Control of cold molecules:

Photoassociation & modified Krotov algorithm

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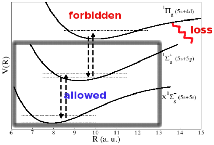
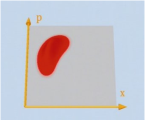
Institut für Theoretische Physik, Freie Universität Berlin, Germany

July 1, 2009

current interests: starting points





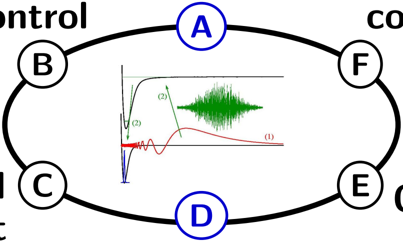
OCT algorithms

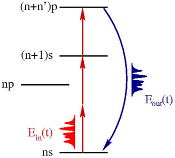
dissipation & control

$\hat{\mathbf{H}}_S + \hat{\mathbf{H}}_B + \hat{\mathbf{H}}_{SB}$

cooling

$\mathrm{Tr}[\hat{\rho}] \rightarrow 1$

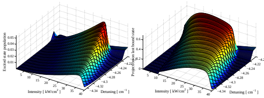


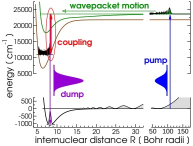


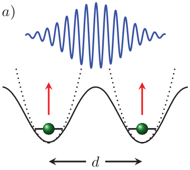
control of light

OCT 4 QI

opt. formation



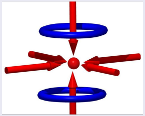




ultracold & ultrafast

temperatures 10^{-4} **to** 10^{-9} **K**

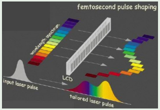
- extreme quantum limit
  $\rightsquigarrow$ coherent matter



- CW lasers, magnetic fields

timescales 10^{-15} **to** 10^{-12} **s**

- coherence of laser light
  $\rightsquigarrow$ *control*



- hot samples, 'black-box' algorithms

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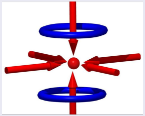

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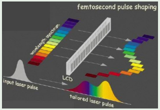
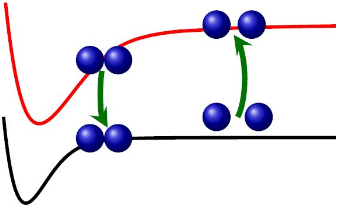


- hot samples, 'black-box' algorithms

ultracold chemistry
= photochemistry

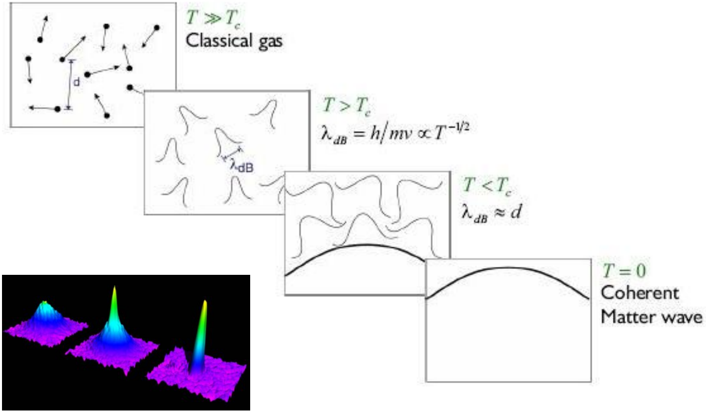
→ photoassociation

⟶ and beyond?



what means ultracold ?

ultracold: $T \leq 100\,\mu$K → a single quantum state
(or very few)



Bose-Einstein condensation

why ultracold molecules ?

- internal degrees of freedom, permant dipole moment
- interesting applications:
  - molecular Bose-Einstein condensate
  - quantum computer
    - cold $\triangleq$ little decoherence
  - precision measurements & tests of fundamental symmetries
    - cold $\triangleq$ long observation times

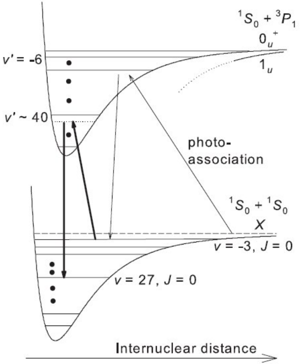
why ultracold molecules ?

- internal degrees of freedom, permant dipole moment
- interesting applications:

- example: precision measurements
  - ultracold Sr_2 molecules
  - time dependence of $\mu = m_e/m_p$

$$\frac{\Delta\mu}{\mu} = \frac{\Delta\nu}{\nu}$$

ν transition frequencies



Zelevinsky, Kotochigova, Ye,
PRL 100, 043201 (2008)

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    - cold $\triangleq$ little decoherence
  - precision measurements & test of fundamental symmetries
    - cold $\triangleq$ long observation times
  - ultracold collisions / reactions
    - cold $\triangleq$ tunneling & resonances

  ➔ **coherent control**

wanted: ultracold molecules

- ultracold : $T \leq 100\,\mu\text{K} \longrightarrow$ single (or very few) quantum state(s)

why ?

- molecular BEC
- ultracold chemistry
- quantum computing
- high precision measurements
- ...

helpers

to associate atoms to molecules

- magnetic fields
- laser fields

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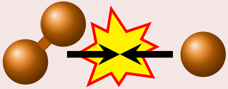
but really, you need $v = 0$

collisions!



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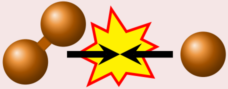
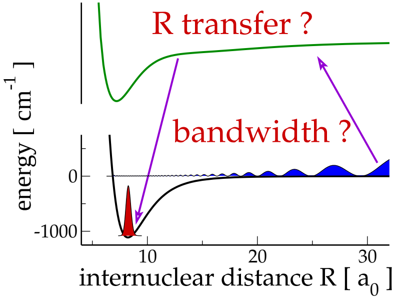
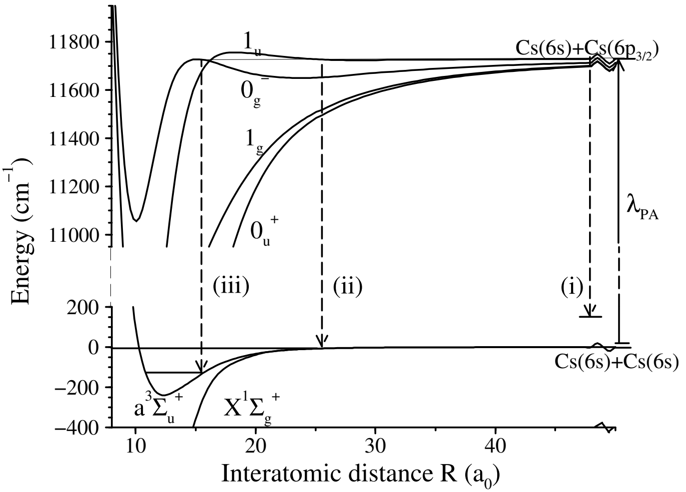




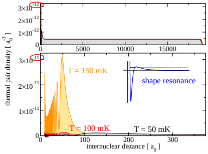
photoassociation

making molecules with laser light

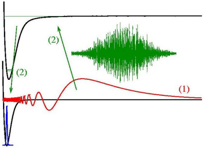
Masnou-Seeuws & Pillet, Adv. At. Mol. Opt. Phys. 47, 53 (2001)



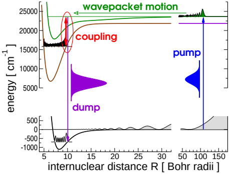
overview



1. photoassociation: an open control problem



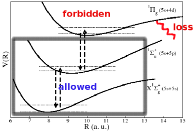
2. molecules in their ground state: we need OCT



3. 'shaping' the potentials: resonant coupling by an external field

4. outlook

5. modified Krotov algorithm: keeping the population in a subspace



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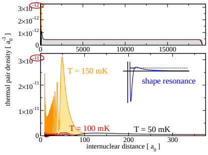

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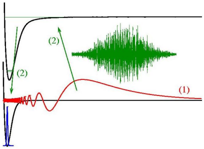
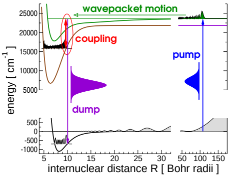
4. **outlook**





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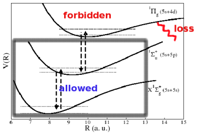
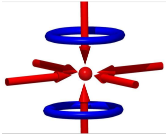
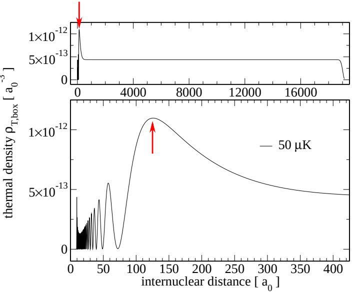
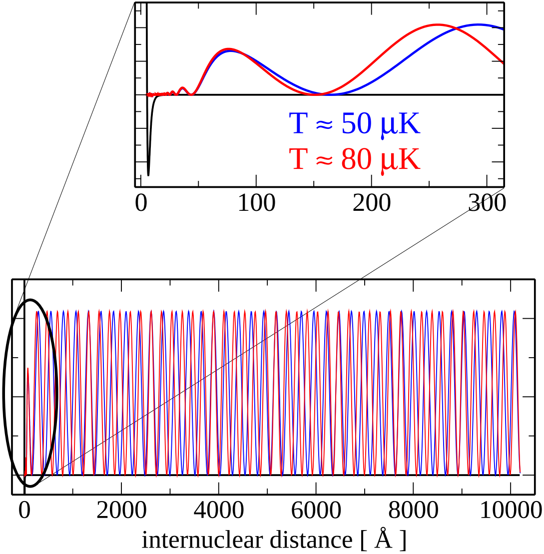


photoassociation is difficult

cold collisions

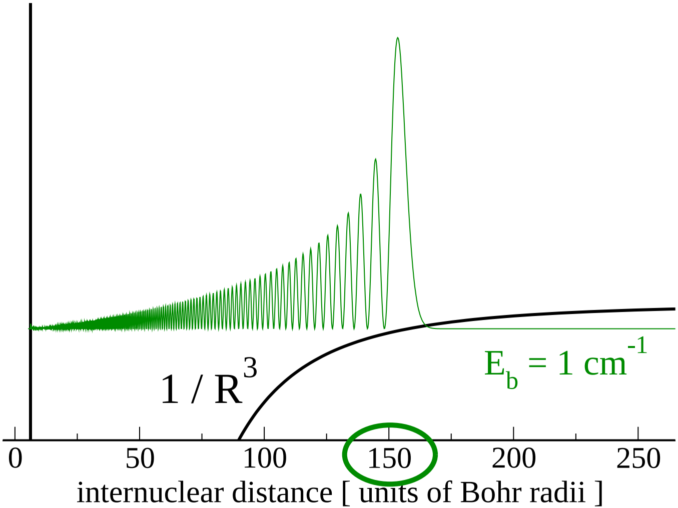






CPK, Kosloff, Luc-Koenig, Masnou-Seeuws, Crubellier, J Phys B 39, S1017 (2006)

why is making molecules possible ?



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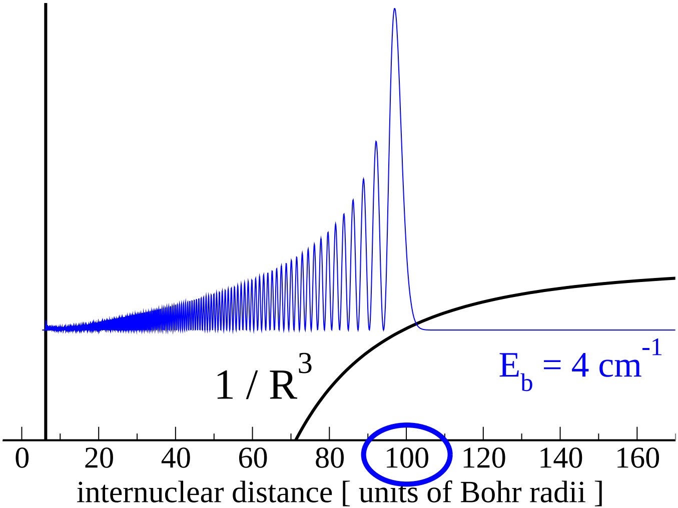
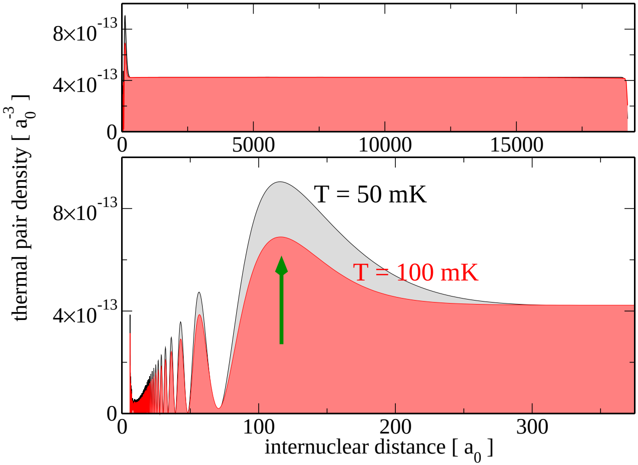
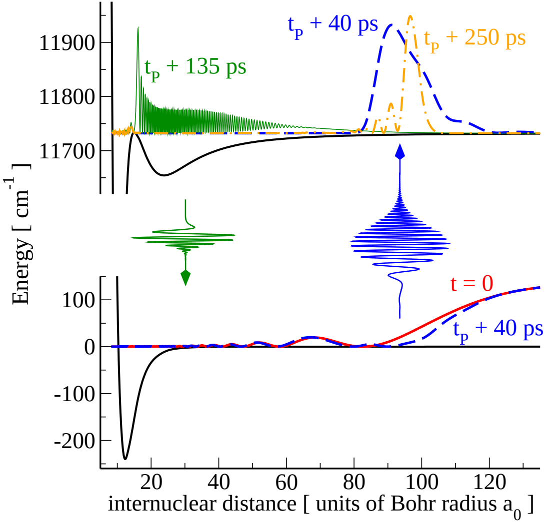


why is photoassociation difficult ?

- our starting point: thermal ensemble, large R
- our goal: $v = 0$ pure state, small R



coherent photoassociation



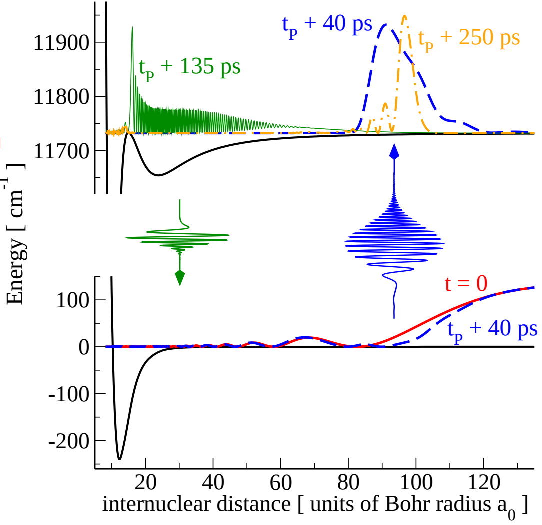
CPK, Luc-Koenig, Masnou-Seeuws, PRA 73, 033408 (2006)

coherent photoassociation

what is different from previous pump-probe schemes?

- initial state
- timescales

  ↷ bandwidths

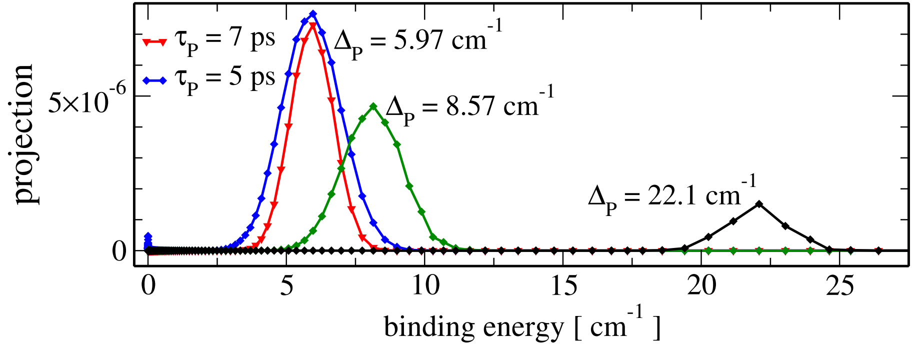


CPK, Luc-Koenig, Masnou-Seeuws, PRA 73, 033408 (2006)

choice of pulse parameters

role of laser detuning and spectral bandwidth

projection of $\Psi_{exc}(R, t_{final})$ onto vibrational levels of $\hat{\mathbf{H}}_e(R)$, $^{87}Rb_2$



CPK, Kosloff, Masnou-Seeuws, PRA 73, 043409 (2006)

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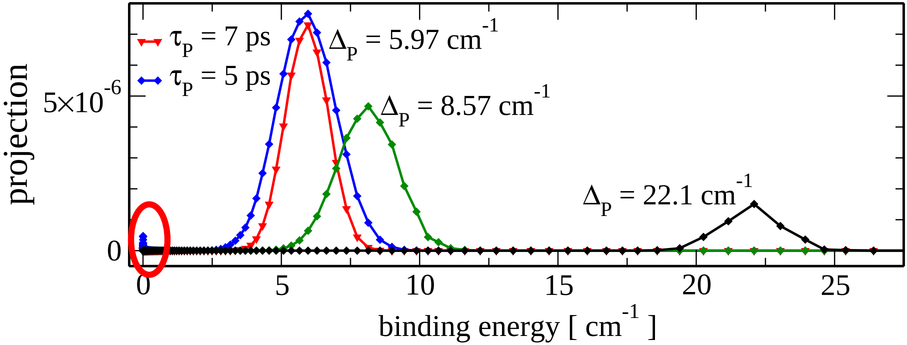

→ **narrow band pulses**

CPK, Kosloff, Masnou-Seeuws, PRA 73, 043409 (2006)

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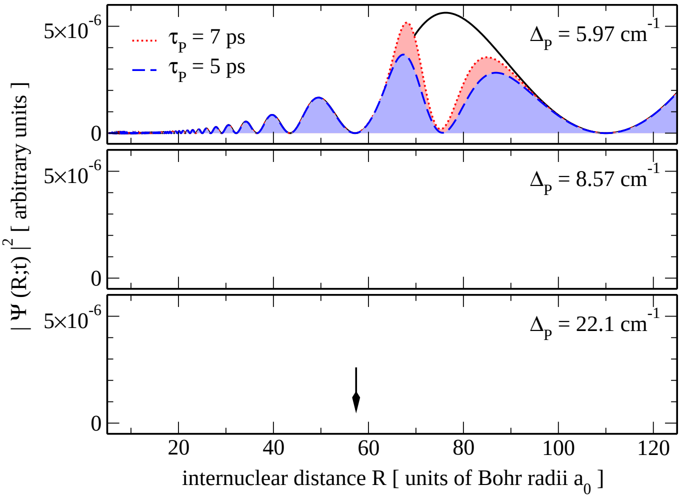

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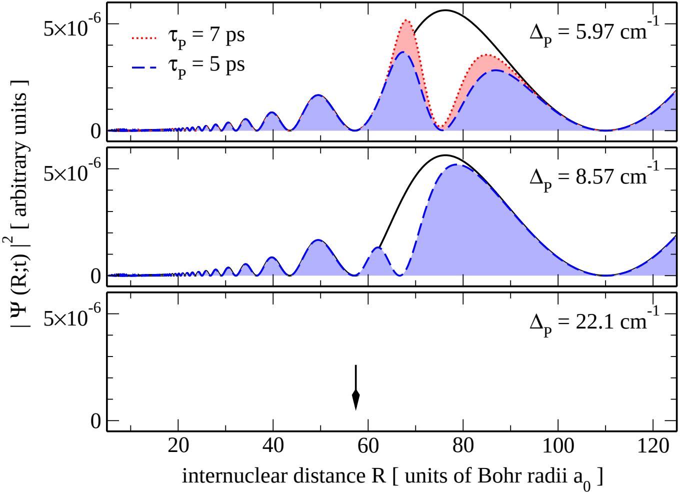

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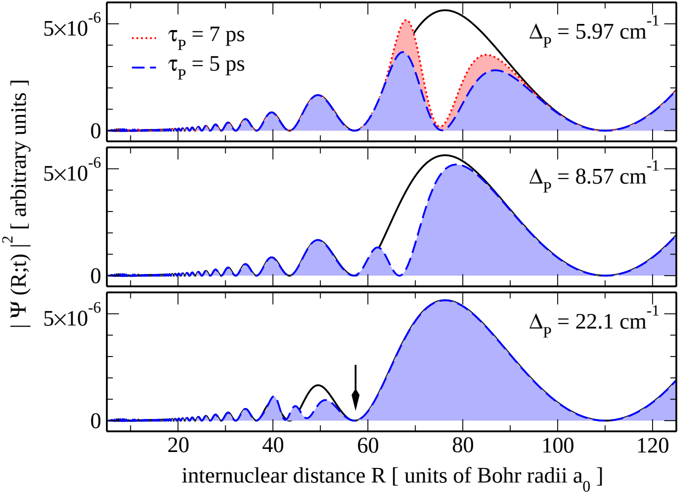
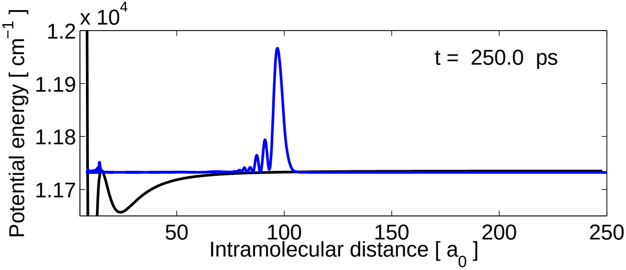
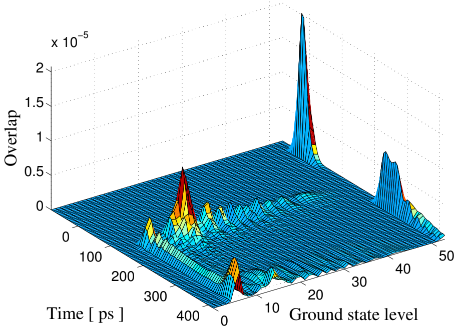


photoassociation dynamics



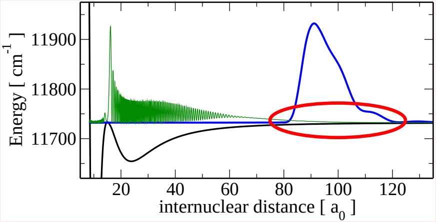


time-dependent 'Franck-Condon factors' ➔ pump-dump-delay

pump-dump efficiency

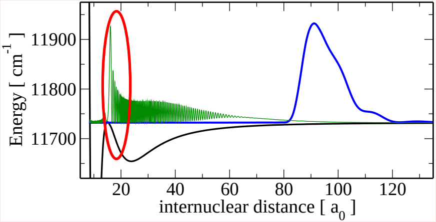
the best example: $Cs_2\ 0_g^-(P_{3/2})$

1. excitation (pump)



- at long range : $1/R^3$ (small Δ_P)

2. stabilization (dump)



- at short range : $-1/R^3$ ($\Delta_D < 0$)

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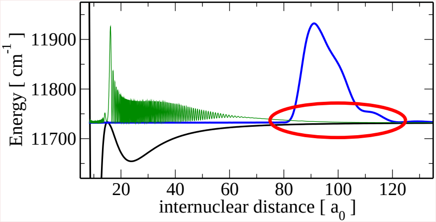

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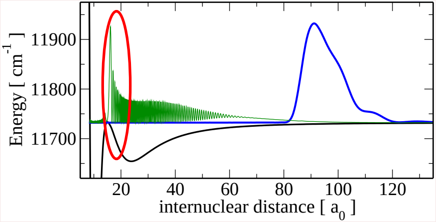
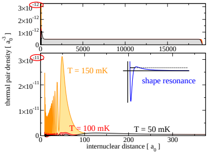


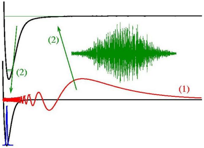
- at short range : $-1/R^3$ ($\Delta_D < 0$)

and at most you get $E_{bind} \approx 100\,\mathbf{cm}^{-1}$

overview



1. photoassociation: an open control problem



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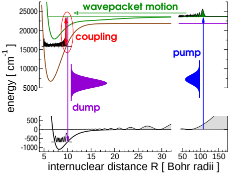

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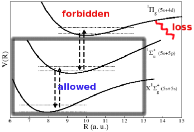
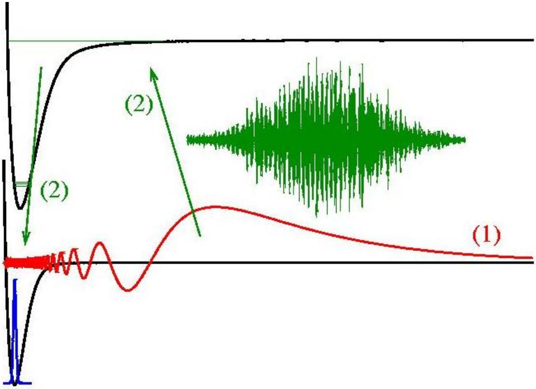


how to get ultracold stable molecules ?

1. formation of *loosely* bound molecules with Feshbach resonance or photoassociation
2. transfer of population to $v = 0$ using shaped laser pulses (vibrational stabilization)



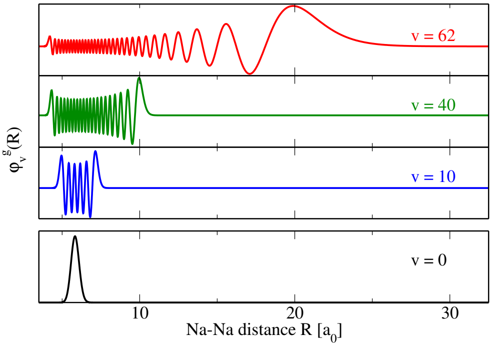
CPK, Palao, Kosloff, Masnou-Seeuws, PRA 70, 013402 (2004)

why is this a difficult control task ?

constraints :

- in a time short compared to collisional decay ($\sim$ ms)
- immediate transfer to lowest levels to avoid energy pooling
- faster than spontaneous emission ($\sim$ 10 ns)

qualitatively different wave functions:



OCT: Solving the inverse problem

Statement of the problem

- Given the initial state
  the molecule in a highly excited level v of the electronic ground state
- and the objective
  the molecule in the vibrational level $v = 0$ of the electronic ground state
- and the equations describing the evolution of the system
  the Schrödinger equation for the molecule coupled to a laser field
- Find the time-dependent field which leads to the objective !

S. E. Sklarz and D. J. Tannor, Phys. Rev. A 66, 053619 (2002)
J. P. Palao and R. Kosloff, Phys. Rev. A 68, 062308 (2003)

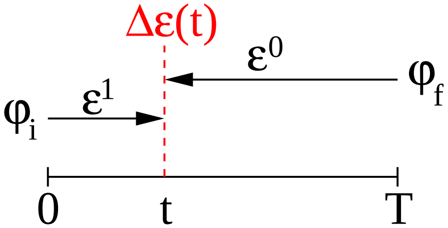
OCT: Krotov method

improve the field by

$$\Delta\epsilon(t) =$$

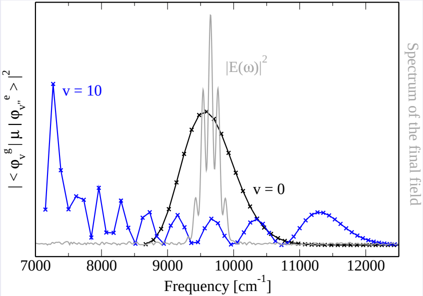
$$\frac{S(t)}{\lambda_0}\,\mathfrak{Im}\left[\underbrace{\langle\varphi_i|\,\hat{\mathbf{U}}^{+}(T,0;\varepsilon^0)}_{\substack{\text{forward}\\ \text{propagation}\\ (1)}}\,|\varphi_f\rangle\,\underbrace{\langle\varphi_f|\,\hat{\mathbf{U}}^{+}(t,T;\varepsilon^0)}_{\substack{\text{backward}\\ \text{propagation}\\ (2)}}\,\hat{\boldsymbol{\mu}}\,\underbrace{\hat{\mathbf{U}}(t,0;\varepsilon^1)\,|\varphi_i\rangle}_{\substack{\text{forward}\\ \text{propagation}\\ (3)}}\right]$$

Interference between past and future events



there is always a solution but ...

easy task



Franck-Condon overlap for intial and target state in same spectral region ⟶ comp. weak pulse with one central frequency sufficient

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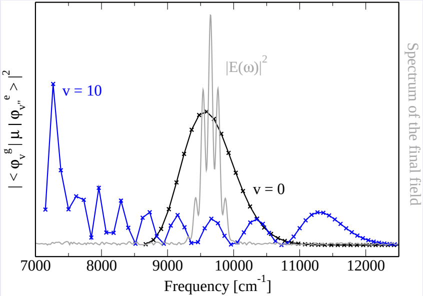
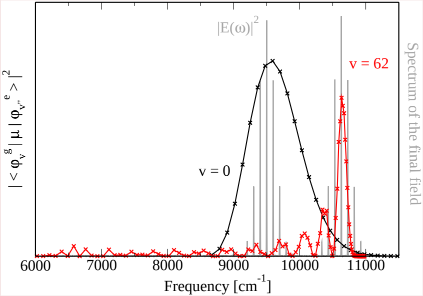


Franck-Condon overlap for intial and target state in same spectral region ⟶ comp. weak pulse with one central frequency sufficient

difficult task

Franck-Condon overlap for intial and target state in different spectral regions ⟶ splice together two comp. strong pulses with different ω_L

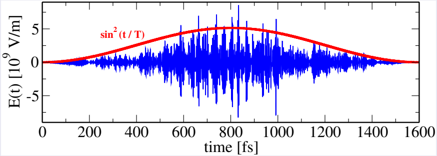


... there is a price

intensity

$$\mathcal{E}_P = \epsilon_0\, c\, A \int_0^T \|E(t)\|^2\, \mathrm{d}t$$

assuming $A = \pi R^2$ with $R = 300\mu$m



minimum intensities necessary for convergence

v_{initial}	10	40	62
$\mathcal{E}_{\text{pulse}}$	60 μJ	1.5 mJ	3.9 mJ

CPK, Palao, Kosloff, Masnou-Seeuws, PRA 70, 013402 (2004)

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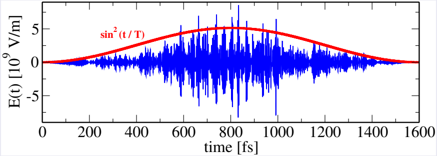


minimum intensities necessary for convergence

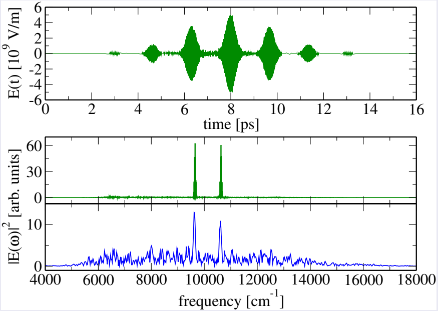
v_{initial}	10	40	62
$\mathcal{E}_{\text{pulse}}$	60 μJ	1.5 mJ	3.9 mJ

time

$T^* = \frac{2\pi h}{\Delta E}$ → $T \geq 2T^*$

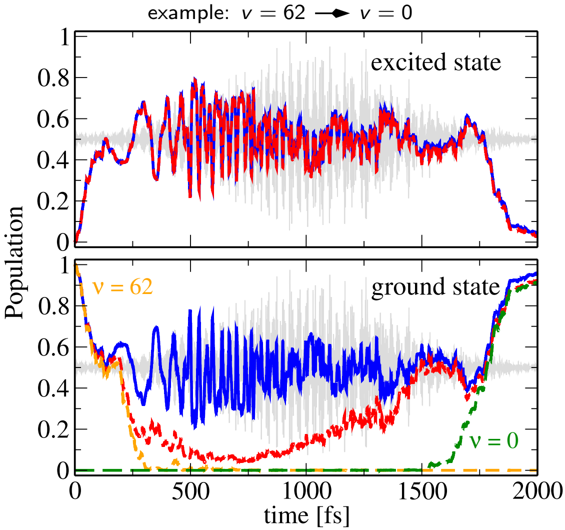
↷ large range of energies

≙ large range of time scales



CPK, Palao, Kosloff, Masnou-Seeuws, PRA 70, 013402 (2004)

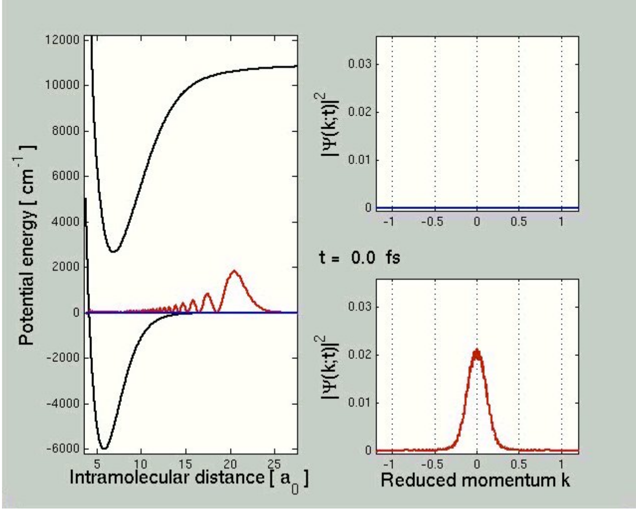
dynamics induced by the optimal field



population of ground and excited state

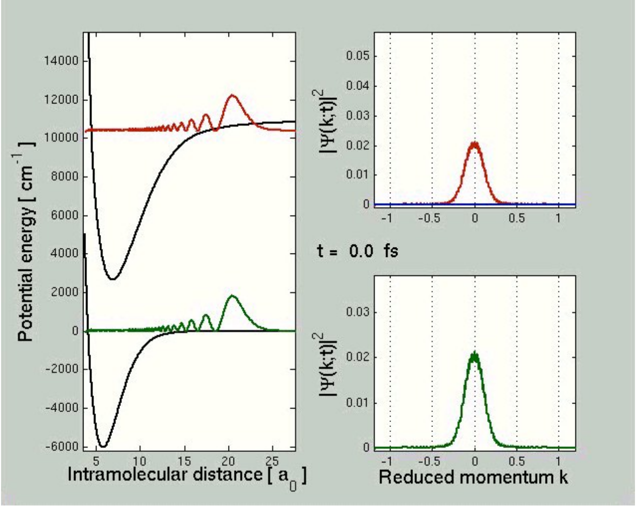
beating the natural timescale

long pulse

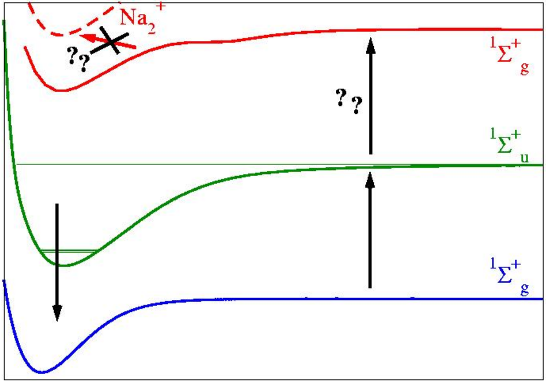


beating the natural timescale

shortened pulse & natural exc. state dynamics



a problem with very strong fields

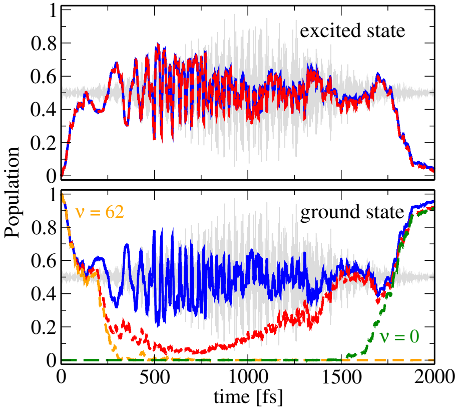


→ **tell algorithm to avoid door-way states**

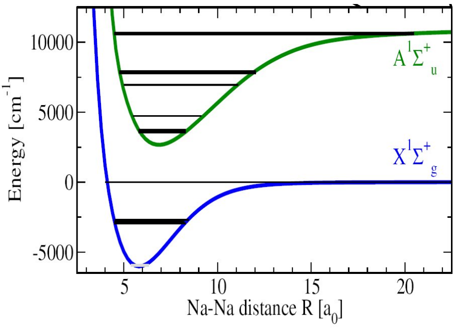
last part of talk

route to $v = 0$ (generic case) : OCT

strong fields and / or many Raman transitions



example: Na_2, $v_{initial} = 62$ ($v_{last} = 65$)



→ **required pulse energy ~ 4 mJ**

CPK, Palao, Kosloff, Masnou-Seeuws, PRA 70, 013402 (2004)

see also: Pe'er, Shapiro, Stowe, Shapiro, Ye, PRL 98, 113004 (2006)

route to $v = 0$ (generic case) : OCT

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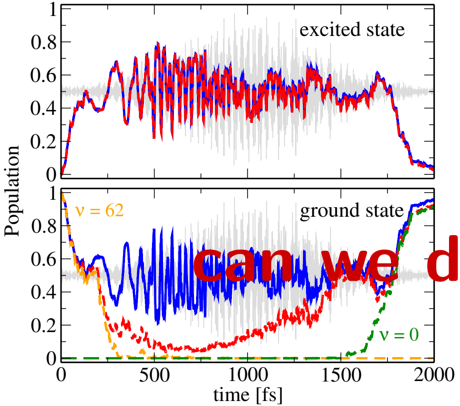
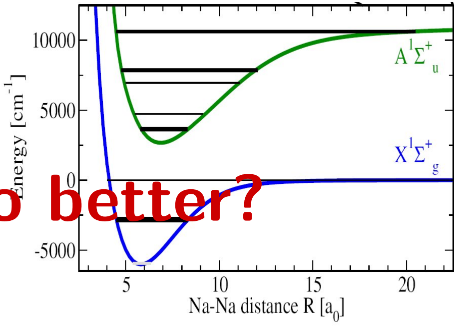


can we do better?

example: Na_2, $v_{initial} = 62$ ($v_{last} = 65$)

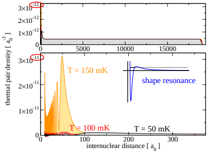


⟶ **required pulse energy ∼ 4 mJ**

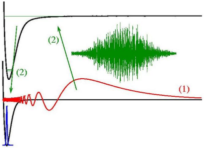
CPK, Palao, Kosloff, Masnou-Seeuws, PRA 70, 013402 (2004)

see also: Pe'er, Shapiro, Stowe, Shapiro, Ye, PRL 98, 113004 (2006)

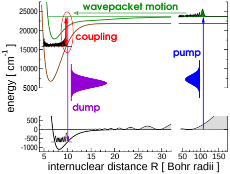
overview



1. **photoassociation: an open control problem**



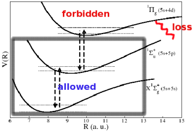
2. **molecules in their ground state: we need OCT**



3. **'shaping' the potentials: resonant coupling by an external field**

4. **outlook**

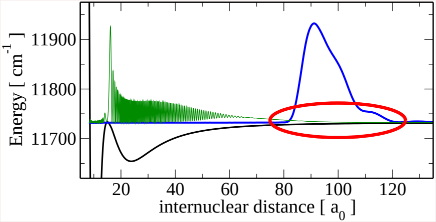
5. **modified Krotov algorithm: keeping the population in a subspace**



pump-dump efficiency

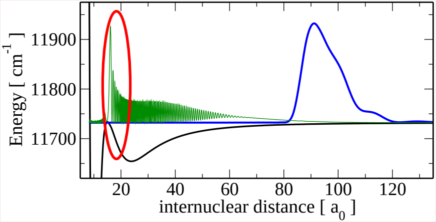
the best example: $Cs_2\ 0_g^-(P_{3/2})$

1. excitation (pump)



- at long range : $1/R^3$ (small Δ_P)

2. stabilization (dump)



- at short range : $-1/R^3$ ($\Delta_D < 0$)

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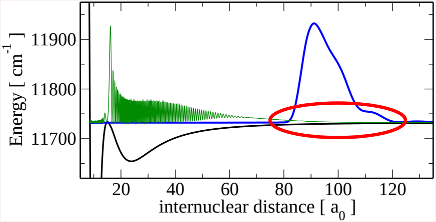

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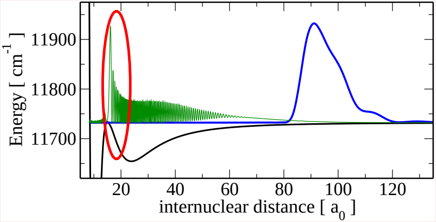
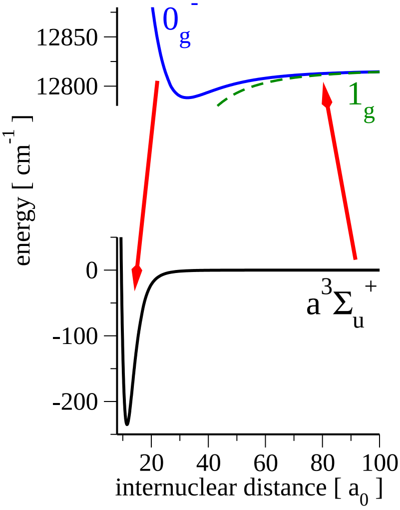


- at short range : $-1/R^3$ ($\Delta_D < 0$)

and at most you get $E_{bind} \approx 100\,\mathbf{cm}^{-1}$

possible R-transfer mechanisms

softly repulsive wall



Dion, Drag, Dulieu, Laburthe Tolra, Masnou-Seeuws, Pillet, PRL 86, 2253 (2001)

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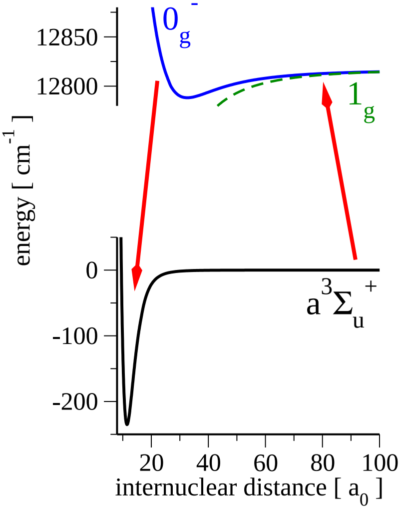
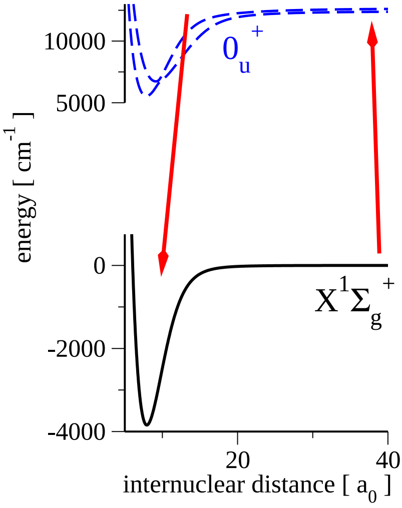


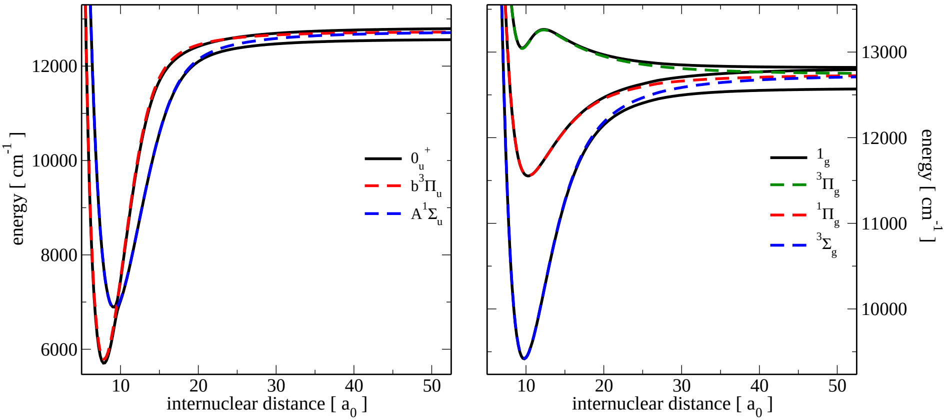
resonant coupling



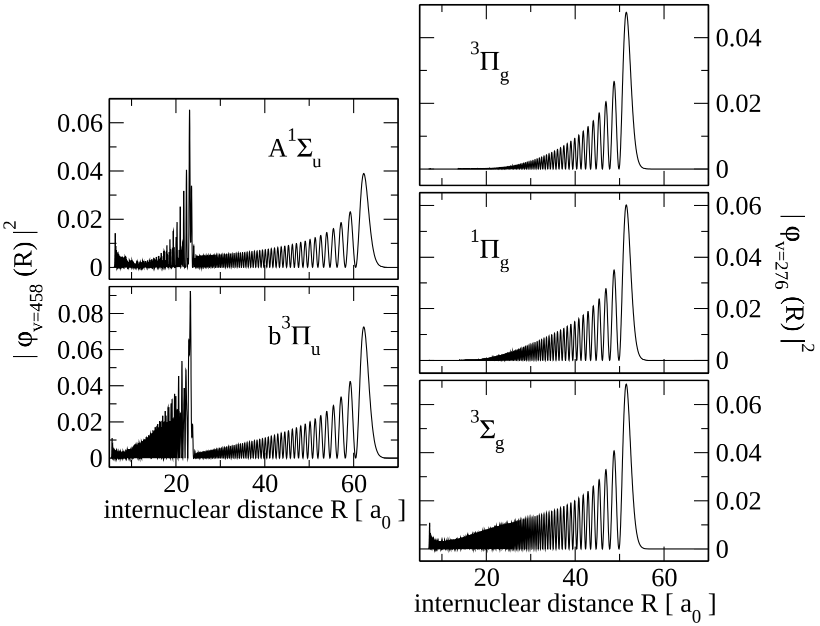
Dion, Drag, Dulieu, Laburthe Tolra, Masnou-Seeuws, Pillet, PRL 86, 2253 (2001)

spin-orbit coupling

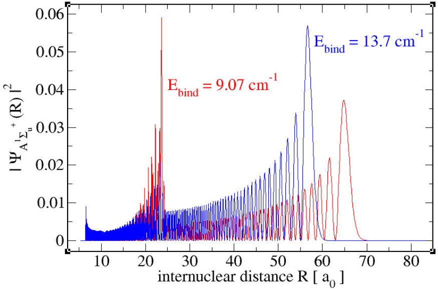
resonant coupling non-resonant coupling

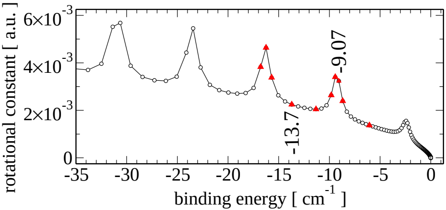


resonant vs non-resonant coupling



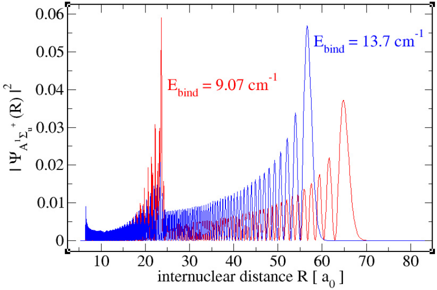
resonant spin-orbit coupling

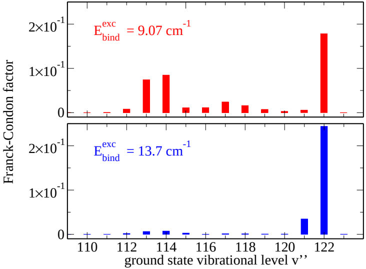


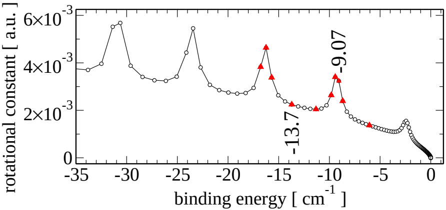


Pechkis, Wang, Eyler, Gould, Stwalley, CPK, PRA 76, 022504 (2007)

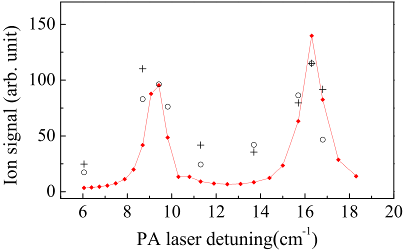
resonant spin-orbit coupling







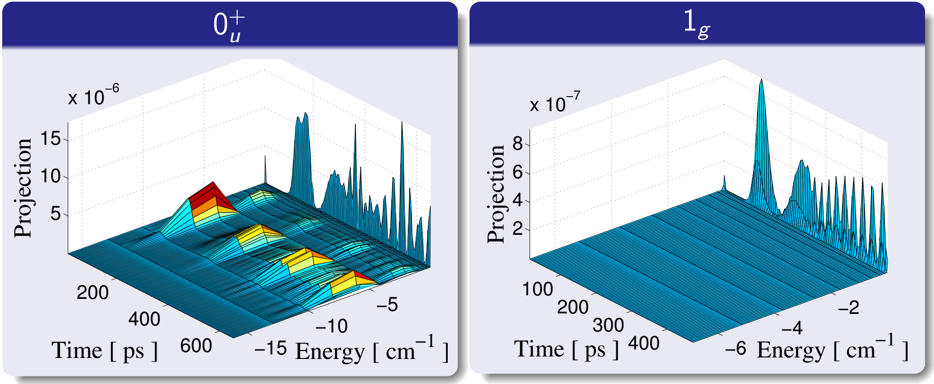
comparison theory-experiment



Pechkis, Wang, Eyler, Gould, Stwalley, CPK, PRA 76, 022504 (2007)

stabilization to the ground state

time-dependent FC factors

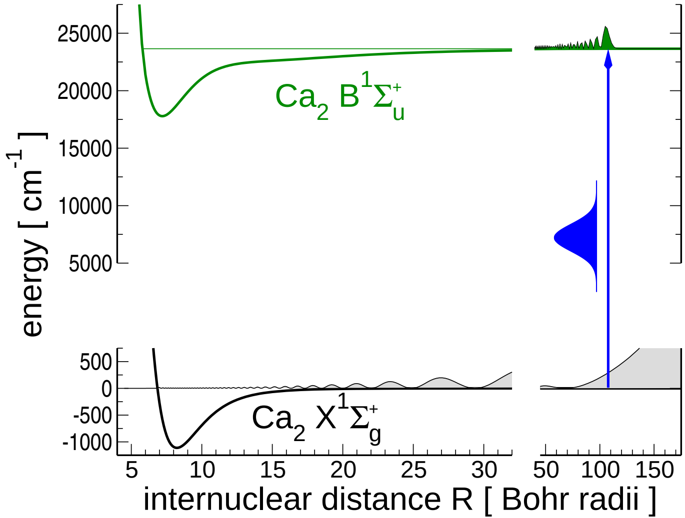


$$\Delta_P = 4.1\,\mathrm{cm}^{-1}$$

CPK, Kosloff, Masnou-Seeuws, PRA 73, 043409 (2006)

field-induced resonant coupling

CPK & R. Moszyński, PRA 78, 043417 (2008)



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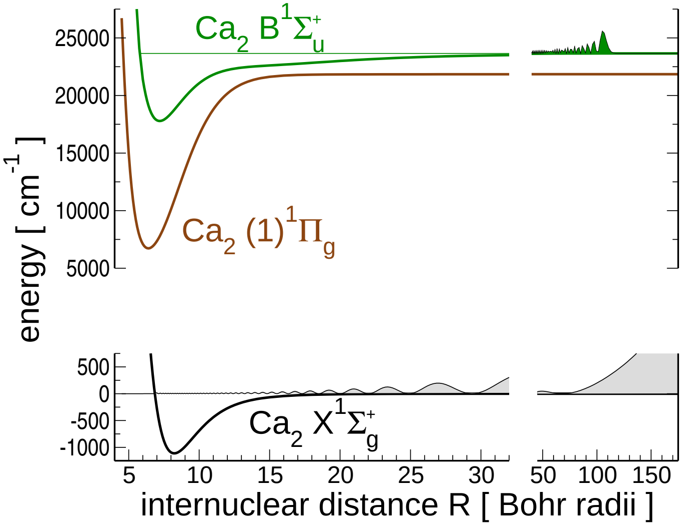

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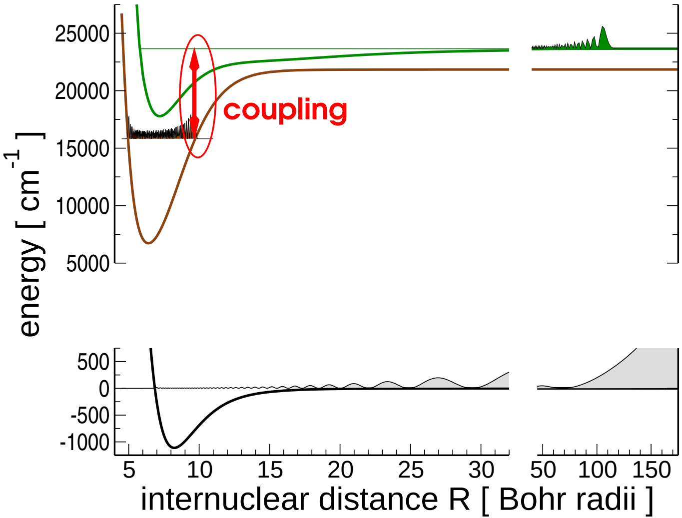

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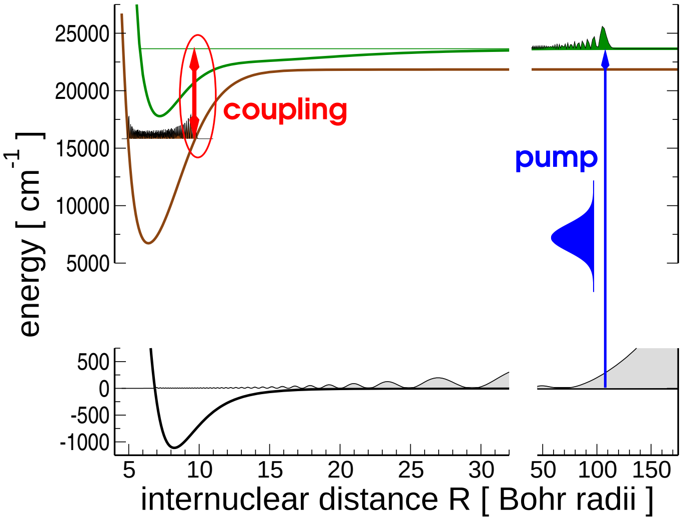

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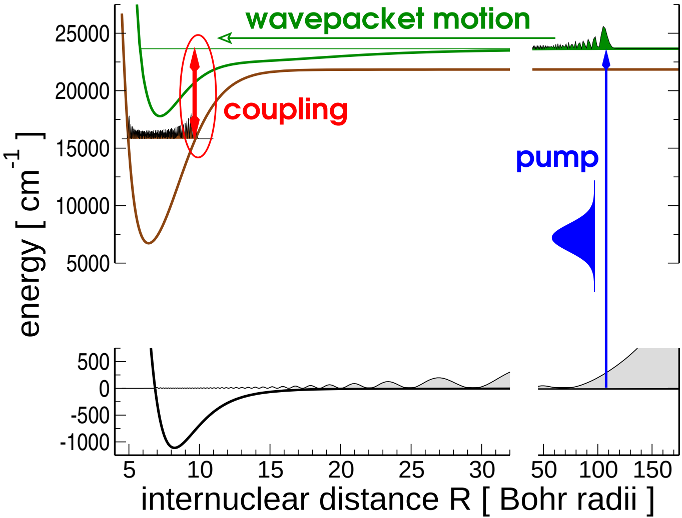

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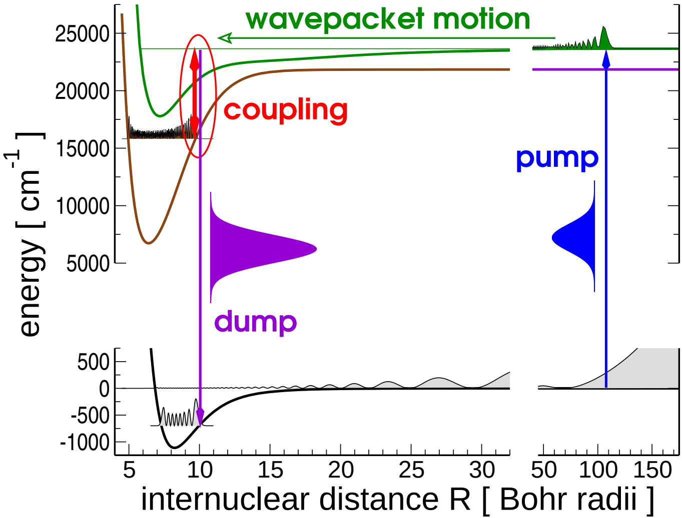

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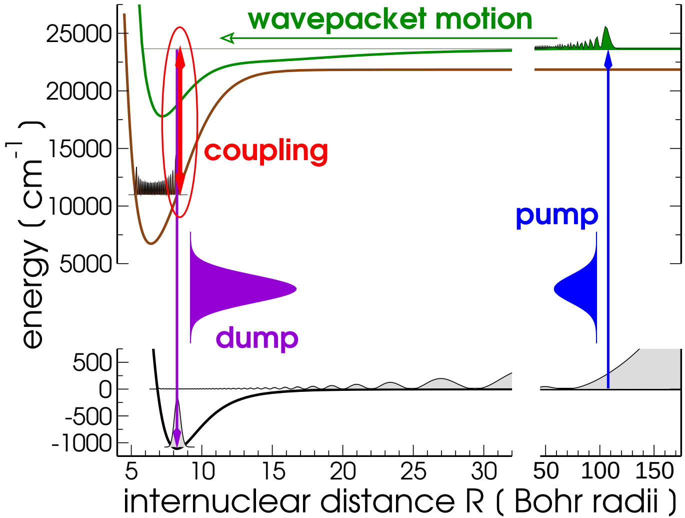
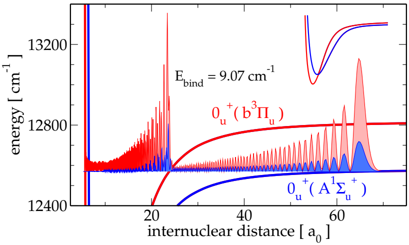
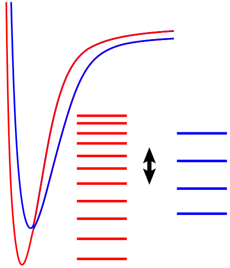


when is the coupling resonant?





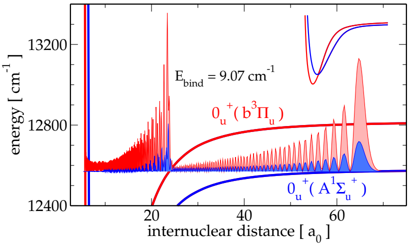
$$\hat{\mathbf{H}} = \begin{pmatrix} \hat{\mathbf{T}} + V_1(\hat{\mathbf{R}}) & \hat{\mathbf{W}} \\ \hat{\mathbf{W}} & \hat{\mathbf{T}} + V_2(\hat{\mathbf{R}}) \end{pmatrix}$$

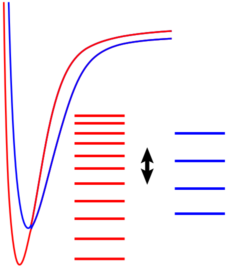
spin-orbit coupling:

$$\hat{\mathbf{W}} = V_{SO}(\hat{\mathbf{R}}) \longrightarrow 237.6\,\mathrm{cm}^{-1}$$

for Rb

when is the coupling resonant?





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for Rb

field-induced coupling:

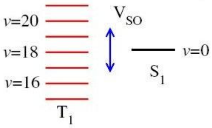
$$\hat{\mathbf{W}} = \hbar\Omega = \frac{1}{2}\mu(\hat{\mathbf{R}}) \cdot E(t)$$

if $\mu(\hat{\mathbf{R}}) \sim 1\,\mathrm{at.u.}$

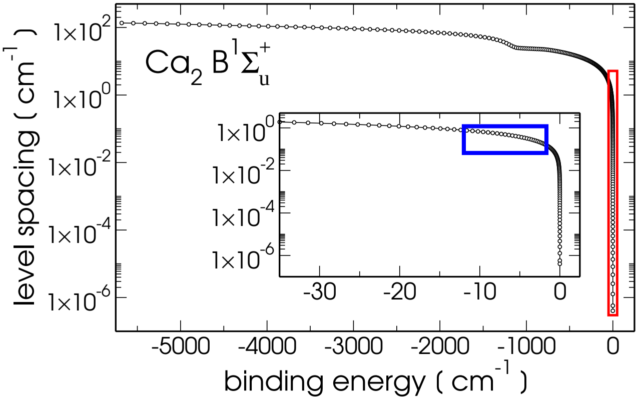
then $E_0 \sim 1.0 \times 10^7$ V/cm

$I \sim 1.4 \times 10^{11}\,\mathrm{W/cm}^2$

res. coupling & photoassociation



coupling needs to be comparable to level spacings



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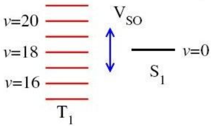

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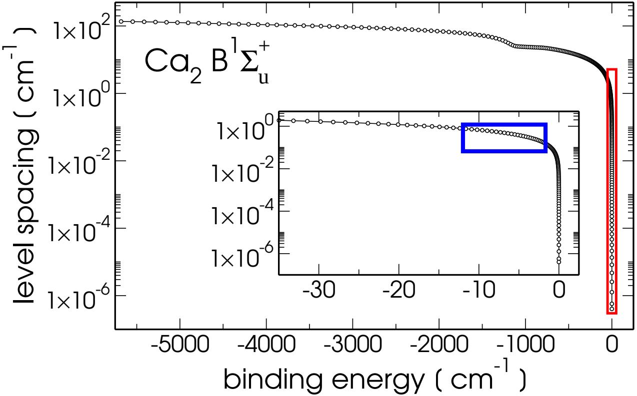
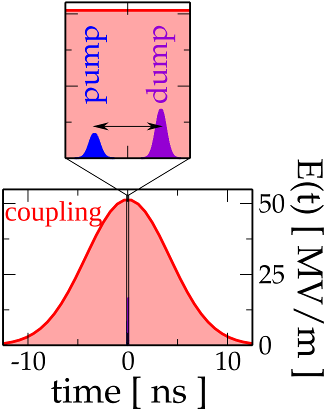


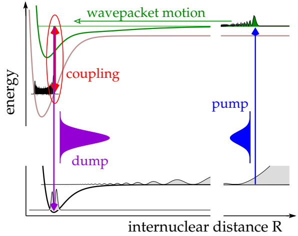
level spacings drop to $\sim 1\,\mathrm{cm}^{-1}$ in range of PA detunings

choice of coupling laser

$I = 3.5 \times 10^8\ \mathrm{W/cm^2}\ - 3.2 \times 10^9\ \mathrm{W/cm^2}$

$\omega_2 = 11351\,\mathrm{cm}^{-1}$ (881 nm) $\curvearrowright$ target $X^1\Sigma_g^+$ level: $v'' = 1$





10 ns pulse is constant on timescale of 100 ps

CPK & Moszyński, PRA 78, 043417 (2008)

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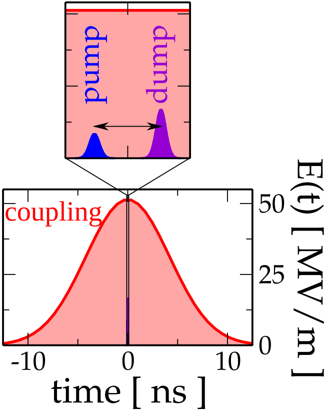
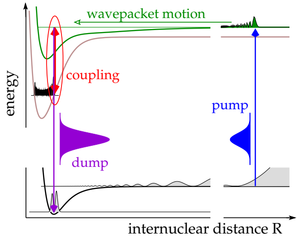


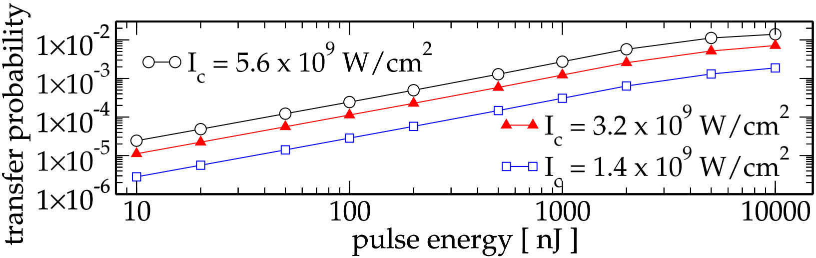


10 ns pulse is constant on timescale of 100 ps

→ **feasible & robust**

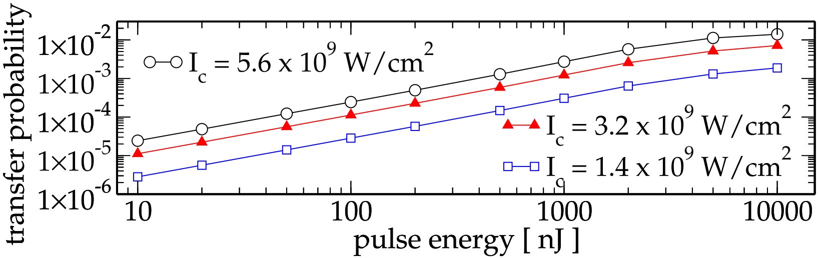
CPK & Moszyński, PRA 78, 043417 (2008)

how many molecules?



typical MOT conditions: $N_{mol} = 12.5$, 10 kHz rep.rate: 1 mol/ms

how many molecules?



typical MOT conditions: $N_{mol} = 12.5$, 10 kHz rep.rate: 1 mol/ms

accumulate molecules over many pump-dump cycles

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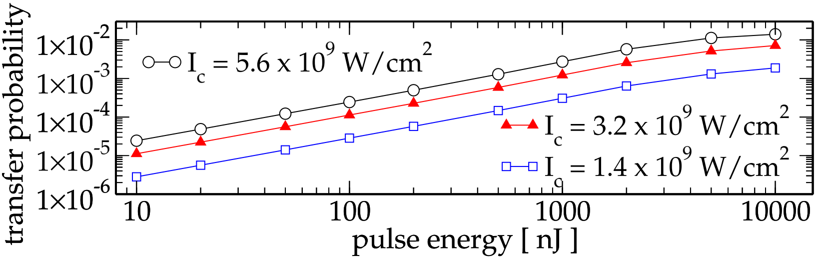
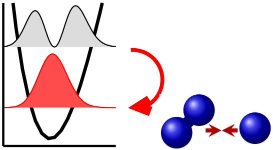


typical MOT conditions: $N_{mol} = 12.5$, 10 kHz rep.rate: 1 mol/ms

accumulate molecules over many pump-dump cycles

employ dissipation to achieve unidirectionality

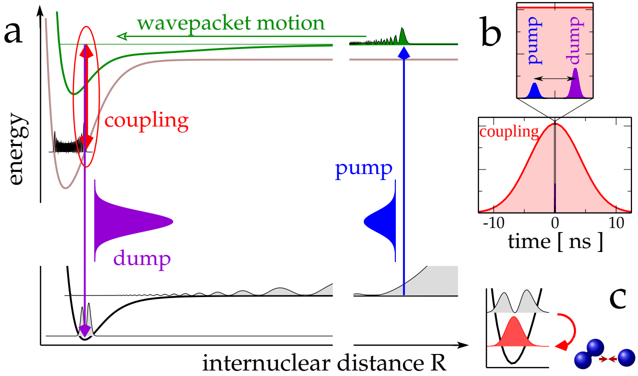
collisional decay to $v = 0$ within 1 ms if $\rho \sim 10^{-13}\,\mathrm{cm}^{-3}$



can be improved: flux enhancement & speed up of decay

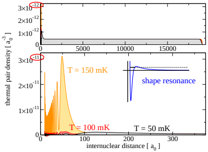
field-induced resonant coupling

'shaping' the potentials by hand



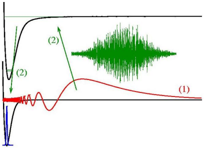
- ➔ **qualitative & substantial change of dynamics**
- ➔ **implementing resonance phenomenon of cold molecules via coherent control**

overview

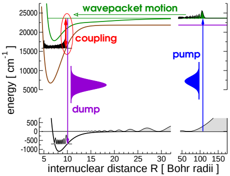


1. **photoassociation: an open control problem**

2. **molecules in their ground state: we need OCT**

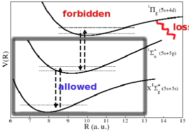


3. **'shaping' the potentials: resonant coupling by an external field**

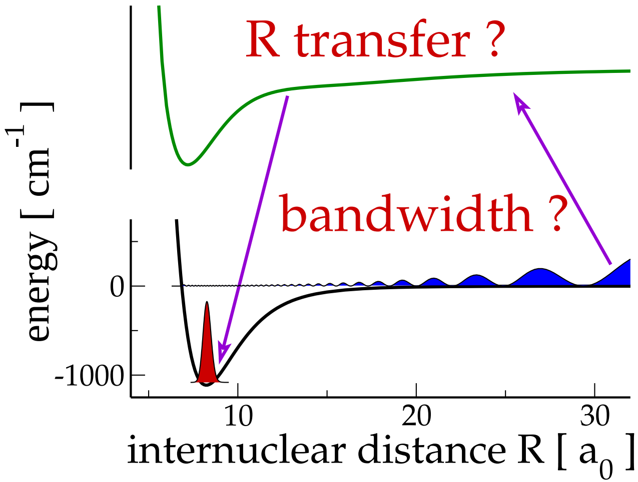


4. **outlook: photoassociation – an open control problem**

5. **modified Krotov algorithm: keeping the population in a subspace**

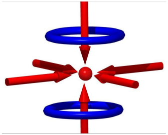


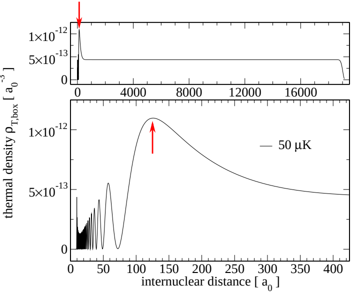
open questions in photoassociation

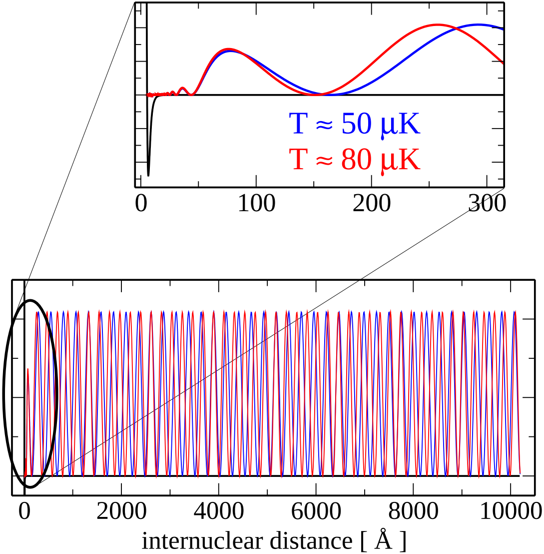


why is photoassociation difficult ?

cold collisions





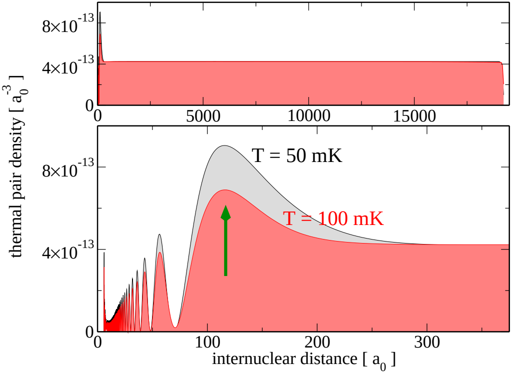


CPK, Kosloff, Luc-Koenig, Masnou-Seeuws, Crubellier, J Phys B 39, S1017 (2006)

why is photoassociation difficult

coherent control out of a continuum!

- continuum is almost structureless
- only atom pairs at short range are correlated



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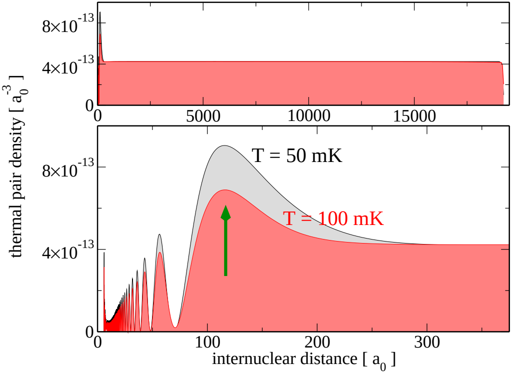
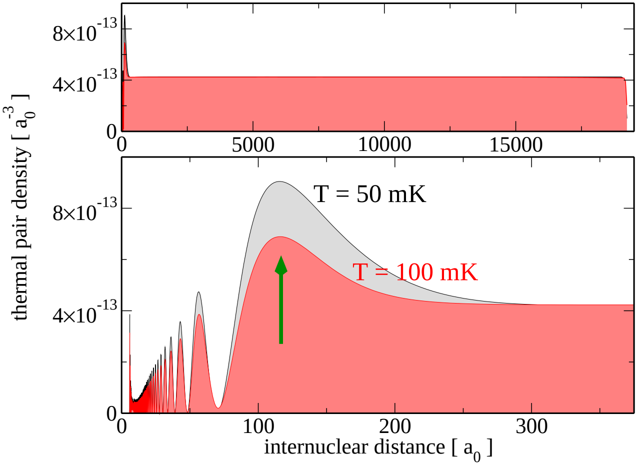


problem of controllability !

resonances change continuum structure

and enhance controllability



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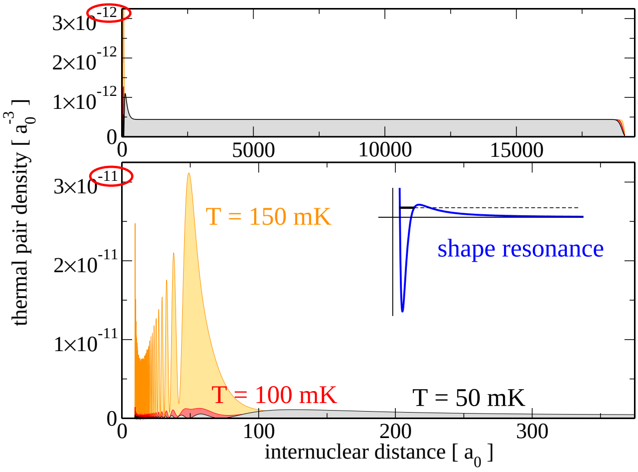

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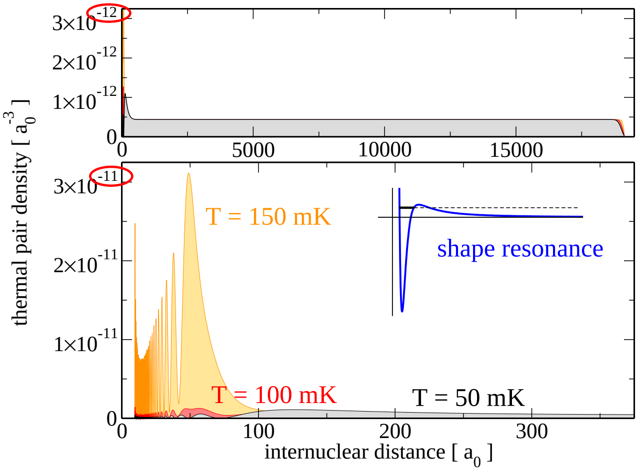
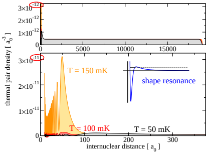


→ **study PA in terms of control out of continuum**

overview



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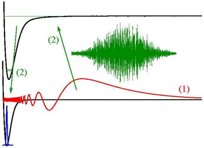

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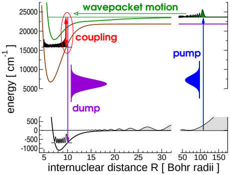
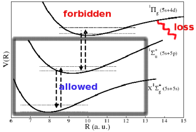
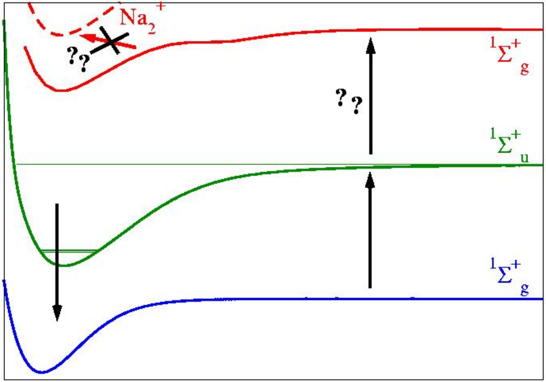


4. outlook

5. **modified Krotov algorithm: keeping the population in a subspace**



a problem with very strong fields



→ **tell algorithm to avoid door-way states**

OCT for quantum information

$$t = 0 \qquad\qquad t = T$$

$$|\varphi_{i,n}\rangle \quad n = 1, \ldots, N \qquad \longrightarrow \qquad |\varphi_{f,n}\rangle \quad n = 1, \ldots, N$$

1. N simultaneous state-to-state transitions

   Tesch & de Vivie-Riedle, PRL 89, 157901 (2002)

2. optimization of unitary transformation

   $$\frac{\partial \hat{\mathbf{U}}(t)}{\partial t} = -\frac{i}{\hbar}\hat{\mathbf{H}}(t)\hat{\mathbf{U}}(t) \qquad \hat{\mathbf{U}}(T) = e^{i\phi}\hat{\mathbf{O}}$$

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functionals

1. $\eta = \sum_{k=1}^{N} \left| \langle k|\hat{\mathbf{O}}^{+}\hat{\mathbf{U}}(T,0;\varepsilon)|k\rangle \right|^2 = F_{ss}$

2. $\tau = \sum_{k=1}^{N} \langle k|\hat{\mathbf{O}}^{+}\hat{\mathbf{U}}(T,0;\varepsilon)|k\rangle$

$$\longrightarrow F_{re} = \mathfrak{Re}\left[\tau\right] \text{ or } F_{sm} = -\left|\tau\right|^2$$

Palao & Kosloff, PRA 68, 062308 (2003)

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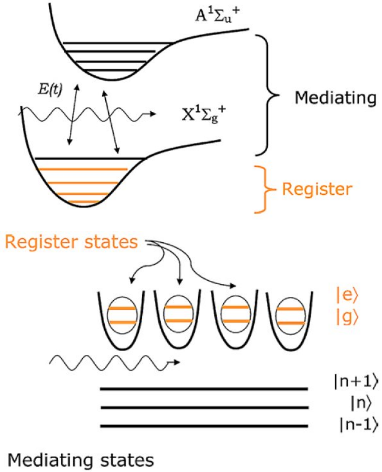
⟶ **optimization of gate operations**

QI: avoid leakage from qu register



Sklarz & Tannor,
arXiv:quant-ph/0404081 &

Chem. Phys. 322, 87 (2006)

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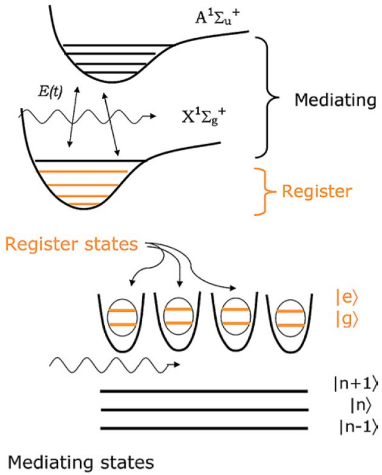

Sklarz & Tannor, arXiv:quant-ph/0404081 &

Chem. Phys. 322, 87 (2006)

local control

$$\dot{J} = \frac{d}{dt}\left| \mathrm{Tr}[\hat{\mathbf{O}}_r^+ \hat{\mathbf{U}}_r] \right|^2 \leq 0$$

$$\dot{C} = \frac{d}{dt} \mathrm{Tr}[\hat{\mathbf{U}}_r^+ \hat{\mathbf{U}}_r] = 0$$

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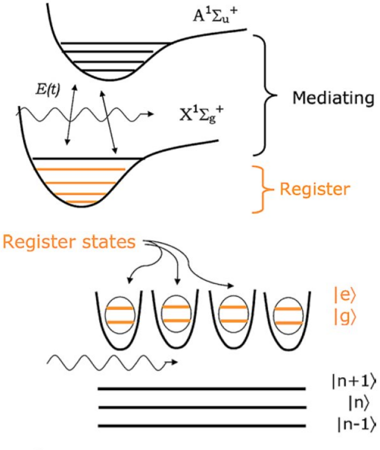

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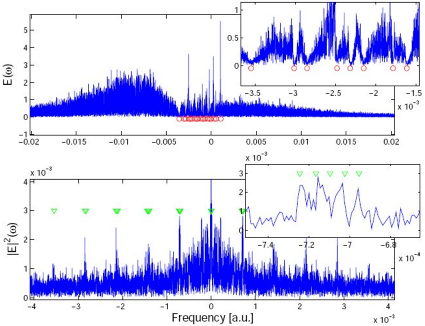
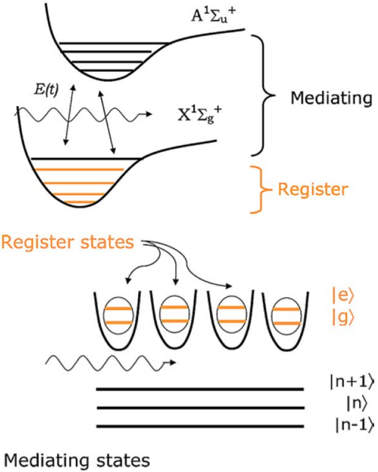


FIG. 5: The spectrum $\tilde{E}(\omega)$ of the field with one-photon transition frequencies marked by (red) circles (top) and the spectrum of the field intensity $|\tilde{E}|^2(\omega)$ with two-photon transition frequencies marked by (green) triangles (bottom).

QI: avoid leakage from qu register



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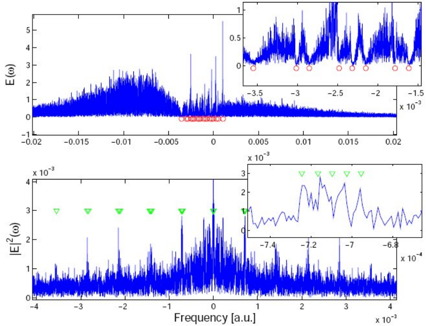

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→ **can we combine restriction to subspace w/ (global) optimal control?**

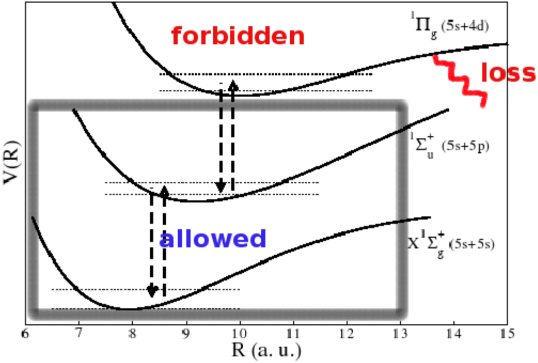
optimal control in a subspace

strong laser fields: undesired effects

avoidable?

optimal control in a subspace

strong laser fields: undesired effects
avoidable?



Palao, Kosloff, CPK, PRA 77, 063412 (2008)

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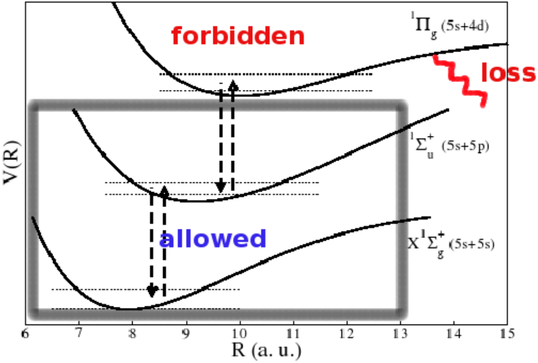

$|\varphi(t)\rangle$ **shall have components only in the allowed subspace:** $\hat{P}_{allow}$

Palao, Kosloff, CPK, PRA 77, 063412 (2008)

control in a subspace: functional

$$J[\varphi, \varphi^{+}, \varepsilon] \quad = \quad J_0[\varphi_T, \varphi_T^*] + J_a[\varepsilon] + J_b[\varphi, \varphi^*]$$

control in a subspace: functional

$$
\begin{aligned}
J[\varphi, \varphi^{+}, \varepsilon] &= J_0[\varphi_T, \varphi_T^*] + J_a[\varepsilon] + J_b[\varphi, \varphi^*] \\
J_0[\varphi_T, \varphi_T^*] &= -\lambda_0 \langle \varphi(T) | \hat{\mathbf{D}} | \varphi(T) \rangle \\
J_a[\varepsilon] &= \int_0^T g_a[\varepsilon]\, dt \\
J_b[\varphi, \varphi^*] &= \int_0^T g_b[\varphi, \varphi^*]\, dt
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with

$$\begin{aligned} g_a[\varepsilon] &= \lambda_a(t)\, [\varepsilon(t) - \varepsilon_r(t)]^2 \\ g_b[\varphi, \varphi^{+}] &= \lambda_b \langle \varphi(t) | \hat{\mathbf{P}}(t) | \varphi(t) \rangle \end{aligned}$$

Krotov method

- ingredients:
  - target $$J_0[\varphi_T, \varphi_T^*] = \lambda_0 \langle \varphi(T) | \hat{\mathbf{D}} | \varphi(T) \rangle$$
  - (physical) constraints $$J_a[\varepsilon] + J_b[\varphi, \varphi^+]$$
  - equations of motion

$$i\hbar \frac{\partial}{\partial t} |\varphi(t)\rangle = \hat{\mathbf{H}}(t) |\varphi(t)\rangle \quad |\varphi(t_0)\rangle = |\varphi_0\rangle$$

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  - target $J_0[\varphi_T, \varphi_T^*] = \lambda_0 \langle \varphi(T)|\hat{\mathbf{D}}|\varphi(T)\rangle$
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- construction of auxiliary functional L

in order to mix the separate dependences on intermediate and final times in the $J's$

$$L[\varphi, \varphi^*, \varepsilon, \Phi] = J[\varphi, \varphi^*, \varepsilon]$$

choose arbitrary $\Phi(t, \varphi, \varphi^*)$ such that

$$L[\varphi^i, \varphi^{*,i}, \varepsilon^i, \Phi] \geq L[\varphi^{i+1}, \varphi^{*,i+1}, \varepsilon^{i+1}, \Phi]$$

i.e. construct Φ from condition that L is maximum for φ^0, $\varphi^{*,0}$

→ *building in monotonic convergence*

Krotov method

$$
\begin{aligned}
L[\varphi,\varphi^*,\varepsilon,\Phi] &= J_0[\varphi_T,\varphi_T^*]+\Phi[\varphi_T,\varphi_T^*]-\Phi[\varphi_0,\varphi_0^*] \\
&\quad +\int_0^T\left\{J_a[\varepsilon]+J_b[\varphi_t,\varphi_t^*]-\frac{d\Phi}{dt}\right\}dt
\end{aligned}
$$

assuming the equation of motion to hold,
minimization of L is equivalent to minimization of J

Sklarz & Tannor, PRA 66, 053619 (2002)

(i) construct $\Phi(t,\varphi^i,\varphi^{i,*})$ such that L is maximum w.r.t. any choice of $\varphi_t^i,\varphi_t^{i,*}$

(ii) derive ε^{i+1} from maximization of

$$\frac{d\Phi}{dt}-\left(J_a[\varepsilon]+J_b[\varphi_t,\varphi_t^*]\right)$$

Krotov method

- variation of the auxiliary functional $L[\varphi, \varphi^*, \varepsilon, \Phi]$:

$$\varepsilon^{i+1}(t) = \varepsilon^i(t) - \frac{1}{\hbar\,\lambda_a(t)}\mathfrak{Im}\left\{\langle\chi(T)|\hat{\mathbf{U}}^+(t,T;\varepsilon^i)\hat{\boldsymbol{\mu}}\hat{\mathbf{U}}(t,0;\varepsilon^{i+1})|\varphi_0\rangle\right\}$$

$$\frac{\partial}{\partial t}|\chi^i(t)\rangle = -\frac{i}{\hbar}\hat{\mathbf{H}}[\varepsilon^i(t)]\,|\chi(t)\rangle + \lambda_b\hat{\mathbf{P}}(t)|\varphi^i(t)\rangle$$

$$|\chi(T)\rangle = -\lambda_0\,\hat{\mathbf{D}}\,|\varphi^i(T)\rangle$$

$$\frac{\partial}{\partial t}|\varphi^{i+1}(t)\rangle = \frac{i}{\hbar}\hat{\mathbf{H}}[\varepsilon^i(t)]\,|\varphi^{i+1}(t)\rangle$$

$$|\varphi(0)\rangle = |\varphi_0\rangle$$

monotonic convergence

$$
\begin{aligned}
\Delta &\equiv J[\varphi^i, \varphi^{*,i}, \varepsilon^i] - J[\varphi^{i+1}, \varphi^{*,i+1}, \varepsilon^{i+1}] \\
&= \Delta_1 + \int_0^T \left(\Delta_{2a}(t) + \Delta_{2b}(t) \right) dt \leq 0
\end{aligned}
$$

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$$\Delta_1 = -\lambda_0 \langle \zeta(T) | \hat{\mathbf{D}} | \zeta(T) \rangle \leq 0 \qquad |\zeta(t)\rangle = |\varphi^{i+1}(t)\rangle - |\varphi^i(t)\rangle$$

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$$\Delta_{2a}(t) = -g_a[\varepsilon^{i+1}] + g_a[\varepsilon^i] + \left[\frac{\partial g_a}{\partial \varepsilon}\right]_{i+1} \left(\varepsilon^{i+1} - \varepsilon^i\right) \leq 0$$

$$\Delta_{2b}(t) = -\lambda_b \langle\zeta(t)|\hat{\mathbf{P}}(t)|\zeta(t)\rangle \leq 0$$

monotonic convergence

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$\lambda_0 \geq 0 \quad \lambda_b \geq 0 \quad \lambda_a(t) \leq 0$ maximization

$\lambda_0 \leq 0 \quad \lambda_b \leq 0 \quad \lambda_a(t) \geq 0$ minimization

monotonic convergence

constraint: keep population in allowed subspace

minimization of J

$$
\begin{aligned}
(a)\ \ \hat{\mathbf{P}}(t) &= \hat{\mathbf{P}}_{allow} : \quad \lambda_b \leq 0 \\
(b)\ \ \hat{\mathbf{P}}(t) &= \hat{\mathbf{P}}_{forbid} : \quad \lambda_b \geq 0
\end{aligned}
$$

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(a) **and** *(b)* **physically equivalent, but only** *(a)* **yields monotonic convergence**

. . . a slight complication

$$\frac{\partial}{\partial t}|\chi^i(t)\rangle = -\frac{i}{\hbar}\hat{\mathbf{H}}[\varepsilon^i(t)]\,|\chi(t)\rangle + \lambda_b \hat{\mathbf{P}}(t)|\varphi^i(t)\rangle$$

how to propagate an inhomogeneous

Schrödinger eq?

1. diagonalization of $\hat{\mathbf{H}}$
2. split propagator $\curvearrowright$ accuracy!?
3. Chebychev propagator for inhomogeneous Schrödinger equations

   formal solution: $|\psi(t)\rangle_{(m)} = \sum_{j=0}^{m-1} \frac{t^j}{j!}|\lambda^{(j)}(0)\rangle + \hat{\mathbf{F}}_m|\lambda^{(m)}(0)\rangle$

   spectral approx.: $|\psi(t)\rangle_{(m)N} = \sum_{j=0}^{m-1} \frac{t^j}{j!}|\lambda^{(j)}(0)\rangle + P_{N(m)}(\hat{\mathbf{H}})|\lambda^{(m)}(0)\rangle$

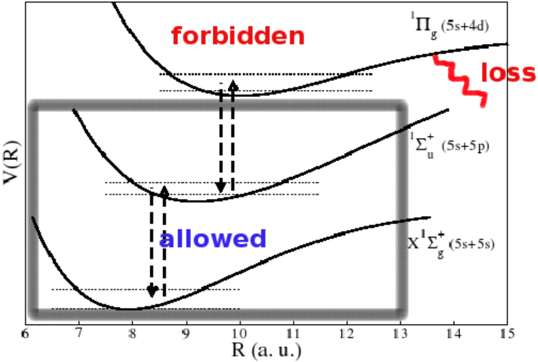
   first order: $|\psi(t)\rangle_{(1)} = e^{-i\hat{\mathbf{H}}t}|\psi_0\rangle + (-i\hat{\mathbf{H}})^{-1}(e^{-i\hat{\mathbf{H}}t} - \mathbb{1})|\Phi_0\rangle$

Ndong, Tal-Ezer, Kosloff, CPK. J. Chem. Phys. 130, 124108 (2009)

optimal control in a subspace

strong laser fields: undesired effects

avoidable?



$|\varphi(t)\rangle$ **shall have components only in the allowed subspace:**

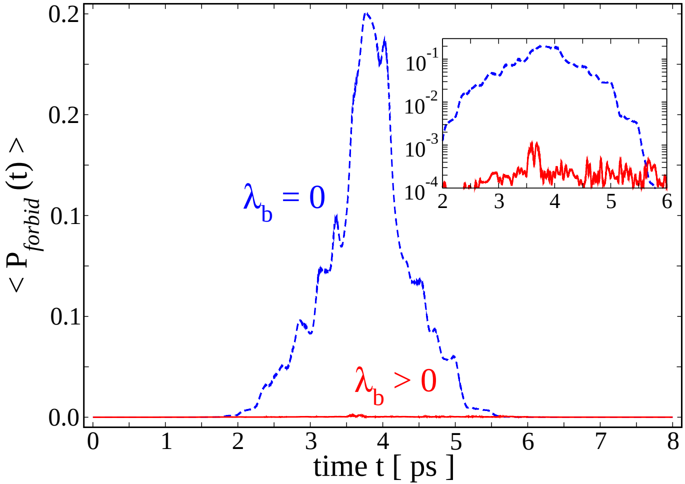
$\hat{\mathsf{P}}_{allow}$

Palao, Kosloff, CPK, PRA 77, 063412 (2008)

optimal control in a subspace

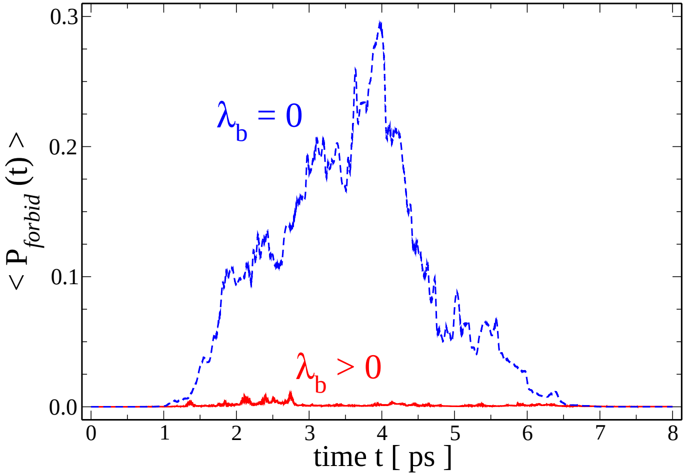
model: 33 vibronic levels of Rb_2, 22 allowed

target: transition $v = 0 \longrightarrow v = 1$



optimal control in a subspace

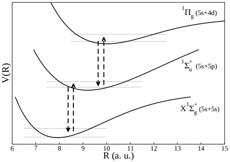
model: 33 vibronic levels of Rb_2, 22 allowed
target: QFFT in $v = 0, \ldots, 3$

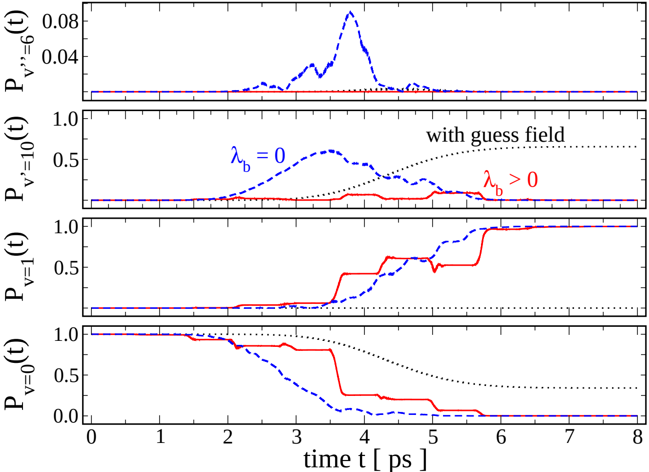


optimal control in a subspace

model: 33 vibronic levels of Rb_2, 22 allowed

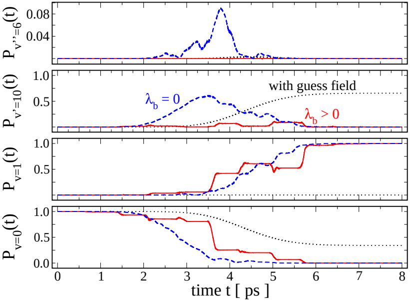
target: transition $v = 0 \longrightarrow v = 1$





optimal control in a subspace

model: 33 vibronic levels of Rb_2, 22 allowed
target: transition $v = 0 \longrightarrow v = 1$

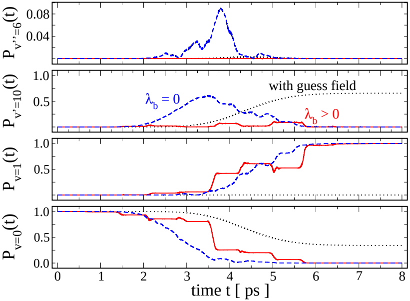


standard OCT:
large amount of population in intermediate state can be further excited to forbidden subspace

OCT w/ state-dep. constraint:
population transfer via ladder-like process $\leftrightarrow$ short subpulses

optimal control in a subspace

model: 33 vibronic levels of Rb_2, 22 allowed
target: transition $v = 0 \longrightarrow v = 1$



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large amount of population in intermediate state can be further excited to forbidden subspace

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population transfer via ladder-like process $\leftrightarrow$ short subpulses

$\longrightarrow$ **state-dep. constraint conveys desired physics to the algorithm**