Introduction to optimal control theory

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Outline

0. Terminology
1. Intuitive control schemes and their experimental realization
2. Controllability of a quantum system
3. Variational approach to quantum control
4. Experimental quantum control: closed learning loops
5. Coherent / optimal control of cold systems?

0. Terminology

Quantum mechanics $\triangleq$ probabilistic, but *deterministic* theory

present wave function $\xrightarrow{\text{Schrödinger equation}}$ future wave function

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Quantum control:

Given an initial wave function at present,
which dynamics ($\triangleq$ which Hamiltonian)
guarantees a particular outcome in the future?

Optimal / Coherent Control

wave properties of atoms/molecules
(Superposition principle)

variation of phase between different, but indistinguishable
quantum pathways

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↓

constructive interference in desired channel

destructive interference in all other channels

Coherent Control

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- Goal: improve outcome of process

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- vary some parameters

$\Downarrow$

simple, intuitive schemes

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- tune "all" available parameters

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complex outcome $\longrightarrow$ discovery of new schemes (usually not accessible by intuition)

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in time or in frequency domain

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complex outcome $\longrightarrow$ discovery of new schemes (usually not accessible by intuition)

in time / frequency "phase space"

global or local in time

1. Intuitive control schemes
&
their experimental realization

Control in frequency domain: Brumer-Shapiro scheme

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analogous to double slit experiment

but IF between quantum pathways

n≥ 2 lasers $\longrightarrow$ n≥ 2 coherent pathways from initial to $degenerate$ final states

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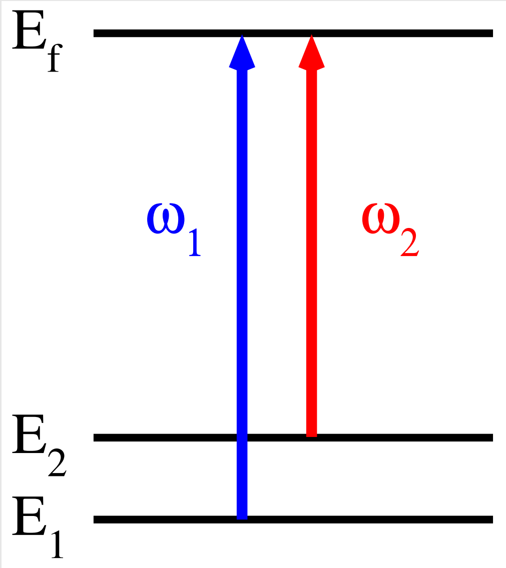
Example: $n = 2$

$$|\Psi(t=0)\rangle = c_1|1\rangle + c_2|2\rangle$$

superposition of 2 eigenstates

n$\geq$ 2 lasers → n$\geq$ 2 coherent pathways from initial to $degenerate$ final states



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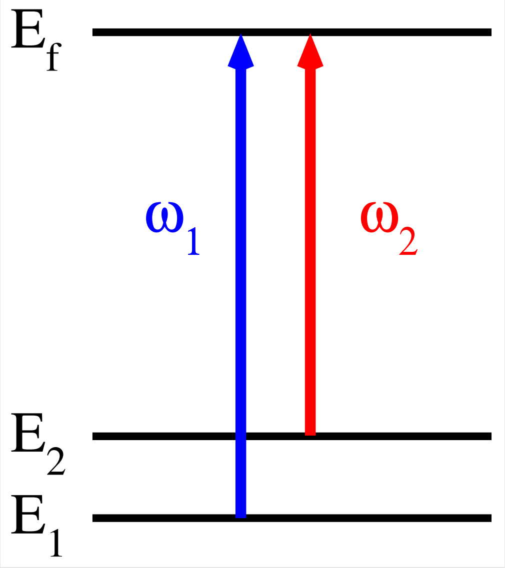


coefficients of final state superposition determined by relative phase and relative amplitude of CW lasers and initial state

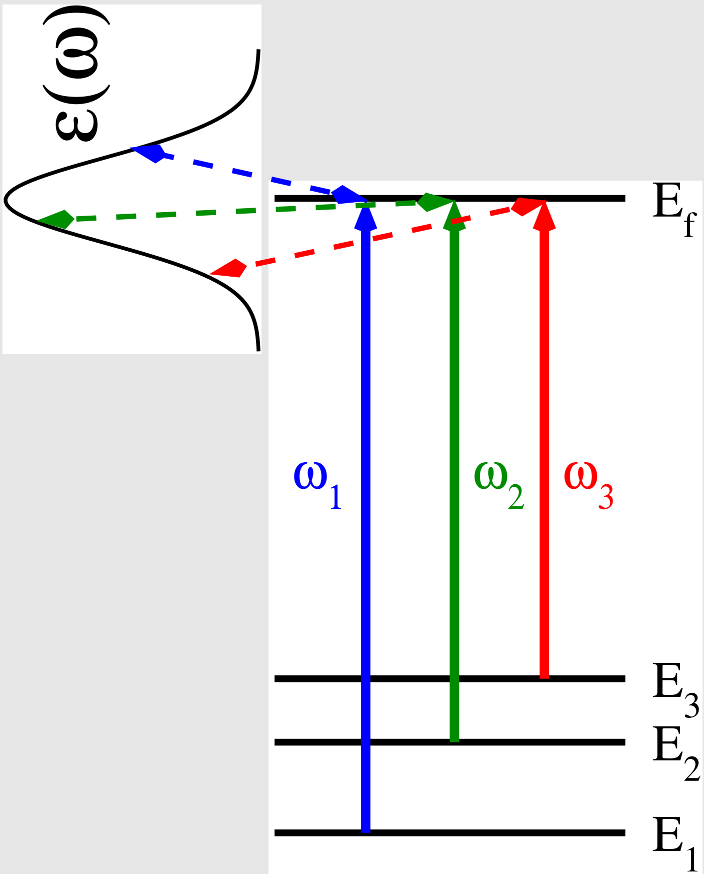
Brumer-Shapiro scheme

With pulsed lasers

1 photon vs. 3 photon absorption

Brumer-Shapiro scheme

With pulsed lasers



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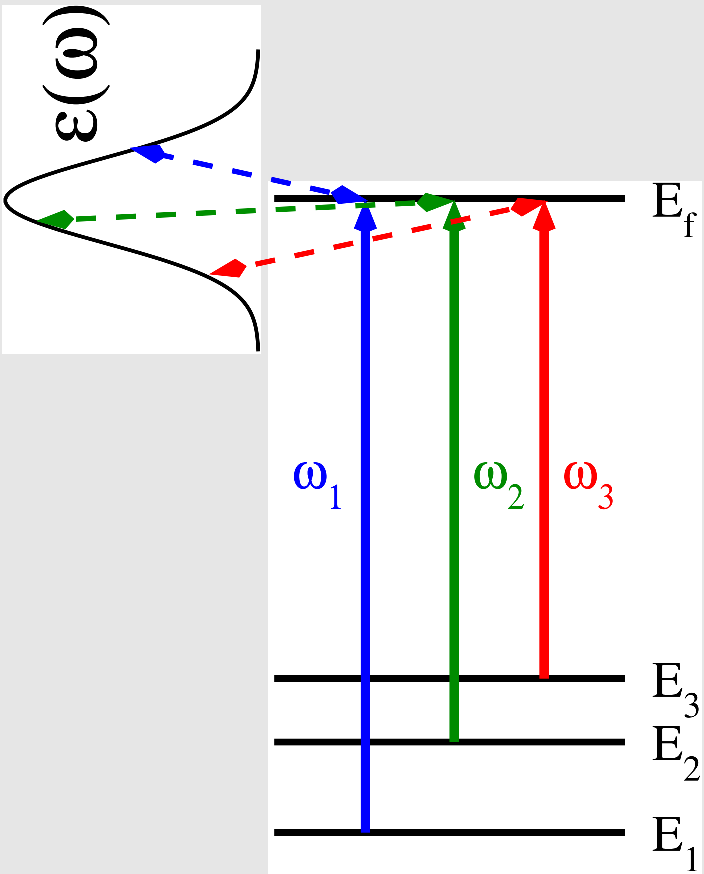

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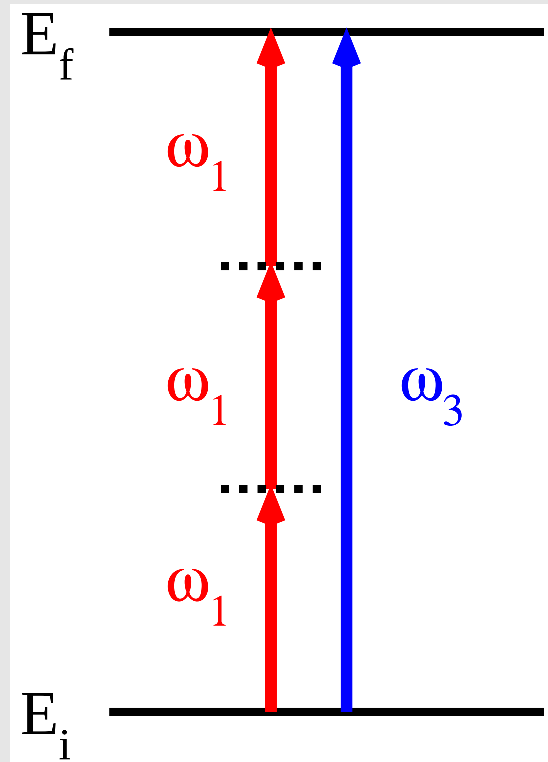

1 photon vs. 3 photon absorption: selective photodissociation of HI molecule

$$\mathrm{HI} \longrightarrow \begin{cases} \mathrm{HI}^{+} + \mathrm{e}^{-} \\ \mathrm{H} + \mathrm{I} \end{cases}$$

L. Zhu, V. Kleiman, X. Li, S. P. Lu, K. Trentelman, R. J. Gordon
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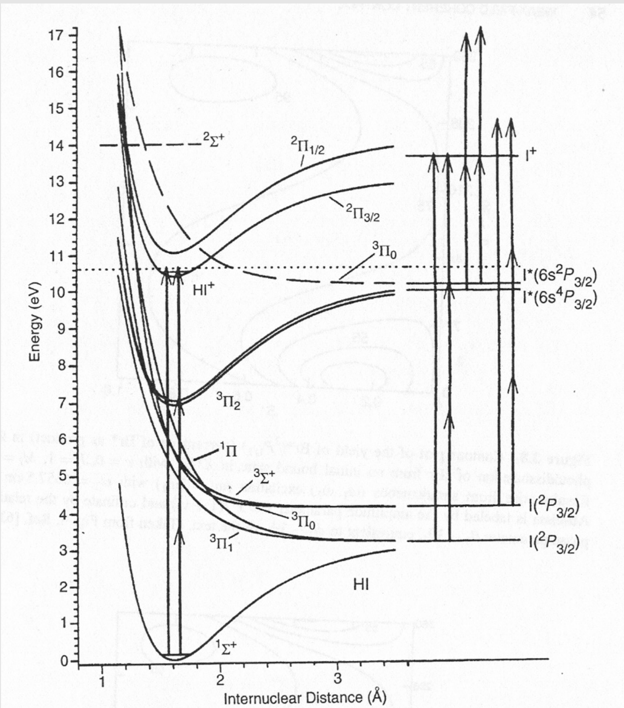

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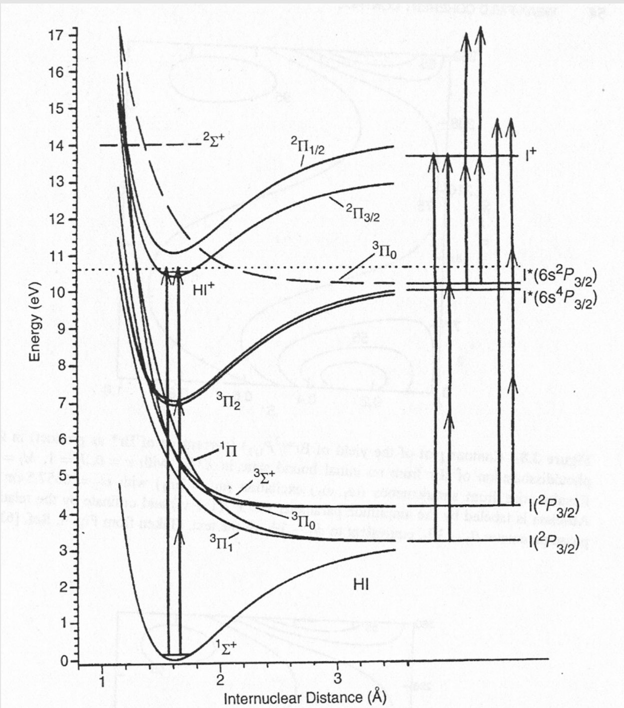

background gas pressure $\longrightarrow$ different refractive indices

changing pressure $\triangleq$ changing phase difference between 1-photon & 3-photon fields

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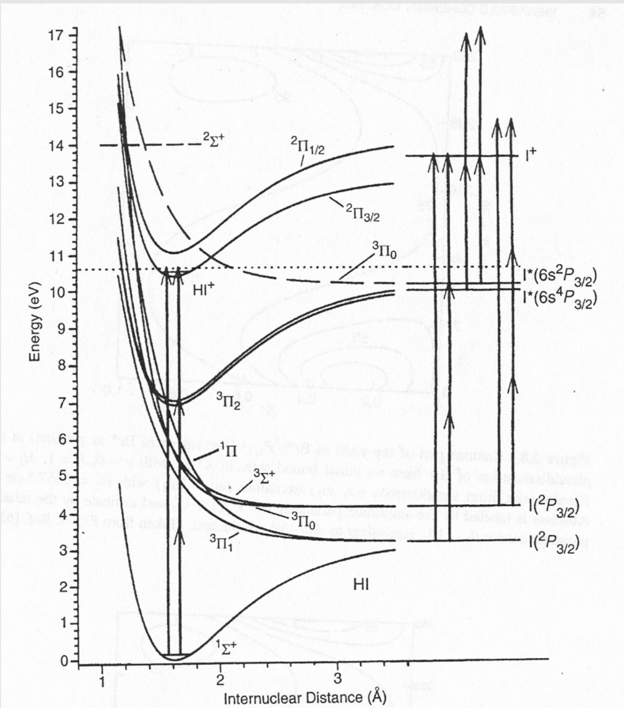

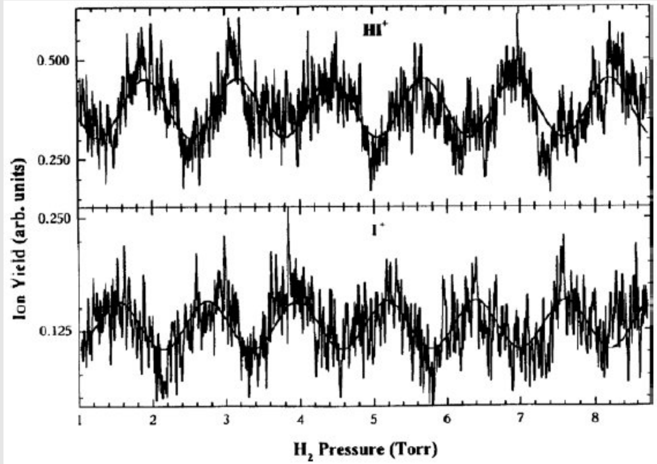

Figure 5. Modulation of the HI+ and I+ signals as a function of the difference between the one- and three-photon phases (proportional to the H_2 pressure in the cell used to phase shift the beams).

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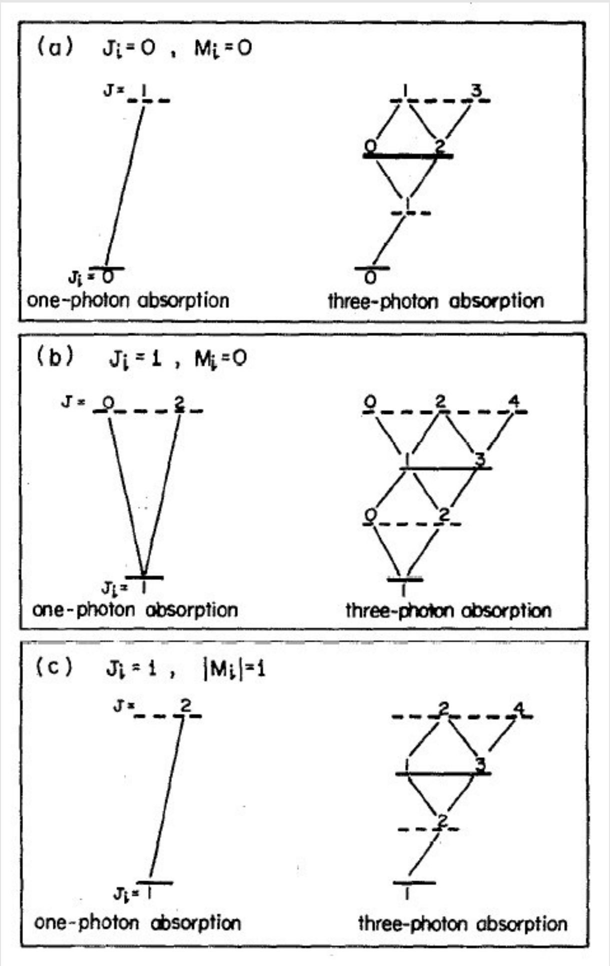
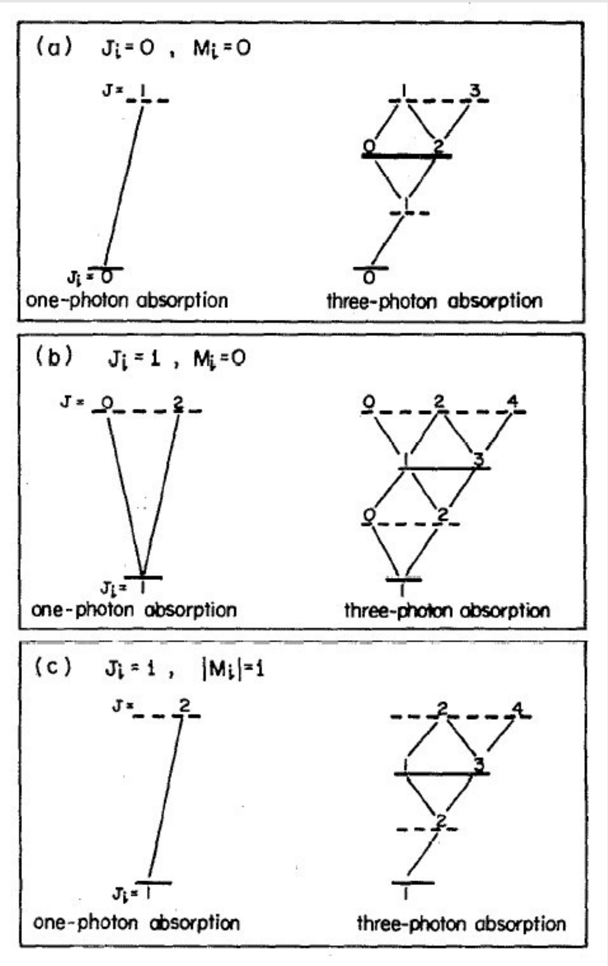


1 photon vs. 3 photon absorption

What about rotations?

- superposition of angular momentum ladders
- *uncontrollable* satellites

M. Shapiro & P. Brumer, *Principles of Quantum Control of Molecular Processes*, Wiley 2003.



Control in the time domain: Tannor-Rice scheme

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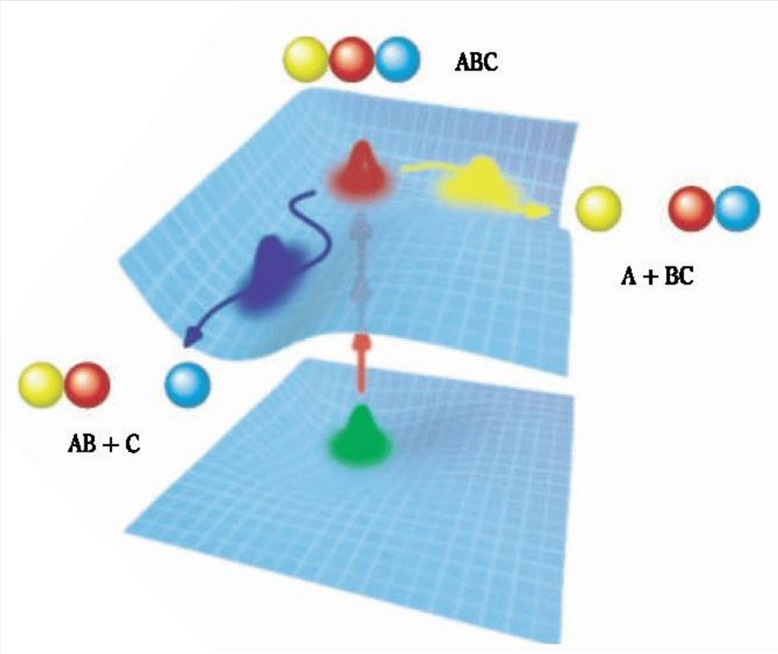

T. Brixner & G. Gerber, Physikal. Blätter April 2001

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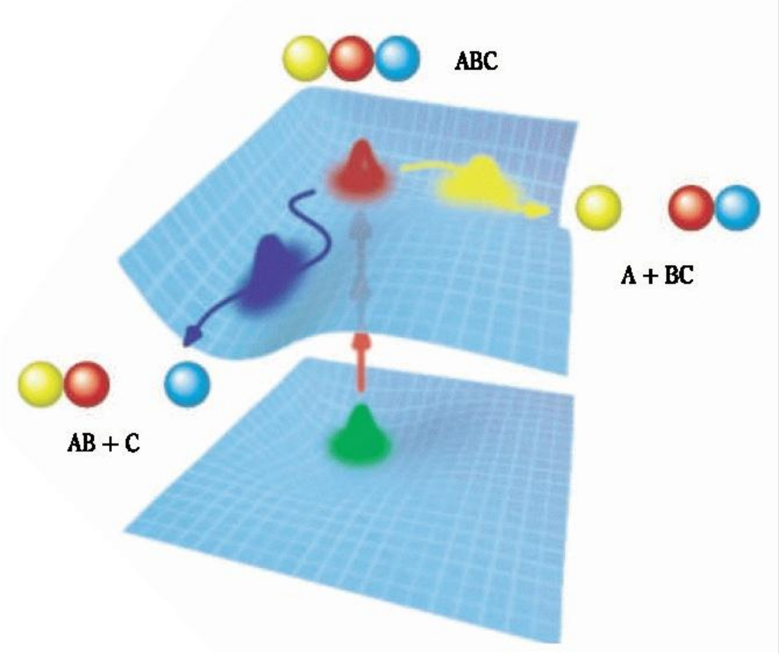
variation of phase between quantum paths $\triangleq$ variation of time delay



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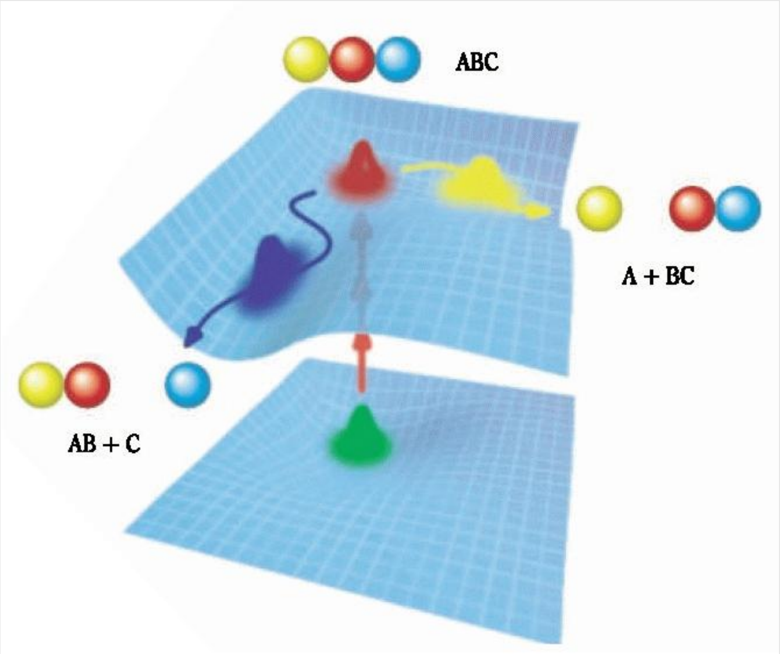
Pump-dump control



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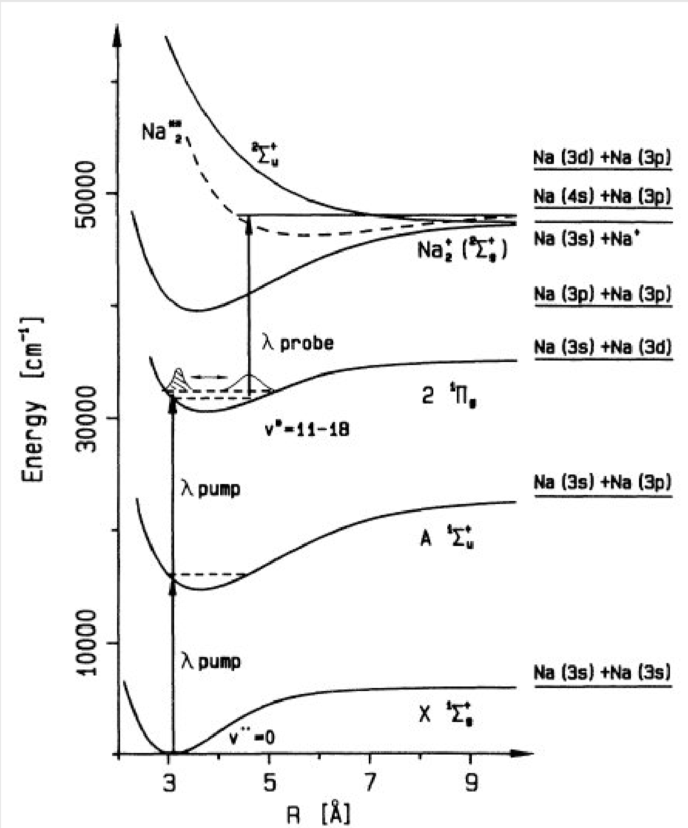
Pump-probe control



competition between ionization & dissociative ionization of Na_2

T. Baumert, M. Grosser, R. Thalweiser, and G. Gerber, *PRL* **67**, 3753 (1991)

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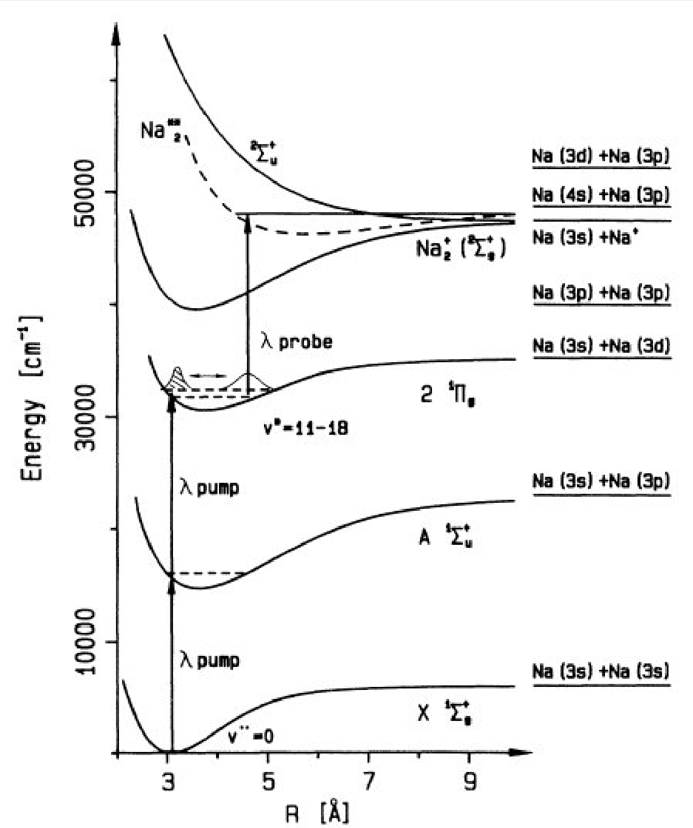

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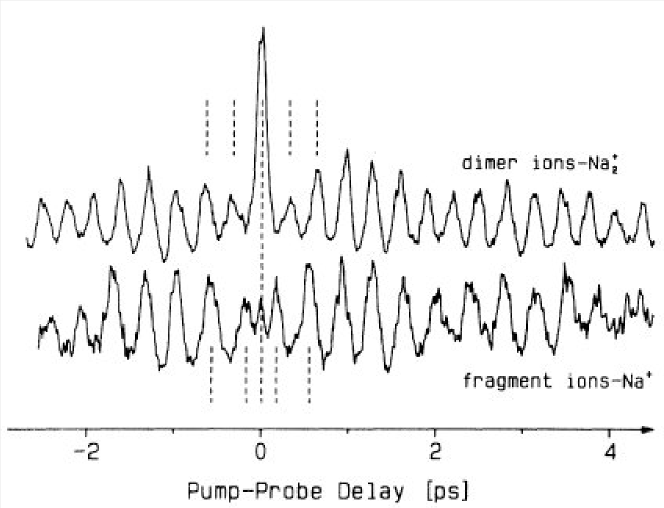

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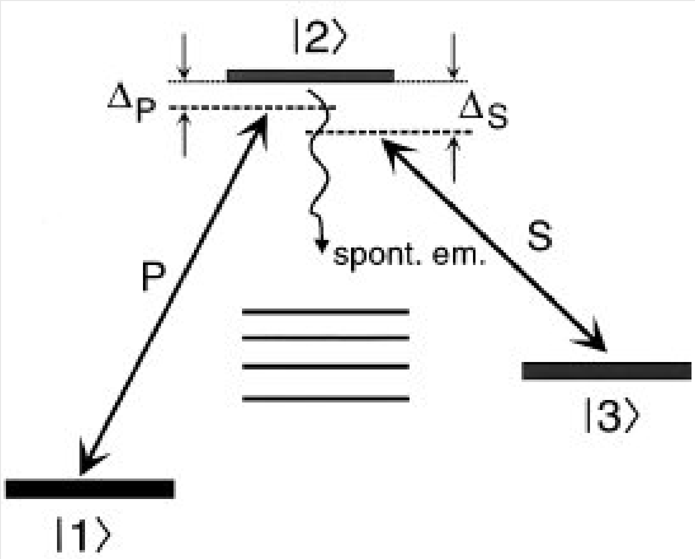
Control via STIRAP



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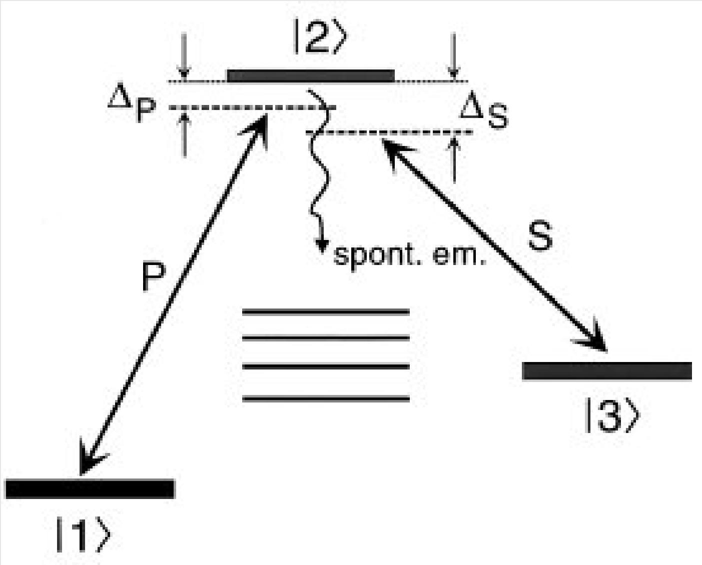

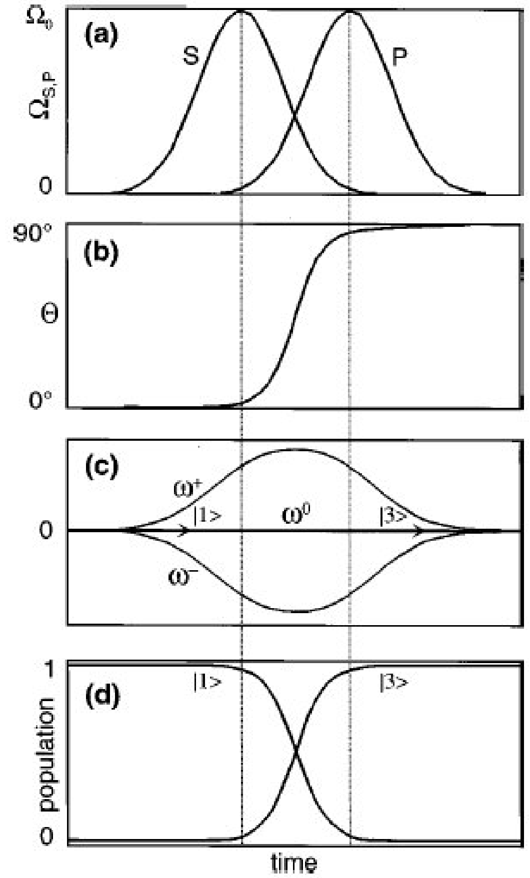

FIG. 3. Time evolution of (a) the Rabi frequencies of the pump and Stokes laser (see Fig. 1); (b) the mixing angle [see Eq. (9)]; (c) the dressed-state eigenvalues [see Eq. (10)]; and (d) the population of the initial level (starting at unity) and the final level (reaching unity).

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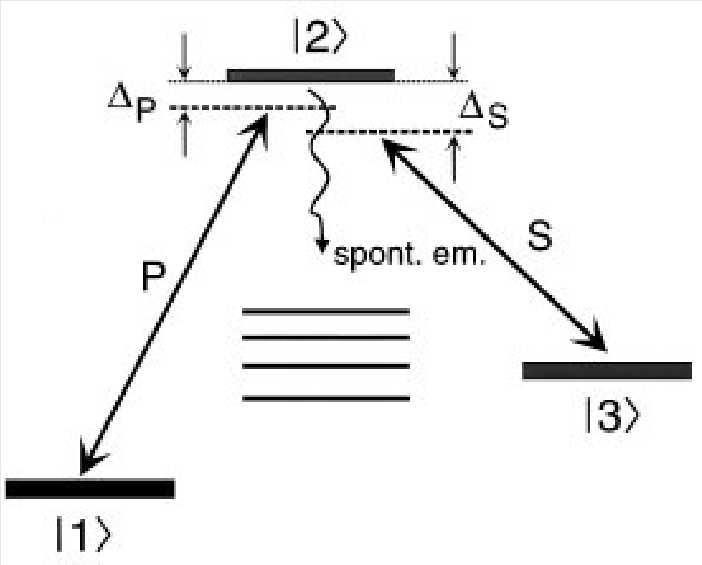
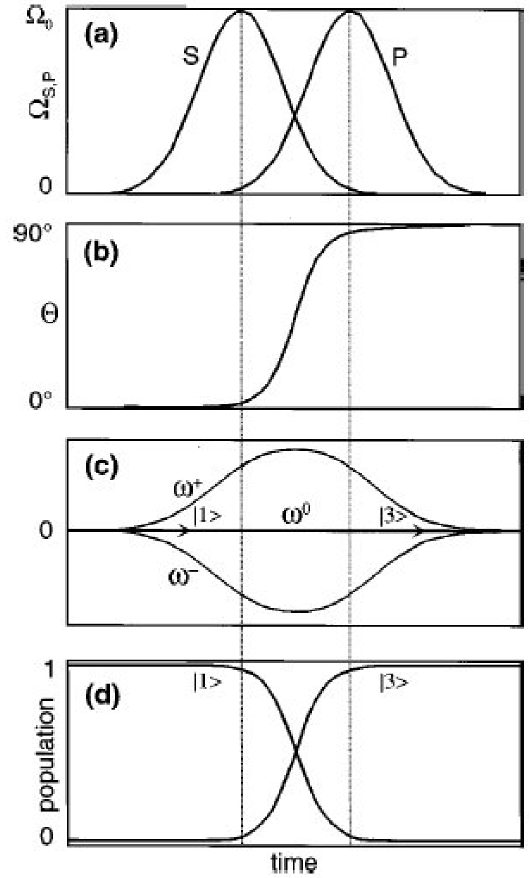


Counterintuitive pulse sequence: S precedes, but overlaps P

⬇

coherent population trapping

K. Bergmann, H. Theuer, and B. W. Shore, *Rev. Mod. Phys.* **70**, 1003 (1998)



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Optimal Control Theory

learning loop experiments

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Optimal Control Theory

learning loop experiments

2. Controllability of a quantum system

Theorem of controllability

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$$\boxed{i\hbar \frac{\mathsf{d}}{\mathsf{d}t}\psi(t) = \left[\hat{\mathbf{H}}_0 + \sum_l \varepsilon_l(t)\hat{\mathbf{V}}_l\right]\psi(t)}$$

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- $\hat{\mathbf{H}}_0$ has a *finite* spectrum
- state-to-state transitions

$\longrightarrow$ It is always possible to completely control evolution from initial to target state

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Controllability retained for system w/ infinitely many states by adjusting an external field (ion trap)

s.t. system separated into finite and infinite subsystem

C. Rangan, A. M. Bloch, C. Monroe, P. H. Bucksbaum, *PRL* **92**, 113004 (2004)

Optimal control theory

Optimal control theory

time/frequency "phase space" picture

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

$$|\varphi_i\rangle \quad \rightsquigarrow \quad |\varphi_f\rangle$$

Inverse problem:
Given the target and the equations of motion,
calculate the field

Optimal control theory

time/frequency "phase space" picture

$$\begin{array}{ccc} t=0 & & t=T \\ |\varphi_i\rangle & \rightsquigarrow & |\varphi_f\rangle \end{array}$$

$$\boxed{\begin{array}{c}\text{Inverse problem:} \\ \text{Given the target and the equations of motion,} \\ \text{calculate the field}\end{array}}$$

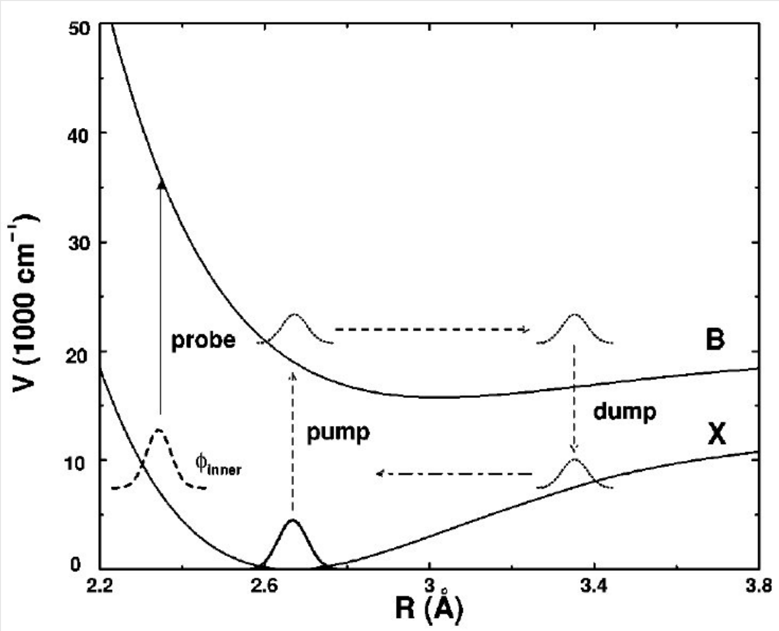
<u>local in time</u>

impose 2 conditions (for phase and amplitude) $\longrightarrow$ derive equations for field

<u>global in time</u>

information from dynamics throughout time interval to reach desired target at final T

Optimal control theory: Example



Z. Shen, V. Engel, R. Xu, J. Cheng, Y.-J. Yan, *J. Chem. Phys.* **117**, 16142 (2002)

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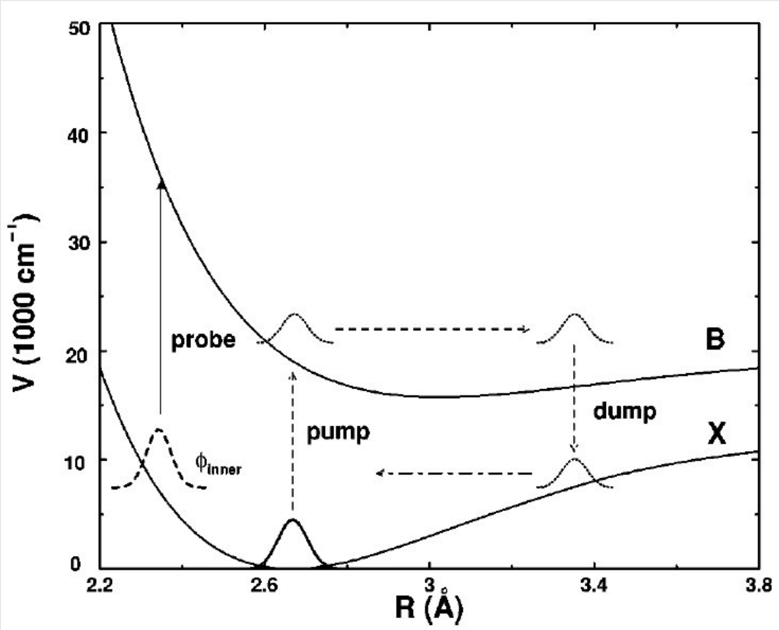

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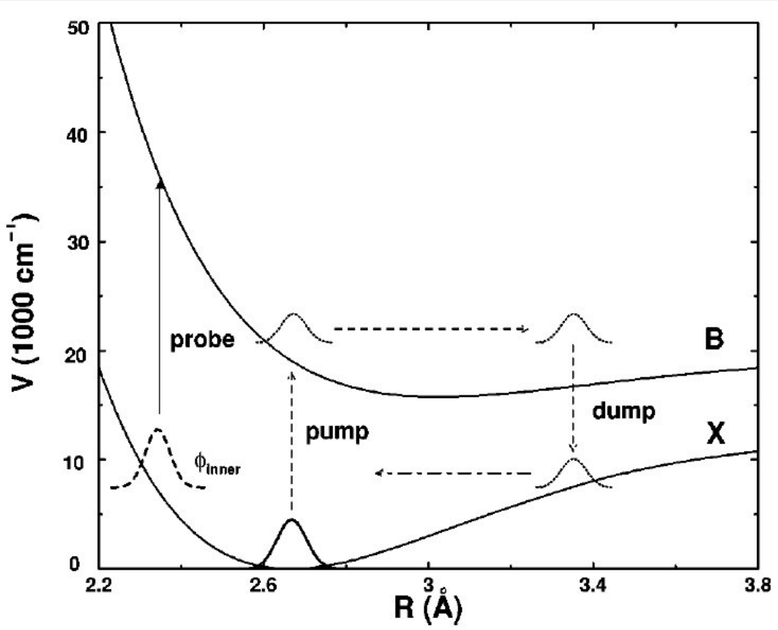

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find the time-dependent pulses which lead to the objective

3. Variational approach to quantum control

OCT: Variational calculus

You can always find a better field $\longrightarrow$ Find it by variation

Define the objective:

$$\boxed{\mathrm{GOAL} \equiv \| \langle \varphi_i | \hat{\mathbf{U}}^{+}(T, 0; \varepsilon) | \varphi_f \rangle \|^2 = F}$$

as a *functional* of the field ε

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Goal depends just on final time T

$\curvearrowright$ new functional J

$$J = F + \int_0^T g(\varepsilon, \varphi)\mathrm{d}t$$

with additional constraints to use information from the dynamics (i.e. intermediate times)

Methods to obtain the optimal field

Rabitz' approach

Krotov method

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Rabitz' approach

- "guess" the right functional
- do the variations to obtain eqs. of motion and eq. for the field
- "guess" the correct time discretization s.t. method converges

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Krotov method

- ingredients: original objective + constraint(s) on the field + EoM of the bare system
- construct auxil. functional with auxil. potential to guarantee monotonic convergence
- derive the field eq. from the minimization of the aux. functional

Sklarz & Tannor, *PRA* **66**, 053619 (2002)
Palao & Kosloff, *PRA* **68**, 062308 (2003)

How to obtain the equation for the optimized field

W. Zhu, J. Botina, H. Rabitz, *J. Chem. Phys.* **108**, 1953 (1998):

$$J = F - 2\mathfrak{Re}\left\{ \langle \varphi(T) | \varphi_f \rangle \int_0^T \mathsf{d}t \underbrace{\left\langle \psi(t) \,\middle|\, \frac{\partial}{\partial t} + \frac{i}{\hbar} \hat{\mathbf{H}}(\boldsymbol{\varepsilon}) \,\middle|\, \varphi(t) \right\rangle} \right\} - \alpha \int_0^T \frac{\boldsymbol{\varepsilon}^2(t)}{S(t)}$$

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original objective

$\left| \langle\varphi(T)|\varphi_f\rangle \right|^2$

minim. pulse energy,

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minim. pulse energy, enforce pulse shape $S(t)$

Variation w.r.t. $|\varphi(t)\rangle$, Lagrange multiplier $\langle\psi(t)|$, $\boldsymbol{\varepsilon}(t)$ keeping only linear terms

Remember: variation of function $f(x)$ w.r.t. x

$$\delta f = \frac{\partial f}{\partial x}\delta x$$

extremum for $\delta f = 0$

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$$\begin{aligned}
\frac{\partial}{\partial t}|\varphi(t)\rangle &= \frac{i}{\hbar}\left[\hat{\mathbf{H}}_0 - \hat{\boldsymbol{\mu}}\boldsymbol{\varepsilon}(t)\right]|\varphi(t)\rangle \quad \text{with} \quad |\varphi(0)\rangle = |\varphi_i\rangle \\
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\end{aligned}$$

$$\boxed{\boldsymbol{\varepsilon}(t) = -\frac{S(t)}{\alpha}\mathfrak{Im}\left\{\langle\varphi(T)\,|\,\varphi_f\rangle\langle\psi(t)\,|\,\hat{\boldsymbol{\mu}}\,|\,\varphi(t)\rangle\right\}}$$

nonlinear set of equations!

Iterative scheme: Step 1

solve first

$$\frac{\partial}{\partial t}|\psi(t)\rangle^{(0)} = \frac{i}{\hbar}\left[\hat{\mathbf{H}}_0 - \hat{\boldsymbol{\mu}}\tilde{\boldsymbol{\varepsilon}}(t)\right]|\psi(t)\rangle^{(0)}$$

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then

$$\frac{\partial}{\partial t}|\varphi(t)\rangle^{(1)} = \frac{i}{\hbar}\left[\hat{\mathbf{H}}_0 - \hat{\boldsymbol{\mu}}\boldsymbol{\varepsilon}^{(0)}(t)\right]|\varphi(t)\rangle^{(1)}$$

with

$$\boldsymbol{\varepsilon}^{(0)}(t) = -\frac{S(t)}{\alpha}\mathfrak{Im}\left\{{}^{(1)}\big\langle\varphi(t)\,\big|\,\psi(t)\big\rangle^{(0)}\,{}^{(0)}\big\langle\psi(t)\,\big|\,\hat{\boldsymbol{\mu}}\,\big|\,\varphi(t)\big\rangle^{(1)}\right\}$$

where we have used $\big\langle\varphi(T)\,\big|\,\varphi_f\big\rangle = \big\langle\varphi(t)\,\big|\,\psi(t)\big\rangle$

Iterative scheme: Step 2

now solve

$$\frac{\partial}{\partial t}|\psi(t)\rangle^{(1)} = \frac{i}{\hbar}\left[\hat{\mathbf{H}}_0 - \hat{\boldsymbol{\mu}}\boldsymbol{\varepsilon}^{(1)}(t)\right]|\psi(t)\rangle^{(1)}$$

then

$$\frac{\partial}{\partial t}|\varphi(t)\rangle^{(2)} = \frac{i}{\hbar}\left[\hat{\mathbf{H}}_0 - \hat{\boldsymbol{\mu}}\boldsymbol{\varepsilon}^{(2)}(t)\right]|\varphi(t)\rangle^{(2)}$$

with

$$\begin{aligned}
\boldsymbol{\varepsilon}^{(1)}(t) &= -\frac{S(t)}{\alpha}\mathfrak{Im}\left\{{}^{(1)}\langle\varphi(t)\,|\,\psi(t)\rangle^{(1)}\,{}^{(1)}\langle\psi(t)\,|\,\hat{\boldsymbol{\mu}}\,|\,\varphi(t)\rangle^{(1)}\right\} \\
\boldsymbol{\varepsilon}^{(2)}(t) &= -\frac{S(t)}{\alpha}\mathfrak{Im}\left\{{}^{(2)}\langle\varphi(t)\,|\,\psi(t)\rangle^{(1)}\,{}^{(1)}\langle\psi(t)\,|\,\hat{\boldsymbol{\mu}}\,|\,\varphi(t)\rangle^{(2)}\right\}
\end{aligned}$$

Iterative scheme

$$\tilde{\boldsymbol{\varepsilon}}(t) \rightarrow |\psi(t)\rangle^{(0)} \rightarrow \boldsymbol{\varepsilon}^{(0)}(t), |\varphi(t)\rangle^{(1)} \longrightarrow \boldsymbol{\varepsilon}^{(1)}(t), |\psi(t)\rangle^{(1)} \rightarrow \boldsymbol{\varepsilon}^{(2)}(t), |\varphi(t)\rangle^{(2)} \longrightarrow \ldots$$

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$$\boldsymbol{\varepsilon}^{(0)}(t) \;=\; -\frac{S(t)}{\alpha}\mathfrak{Im}\Big\{ {}^{(1)}\big\langle \varphi(t) \,\big|\, \psi(t) \big\rangle^{(0)} \, {}^{(0)}\big\langle \psi(t) \,\big|\, \hat{\boldsymbol{\mu}} \,\big|\, \varphi(t) \big\rangle^{(1)} \Big\}$$

$$\boldsymbol{\varepsilon}^{(1)}(t) \;=\; -\frac{S(t)}{\alpha}\mathfrak{Im}\Big\{ {}^{(1)}\big\langle \varphi(t) \,\big|\, \psi(t) \big\rangle^{(1)} \, {}^{(1)}\big\langle \psi(t) \,\big|\, \hat{\boldsymbol{\mu}} \,\big|\, \varphi(t) \big\rangle^{(1)} \Big\}$$

$$\boldsymbol{\varepsilon}^{(2)}(t) \;=\; -\frac{S(t)}{\alpha}\mathfrak{Im}\Big\{ {}^{(2)}\big\langle \varphi(t) \,\big|\, \psi(t) \big\rangle^{(1)} \, {}^{(1)}\big\langle \psi(t) \,\big|\, \hat{\boldsymbol{\mu}} \,\big|\, \varphi(t) \big\rangle^{(2)} \Big\}$$

How does the optimization work?

How does the optimization work?

Rabitz' approach

$$\boldsymbol{\varepsilon}(t) = -\frac{S(t)}{\alpha}\mathfrak{Im}\Big\{\big\langle\varphi(T)\,\big|\,\varphi_f\big\rangle\big\langle\psi(t)\,\big|\,\hat{\boldsymbol{\mu}}\,\big|\,\varphi(t)\big\rangle\Big\}$$

Krotov method

$$\boldsymbol{\varepsilon}(t) = -\frac{S(t)}{\alpha}\mathfrak{Im}\Big\{\langle\varphi_i\,|\;\hat{\mathbf{U}}^{+}(T,0;\boldsymbol{\varepsilon}^0)\;|\,\varphi_f\rangle\;\langle\varphi_f\,|\;\hat{\mathbf{U}}^{+}(t,T;\boldsymbol{\varepsilon}^0)\,\hat{\boldsymbol{\mu}}\,\hat{\mathbf{U}}(t,0;\boldsymbol{\varepsilon}^1)\;|\,\varphi_i\rangle\;\Big\}$$

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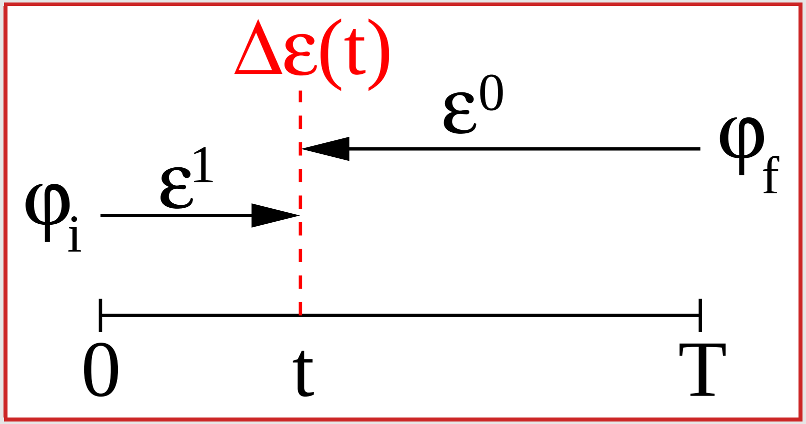
Interference between past and future events



How does the optimization work?

1. set the parameters of the algorithm: optimization time T, weight of constraint α, shape function $S(t)$
2. choose a physically sensible guess field
3. run the iterative scheme
4. **analyze the optimal field**

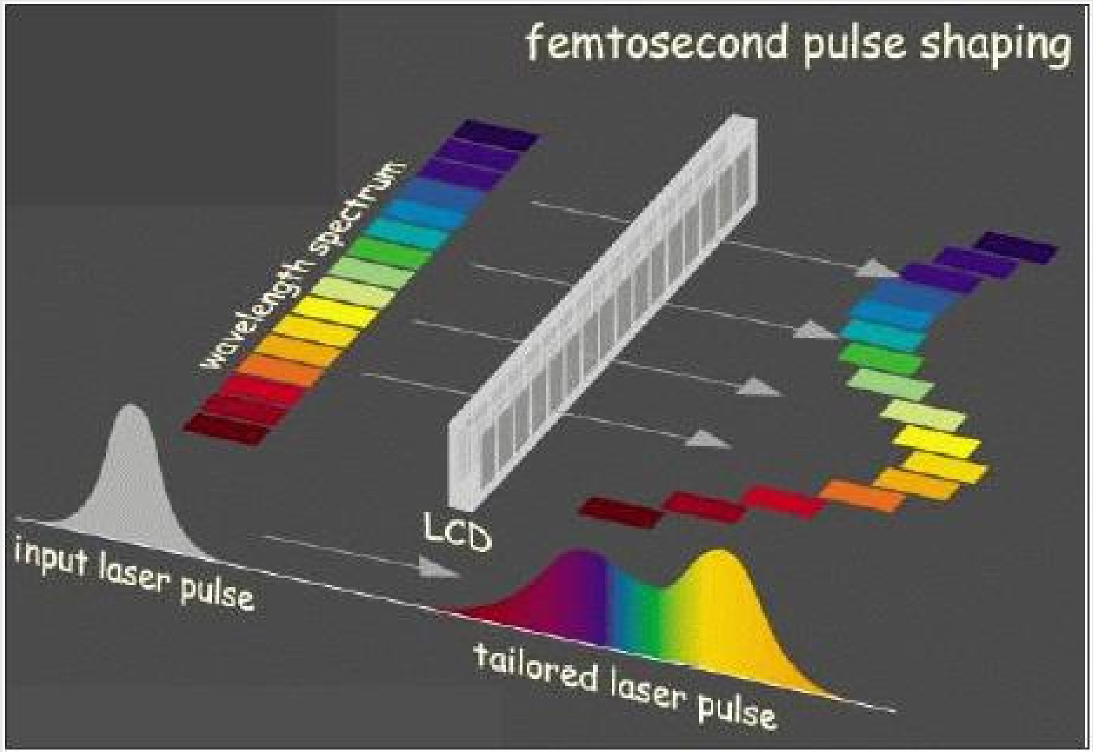
4. Experimental quantum control: Closed learning loops

The versatility of light

frequency domain pulse shaping using AOM's or LCD's

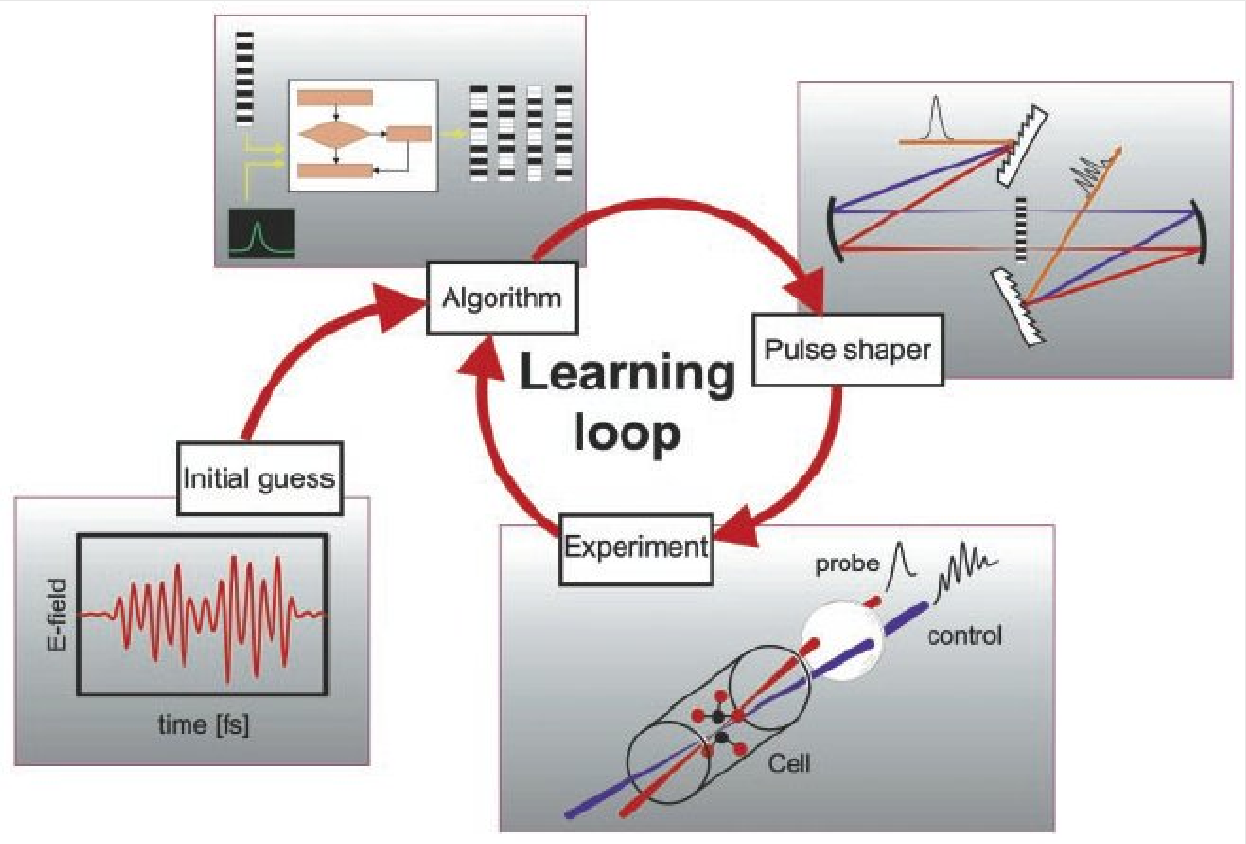
for both amplitude and phase

available in VIS, near-IR



T. Brixner & G. Gerber, *ChemPhysChem*, **4**, 418 (2003)

Control experiments



H. Rabitz, R. de Vivie-Riedle, M. Motzkus, K. Kompa
Science, **288**, 824 (2000)

Control experiments

"Teaching lasers to control molecules"
R. S. Judson, H. Rabitz
PRL **68**, 1500 (1992)

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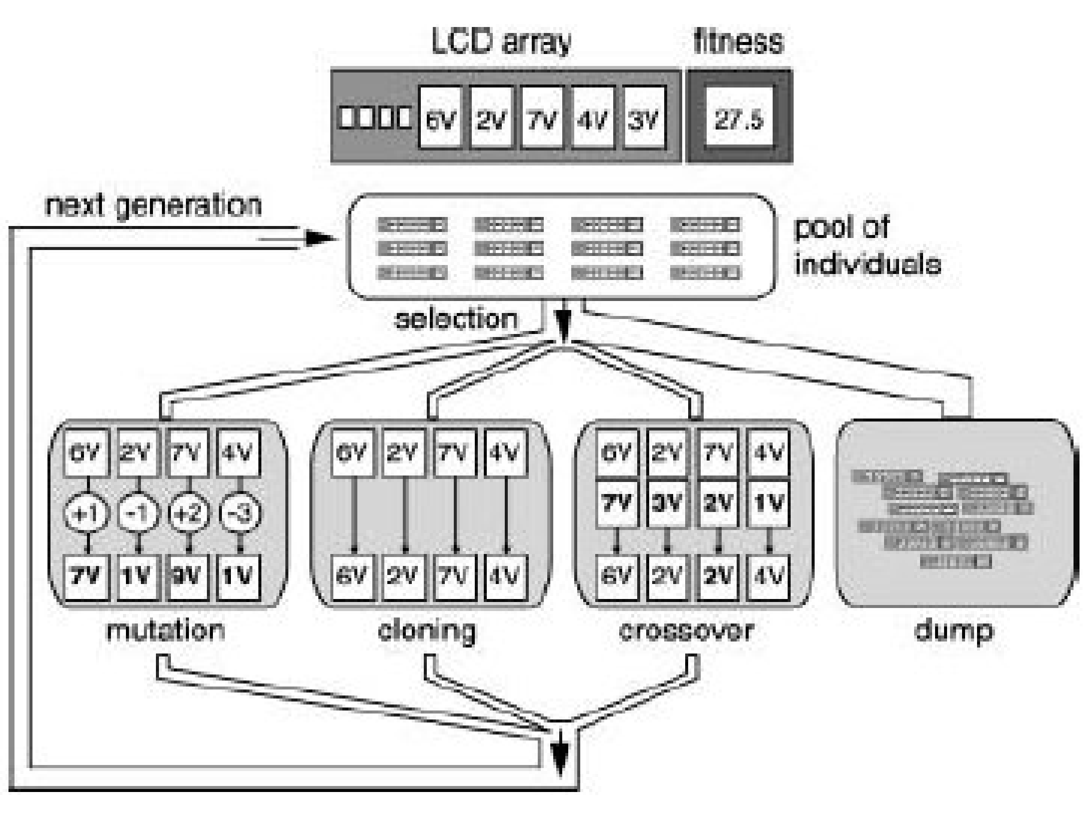

T. Brixner & G. Gerber, *ChemPhysChem* **4**, 418 (2003)

Theory vs. Experiment

Theory

uses information about many different pathways contained in Schrödinger eq.

↓

very efficient,
but optimal pulses extremely complex (additional experimental constraints destroy convergence in currently available algorithms)

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Theory	Experiment
uses information about many different pathways contained in Schrödinger eq.	pulse shapers steered by evolutionary algorithm (closed learning loops)
↓	↓
very efficient, but optimal pulses extremely complex (additional experimental constraints destroy convergence in currently available algorithms)	many repetitions, no insight (combination with ultracold experiments may be difficult)

→ **Comparison between theory and experiment necessary, but not straightforward!**

Theory can provide estimate of **feasibility**, i.e. experimentally required **spectral width and intensity**

5. Coherent / optimal control of cold systems?

Theoretical proposals

Cold systems ideally suited for coherent processes

Theoretical proposals

Cold systems ideally suited for coherent processes

- controlled cold chemistry
  $\longrightarrow$ S. Jørgensen, R. Kosloff
- STIRAP to convert atomic to molecular BEC
  $\longrightarrow$ P. D. Drummond, K. V. Kheruntsyan, D. J. Heinzen & R. H. Wynar, PRA **65**, 063619 (2002)
- coherent photoassociation ?
  (BEC $\longrightarrow$ J. Javanainen, M. Mackie, PRA **59**, R3186 (1999))

Theoretical proposals: Optimal control

Cold systems ideally suited for coherent processes

Theoretical proposals: Optimal control

Cold systems ideally suited for coherent processes

- controlled scattering (for quantum computing with qubits) $\longrightarrow$ T. Calarco U. Dorner, P. Julienne, C. Williams, P. Zoller, quant-ph/0403197
- quantum computing with molecules $\longrightarrow$ C. M. Tesch & R. de Vivie-Riedle, *PRL* **89**, 157901 (2002) , J. P. Palao & R. Kosloff, *PRL* **89**, 188301 (2002)
- coherent stabilization of ultracold molecules $\longrightarrow$ C. P. Koch, J. P. Palao, R. Kosloff & F. Masnou-Seeuws, *PRA* in press

Control experiments with cold systems?

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so far none!

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- STIRAP in optical lattice not (yet?) successful
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- learning loops may not be feasible for cold experiments

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$\downarrow$

- better "overlap" between theory and experiment
  $\longrightarrow$ **mask function**
  T. Hornung, M. Motzkus, R. de Vivie-Riedle, *J. Chem. Phys.* **115**, 3105 (2001)
- combine **"cold"** and **"hot"** experiments ?

Summary

- **It is possible to take advantage of quantum coherences !**
  (constructive IF in desired channel, destructive IF in all other channels)
- A few **intuitive** schemes to control quantum processes exist (limited applicability).
- **Optimal** control can be employed for a general solution of the control problem.
- It relies on the theorem of **controllability**.
- Specific solutions can be found using the **variational principle** and **iterative search schemes**.