# Coherent control of ultracold molecules

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Basics on Quantum Control Summer School, Cargèse
August 21, 2008

# overview

**introduction 1:**
**ultracold molecules** ↘

**introduction 2:**
**coherent control** ↙

**4 examples:**

**making ultracold molecules with laser light**

**shaping the potential energy surfaces**

**quantum information with ultracold molecules**

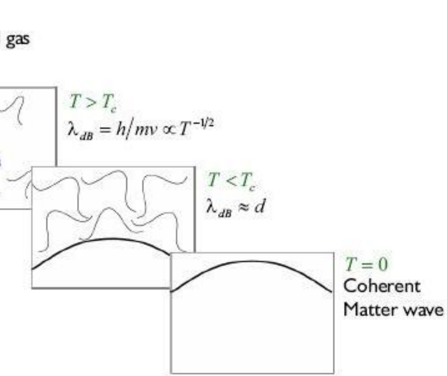
**cooling molecules**

**summary**

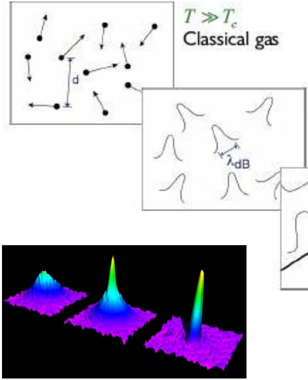
# introduction 1:
# ultracold molecules

# what means ultracold?

ultracold: $T \leq 100\,\mu\text{K}$ → 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

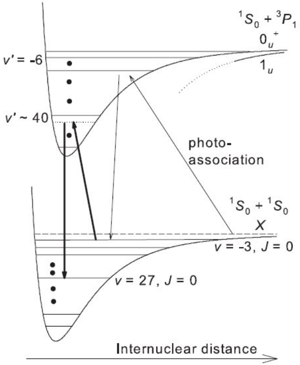
# why ultracold molecules

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

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

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

$\nu$ transition frequencies




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

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# why ultracold molecules

- internal degrees of freedom, permant dipole moment
- interesting applications:
  - molecular Bose-Einstein condensate
  - quantum computer
    - cold ≙ little decoherence
  - precision measurements & test of fundamental symmetries
    - cold ≙ long observation times
  - ultracold collisions / reactions
    - cold ≙ tunneling & resonances

    ➔ **coherent control**

# introduction 2:
# coherent control

# what means coherent control?

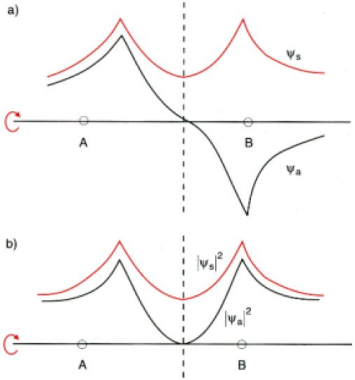
quantum mechanics $\triangleq$ probabilistic,
*deterministic* theory

$|\psi(t=0)\rangle$ Schrödinger equation $\rightsquigarrow$ $|\psi(t>0)\rangle$

For $|\psi(t=0)\rangle$ given,
what dynamics ($\triangleq$ what $\hat{\mathbf{H}}$)
guarantees a certain $|\psi(t>0)\rangle$?

# principle of coherent control

- QM: matter $\sim$ waves $\longrightarrow$ superposition principle




$$\Psi_s = \frac{1}{\sqrt{2}}\phi_s(r_A) + \frac{1}{\sqrt{2}}\phi_s(r_B)$$

$$\Psi_a = \frac{1}{\sqrt{2}}\phi_s(r_A) - \frac{1}{\sqrt{2}}\phi_s(r_B)$$

# principle of coherent control

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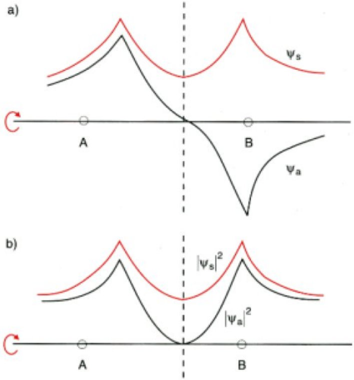


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$$+1 = e^{2\pi i} \quad -1 = e^{\pi i}$$

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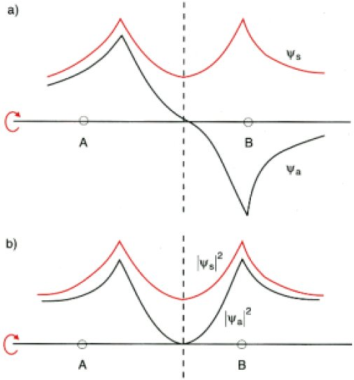


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- manipulation of relative phases of different partial wavepackets $\longrightarrow$ control of interferences

# what is a wavepacket?

$$|\Psi(t)\rangle = \sum_i c_i \underbrace{e^{-\frac{i}{\hbar}E_i t}} |\varphi_i\rangle$$

time-dependent phases

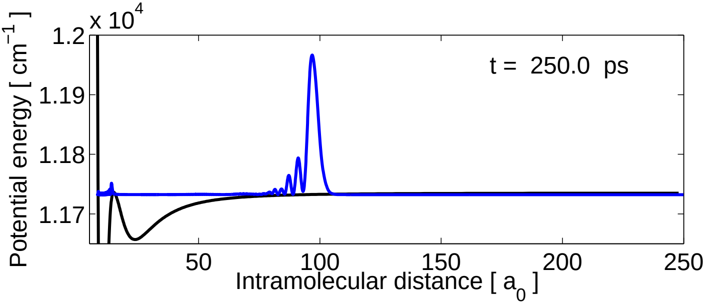
**coordinate representation:**

$$\langle R|\Psi(t)\rangle = \Psi(R,t) = \sum_i c_i e^{-\frac{i}{\hbar}E_i t}\varphi_i(R)$$

**wavepacket dynamics**

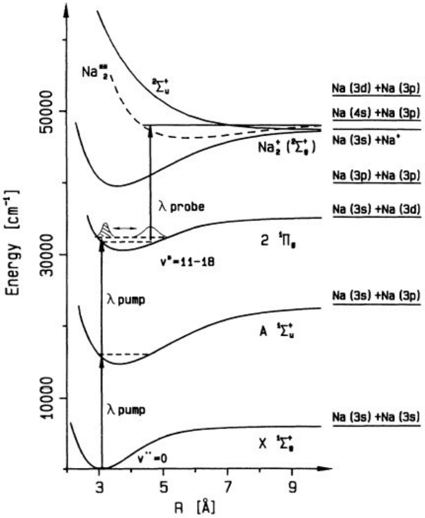
*java applets*

# wavepacket dynamics



# control in time domain

## Tannor & Rice




Baumert, Grosser, Thalweiser, Gerber, PRL 67, 3753 (1991)

- spatially localized wavepackets
- manipulation of phase $\triangleq$ variation of $\Delta t$

# control in time domain

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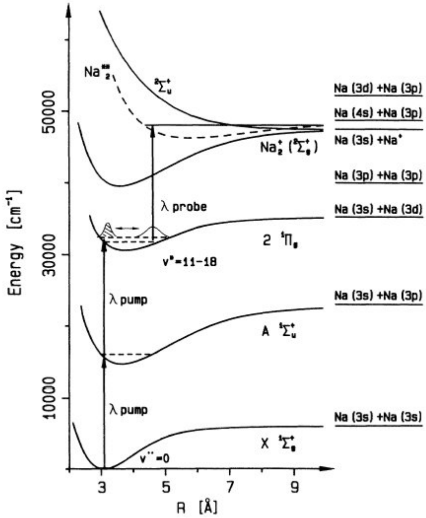


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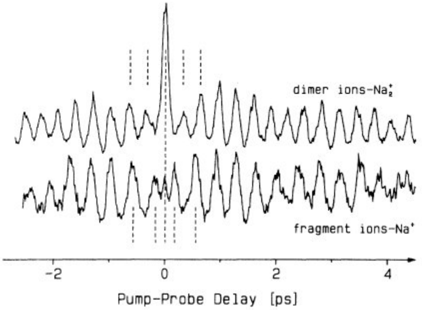


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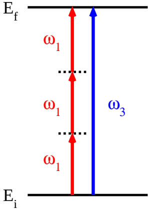
# coherent control

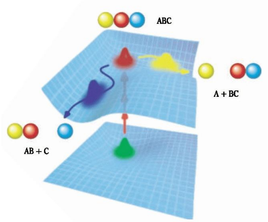
## the classic examples

Brumer & Shapiro




Shapiro & Brumer: Principles of Quantum Control of Molecular Processes. Wiley 2003

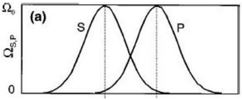
Tannor & Rice




Brixner & Gerber

*Physikal. Blätter*, April 2001

STIRAP




Bergmann, Theuer, Shore

*Rev. Mod. Phys.* **70**, 1003 (1998)

**all realized experimentally in the 1990s**

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## the classic examples

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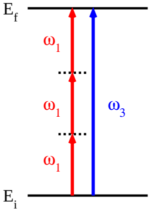


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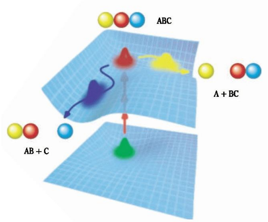


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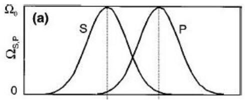


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**all realized experimentally in the 1990s**

**but there's more to control: optimization**

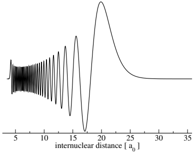
# optimal control

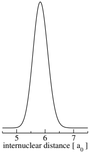
## theory: inversion problem

find the 'potential', which
generates the desired dynamics!

Schrödinger equation

$\mathbf{\Psi}(t = t_0)$  $\mathbf{\Psi}(t = t_0 + T)$




CPK, Palao, Kosloff, Masnou-Seeuws,
*Phys. Rev. A* **70**, 013402 (2004)

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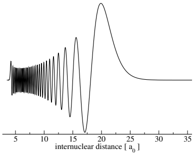


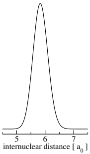


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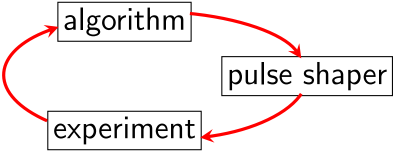
## experiment: feedback-loops




but: black-box optimization of often complex systems

- ➔ *average* over dof
- ➔ thermal *average*

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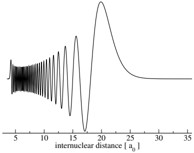


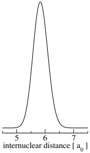


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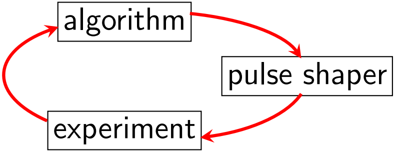



but: black-box optimization of often complex systems

- ➔ *average* over dof
- ➔ thermal *average*

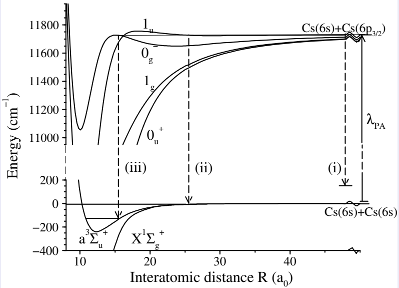
⟶ **ultracold atoms & molecules**

# example 1:
# making ultracold molecules with laser light

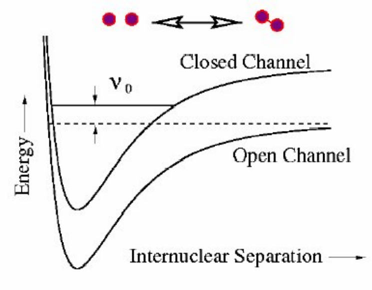
# formation of ultracold molecules

- laser cooling does not work for molecules (to date)!
- alternative cooling methods $\rightsquigarrow$ $T \gtrsim 30$ mK
- laser & evaporative cooling of atoms, then

## photoassociation




## Feshbach resonances




$T \sim 10$ nK $\ldots 100\ \mu$K

# formation of ultracold molecules

## photoassociation

- general (optical transitions)
- first $\mu$K molecules

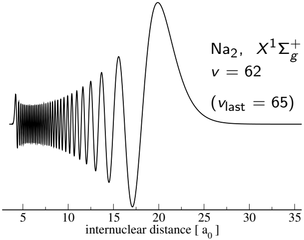
**but** spontaneous emission
$\leftrightarrow$ coherence of BEC

## Feshbach resonances

- first molecules from atomic BEC
- first molecular BEC

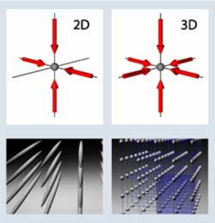
**but** not generally available
not generally feasible

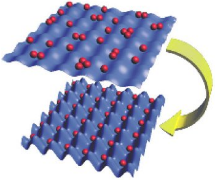
high vibrational excitation, $J \gtrsim 0$

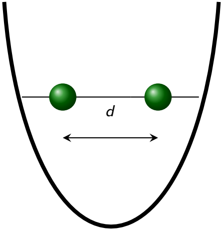


1. can we find an optical analogon of magnetic Feshbach resonances?
2. what about coherent photoassociation?
3. how do we get 'true' molecules?

# optical Feshbach resonances



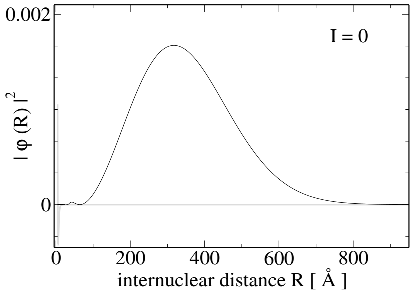






**2 atoms at the same site of an optical lattice**

# optical Feshbach resonances

CPK, Masnou-Seeuws, Kosloff, *Phys. Rev. Lett.* **94**, 193001 (2005)




first trap level (ground state potential)

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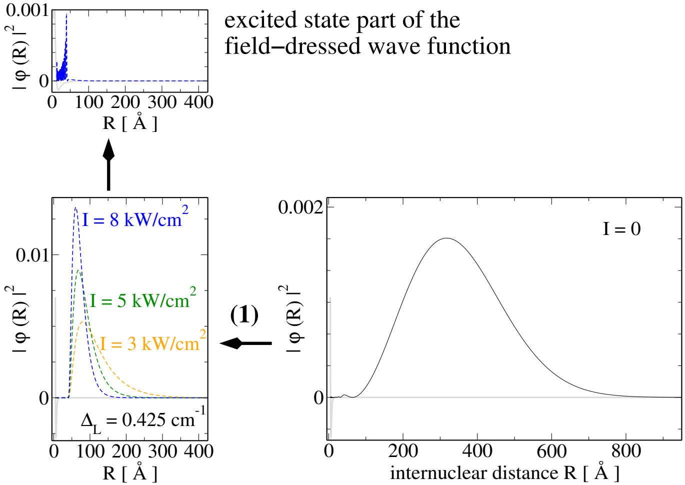


ground state part of the
field–dressed wave functions

first trap level (ground state potential)

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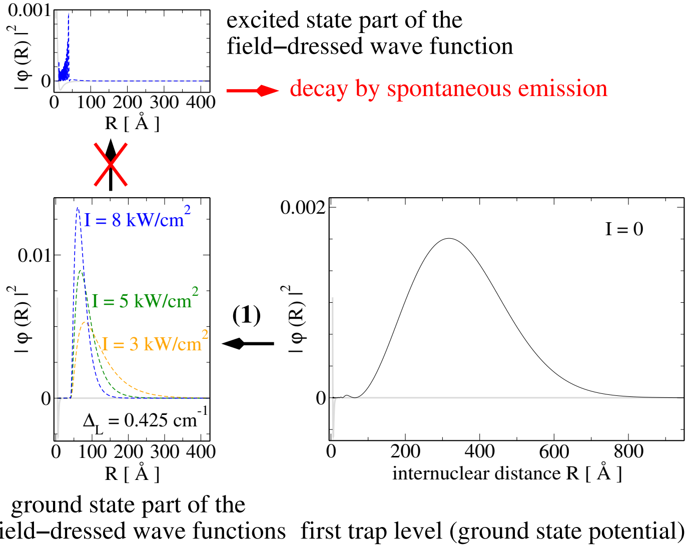

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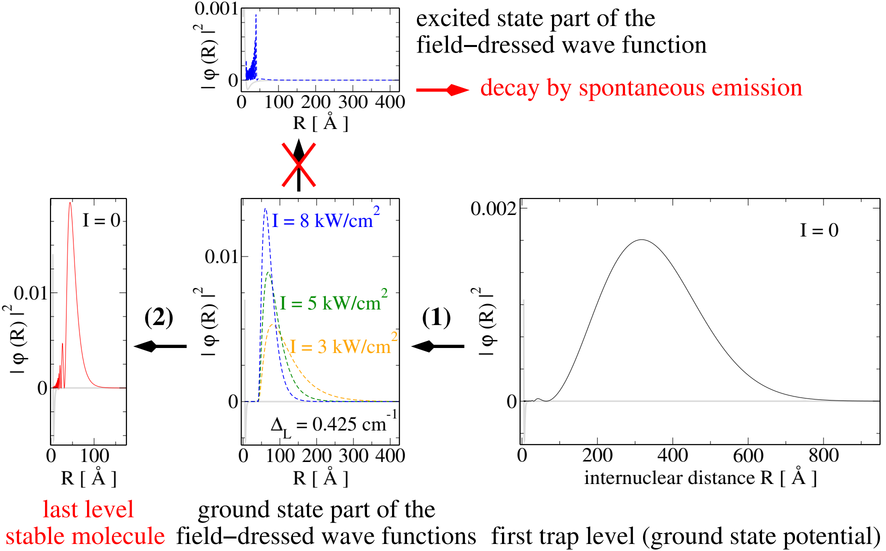
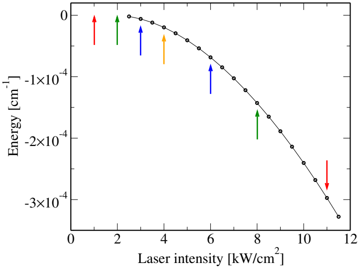


# Feshbach resonance due to intensity

when crossing the resonance, the lowest trap state / continuum state becomes a bound state



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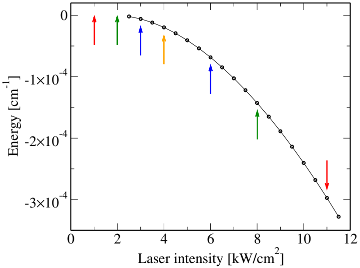


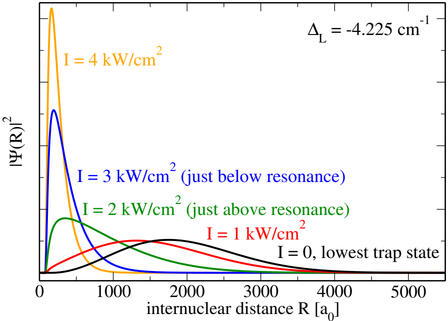

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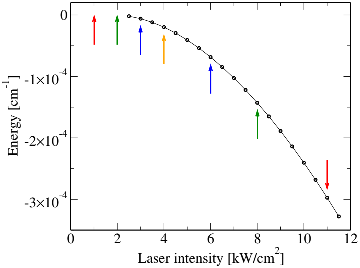


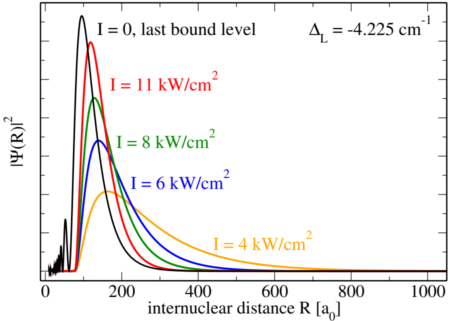


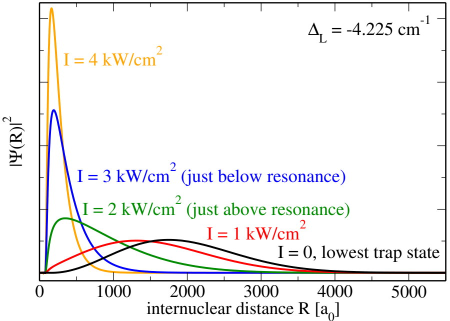

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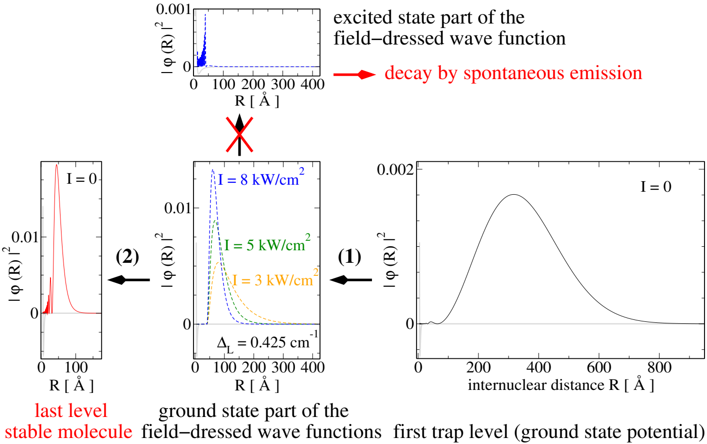
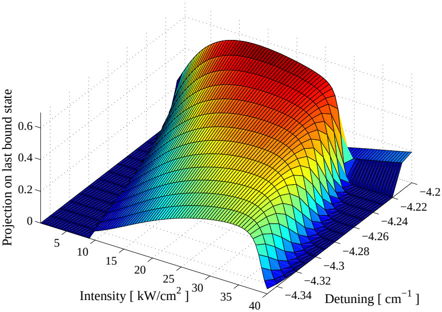



**can we switch on the laser adiabatically while avoiding spontaneous emission?**

# gain vs loss

projection :
$\langle \phi_{\text{bare}} | \varphi_{\text{dressed}} \rangle$
$\doteq$ sudden switchoff of field
**gain**



# gain vs loss

projection :
$\langle \phi_{\text{bare}} | \varphi_{\text{dressed}} \rangle$
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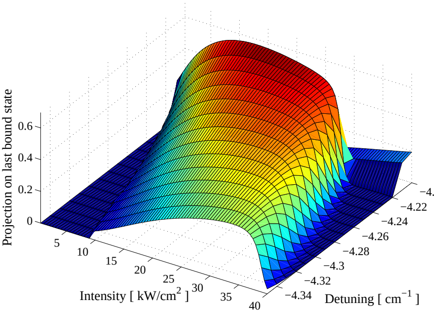
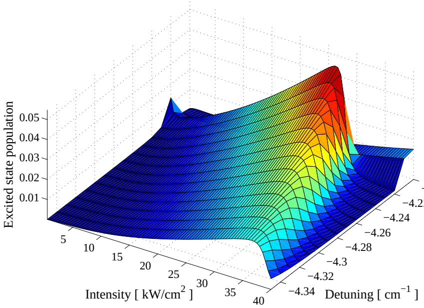
**gain**

excited state part
$P_{\text{exc}} = \langle e | \varphi_{\text{dressed}} \rangle$
$\doteq$ lifetime of $|\varphi_{\text{dressed}}\rangle$
**loss**



# timescales

**constraint of adiabaticity**

$$T \gg T_{\mathrm{ad}} = \frac{\hbar}{E_{\mathrm{trap}}^{\mathrm{ground}}}$$

**constraint of spont. emission**

$$T \ll T_{\mathrm{spont}} = \frac{\tau_{\mathrm{atom}}}{\sqrt{2}P_{\mathrm{exc}}^{\mathrm{dressed}}}$$

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$$T \ll T_{\mathrm{spont}} = \frac{\tau_{\mathrm{atom}}}{\sqrt{2} P_{\mathrm{exc}}^{\mathrm{dressed}}}$$

$\nu_{\mathrm{trap}} = 5$ kHz,
$E_{\mathrm{trap}}^{\mathrm{ground}} \approx 1.7 \cdot 10^{-4}$ cm$^{-1}$
$T_{\mathrm{ad}} \approx 3000$ ns

$\nu_{\mathrm{trap}} = 250$ kHz,
$E_{\mathrm{trap}}^{\mathrm{ground}} \approx 1.3 \cdot 10^{-3}$ cm$^{-1}$
$T_{\mathrm{ad}} \approx 45$ ns

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$\nu_{\mathrm{trap}} = 250$ kHz,
$E_{\mathrm{trap}}^{\mathrm{ground}} \approx 1.3 \cdot 10^{-3}\ \mathrm{cm}^{-1}$

$$T_{\mathrm{ad}} \approx 45\ \mathrm{ns}$$

i.e. gain a factor 100 w.r.t. $\tau_{\mathrm{atom}} \approx 30$ ns by choosing the adiabatic path right!

$$T_{\mathrm{spont}} \approx 3000\ \mathrm{ns}$$

# timescales

**constraint of adiabaticity**

$$T \gg T_{\rm ad} = \frac{\hbar}{E_{\rm trap}^{\rm ground}}$$

**constraint of spont. emission**

$$T \ll T_{\rm spont} = \frac{\tau_{\rm atom}}{\sqrt{2} P_{\rm exc}^{\rm dressed}}$$

$\nu_{\rm trap} = 5$ kHz,
$E_{\rm trap}^{\rm ground} \approx 1.7 \cdot 10^{-4}$ cm$^{-1}$

$T_{\rm ad} \approx 3000$ ns

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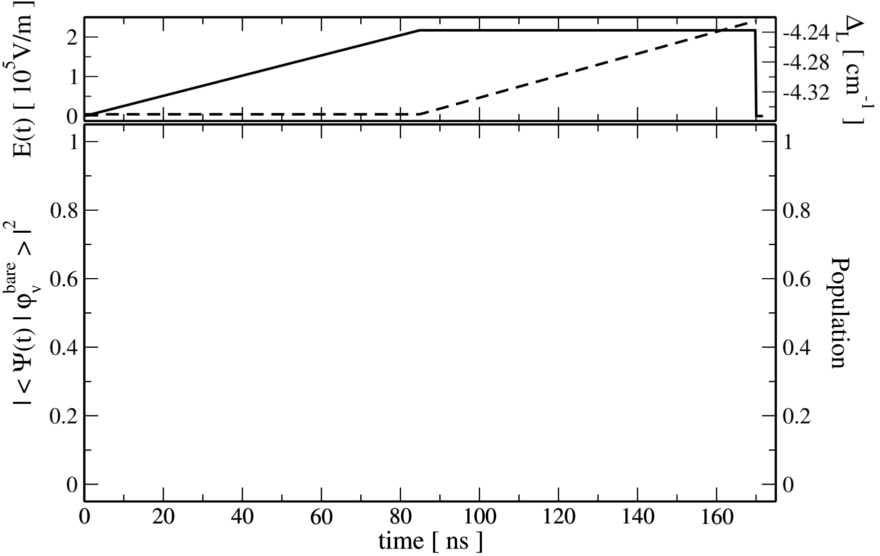
i.e. gain a factor 100 w.r.t. $\tau_{\rm atom} \approx 30$ ns by choosing the adiabatic path right!

$T_{\rm spont} \approx 3000$ ns

**There is a time window allowing the adiabatic formation of molecules via an optical Feshbach resonance while avoiding spontaneous emission for sufficiently tight traps!**

# time-dep. simulations: $\nu = 250$ kHz

ramps of field amplitude & frequency (final $I = 8$ kW/cm$^2$ )



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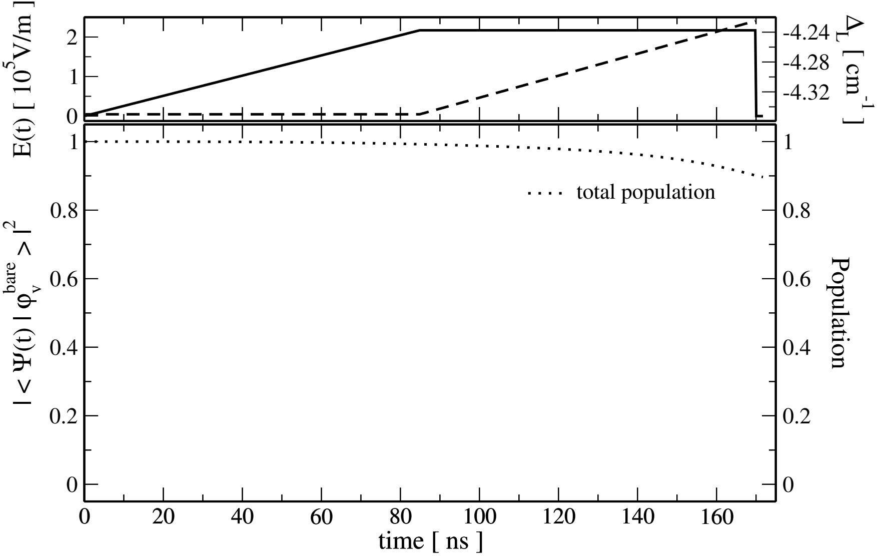

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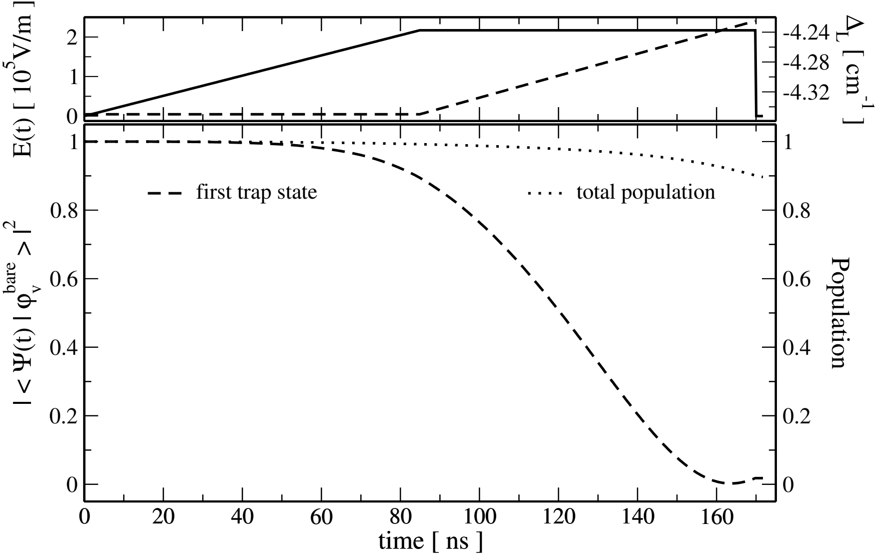
ramps of field amplitude & frequency (final $I = 8$ kW/cm$^2$ )



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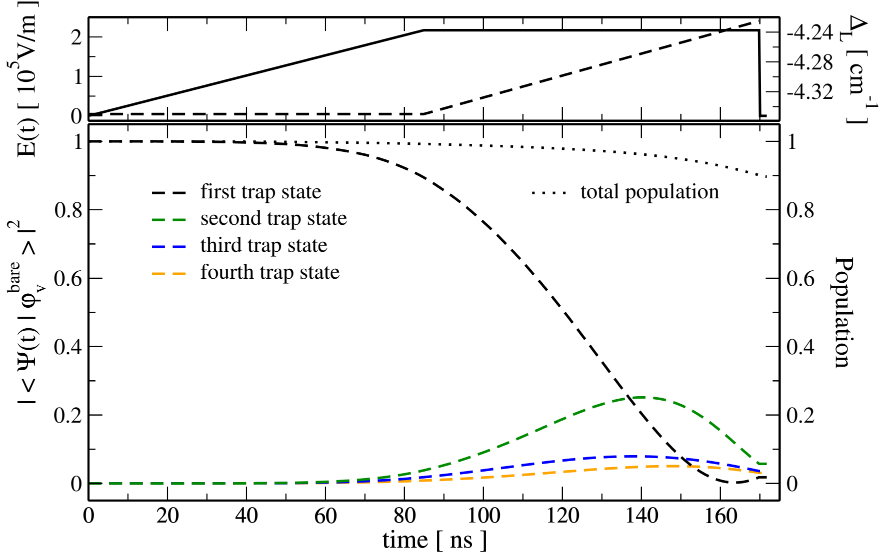

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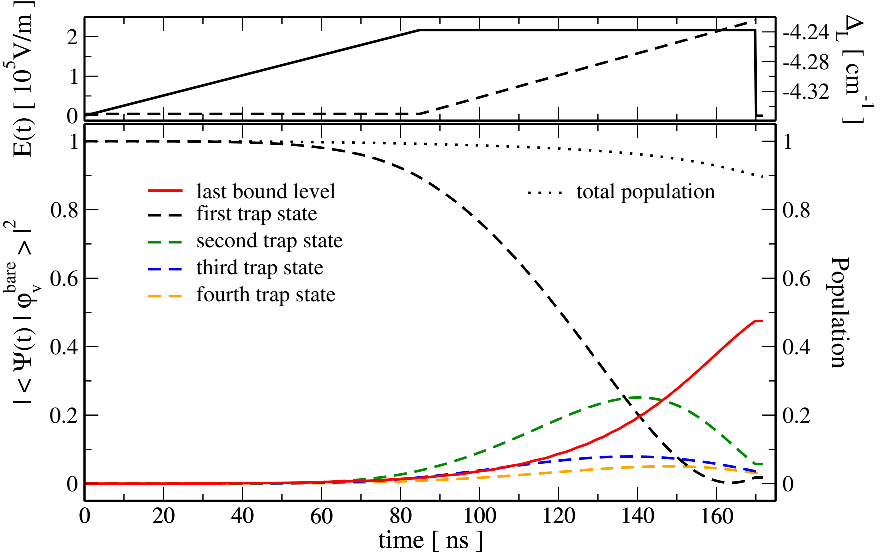
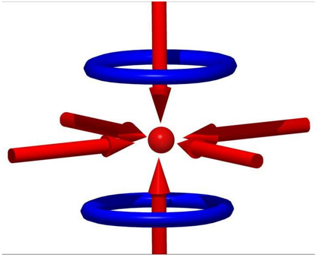
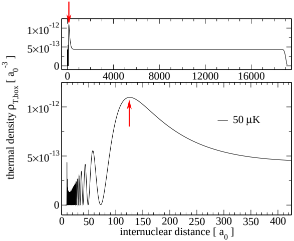


# coherent photoassociation

## atoms in a MOT



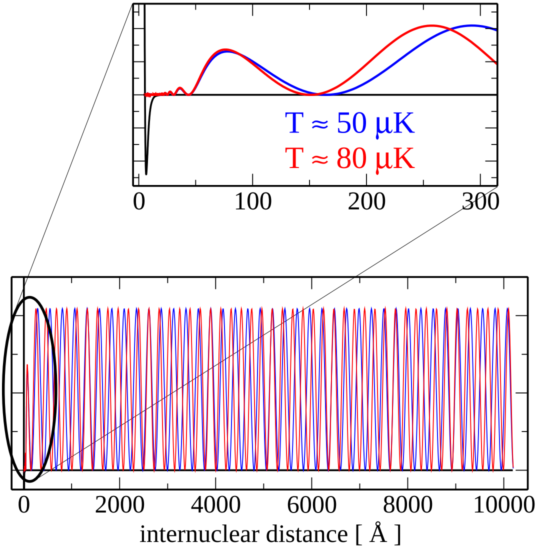
## thermal ensemble




**→ PA is difficult**

# cold thermal ensemble

## cold collisions: s-wave

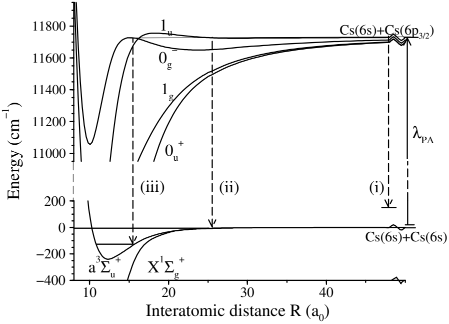



*CPK, Kosloff, Luc-Koenig, Masnou-Seeuws, Crubellier, J Phys B 39, S1017 (2006)*

# 'pump'-'dump' photoassociation

## making molecules with photoassociation:

## cw vs short pulses




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

- spontaneous emission
- time-reversal symmetry

# 'pump'-'dump' photoassociation

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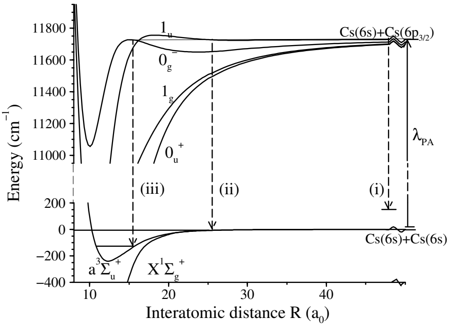
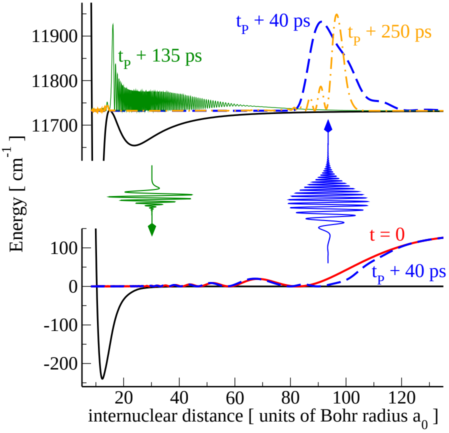


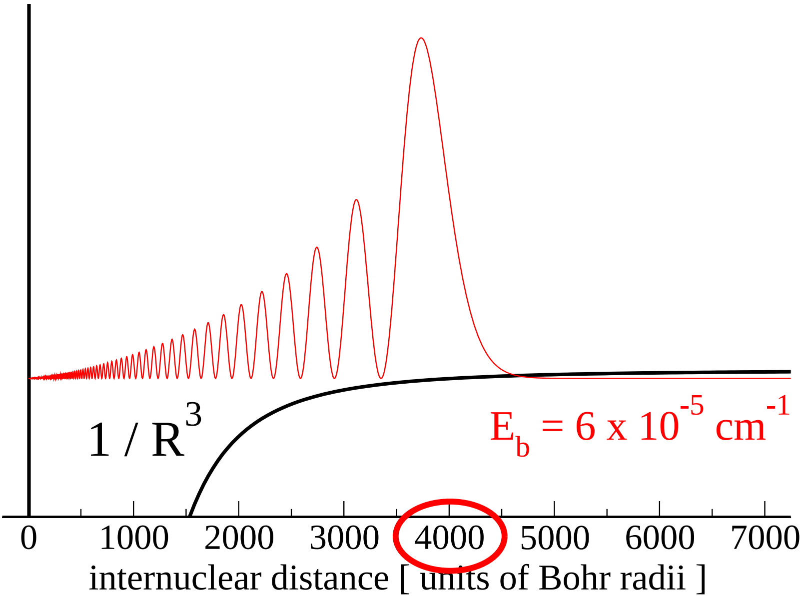

Masnou-Seeuws & Pillet, Adv. At. Mol. Opt. Phys. 47, 53 (2001)




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

- spontaneous emission
- time-reversal symmetry

# why is making molecules possible?



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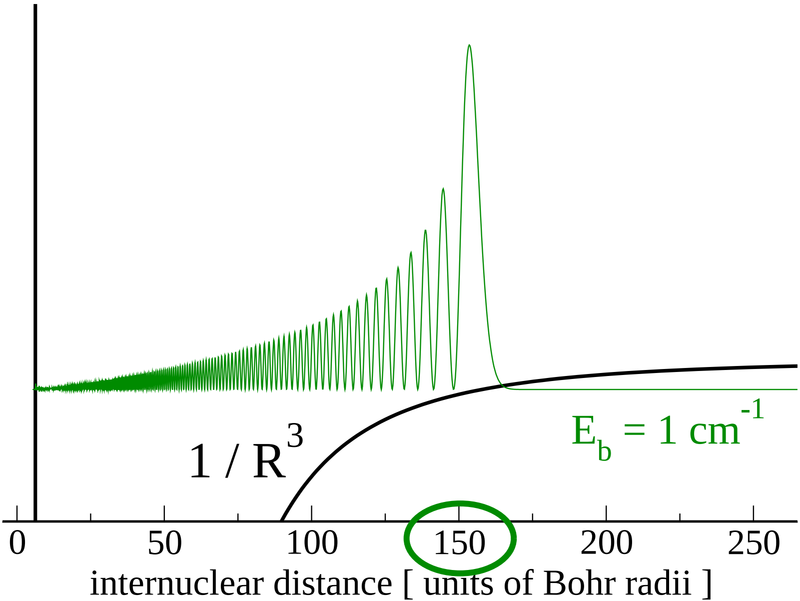

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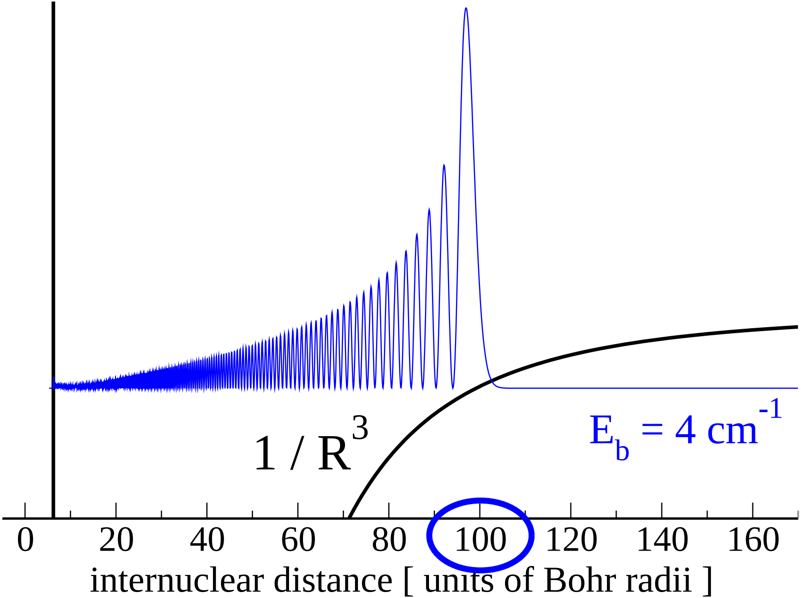
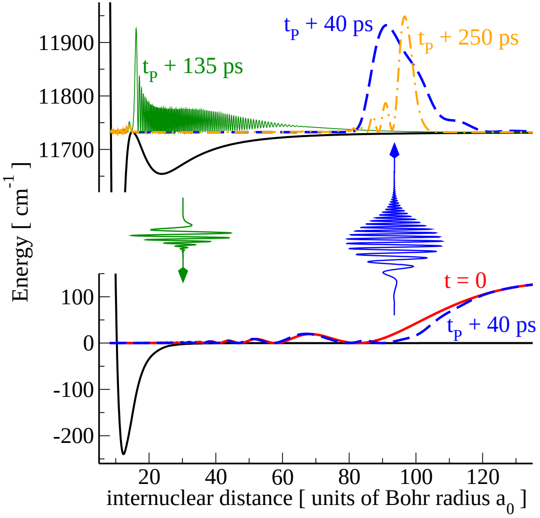


# coherent photoassociation?



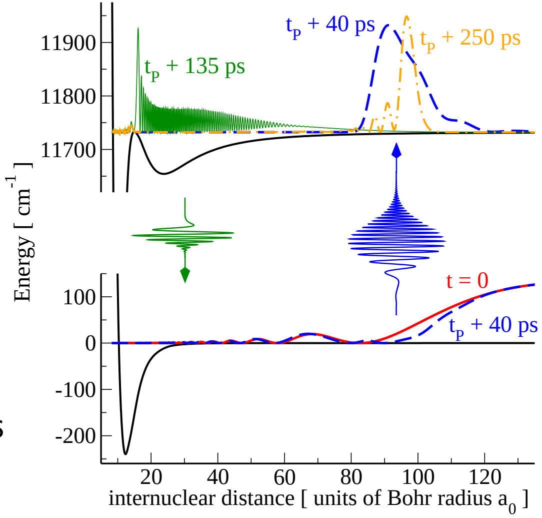

*CPK, Luc-Koenig, Masnou-Seeuws, PRA 73, 033408 (2006)*

# coherent photoassociation?

**what is different from previous pump-probe schemes?**

- initial state
- timescales

↷ bandwidths

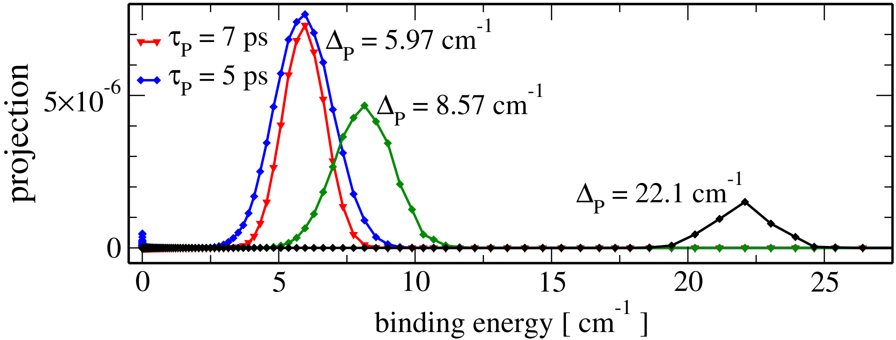



*CPK, Luc-Koenig, Masnou-Seeuws, PRA 73, 033408 (2006)*

# choice of pulses

## role of laser detuning and spectral bandwidth

projection of $\Psi_{exc}(R, t_{final})$ auf Vibrationsniveaus von $\hat{\mathbf{H}}_e(R)$, $^{87}Rb_2$




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

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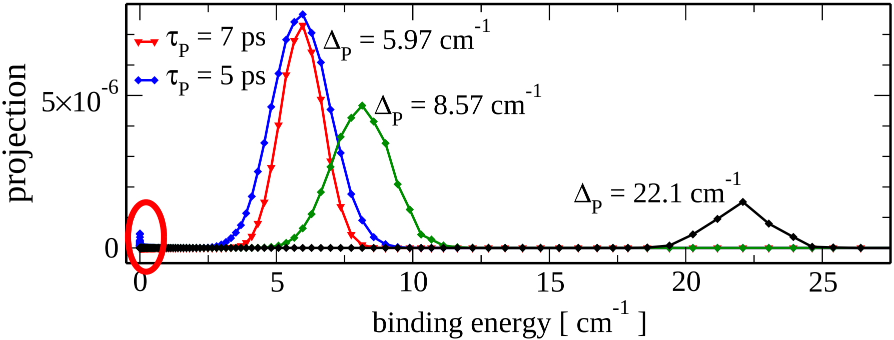


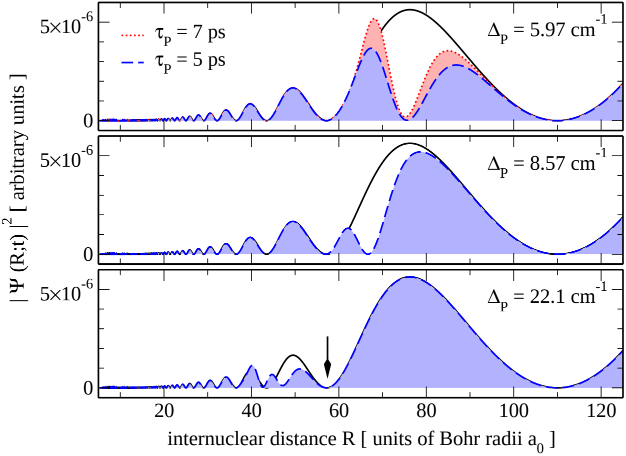

→ **narrow band pulses**

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

# choice of pulses

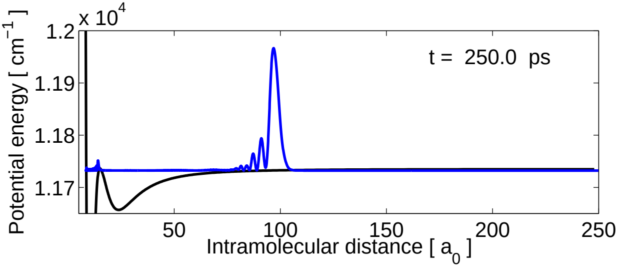
## role of laser detuning, spectral bandwidth & intensity

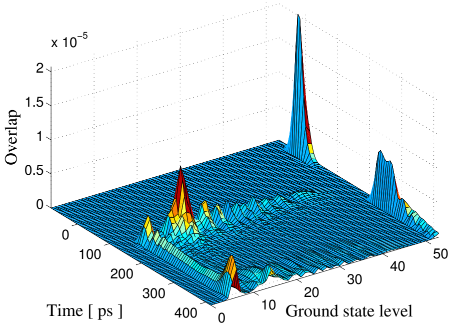
### ground state wavefunction after the pulse




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

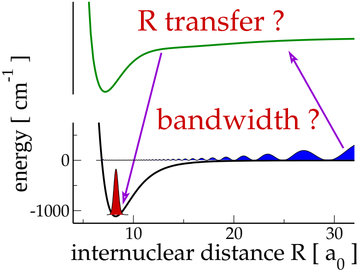
# photoassociation dynamics

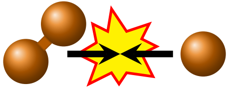


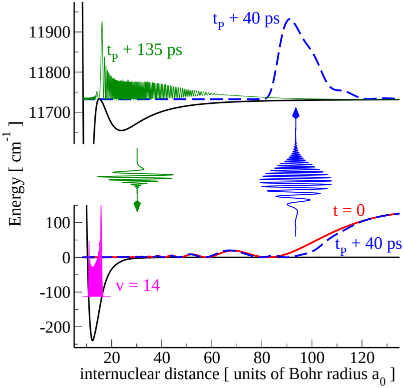




**time-dependent**
**Franck-Condon factors ➔**
**pump-dump-delay**

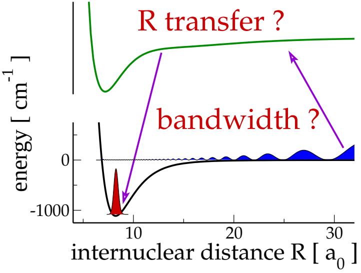
# open questions: PA with pulses

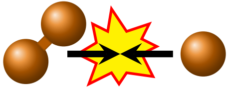


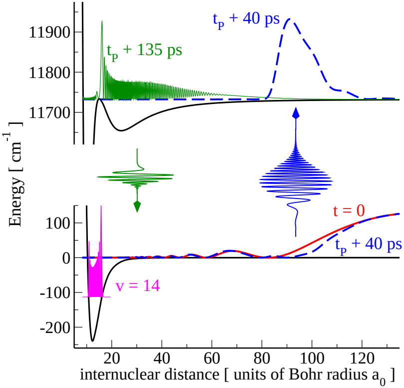





# open questions: PA with pulses

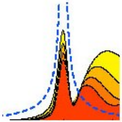





## first experiments

### APS Journals Highlights



**On PRL's Cover**

**June 13, 2008**

Population of a rubidium molecular state as a function of the delay time between two femtosecond laser pulses. The peak at zero and the coherent oscillations agree with the experiment in which ultracold molecules were formed by photoassociation using $10^8$ trapped atoms.

Salzmann et al., PRL 100, 233003 (2008)

et al. = . . . Fabian Weise . . .

# example 2:

# laser induced resonance

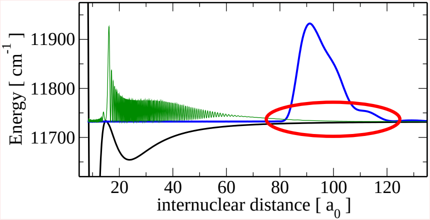
# or

# shaping the potential energy surfaces

# is the pump-dump scheme general?

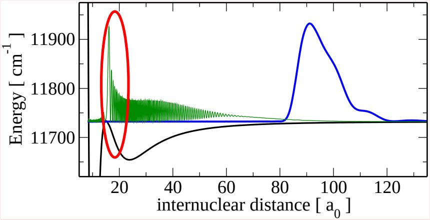
**the perfect example: $Cs_2\ 0_g^-(P_{3/2})$**

## 1. excitation (pump)




- at long range : $1/R^3$ (small $\Delta_P$)

## 2. stabilization (dump)

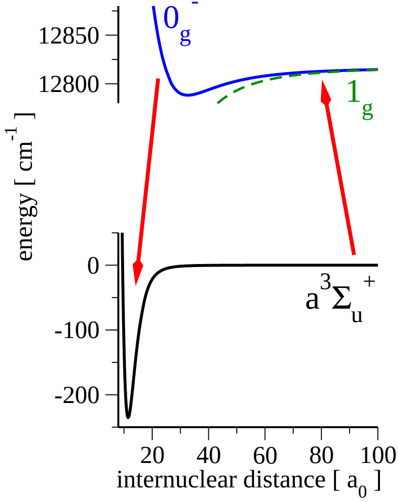



- at short range : $-1/R^3$ ($\Delta_D < 0$)

**efficiency of process**

# two possible dump mechanisms

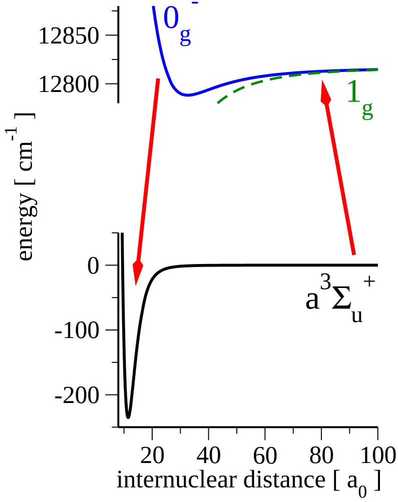
## softly repulsive wall



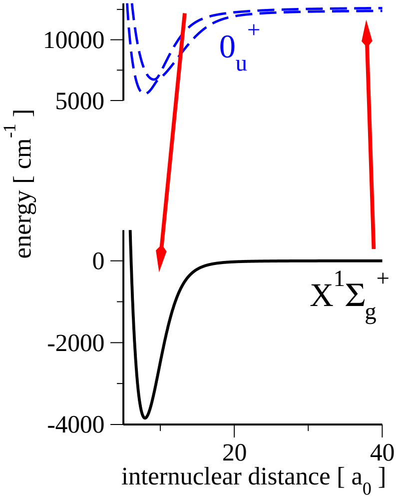

*see also: Dion, Drag, Dulieu, Laburthe Tolra, Masnou-Seeuws, Pillet, PRL 86, 2253 (2001)*

# two possible dump mechanisms

## softly repulsive wall

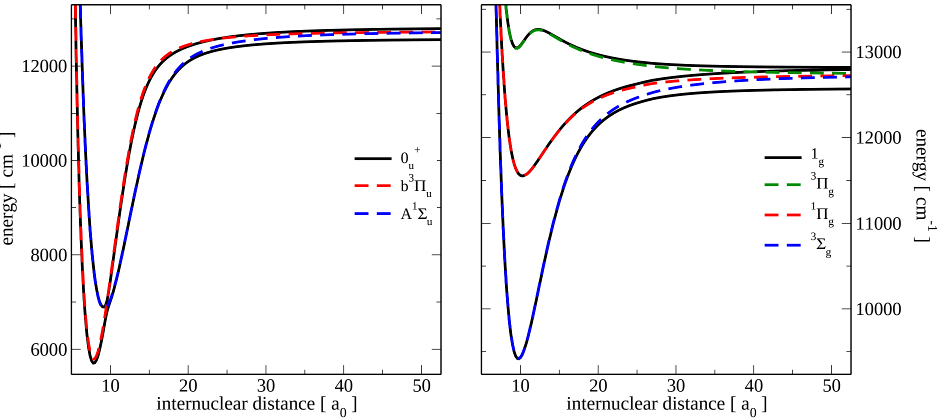



## resonant coupling



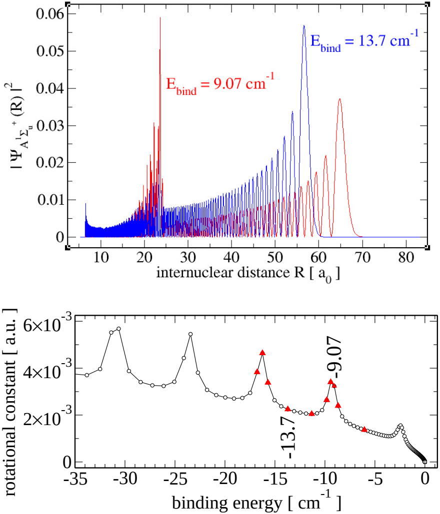

*see also: Dion, Drag, Dulieu, Laburthe Tolra, Masnou-Seeuws, Pillet, PRL 86, 2253 (2001)*

# resonant vs non-res. SO coupling



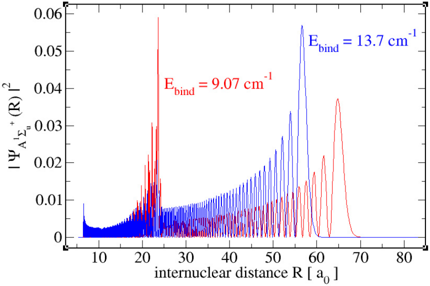

*potentials and dipole moments : M. Aymar & O. Dulieu (Laboratoire Aimé Cotton, Orsay)*

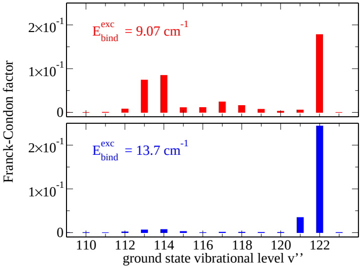
# resonant coupling

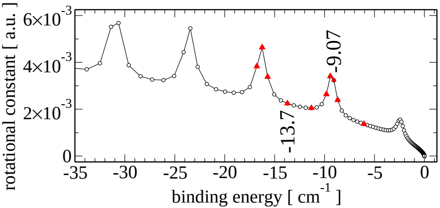



*Pechkis, Wang, Eyler, Gould, Stwalley, CPK, PRA 76, 022504 (2007)*

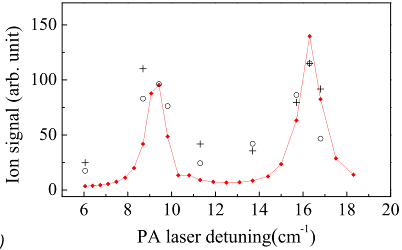
# resonant coupling




## comparison theory-experiment

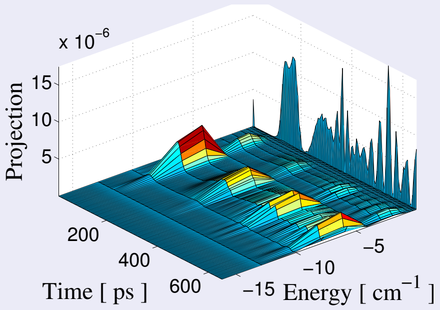



*Pechkis, Wang, Eyler, Gould, Stwalley, CPK, PRA 76, 022504 (2007)*

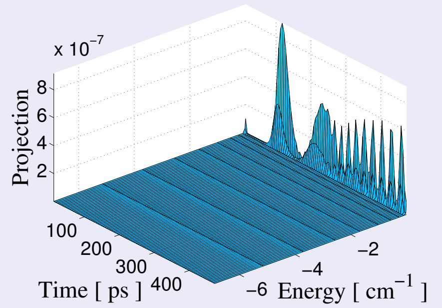
# stabilization to the ground state?

## time-dependent FC factors

$0_u^+$



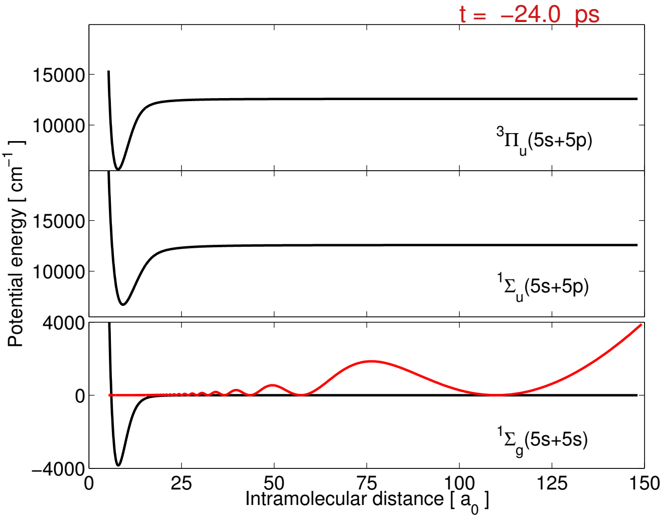

$1_g$



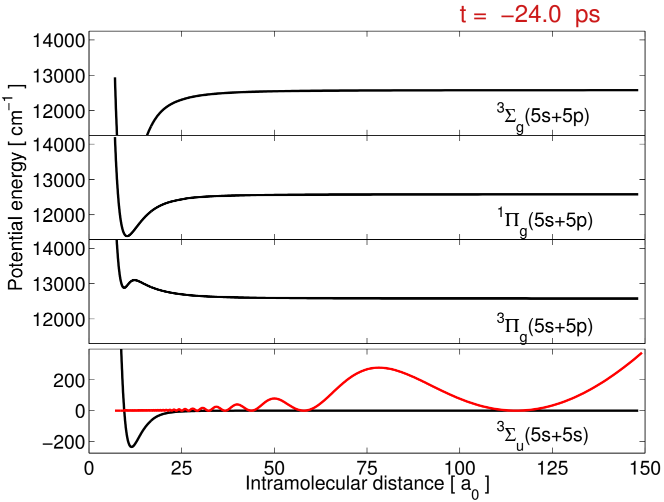

$$\Delta_P = 4.1\,\mathrm{cm}^{-1}$$

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

# dynamics with resonant SO coupling

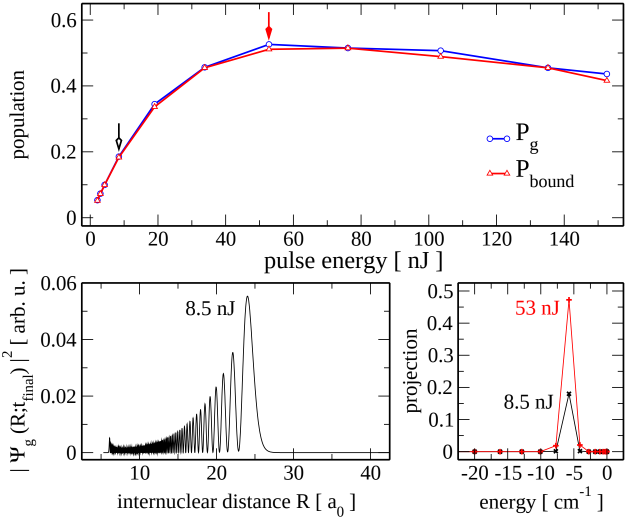


# dynamics w/o resonant SO coupling



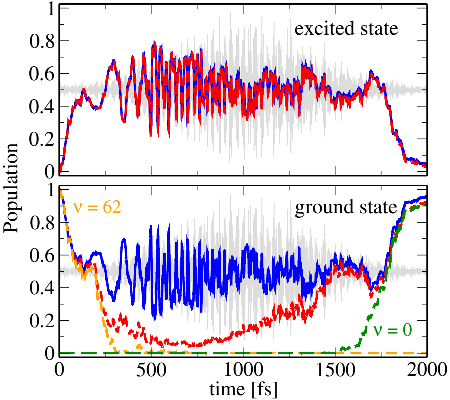
# stabilization to the ground state

with resonant spin-orbit coupling



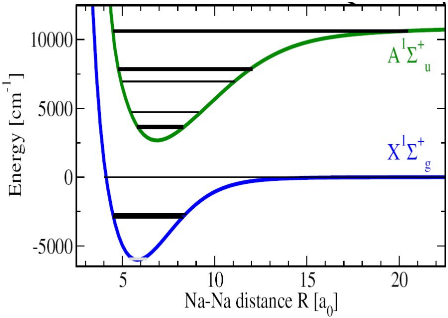
# route to $v = 0$ (generic case) : OCT

## strong fields and / or many Raman transitions




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

*potentials: Tiemann group PRA (2000), Z Phys D (1996)*




→ **required pulse energy ∼ 4 mJ**

*Koch, 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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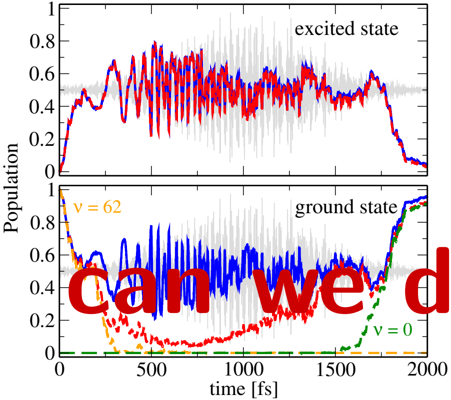


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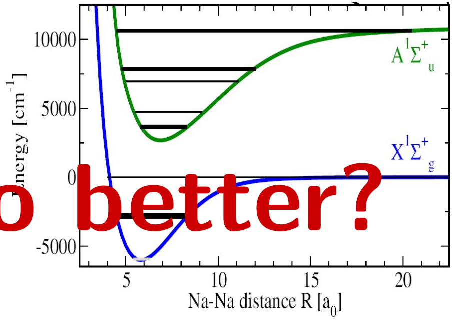



**can we do better?**

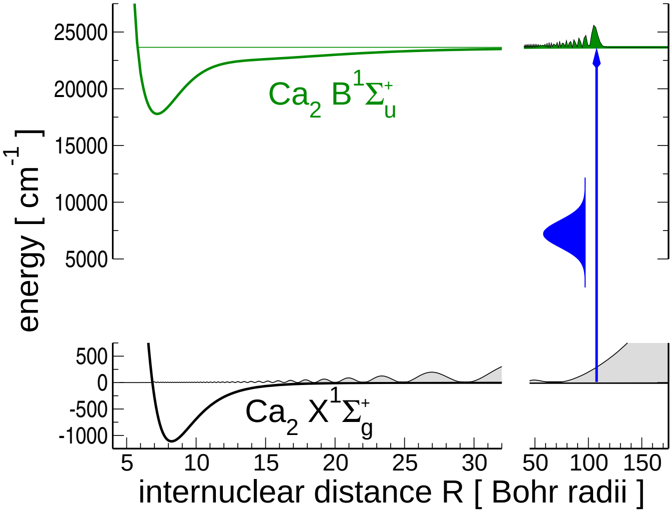
→ **required pulse energy $\sim 4\,$mJ**

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

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

# field-induced resonant coupling

*in collaboration with R. Moszyński*



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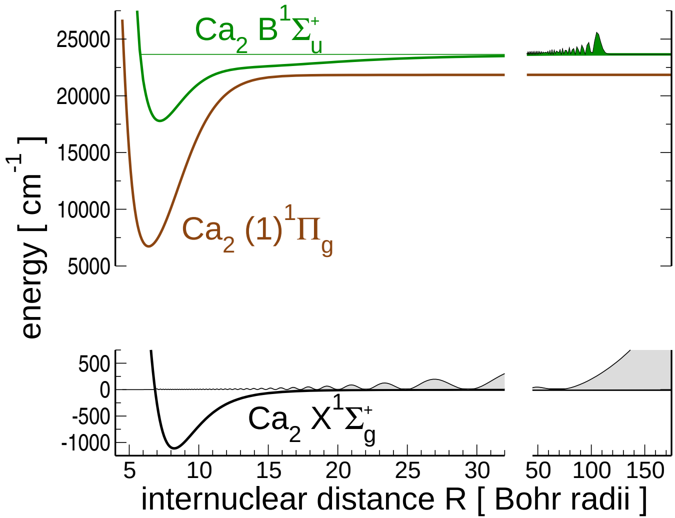

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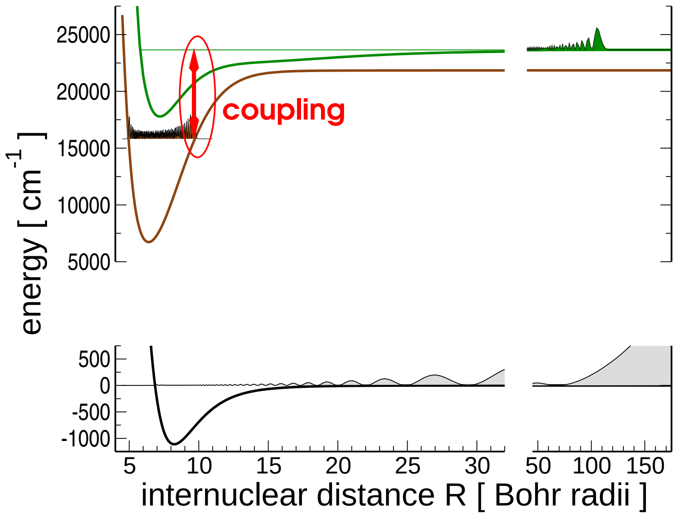

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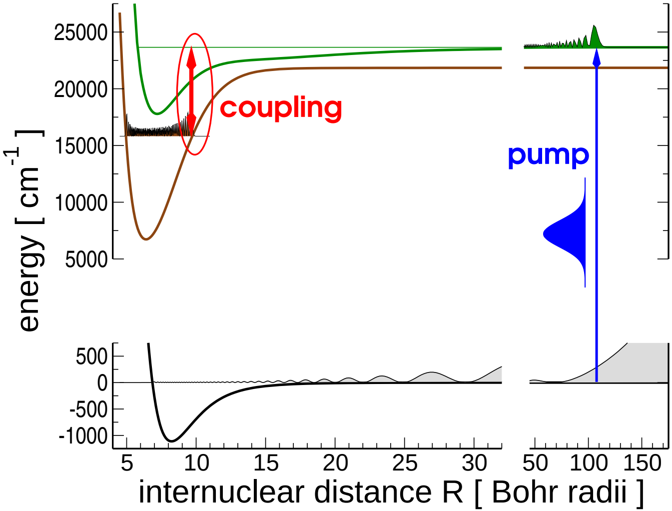

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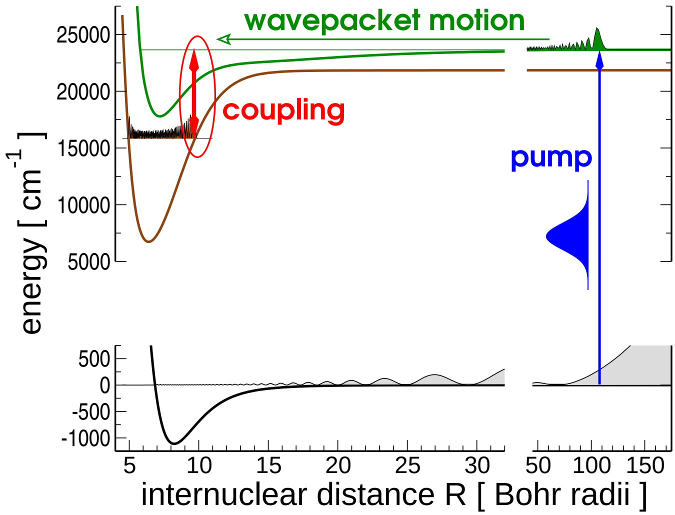

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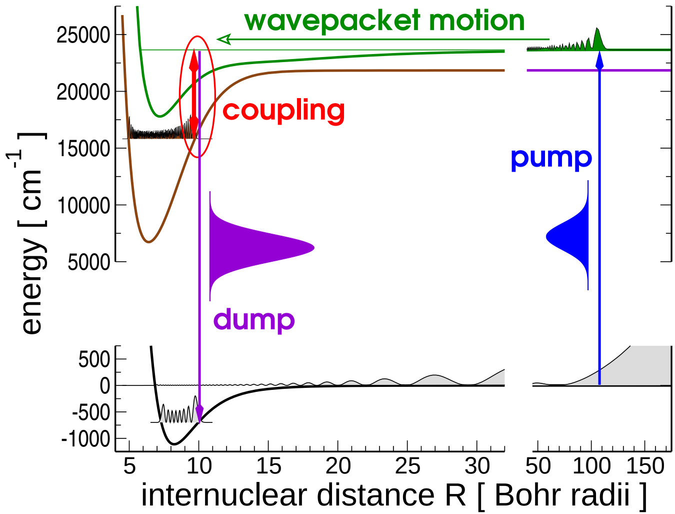

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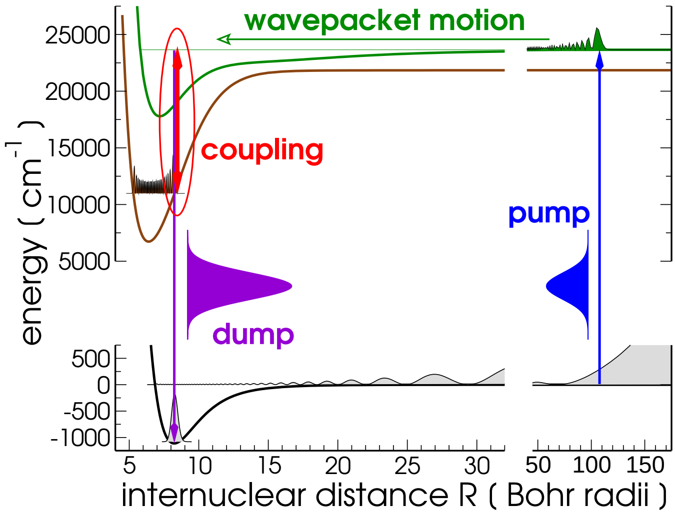

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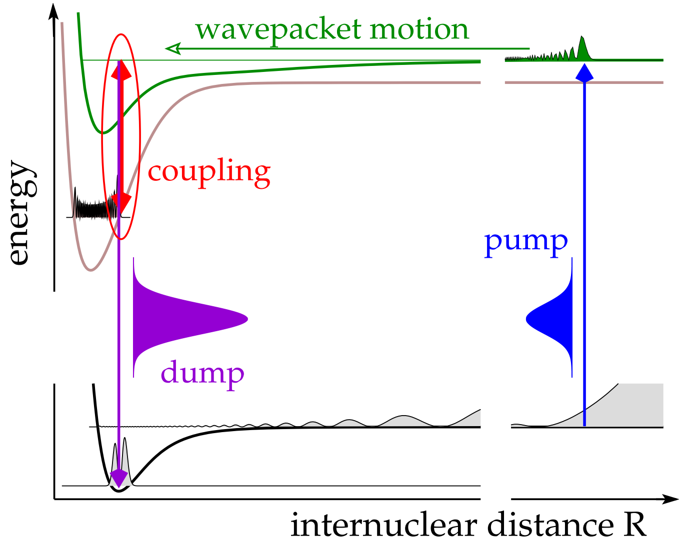
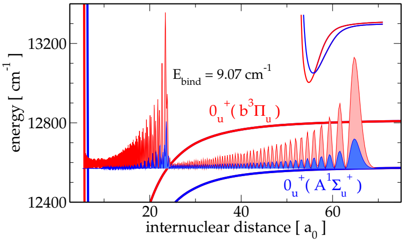


# when is the coupling resonant?

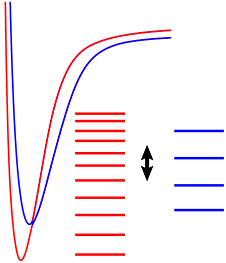



$$\hat{\mathbf{H}} = \begin{pmatrix} \hat{\mathbf{T}} + V_1(\hat{\mathbf{R}}) & \hat{\mathbf{W}} \\ \hat{\mathbf{W}} & \hat{\mathbf{T}} + V_2(\hat{\mathbf{R}}) \end{pmatrix}$$

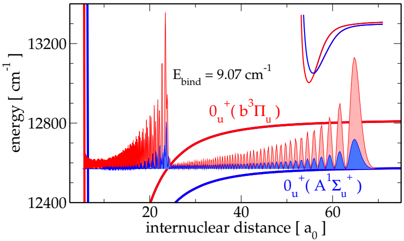
spin-orbit coupling:

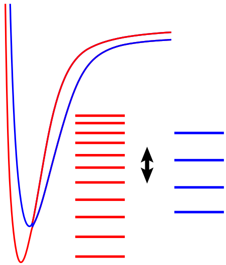
$$\hat{\mathbf{W}} = V_{SO}(\hat{\mathbf{R}}) \longrightarrow 237.6\,\mathrm{cm}^{-1}$$

for Rb



# when is the coupling resonant?



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

spin-orbit coupling:

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

for Rb

field-induced coupling:

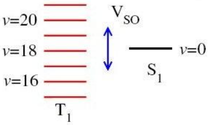
$$\hat{\mathbf{W}} = \hbar\Omega = \frac{1}{2}\mu(\hat{\mathbf{R}}) \cdot E(t)$$

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

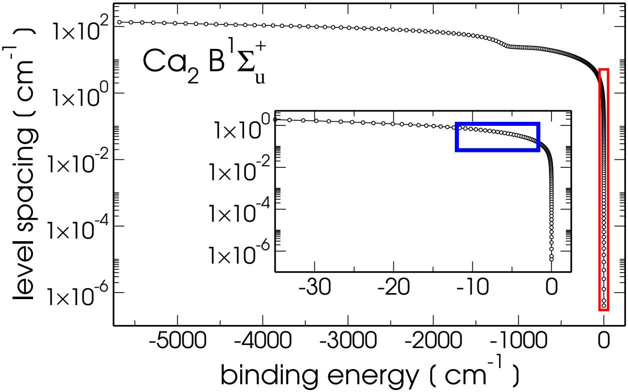
then $E_0 \sim 1.0 \times 10^7$ V/cm

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

# res. coupling & photoassociation




*coupling needs to be comparable to level spacings*



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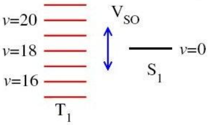


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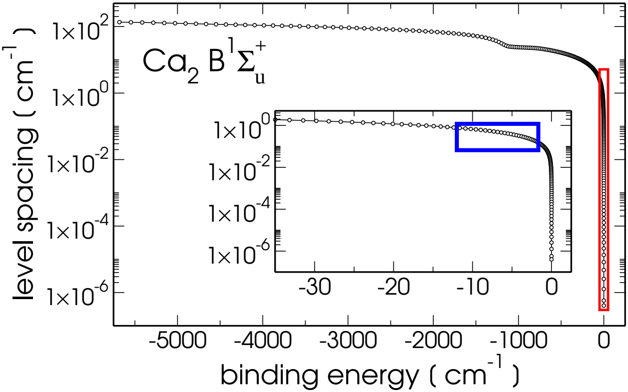
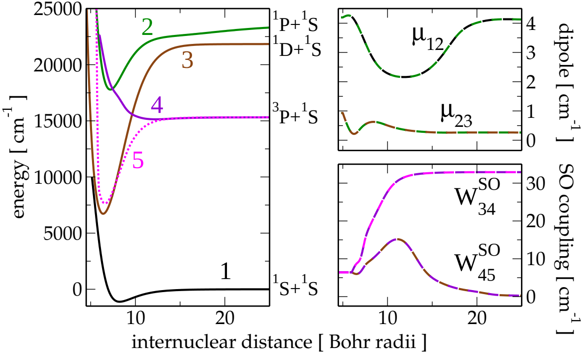



**level spacings drop to $\sim 1\,\mathrm{cm}^{-1}$ in range of PA detunings**

# minimal model for $Ca_2$

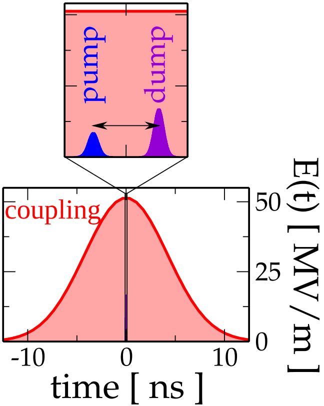
$$\hat{\mathbf{H}}_{\mathrm{Ca}_2} = \begin{pmatrix} \hat{\mathbf{H}}_{X^1\Sigma_g^+(^1S+^1S)} & \mu_{12}(\hat{\mathbf{R}})\cdot E_1(t) & 0 & 0 & 0 \\ \mu_{12}(\hat{\mathbf{R}})\cdot E_1^*(t) & \hat{\mathbf{H}}_{B^1\Sigma_u^+(^1P+^1S)} & \mu_{23}(\hat{\mathbf{R}})\cdot E_2(t) & 0 & 0 \\ 0 & \mu_{23}(\hat{\mathbf{R}})\cdot E_2^*(t) & \hat{\mathbf{H}}_{(1)^1\Pi_g(^1D+^1S)} & \xi(\hat{\mathbf{R}}) & 0 \\ 0 & 0 & \xi(\hat{\mathbf{R}}) & \hat{\mathbf{H}}_{(1)^3\Sigma_g^+(^3P+^1S)} & \zeta(\hat{\mathbf{R}}) \\ 0 & 0 & 0 & \zeta(\hat{\mathbf{R}}) & \hat{\mathbf{H}}_{(1)^3\Pi_g(^3P+^1S)} \end{pmatrix}$$

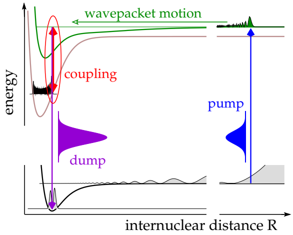


# choice of coupling laser

$I = 3.5 \times 10^8$ W/cm$^2$ $-3.2 \times 10^9$ 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, submitted

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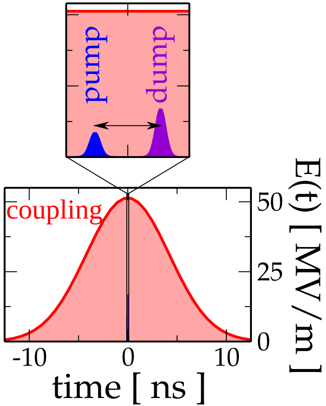
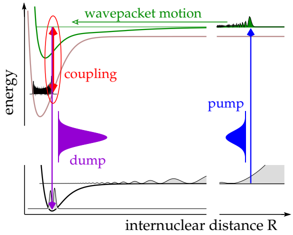


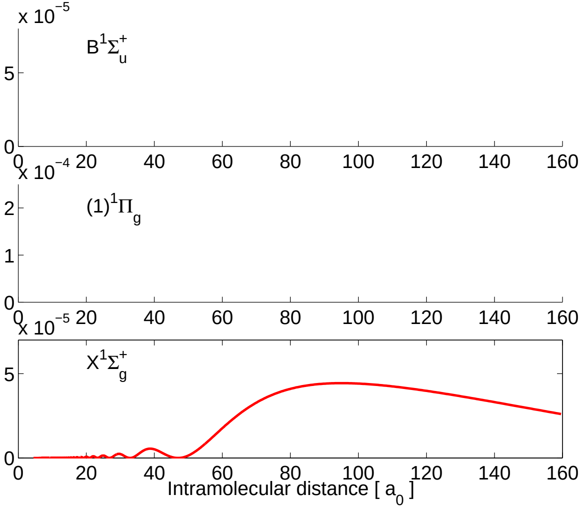

**10 ns pulse is constant on timescale of 100 ps**

⟶ **feasible & robust**

CPK & Moszyński, submitted

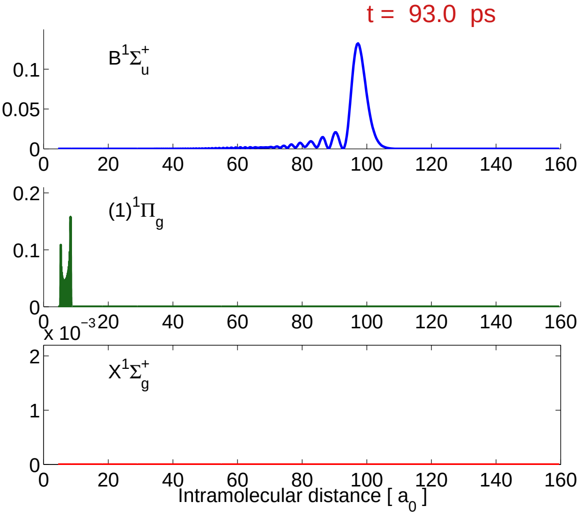
# dynamics w/ induced res. coupling

## photoassociation (pump) pulse

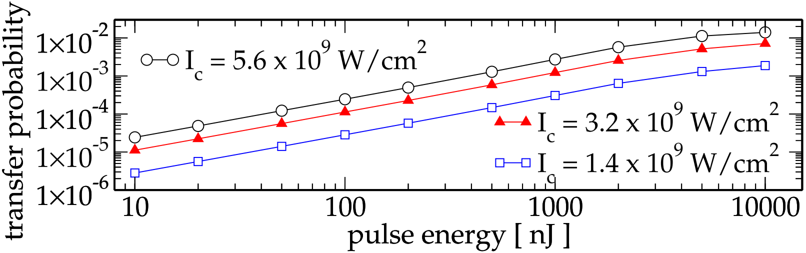


# dynamics w/ induced res. coupling

## stabilization (dump) pulse

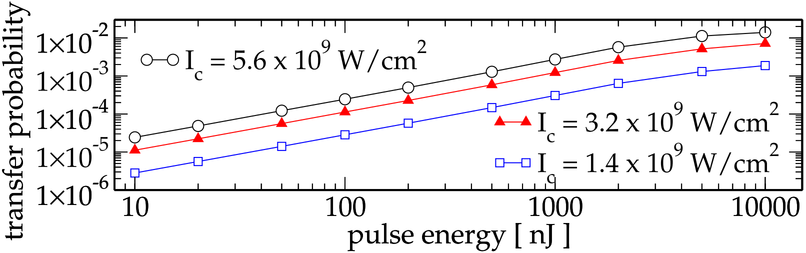


# how many molecules?




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

# how many molecules?

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**accumulate molecules over many pump-dump cycles**

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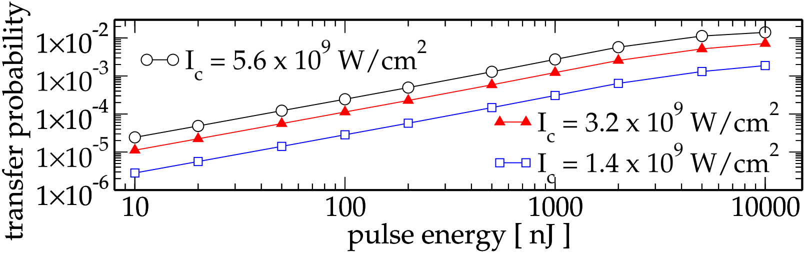


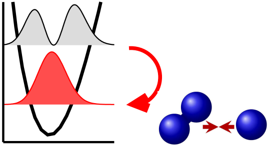

typical MOT conditions: $N_{mol} = 12.5$, 10 kHz rep.rate: 1 mol/ms

**accumulate molecules over many pump-dump cycles**

**employ dissipation to achieve unidirectionality**

collisional decay to $v = 0$ within 1 ms if

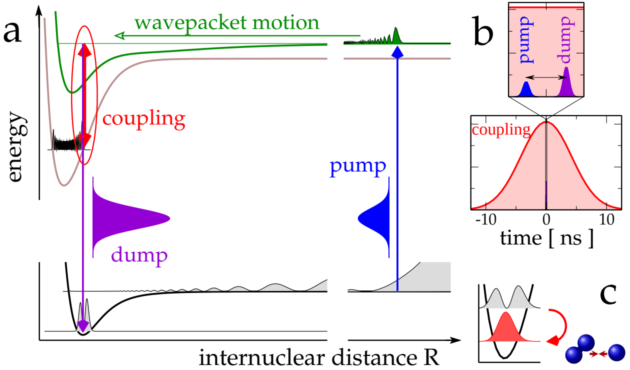
$\rho \sim 10^{-13}\,\mathrm{cm}^{-3}$



**can be improved: flux enhancement & speed up of decay**

# field-induced resonant coupling

## 'shaping' the potentials




➔ **qualitative & substantial change of dynamics**

➔ **implementing resonance phenomenon of cold molecules via coherent control**

# acknowledgements


## people

*work on alkalis*

- **Françoise Masnou-Seeuws & Eliane Luc-Koenig, Orsay (France)**
- **Ronnie Kosloff, Jerusalem (Israel)**

*work on $Ca_2$*

- **Robert Moszyński, Warsaw (Poland)**

## funding

- Deutsche Forschungsgemeinschaft

Emmy Noether-Programm
Deutsche Forschungsgemeinschaft
DFG

# example 3:
# quantum information with ultracold molecules

# quantum information

**= squaring the circle**

# quantum information

= squaring the circle

particles with no interaction
➔ little decoherence

controlled interaction
➔ two-qubit gates

scalability

# quantum information

**= squaring the circle**

**particles with no interaction**
**➔ little decoherence**

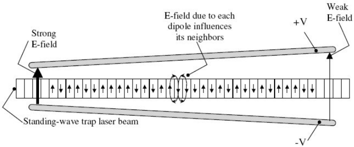
**controlled interaction**
**➔ two-qubit gates**

**scalability**

## polar molecules in periodic arrangements

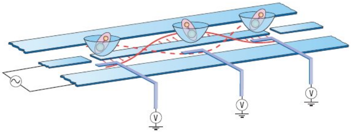
**polar molecules = dipole-dipole interaction**

in a static electric field




DeMille, PRL 88, 067901 (2002)

electrostatic trap on a chip



Côté, Nat. Phys. 2, 583 (2006)

# coherent control: shielding

PRL **101**, 073201 (2008) PHYSICAL REVIEW LETTERS week ending 15 AUGUST 2008

**Suppression of Inelastic Collisions Between Polar Molecules With a Repulsive Shield**

A. V. Gorshkov,[1] P. Rabl,[1,2] G. Pupillo,[3,4] A. Micheli,[3,4] P. Zoller,[3,4] M. D. Lukin,[1,2] and H. P. Büchler[5]

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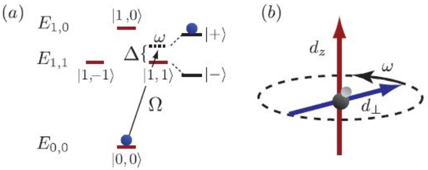
$$H = \frac{\mathbf{P}^2}{4m} + \frac{\mathbf{p}^2}{m} + \frac{\mathbf{d}_1\mathbf{d}_2 - 3(\mathbf{d}_1\hat{\mathbf{r}})(\mathbf{d}_2\hat{\mathbf{r}})}{r^3} + \sum_{i=1}^{2} H_{\mathrm{rot}}^{(i)}, \qquad (1)$$




$$H_{\mathrm{rot}}^{(i)} = B\mathbf{J}_i^2 - \mathbf{d}_i \cdot \mathbf{E}_{\mathrm{dc}} - \mathbf{d}_i \cdot \mathbf{E}_{\mathrm{ac}}(t)$$

**cancel dipole-dipole interaction with external field control in rotational Hamiltonian**

# coherent control: shielding

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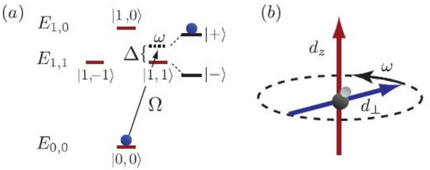


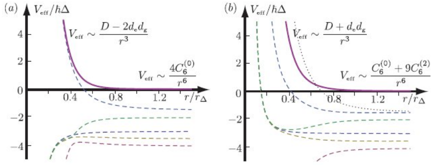




FIG. 2 (color online). Born-Oppenheimer potentials in the limit $r \gg r_B$: (a) $\theta = 0$ and (b) $\theta = \pi/2$. The effective potential $V_{\mathrm{eff}}(\mathbf{r})$ (solid line) is repulsive for all angles $\theta$. The dotted line denotes the antisymmetric level relevant during a three-body collision.

$$H_{\mathrm{rot}}^{(i)} = B\mathbf{J}_i^2 - \mathbf{d}_i \cdot \mathbf{E}_{\mathrm{dc}} - \mathbf{d}_i \cdot \mathbf{E}_{\mathrm{ac}}(t)$$

**cancel dipole-dipole interaction with external field control in rotational Hamiltonian**

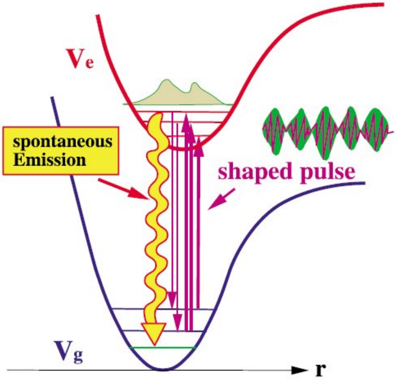
# example 4:
# vibrational cooling of ultracold molecules

# vibrational cooling – theory

**Laser cooling of internal degrees of freedom of molecules by dynamically trapped states**

David J. Tannor,[a] Ronnie Kosloff[b] and Alon Bartana[b]

*Faraday Discuss.*, 1999, **113**, 365–383

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Chemical Physics 267 (2001) 195–207

Chemical Physics

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## Laser cooling of molecules by dynamically trapped states

Allon Bartana [a], Ronnie Kosloff [a,*], David J. Tannor [b]

[a] *Department of Chemistry and the Fritz Haber Institute for Molecular Dynamics, Hebrew University of Jerusalem, 91904 Jerusalem, Israel*

[b] *Department of Chemical Physics, Weizmann Institute of Science, 76100 Rehovot, Israel*

Received 30 August 2000

**Abstract**

Optimal control theory (OCT) is applied to laser cooling of molecules. The objective is to cool vibrations, using shaped pulses synchronized with the spontaneous emission. An instantaneous in time optimal approach is compared to solution based on OCT. In both cases the optimal mechanism is found to operate by a "vibrationally selective coherent population trapping". The trapping condition is that the instantaneous phase of the laser is locked to the phase of the transition dipole moment of $v = 0$ with the excited population. The molecules that reach $v = 0$ by spontaneous emission are then trapped, while the others are continually repumped. For vibrational cooling to $v = 2$ and rotational cooling, a different mechanism operates. The field completely changes the transient eigenstates of the Hamiltonian creating a superposition composed of many states. Finally this superposition is transformed by the field to the target energy eigenstate. 

# vibrational cooling – theory

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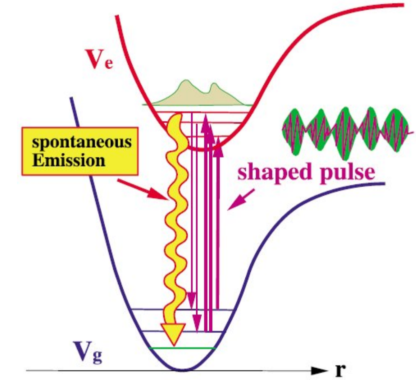


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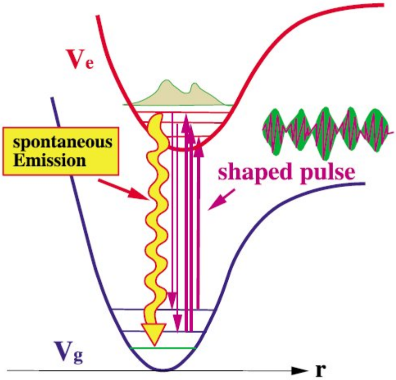


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$$\hat{\rho} = \hat{\rho}_{\mathrm{g}} \otimes \widehat{\mathbf{P}}_{\mathrm{g}} + \hat{\rho}_{\mathrm{e}} \otimes \widehat{\mathbf{P}}_{\mathrm{e}} + \hat{\rho}_{\mathrm{c}} \otimes \widehat{\mathbf{S}}_{+} + \hat{\rho}_{\mathrm{c}}^{\dagger} \otimes \widehat{\mathbf{S}}_{-} = \begin{pmatrix} \hat{\rho}_{\mathrm{e}} & \hat{\rho}_{\mathrm{c}} \\ \hat{\rho}_{\mathrm{c}}^{\dagger} & \hat{\rho}_{\mathrm{g}} \end{pmatrix}, \tag{2.1}$$

$$\frac{\partial \hat{\rho}}{\partial t} = -\frac{\mathrm{i}}{\hbar}[\widehat{\mathbf{H}}, \hat{\rho}] + \mathscr{L}_D(\hat{\rho}), \tag{2.2}$$

$$\mathscr{L}_D(\hat{\rho}) = \widehat{\mathbf{F}}\hat{\rho}\widehat{\mathbf{F}}^{\dagger} - \frac{1}{2}\{\widehat{\mathbf{F}}^{\dagger}\widehat{\mathbf{F}}, \hat{\rho}\}, \tag{2.5}$$

# vibrational cooling – theory

## the cooling target

### dark ground state

$$\frac{\mathrm{d}\langle |k\rangle\langle k| \otimes \widehat{\mathbf{P}}_{\mathrm{g}}\rangle}{\mathrm{d}t} = \frac{\mathrm{i}}{\hbar}\langle[\widehat{\mathbf{H}}, |k\rangle\langle k| \otimes \widehat{\mathbf{P}}_{\mathrm{g}}]\rangle + \langle \mathscr{L}_D^{\dagger}(|k\rangle\langle k| \otimes \widehat{\mathbf{P}}_{\mathrm{g}})\rangle. \tag{3.1}$$

$$2\,\mathrm{Imag}\{\langle \hat{\mu}|k\rangle\langle k| \otimes \widehat{\mathbf{S}}_{+}\rangle \cdot \epsilon(t)\} = 0. \tag{3.2}$$

$$2|\langle \hat{\mu}|k\rangle\langle k| \otimes \widehat{\mathbf{S}}_{+}\rangle||\epsilon(t)|\sin(\phi_{\mu|k\rangle\langle k|} + \phi_{\epsilon}) = 0. \tag{3.3}$$

### maximum excitation

$$\frac{\mathrm{d}\langle \widehat{\mathbf{P}}_{\mathrm{g}}\rangle_{H}}{\mathrm{d}t} = 2\,\mathrm{Imag}\{\langle \hat{\mu} \otimes \widehat{\mathbf{S}}_{+}\rangle \cdot \epsilon(t)\}. \tag{3.5}$$

## solutions

# vibrational cooling – theory

## the cooling target

### dark ground state

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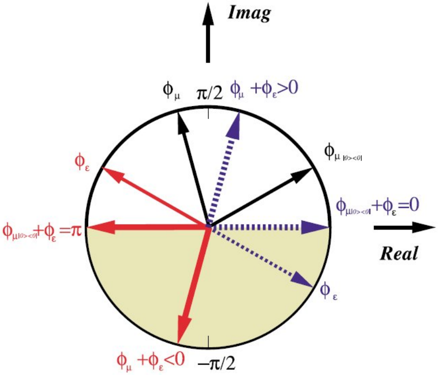
## solutions



# vibrational cooling – theory

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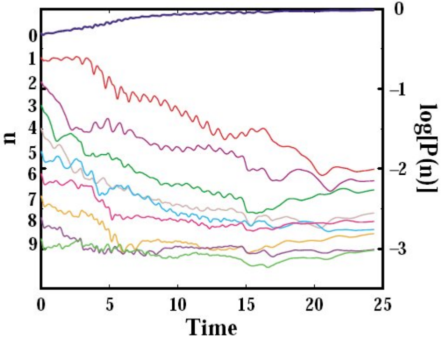
## solutions



# vibrational cooling – theory

## the cooling target

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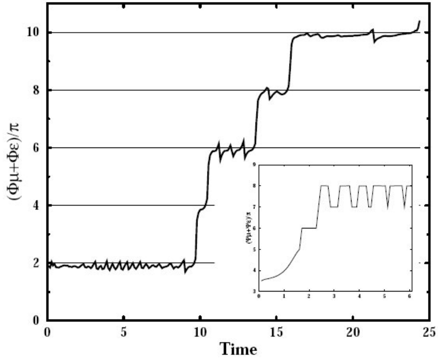
$$\frac{\mathrm{d}\langle \widehat{\mathbf{P}}_{\mathrm{g}}\rangle_H}{\mathrm{d}t} = 2\,\mathrm{Imag}\{\langle \hat{\mu} \otimes \widehat{\mathbf{S}}_{+}\rangle \cdot \epsilon(t)\}. \tag{3.5}$$

## solutions



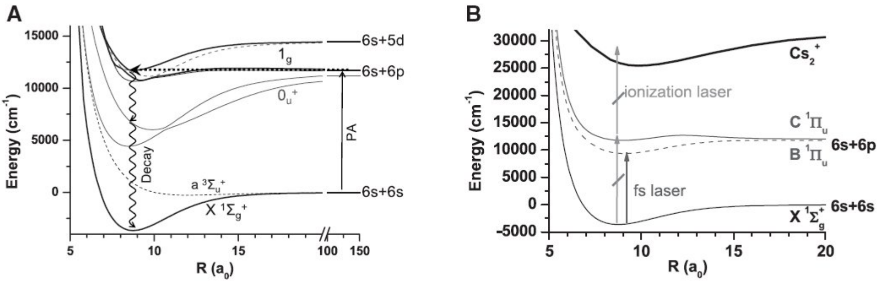

**cooling = maintaining a dark ground state**

# vibrational cooling – exp.

## Optical Pumping and Vibrational Cooling of Molecules

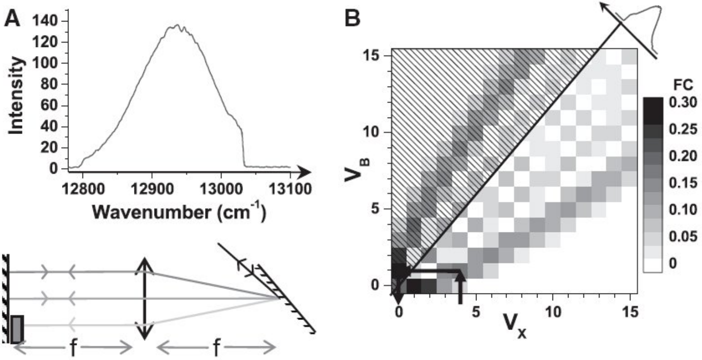
Matthieu Viteau,[1] Amodsen Chotia,[1] Maria Allegrini,[1,2] Nadia Bouloufa,[1] Olivier Dulieu,[1] Daniel Comparat,[1] Pierre Pillet[1*]

11 JULY 2008 VOL 321 SCIENCE www.sciencemag.org



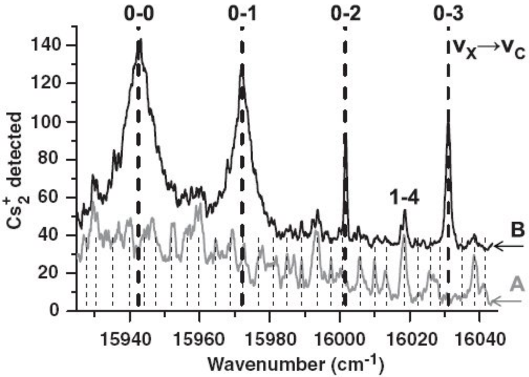
# vibrational cooling – exp.

## a crude way of maintaing a dark ground state . . .



# vibrational cooling – exp.

. . . **but it works!**



# summary

- **pump-dump photoassociation: initial state & timescales**
- shaping the potentials 1: mimic resonant coupling with an external field
- shaping the potentials 2: shield molecules from each other by manipulation with ac or dc electric fields
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$\longrightarrow$ ultracold & ultrafast = pretty cool