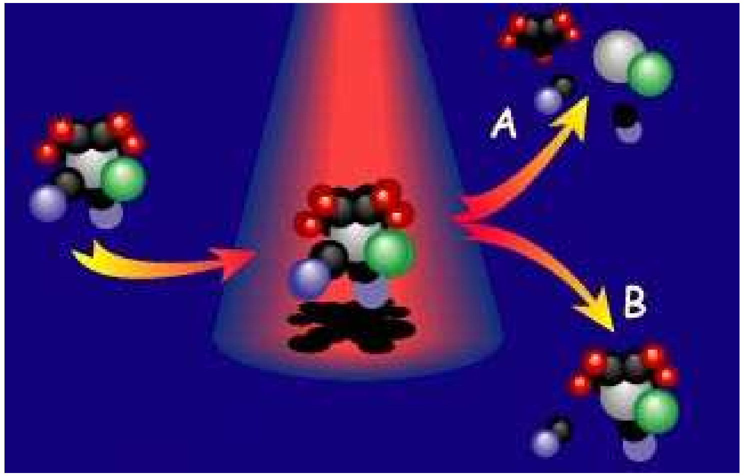


Quantum control: goals and examples

The goal of quantum control is to manipulate a quantum system in a desired way. This is a prerequisite for many experiments in fundamental research and for future technologies.

Examples are:

- The control of chemical reactions to do chemistry in a clean, non-statistical, cold, and thus energetically efficient way



http://wep1101.physik.uni-wuerzburg.de/

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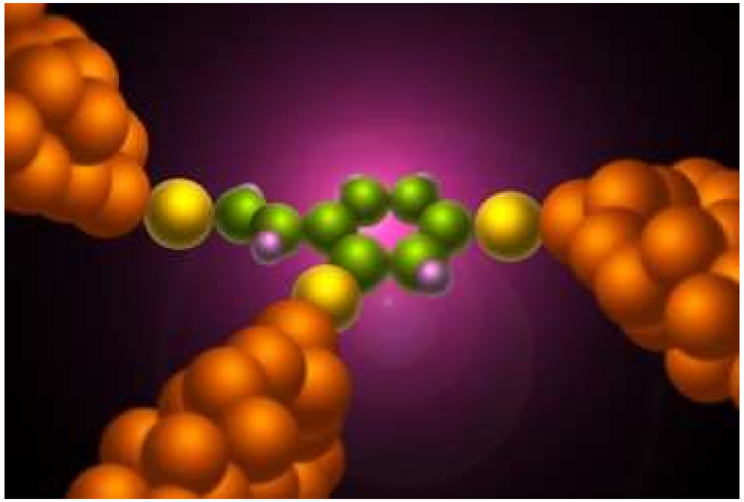

D. M. Cardamone *et al.*, Nanoletters, **6**, 2422 (2006)

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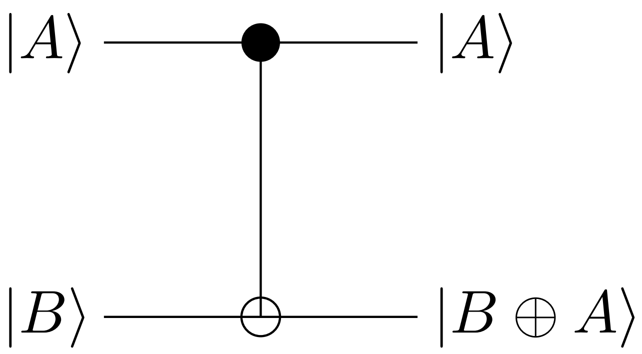

$$|00\rangle \to |00\rangle; \ |01\rangle \to |01\rangle$$

$$|10\rangle \to |11\rangle; \ |11\rangle \to |10\rangle$$

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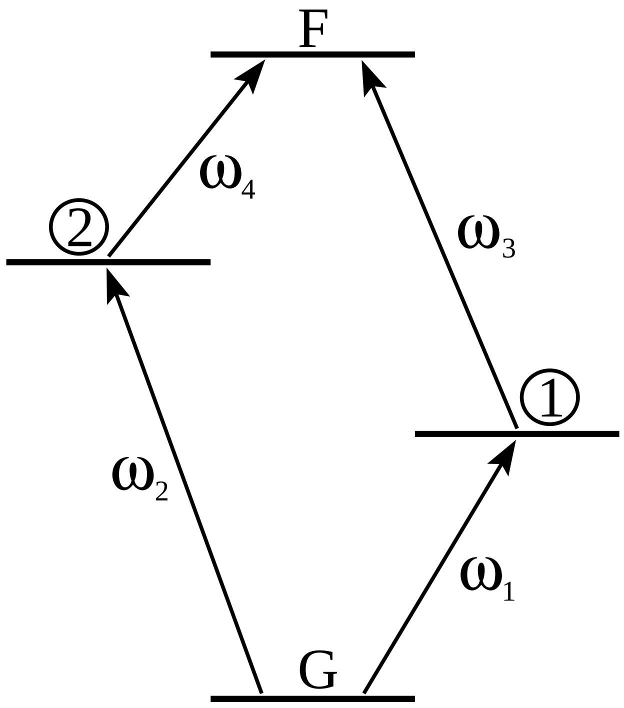
- The creation of special quantum states, e.g., Bell states, for quantum computing and fundamental tests of quantum mechanics
- Quantum state tomography

Quantum control: schemes and techniques

Several schemes have been developed for different control scenarios, e.g.,

- Brumer-Shapiro coherent control. Idea: Use interferences between different pathways to control processes



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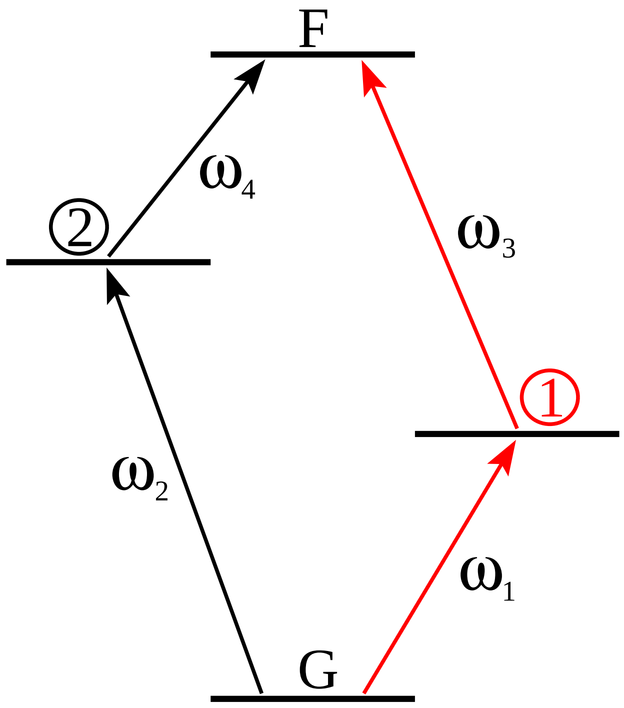

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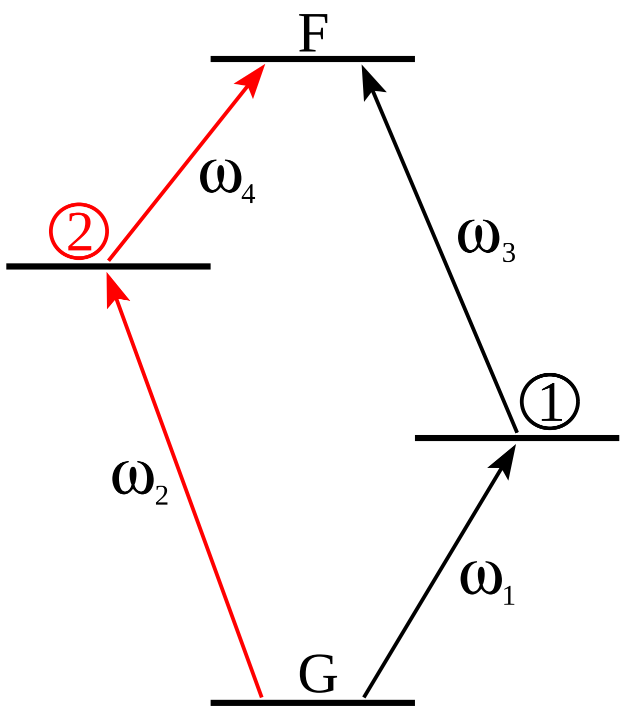

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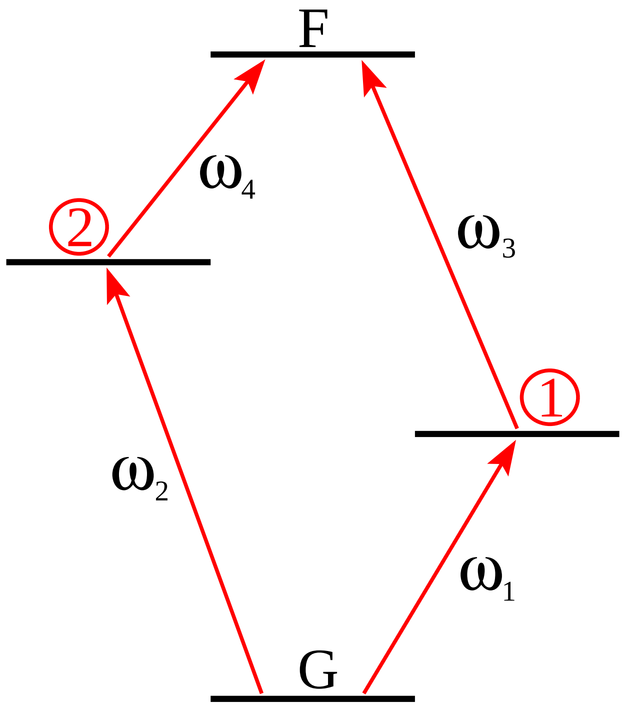

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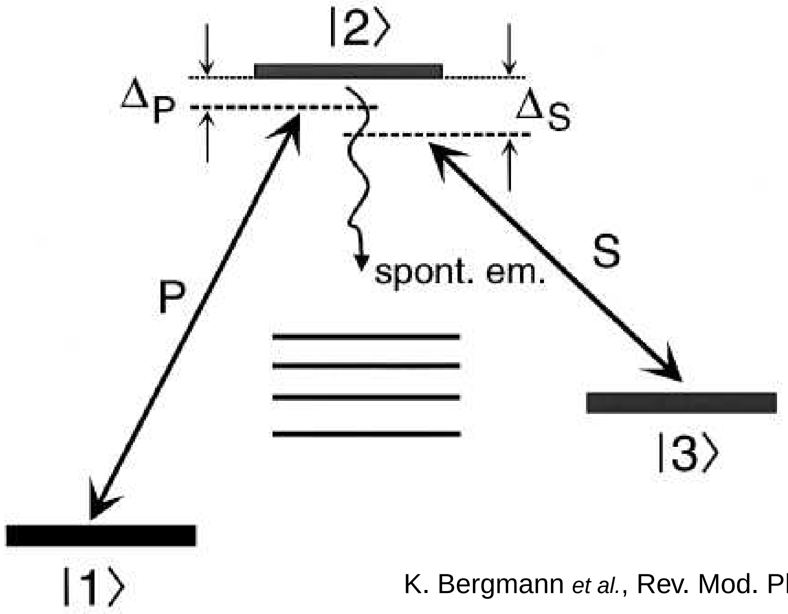

K. Bergmann *et al.*, Rev. Mod. Phys. **70**, 1003 (1998)

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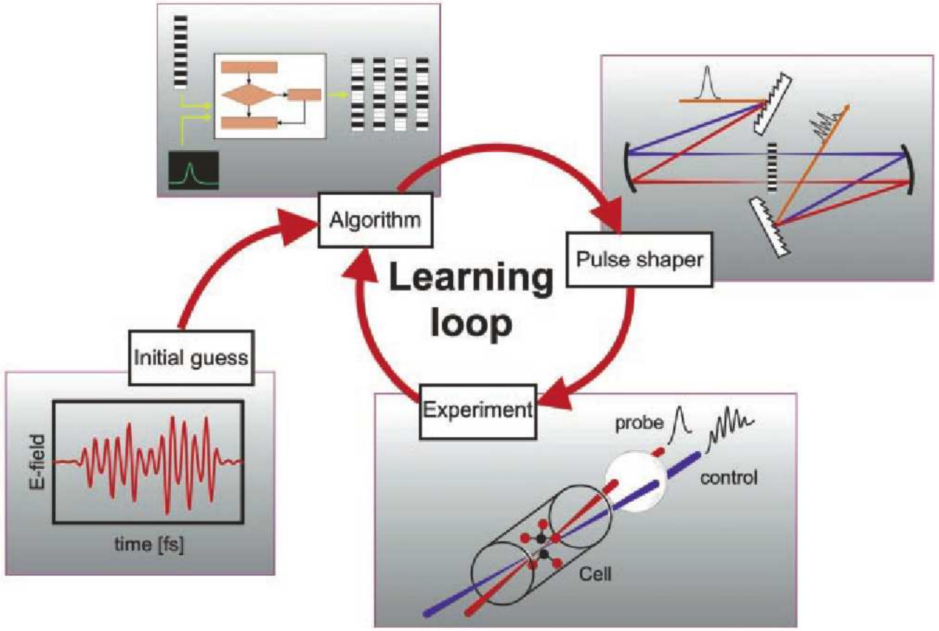
- Genetic/Learning schemes



H. Rabitz *et al.*, Science **288**, 824 (2000)

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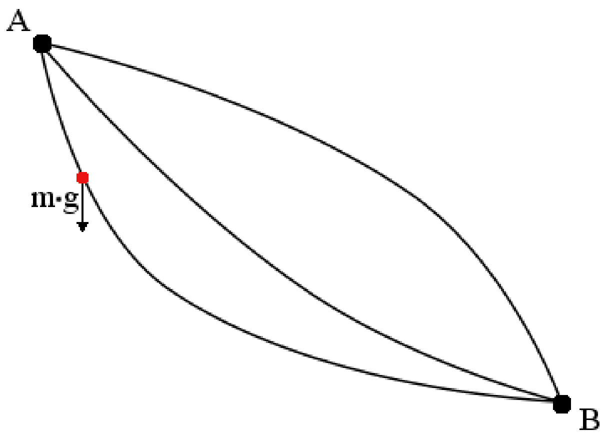
- Optimal control theory

The Brachistochrone problem (J. Bernoulli, 1696)

Question: Given a bead on a wire that connects two points A and B. What is the profile of the wire that minimizes the time the bead needs to go from A to B under the influence of a gravitational force $m \cdot g$?



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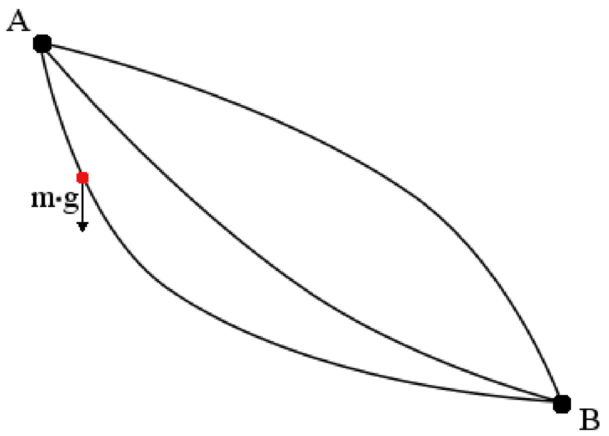

Three basic ingredients:

1.) Objective: fastest way from A to B

2.) Control: shape/angle of the wire

3.) Equation-of-motion: Newton's laws

Quantum optimal control theory

Examples for objectives:

- Population transfer from initial state $|a\rangle$ to state $|b\rangle$, i.e., maximization of $|\langle\psi(T)|b\rangle|^2$
- Optimization of high-harmonic generation / ionization yields
- Control of chemical reactions

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Examples for controls:

- Laser parameters: amplitude, frequencies, polarization, . . .
- Distance of ions in a trap
- Coupling to environment/measurement

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Examples for equation-of-motion:

- Schrödinger equation (TDSE), density-functional theory,...
- Approximations, e.g., perturbation theory,...

Quantum optimal control: fundamental equations

Example: Interacting particles in a laser field

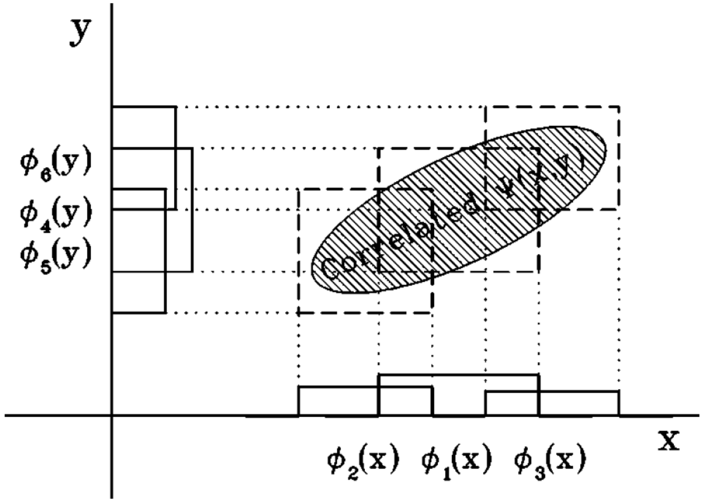
$$\hat{H} = \hat{T} + \hat{V}_{\text{ext}} + \hat{V}_{pp} + \sum_{k=1}^{3} \epsilon_k(t)\hat{\mu}_k$$

Optimal field to maximize, e.g., $\langle\psi(T)|\hat{A}|\psi(T)\rangle$

$$\begin{aligned} i\,\hbar\,\partial_t|\psi(t)\rangle &= \hat{H}\,|\psi(t)\rangle \\ i\,\hbar\,\partial_t|\chi(t)\rangle &= \hat{H}\,|\chi(t)\rangle, \qquad |\chi(T)\rangle = \hat{A}|\psi(T)\rangle \\ \epsilon_k(t) &\sim \operatorname{Im}\langle\chi(t)|\hat{\mu}_k|\psi(t)\rangle, \qquad k = 1, 2, 3 \end{aligned}$$

$\Longrightarrow$ Interacting TDSE must be solved!

The multi-configuration time-dependent Hartree-Fock method



taken from J. Caillat, J. Zanghellini, M. Kitzler, O. Koch, W. Kreuzer, and A. Scrinzi, Phys. Rev. A **71**, 012712 (2005).

Multi-configuration time-dependent Hartree-Fock

Idea: Reduce the number of degrees of freedom.

Starting point: Ansatz for the wavefunction $|\psi(t)\rangle$ of the form

$$|\psi(t)\rangle = \sum_{j_1=1}^{N_O} \cdots \sum_{j_N=1}^{N_O} c_{j_1 \ldots j_N}(t) \prod_{k=1}^{N} |\varphi_{j_k}(t)\rangle$$

Dirac-Frenkel variational principle determines the time evolution

$$\langle \delta\psi(t)| i\,\hbar\,\partial_t \;-\; \hat{H}|\psi(t)\rangle = 0\;,$$

i. e.,

$\Longrightarrow |\psi(t)\rangle$ does not satisfy the Schrödinger equation!

see, e.g., M. H. Beck, A. Jäckle, G. A. Worth, and H.-D. Meyer, Phys. Rep. **324**, 1 (2000).

The MCTDHF equations

The variational principle leads to the following coupled equations

$$
\begin{aligned}
i\,\hbar\,\dot{c}_J(t) &= \sum_L \langle \Phi_J | \hat{V}_{pp} | \Phi_L \rangle \, c_L(t) \\
i\,\hbar\,\partial_t \vec{\varphi}(t) &= [\hat{h} + (1 - \hat{P})\,\rho^{-1} \langle \hat{V}_{pp} \rangle] \vec{\varphi}(t)
\end{aligned}
$$

with

- $\vec{\varphi}(t) = (|\varphi_1\rangle \dots |\varphi_n\rangle)^T$,
- $|\Phi_J\rangle = \prod_{k=1}^{N} |\varphi_{j_k}(t)\rangle$ and $J = j_1, \dots, j_N$,
- projector $\hat{P} = \sum_j |\varphi_j\rangle\langle\varphi_j|$ and single-particle Hamiltonian $\hat{h}$,
- density matrix ρ_{jl} and mean-fields $\langle \hat{V}_{pp} \rangle_{jl}$ ($\longrightarrow$ nonlinearity).

see, e.g., M. Nest, T. Klamroth, and P. Saalfrank, J. Chem. Phys. **122**, 124102 (2005).

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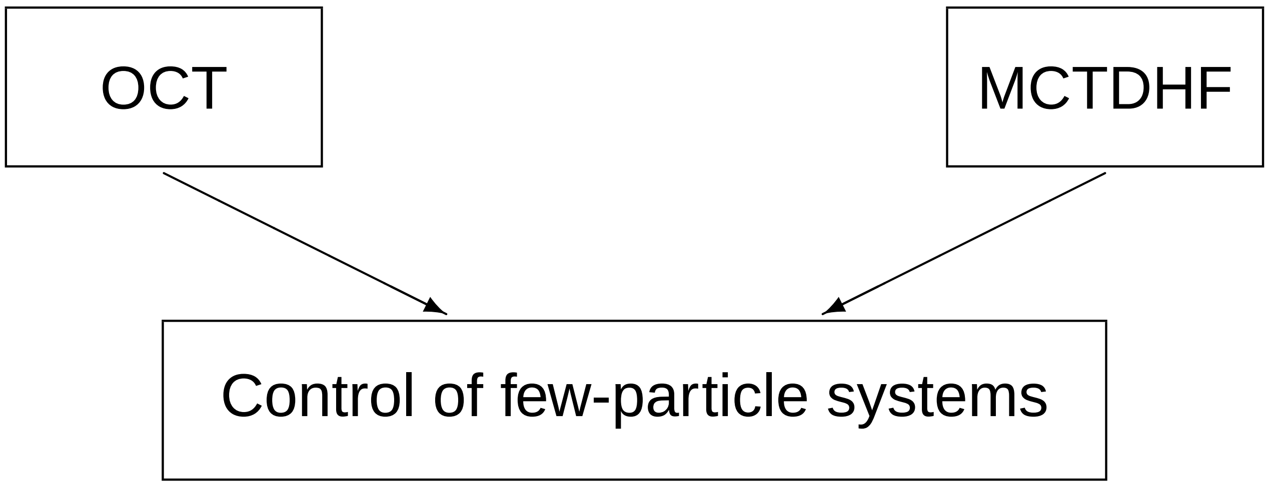
Advantages:

- Convergence towards the exact result for increasing N_{O}
- First-principle approach (no model parameters,...)
- Non-perturbative $\longrightarrow$ access to strong-field phenomena, e.g., high-harmonic generation
- Description of bound and continuum states

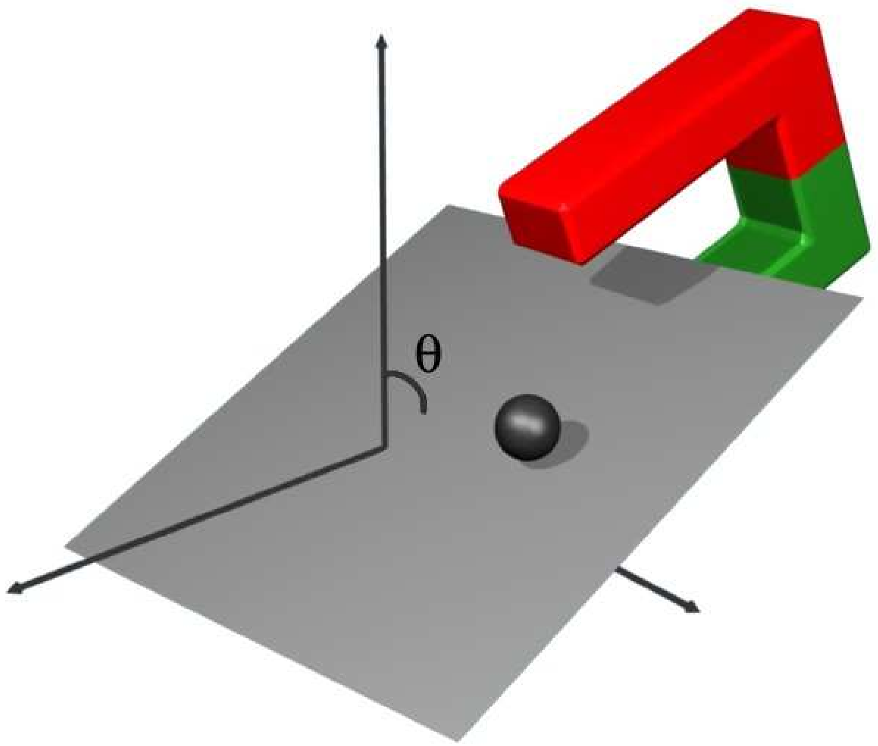
Combining OCT and MCTDHF



MCTDHF+OCT: Controlling a subspace

The MCTDHF state is always an element of the subspace spanned by the orbitals $|\varphi_n\rangle$.

$\Longrightarrow$ we have to control the dynamics inside the subspace and the dynamics of the subspace!



Approach I: nonlinear control theory

Starting point: Control Hamiltonian with 'adjoint' orbitals $\chi_j(t)$, coefficients $\gamma_J(t)$, and field penalty $p(\epsilon)$

$$\mathrm{H} = \sum_j \langle \chi_j(t) | f_j(t) \rangle + c.c. + \sum_J \gamma_J^*(t)\, g_J(t) + c.c. - p(\epsilon)$$

with

$$g_J(t) = \dot{c}_J(t), \qquad \text{and} \qquad |f_j(t)\rangle = \partial_t |\varphi_j(t)\rangle \ .$$

The resulting control equations are

$$\dot{\gamma}_J(t) = -\frac{\delta \mathrm{H}}{\delta c_J^*(t)}, \qquad \partial_t\, \chi_j(t) = -\frac{\delta \mathrm{H}}{\delta \varphi_j^*(t)}, \qquad \frac{\delta \mathrm{H}}{\delta \epsilon(t)} = 0$$

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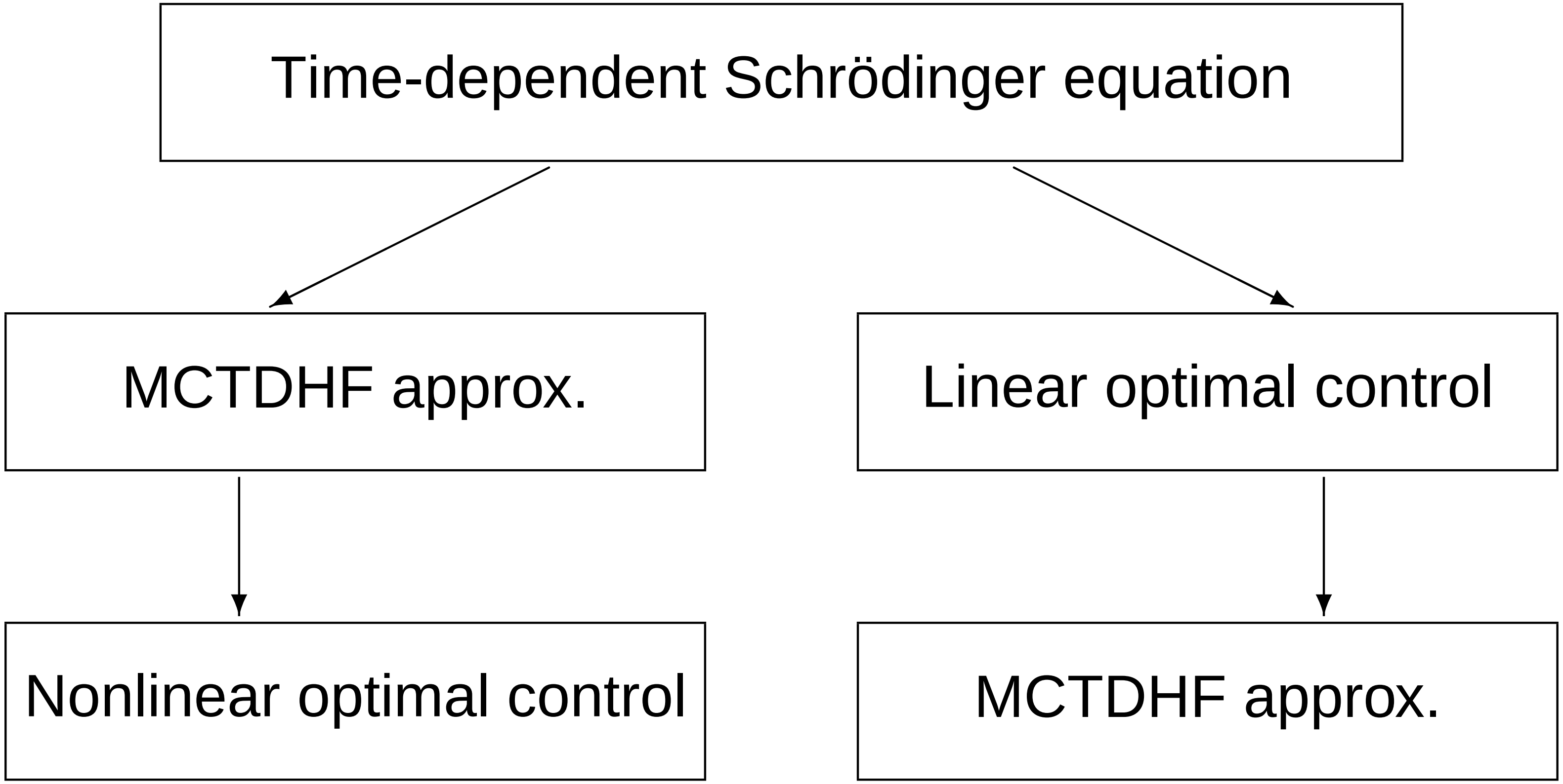
Problems:

- Control equations very involved due to strong nonlinearity
- Difficult numerics
- Control equations do not reduce to linear control equations for $N_{\mathrm{O}} \longrightarrow \infty$

Approach II: linear control theory

Idea: Using the MCTDHF method as an efficient tool to solve the Schrödinger equation, i.e., first derive the control equations and then solve them approximately using the MCTDHF approach[1].



[1] similar to Wang *et al.*, J. Chem. Phys. **125**, 014102 (2006)

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Advantages:

- Only linear control is required $\longrightarrow$ many known properties of linear control can be used
- Numerical implementation is straight-forward
- Requires less computational efforts

Disadvantages:

- Monotonic change of the objective not guaranteed for all N_{O}
- May require large numbers of orbitals N_{O}
- Approximate results for small N_{O}, e.g., Hartree-Fock results, cannot be obtained

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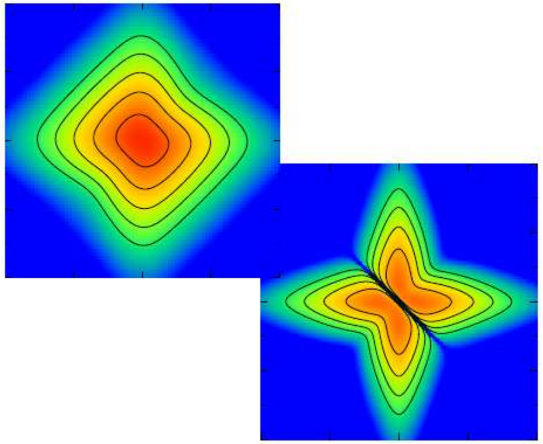
Open question: How severe are the disadvantages?

1. Application: one-dimensional He atom

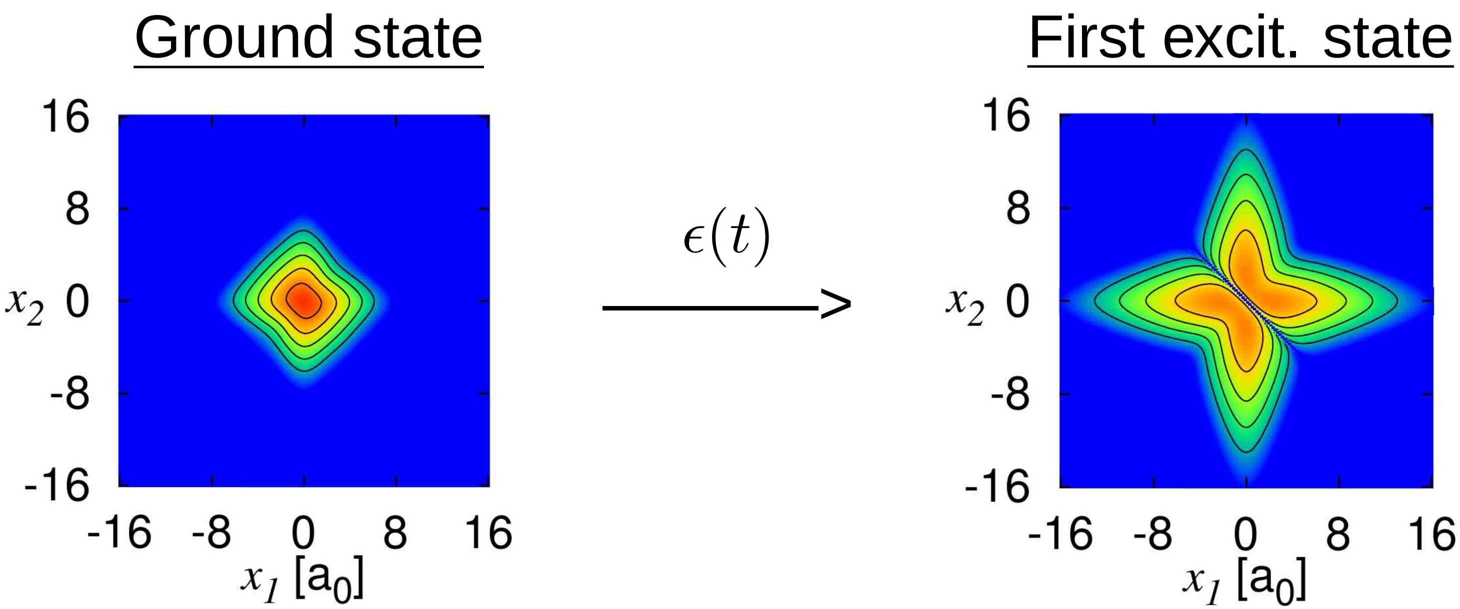


The model: one-dimensional He atom

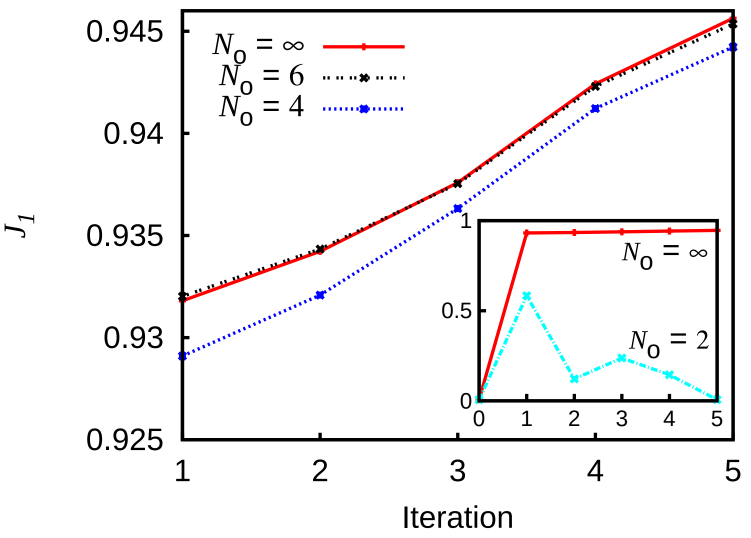
The system is described by the Hamiltonian

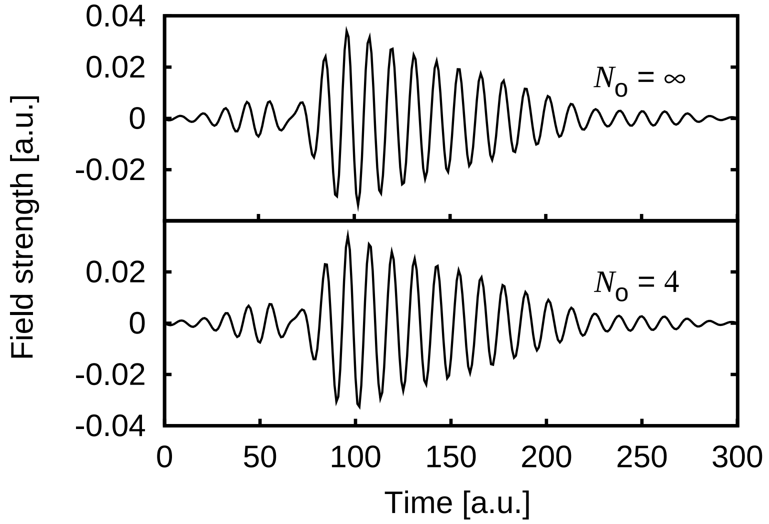
$$\hat{H} = \sum_{j=1}^{2} \frac{\hat{p}_j^2}{2m} - \frac{2}{\sqrt{(\hat{x}_j)^2 + 1}} + \epsilon(t)\hat{x}_j + \frac{1}{\sqrt{(\hat{x}_1 - \hat{x}_2)^2 + 1}}$$

and the objective is to maximize $J_1 = |\langle\psi(T)|\psi_1\rangle|^2$ at time $T = 300$ a.u. starting from the ground state $|\psi_0\rangle$



He atom: optimization results for $\psi_0 \rightarrow \psi_1$





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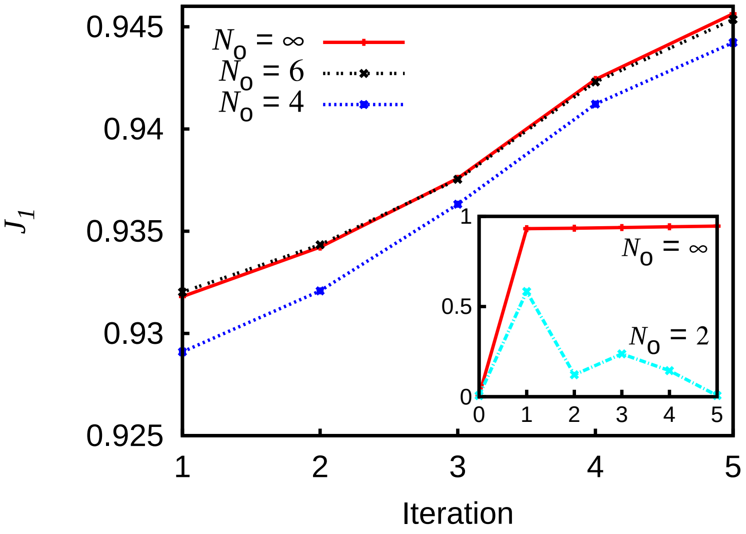
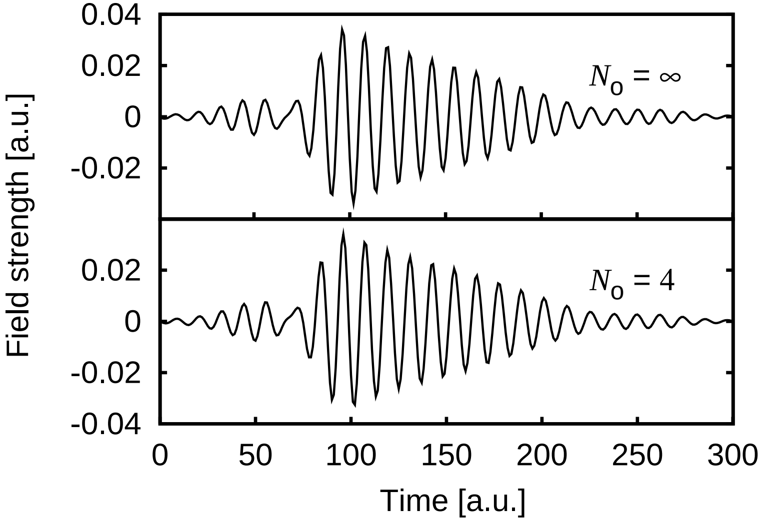
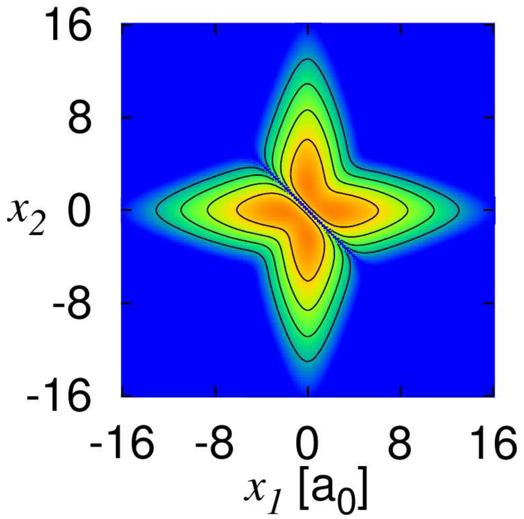
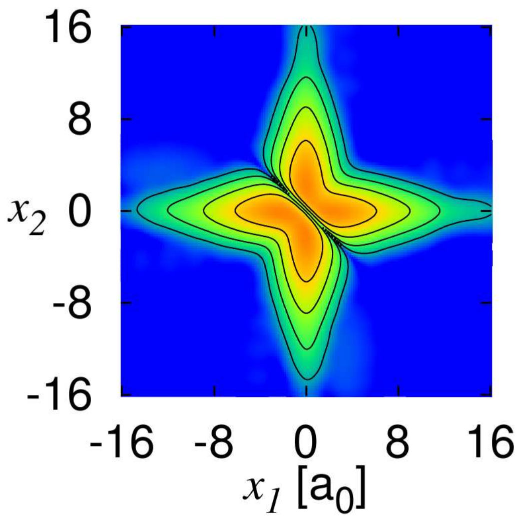




Target state

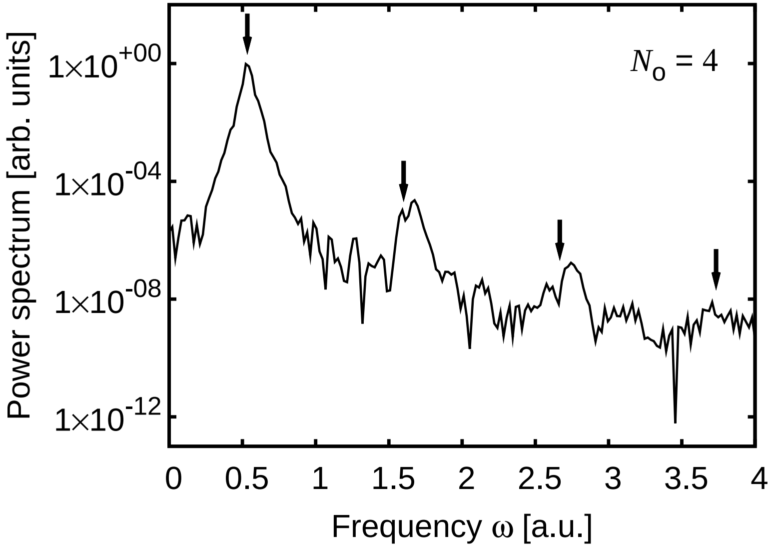


Optimized state

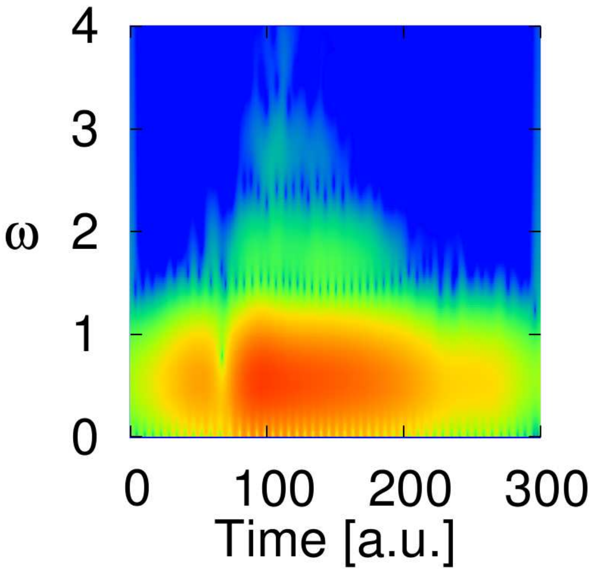


He atom: transition mechanism

Powerspectrum of $\epsilon(t)$



Husimi plot



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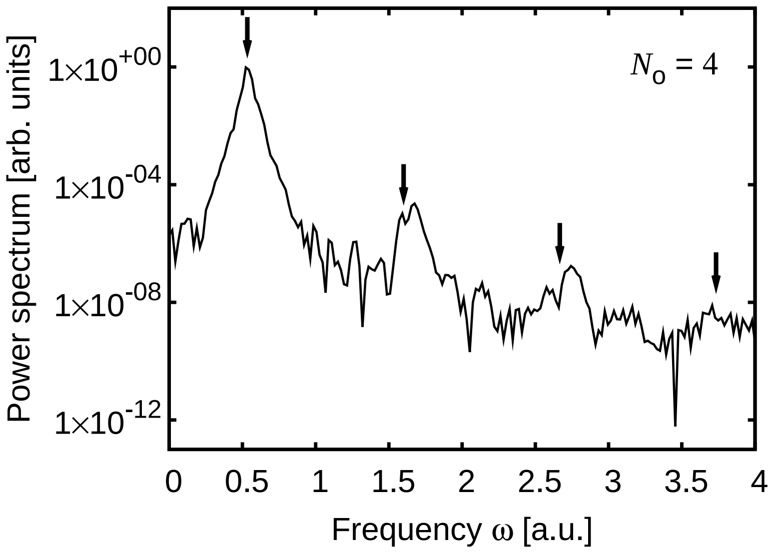

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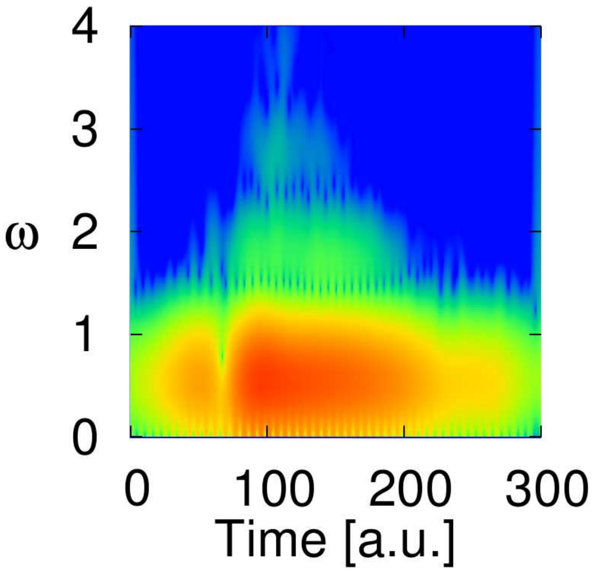

$\Longrightarrow$ Main mechanism: resonant transition from $|\psi_0\rangle$ to $|\psi_1\rangle$

$\longrightarrow$ Optimization corresponds to a large extent to an optimization in a two-level system

Performance of simple π-pulses

In a two-level system the resonant pulse $\epsilon(t) = A(t)\,\sin(\omega_{01} t)$ must satisfy the pulse-area theorem

$$\mu \int_0^T A(t)\,dt \;=\; \pi$$

for a complete population transfer. μ is the coupling dipole matrix element.

For $A(t) = 0.034\,\sin^2(\pi t/T)$ one obtains $T \approx 170$ and

$$\boxed{N_{\mathrm{O}} = 4\text{:} \longrightarrow J_1 \;=\; 0.92}$$

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For $A(t) = 0.034\,\sin^2(\pi t/T)$ one obtains $T \approx 170$ and

$$N_{\mathrm{O}} = 2\text{:} \longrightarrow J_1 \;=\; 0.06$$

Despite $\langle \psi_1^{N_{\mathrm{O}}=2} | \psi_1^{N_{\mathrm{O}}=4} \rangle = 0.997$ and $|E_1^{N_{\mathrm{O}}=2} - E_1^{N_{\mathrm{O}}=4}| < 0.1$ eV

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$$N_{\mathrm{O}} = 2\text{:} \longrightarrow J_1 \;=\; 0.06$$

$\Longrightarrow$ Wrong dynamics of the state caused by the violation of the superposition principle due to the nonlinearity!

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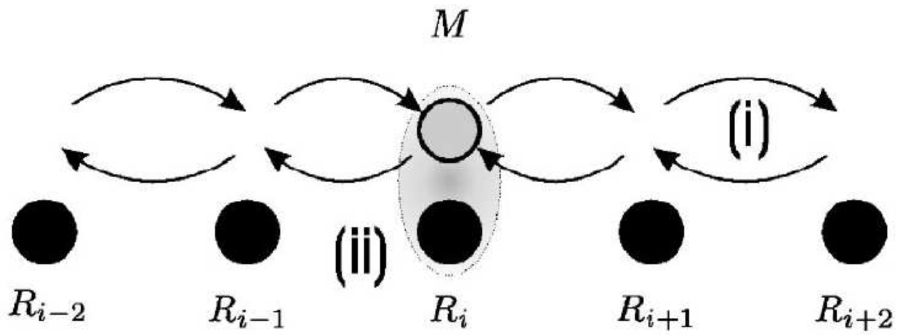
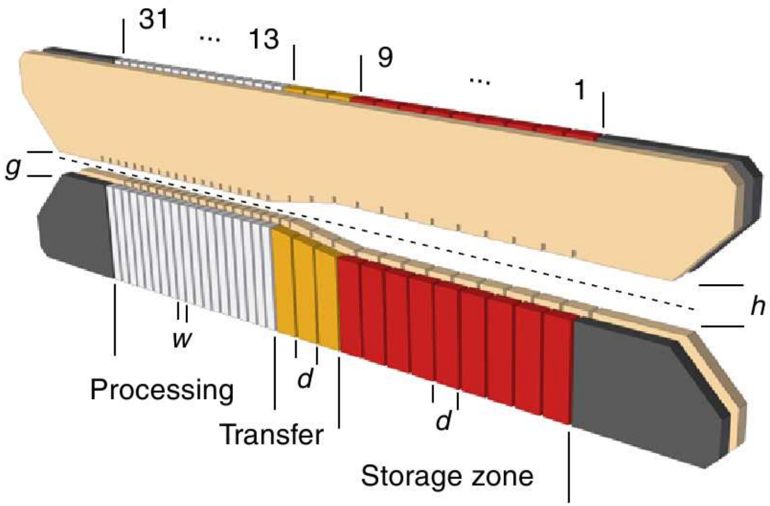
‘Nonlinear’ quantum control meaningful?

2. Application: transport in an optical lattice



T. Calarco, *et al.*, Phys. Rev. A, **70**, 012306 (2004).

2. Application: transport in an optical lattice



http://www.uni-ulm.de/nawi/nawi-qiv/forschung.html

Transport of cold Rb atoms in an optical lattice

The Hamiltonian of the system is given by

$$\hat{H} = \sum_{j=1}^{2} \frac{\hat{p}_j^2}{2M_{\mathrm{Rb}}} + V_{\mathrm{ext}}(\hat{x}_j, V_0(t), \beta(t), \theta(t)) + g\,\delta(\hat{x}_1 - \hat{x}_2)$$

with the optical lattice

$$\begin{aligned} V_{\mathrm{ext}}(\hat{x}_j, V_0(t), \beta(t), \theta(t)) = \\ - \quad V_0(t)\Big\{ \cos^2\left(\frac{\beta(t)}{2}\right)\left(1 + \cos^2\left(k_{\mathrm{L}}\hat{x}_j - \pi/2\right)\right) \\ + \quad \sin^2\left(\frac{\beta(t)}{2}\right)\left[1 + \cos\left(k_{\mathrm{L}}\hat{x}_j - \theta(t) - \pi/2\right)\right]^2 \Big\}\,. \end{aligned}$$

G. De Chiara, *et al.*, Phys. Rev. A, **77**, 052333 (2008)

containing the controls $V_0(t)$, $\beta(t)$, and $\theta(t)$.

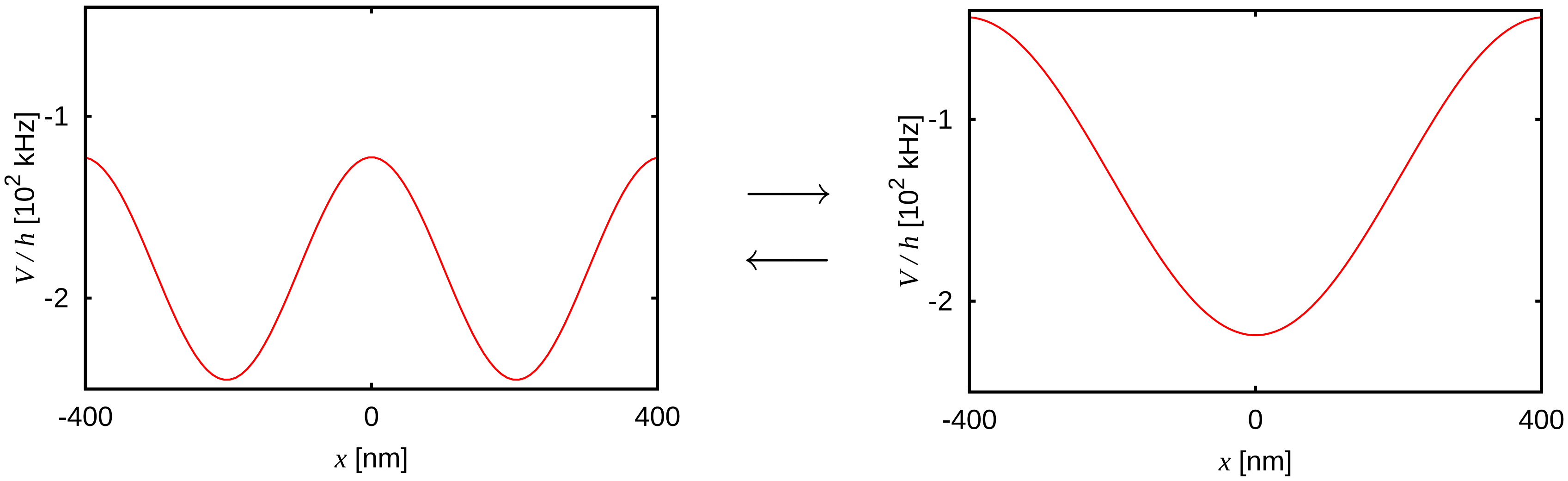
Transport of cold Rb atoms in an optical lattice

The Hamiltonian of the system is given by

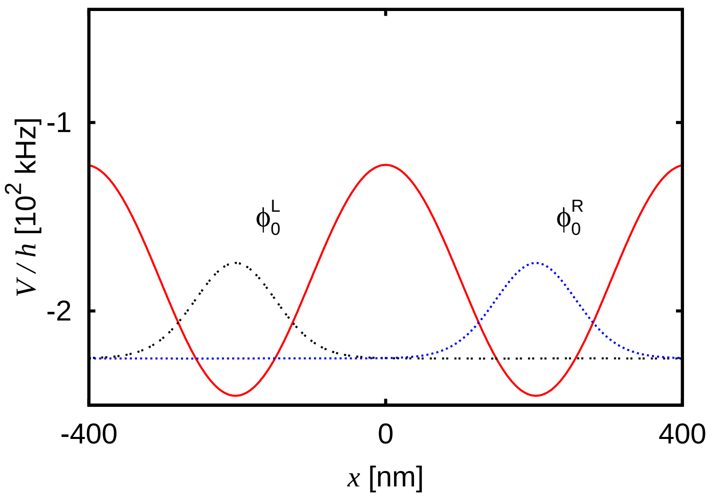
$$\hat{H} = \sum_{j=1}^{2} \frac{\hat{p}_j^2}{2M_{\mathrm{Rb}}} + V_{\mathrm{ext}}(\hat{x}_j, V_0(t), \beta(t), \theta(t)) + g\,\delta(\hat{x}_1 - \hat{x}_2)$$

with the optical lattice

$V_0 = 122.5,\ \beta = 0$ $\qquad$ $V_0 = 87.5,\ \beta = \pi/2 = -\theta$



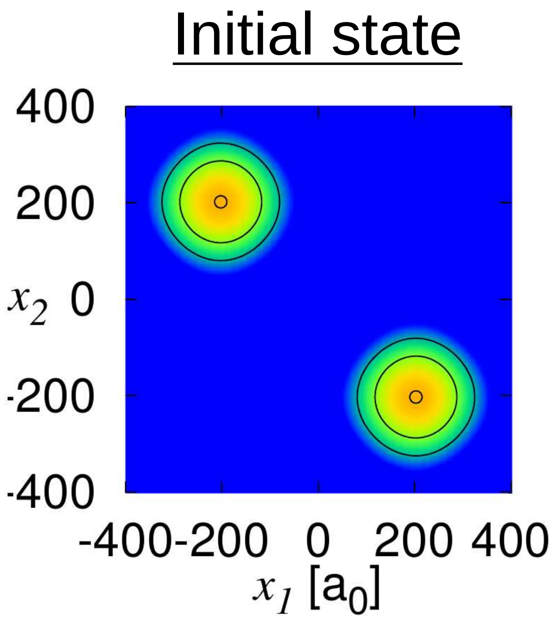
Cold atom transport: creation of an entangled state

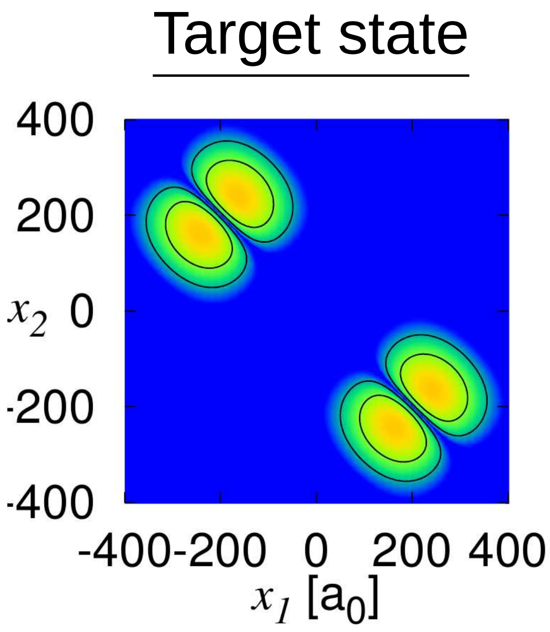


$$\psi^{\mathrm{I}}(x_1, x_2) \;=\; \mathcal{S}\,(\,\phi_0^{\mathrm{L}}(x_1)\,\phi_0^{\mathrm{R}}(x_2)\,)/\sqrt{2}$$

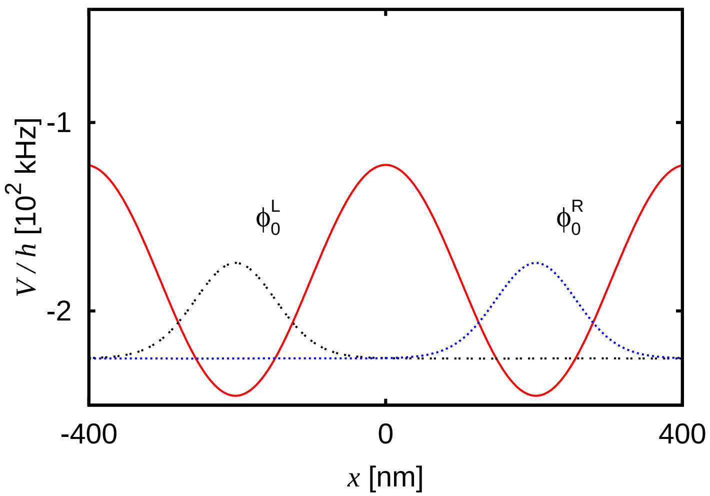
$$\psi^{\mathrm{T}}(x_1, x_2) \;=\; \mathcal{S}\,(\,\phi_0^{\mathrm{L}}(x_1)\,\phi_1^{\mathrm{R}}(x_2) + \phi_1^{\mathrm{L}}(x_1)\,\phi_0^{\mathrm{R}}(x_2)\,)/2$$

$$\phi_1^{\mathrm{L/R}} \;=\; C\,(x - x_0^{\mathrm{L/R}})\,\phi_0^{\mathrm{L/R}}$$





Cold atom transport: creation of an entangled state



$$\psi^{\mathrm{I}}(x_1, x_2) = \mathcal{S}\,(\,\phi_0^{\mathrm{L}}(x_1)\,\phi_0^{\mathrm{R}}(x_2)\,)/\sqrt{2}$$

$$\psi^{\mathrm{T}}(x_1, x_2) = \mathcal{S}\,(\,\phi_0^{\mathrm{L}}(x_1)\,\phi_1^{\mathrm{R}}(x_2) + \phi_1^{\mathrm{L}}(x_1)\,\phi_0^{\mathrm{R}}(x_2)\,)/2$$

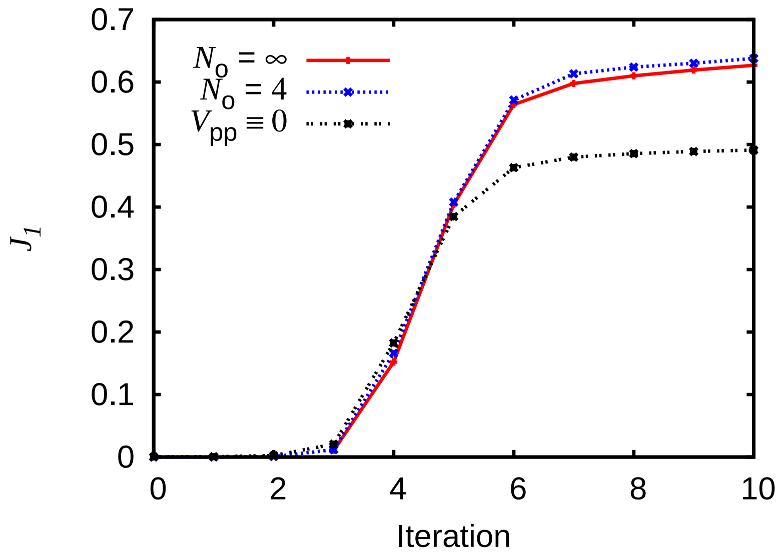
$$\phi_1^{\mathrm{L/R}} = C\,(x - x_0^{\mathrm{L/R}})\,\phi_0^{\mathrm{L/R}}$$

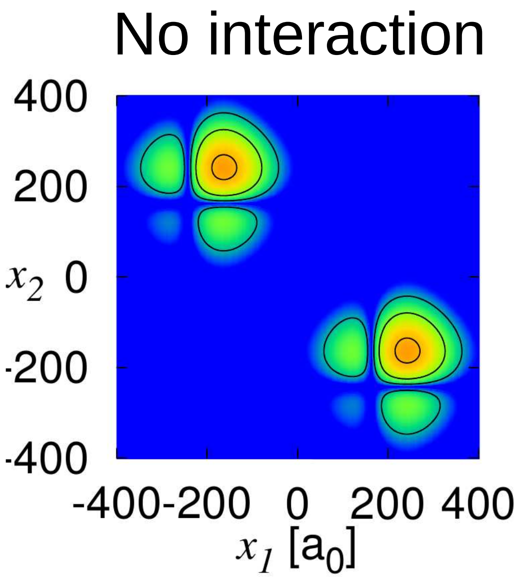
This process is a severe test for the MCTDHF method because the particle-particle interaction is crucial for the process, but can only be controlled indirectly.

$$i\,\hbar\,\dot{c}_J(t) = \sum_L \langle \Phi_J | \hat{V}_{pp} | \Phi_L \rangle\, c_L(t)$$

$$i\,\hbar\,\partial_t \vec{\varphi}(t) = [\hat{h} + (1 - \hat{P})\,\rho^{-1}\langle \hat{V}_{pp} \rangle]\vec{\varphi}(t)$$

Cold atom transport: optimization results

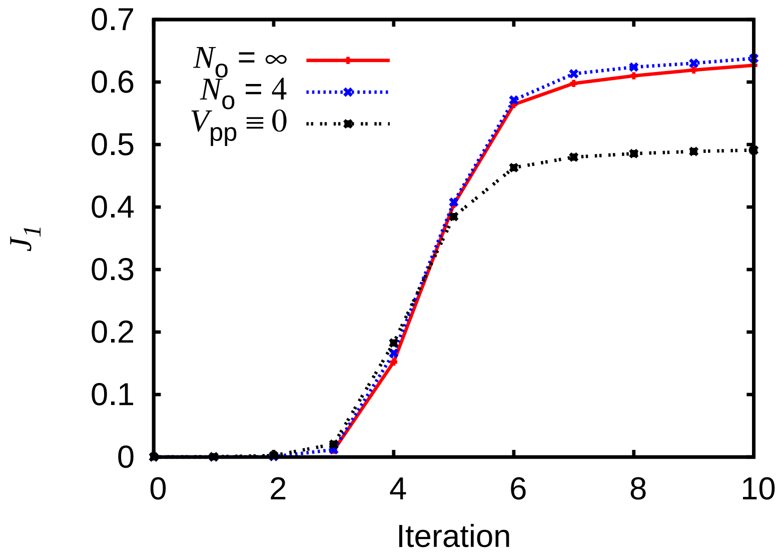


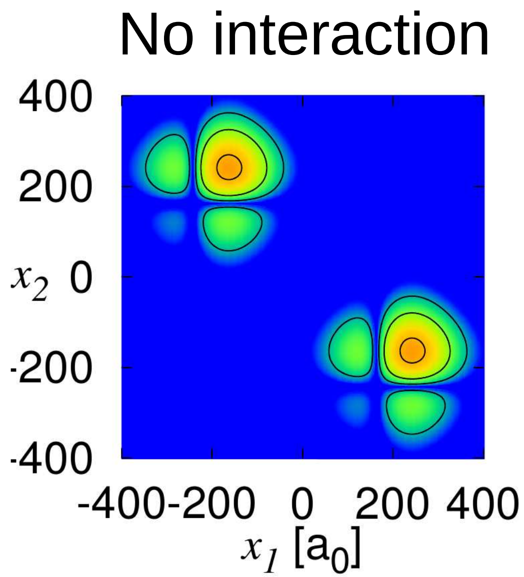


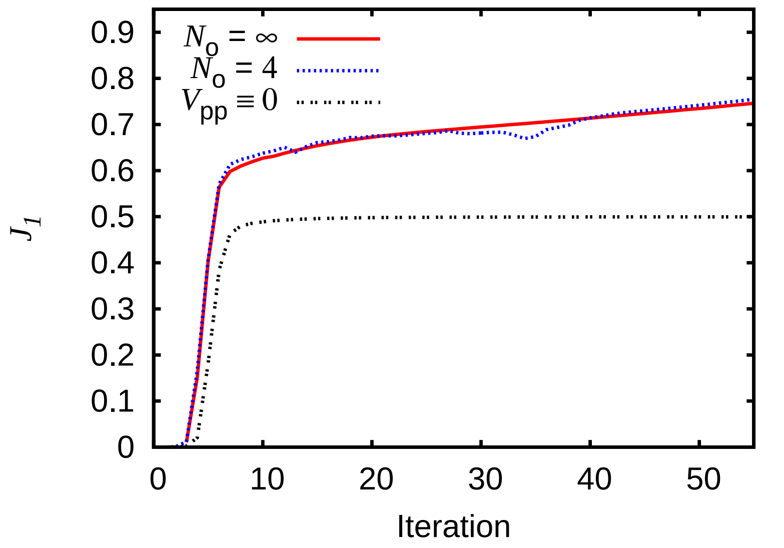
$$\psi(x_1,x_2) = \mathcal{S}\left(\varphi^{\rm L}(x_1)\,\varphi^{\rm R}(x_2)\right)/2$$

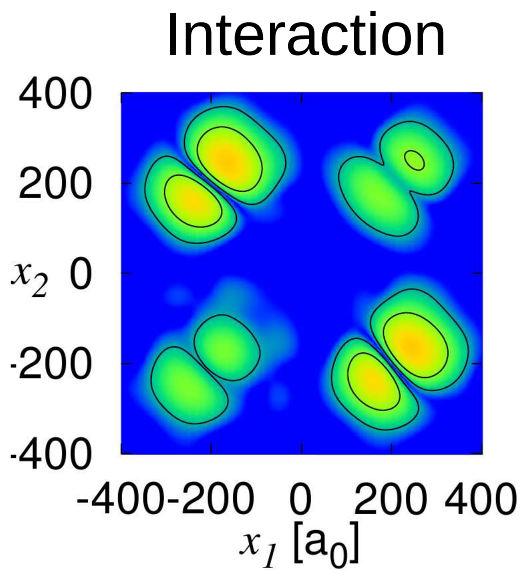
$$\varphi^{{\rm L}/R}(x) = \phi_0^{{\rm L}/R}(x) + \phi_1^{{\rm L}/R}(x)$$

Cold atom transport: optimization results

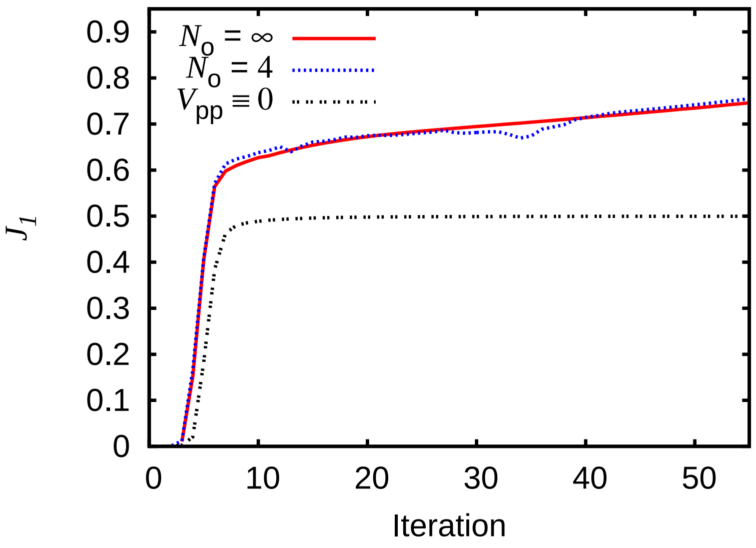


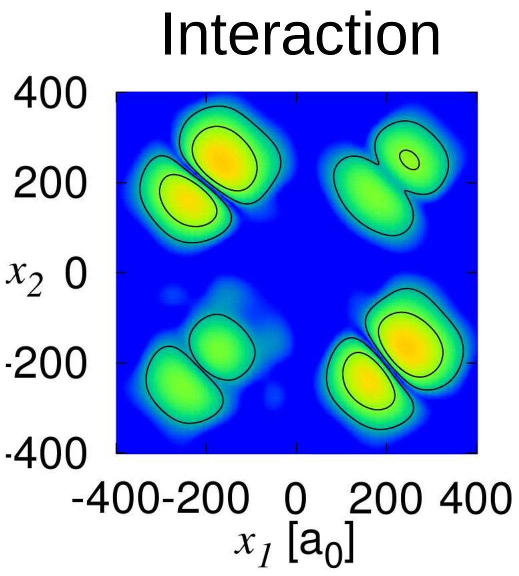






Cold atom transport: optimization results





- MCTDHF approach is approx. 8 × faster
- MCTDHF approach can be used also in 3-dim. problems

$\longrightarrow$ Linear OCT + MCTDHF is very promising!

Summary

- Quantum control plays a crucial role for both fundamental research and future technologies
- Optimal control theory is a natural candidate for quantum control due to its generality
- Combining OCT with the MCTDHF method requires in general nonlinear OCT
- The combination of linear OCT with the MCTDHF method as tool to solve the control equations offers an efficient approach for controlling interacting few-particle systems

Thank you for your attention!