Training Schrödinger’s Cat: Quantum Control in Molecular Physics and Quantum Information Science

Christiane Koch
last year so many things were different . . .
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quantum control
— the superposition principle at work —


quantum control:
build interferences at will using external fields

Shapir o & Brumer: Principles of Quantum Control of Molecular Processes.
quantum control — the superposition principle at work —

quantum mechanics:

a molecule can interfere with itself

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— the superposition principle at work —

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a molecule can interfere with itself

quantum control:
built interferences at will using external fields

Shapiro & Brumer: Principles of Quantum Control of Molecular Processes.
quantum optimal control
quantum optimal control

current applications

(laser) control of atoms & molecules

NMR

quantum information

courtesy: R. de Vivie-Riedle

courtesy: S. Glaser

courtesy: F. Jelezko
quantum optimal control

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quantum optimal control
determining the fields that build interferences at will


define the objective:

\[
\text{GOAL} \equiv \| \langle \psi_{\text{ini}} | \hat{U}^{+} (T, 0; \epsilon) | \psi_{\text{target}} \rangle \|^2 = -J_{T}
\]

as a functional of the external field \( \epsilon \)
quantum optimal control
determining the fields that build interferences at will


define the objective:

\[
\text{GOAL} \equiv \| \langle \varphi_{\text{ini}} | \hat{U}^+ (T, 0; \epsilon) | \varphi_{\text{target}} \rangle \|^2 = -J_T
\]

as a functional of the external field \( \epsilon \)

include additional constraints:

\[
J = J_T + \int_0^T J_t (\epsilon, \varphi) \, dt
\]
quantum optimal control
determining the fields that build interferences at will


define the objective:

$$\text{GOAL} \equiv \| \langle \varphi_{\text{ini}} | \hat{U}^+(T, 0; \epsilon) | \varphi_{\text{target}} \rangle \|^2 = -J_T$$

as a functional of the external field $\epsilon$

include additional constraints:

$$J = J_T + \int_0^T J_t(\epsilon, \varphi) \, dt$$

optimize $J$ by demanding:

$$\partial_\epsilon J = 0 \quad \partial_{\varphi(t)} J = 0 \quad \partial_\epsilon^2 J > 0$$

$$|\varphi(t)\rangle = \hat{U}(t, 0; \epsilon) |\varphi_{\text{ini}}\rangle \quad \hat{H} = \hat{H}_0 + \epsilon(t) \hat{H}_1$$

or by searching for best parametrization of $\epsilon(t)$
quantum optimal control
determining the fields that build interferences at will


define the objective:

\[ \text{GOAL} \equiv \| \langle \varphi_{\text{ini}} | \hat{U}^+(T, 0; \epsilon) | \varphi_{\text{target}} \rangle \|^2 = -J_T \]

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or by searching for best parametrization of \( \epsilon(t) \)

‘easily’ generalized to open quantum systems, target operations, expectation values, or other properties
an example
a first control problem
fast & accurate preparation of circular state

Rb valence electron in
$|n, (l), m_l^{\text{max}} \rangle$
a first control problem

fast & accurate preparation of circular state

Rb valence electron in $|n,(l),m_l^{\text{max}}\rangle$
control problem

fast & accurate preparation of circular state

\[ |n, (l), m^\text{max}_l \rangle \]
preparation of circular state

why?
preparation of circular state
why?

- super long lifetime
- qu sensing
  - cavity QED
- qu interfaces
  - qu computing
preparation of circular state
why?
• super long lifetime
  ⇓ qu sensing ⇓ cavity QED
  ⇓ qu interfaces ⇓ qu computing
preparation of circular state

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preparation of circular state

after laser excitation, use RF drive: $\pi$-pulse

$\Delta E_{51}$

Hydrogen
Rubidium

initial state

|51C⟩

target state

$\ell m$

0 1 2 3 47 48 49 50
preparation of circular state

problem for speed up:
larger amplitudes drive unwanted transitions to lower $m_\ell$
preparation of circular state

optimization: fast & accurate state transfer

optim. RF field: \( \sim 99\% \)
preparation of circular state

Patsch, Reich, Raimond, Brune, Gleyzes, CPK, Phys Rev A 97, 053418 (2018)

how does the optimized RF pulse work?
optimized pulse in experiment @ ENS Paris

using theoretical pulse as is

fidelity: 94%
optimized pulse in experiment

improved calibration & simplified pulse

![Graph showing improved calibration and simplified pulse with 97% efficiency]

Larrouy, Patsch, Richaud, Raimond, Brune, CPK, Gleyzes, Phys Rev X 10, 021058 (2020)
a more challenging target

Larrouy, Patsch, Richaud, Raimond, Brune, CPK, Gleyzes, Phys Rev X 10, 021058 (2020)

superposition of $|m_l = 1\rangle$ and $|m_l = m_l^{max}\rangle$ no known protocol!
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but we can “train Schrödinger’s cat”
a more challenging target

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but we can “train Schrödinger’s cat”
optimal control for dissipative generation of entanglement
driven dissipative evolution

— quantum reservoir engineering —

\[ \partial_t \rho = \mathcal{L}(\rho) = -i[H, \rho] - + \mathcal{L}_D(\rho) \]

steady state \[\longleftrightarrow\] \[ \partial_t \rho = 0 \]

dissipation as friend, not foe
Driven dissipative evolution

— quantum reservoir engineering —

\[ \partial_t \rho = \mathcal{L}(\rho) = -i[H, \rho]_\text{−} + \mathcal{L}_D(\rho) \]

steady state \iff \partial_t \rho = 0

dissipation as friend, not foe

Use external fields + unidirectional evolution due to \( \mathcal{L}_D \) to create non-trivial steady state
driven dissipative evolution

— quantum reservoir engineering —

\[ \partial_t \rho = \mathcal{L}(\rho) = -i[H, \rho] - + \mathcal{L}_D(\rho) \]

steady state \( \leftrightarrow \) \( \partial_t \rho = 0 \)

dissipation as friend, not foe

use external fields + unidirectional evolution due to \( \mathcal{L}_D \) to create non-trivial steady state
Boulder experiment

Boulder experiment


microwave

\[ |e\rangle \]
\[ |↑\rangle \]
\[ \Omega_e \]
\[ |↓\rangle \]
\[ |a\rangle \]

repump

\[ \Omega_e \]
\[ γ_{e,↑}, γ_{e,↓}, γ_{e,a} \]
\[ |e\rangle \]
\[ |↑\rangle \]
\[ |↓\rangle \]
\[ |a\rangle \]

sideband

\[ Δ \]
\[ E_{blue} \]
\[ E_{red} \]
\[ |e\rangle \]
\[ |n = 1\rangle \]
\[ |↑, n = 0\rangle \]
\[ |↓, n = 0\rangle \]
\[ |n = 0\rangle \]
\[ |a, n = 0\rangle \]

plus laser cooling on Mg$^+$
coherent couplings & incoherent decays

population flow for entanglement generation
coherent couplings & incoherent decays

population flow for entanglement generation

- laser cooling
- repump
- microwave
- sideband
coherent couplings & incoherent decays

population flow for entanglement generation

→ laser cooling

→ repump

← microwave

← sideband
coherent couplings & incoherent decays

population flow for entanglement generation

laser cooling
repump
microwave
sideband
coherent couplings & incoherent decays

population flow for entanglement generation

\\begin{itemize}
  \item \textbf{laser cooling}
  \item \textbf{repump}
  \item \textbf{microwave}
  \item \textbf{sideband}
\\end{itemize}
coherent couplings & incoherent decays

population flow for entanglement generation

---

laser cooling

---

repump

---

microwave

---

sideband
all fields continuously on: $\mathcal{F} = 0.75$
with optimized parameters

\[ F = 0.75 \rightarrow 0.88 \]
quantum optimal control

to ensure the proper steady state
quantum optimal control

replace cooling on Mg$^+$ by second sideband

keep blue sideband

replace cooling on Mg$^+$ by second sideband to ensure the proper steady state

no transitions out of target state!
quantum optimal control

choose a different microwave transition

to ensure the proper steady state

choose a different micro wave transition

keep blue sideband
replace cooling on Mg by second sideband
keep repump to ensure the proper steady state

no transitions out of target state!
only remaining source of error: anomalous heating
quantum optimal control

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choose a different microwave transition

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no transitions out of target state!
only remaining source of error: anomalous heating
comparison

two sidebands vs original scheme

\[ \text{population} = 2\pi \times 10 \text{s}^{-1} \]

\[ \text{pol. optimised} \]

\[ \text{original scheme} \]
comparison

two sidebands vs original scheme

\[ \frac{2\pi}{1s} \times 1 \]
\[ \frac{2\pi}{10s} \times 10 \]
\[ \frac{2\pi}{100s} \times 100 \]

pol. optimised
original scheme
ultimately attainable error

Horn, Reiter, Lin, Leibfried, CPK, New J Phys 20, 123010 (2018)

\[
\epsilon = 1 - F
\]

\[
P \text{ (mW) per beam}
\]

\[
E_{r/b} \text{ (V/m) per beam}
\]

\[
\kappa_h = 2\pi \times 10^2 \text{s}^{-1}
\]
ultimately attainable error
ultimately attainable error

Horn, Reiter, Lin, Leibfried, CPK, New J Phys 20, 123010 (2018)
which fields are needed to reach a control target?
which fields are needed to reach a control target?

is there a systematic way to find out?

what chiral molecules can teach us about quantum control
which fields are needed to reach a control target?

is there a systematic way to find out?

what chiral molecules can teach us about quantum control
chiral molecules

chirality = handedness
chirality in pharmacology

(S)-ibuprofen

(S)-(-)-Thalidomid

(R)-(+-)-Thalidomid
chirality in pharmacology

(S)-ibuprofen

(S)-(-)-Thalidomid

(R)-(+)-Thalidomid

(S)-ibuprofen
separate enantiomers with electric fields?

Yes! with three-wave mixing microwave spectroscopy.

Patterson, Schnell, Doyle, Nature 497, 475 (2013)

This works for randomly oriented molecules and requires only electric-dipole transitions!
separate enantiomers with electric fields?
can we distinguish left/right-handed molecules with el. fields?

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Can we distinguish left/right-handed molecules with electric fields?

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enantiomer-selective excitation?

enantiomers have identical spectra except for parity violation-related energy shifts
enantiomer-selective excitation!
enantiomer-selective excitation!

\[ \mu = \begin{pmatrix} \mu_a \\ \mu_b \\ \mu_c \end{pmatrix} \]

\[ \begin{align*} 
\mu_a^{(S)} &= \mu_a^{(R)} \\
\mu_b^{(S)} &= \mu_b^{(R)} \\
\mu_c^{(S)} &= -\mu_c^{(R)} 
\end{align*} \]

\[ H_{int} \sim \mu_a E_z(t) \]

\[ \sim \mu_b E_y(t) \]

\[ \sim \mu_c E_x(t) \]
enantiomer-selective excitation!

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\[ \mu_a^{(S)} = \mu_a^{(R)} \]
\[ \mu_b^{(S)} = \mu_b^{(R)} \]
\[ \mu_c^{(S)} = -\mu_c^{(R)} \]

- two excitation pathways from \( |0\rangle \) to \( |2\rangle \)
- constructive vs destructive interference
enantiomer-selective excitation!

\[ \mu = \begin{pmatrix} \mu_a \\ \mu_b \\ \mu_c \end{pmatrix} \]

- \( \mu_a^{(S)} = \mu_a^{(R)} \)
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- two excitation pathways from \( |0\rangle \) to \( |2\rangle \)
- constructive vs destructive interference
enantiomer-selective excitation
and it doesn’t even matter how exactly you do it

adiabatic ($\Phi = 0$)

but you do need
three mutually orthogonal polarization directions
enantiomer-selective excitation
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enantiomer-selective excitation
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adiabatic ($\Phi = 0$)

nonadiabatic ($\Phi = \pi / 2$)

but you do need
three mutually orthogonal polarization directions
enantiomer-specific state transfer
a first step towards separating enantiomers

Eibenberger, Patterson, Doyle, Phys Rev Lett 118, 123002 (2017)
enantiomer-specific state transfer
so far limited by temperature and (!) M-degeneracy

Perez, Steber, Domingos, Krin, Schmitz, Schnell, Angewandte Chemie 56, 12512 (2017)
what's the problem?

$M$-degeneracy

Initial states: $1_{01}$

Temperature

<table>
<thead>
<tr>
<th>M</th>
<th>-2</th>
<th>-1</th>
<th>0</th>
<th>1</th>
<th>2</th>
</tr>
</thead>
<tbody>
<tr>
<td>1_01</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
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</table>

![Diagram of initial states and population over time](image)

Loss due to forbidden transition
what’s the problem?

$M$-degeneracy

Temperature

Initial states: $1_{01}$

Thermal distribution 1K

all states are initially populated

loss due to forbidden transition

approx 3% selectivity
problem of temperature

example of menthone

thermal occupation of rotational states at $T=1K$
problem of temperature

example of menthone

✉ use thermally unpopulated states!

e.g. vibrationally excited states

problem of $M$-degeneracy controllability of degenerate levels?
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problem of $M$-degeneracy
controllability of degenerate levels?

- Different Rabi-frequencies
- Forbidden transitions
- Multiple transitions

$\Delta J = 0$

Transitions between states with $M=0$ are forbidden
problem of $M$-degeneracy controllability of degenerate levels?
problem of $\mathcal{M}$-degeneracy: how bad is it?

problem of $M$-degeneracy: how bad is it?

but a solution exists!
controllability

Molecular Hamiltonian

\[ H_0 = \begin{pmatrix} E_1 & 0 & 0 \\ 0 & \ddots & 0 \\ 0 & 0 & E_N \end{pmatrix} \]

Interaction Hamiltonian

\[ H_{\text{int}} = \sum_{i=1}^{K} u_i(t) H_i \]

Lie-algebra

\[ H_0, H_1, \ldots, H_K \]

\[ [H_0, H_i], [H_i, H_j] \]

\[ [H_0, [H_i, H_j]], [H_k, [H_i, H_j]] \]

\[ \vdots \]

number of linear independent operators = dimension of Lie-algebra

N-state system is controllable if dimension of Lie-algebra is equal or larger than \( N^2 - 1 \)

Two enantiomers of a chiral N-level system: 2N-level system

\[ H_{0}^{chiral} = \begin{pmatrix} H_0 & 0 \\ 0 & H_0 \end{pmatrix} \quad H_{int}^{chiral} = \begin{pmatrix} H_{int}^{(R)} & 0 \\ 0 & H_{int}^{(S)} \end{pmatrix} \]

Without conversion between enantiomers:

The system is enantio-selective controllable if the dimension of the Lie-algebra is equal or larger then \( 2(N^2 - 1) \)
enantiomer-selective controllability

Two enantiomers of a chiral N-level system: 2N-level system

\[ H_{0}^{\text{chiral}} = \begin{pmatrix} H_{0} & 0 \\ 0 & H_{0} \end{pmatrix} \quad H_{\text{int}}^{\text{chiral}} = \begin{pmatrix} H_{\text{int}}^{(R)} & 0 \\ 0 & H_{\text{int}}^{(S)} \end{pmatrix} \]

Without conversion between enantiomers:

The system is enantio-selective controllable if the dimension of the Lie-algebra is equal or larger than \(2(N^2 - 1)\)
enantiomer-selective controllability


- can be proven for any rotational subspace with a finite number of $J$’s
- requires 3 orthogonal polarization directions & 3 dipole moment projections
- (exceptions have measure zero)

\[
\begin{align*}
\begin{array}{cccccccc}
2,2 & 2,1 & 3,-1 & 3,-2 & 3,-3 & 2,0 & 2,-1 & 1,-1 \\
\hline
\end{array}
\end{align*}
\]

\[
\begin{align*}
\begin{array}{cccccccc}
M= & -3 & -2 & -1 & 0 & 1 & 2 & 3 \\
\hline
\end{array}
\end{align*}
\]
how many fields are needed?


single enantiomer controllability

\[
\begin{align*}
\omega_2, \mu_a & \quad \begin{array}{c}
\begin{array}{c}
\text{z} \\
\text{y} \\
\text{y} \\
\text{z}
\end{array}
\end{array} \\
\omega_1, \mu_b & \quad \begin{array}{c}
\begin{array}{c}
\text{x} \\
\text{z} \\
\text{x}
\end{array}
\end{array}
\end{align*}
\]

\[M = \begin{bmatrix} -1 & 0 & 1 \end{bmatrix}\]

four

enantiomer-selective controllability

\[
\begin{align*}
\omega_3, \mu_c & \quad \begin{array}{c}
\begin{array}{c}
\text{z} \\
\text{y} \\
\text{y} \\
\text{z}
\end{array}
\end{array} \\
\omega_3, \mu_c & \quad \begin{array}{c}
\begin{array}{c}
\text{x} \\
\text{z} \\
\text{x}
\end{array}
\end{array}
\end{align*}
\]

\[M = \begin{bmatrix} -1 & 0 & 1 \end{bmatrix}\]

five
pulse design — step 1: separate $\pm M$

pulse design — step 1: separate $\pm M$ 

pulse design — step 2: separate $R/S$

pulse design — step 2: separate $R/S$

a much simpler solution

a much simpler solution

feasible with current microwave technology
a much simpler solution

feasible with current microwave technology
summary

quantum optimal control is a versatile tool

in qu info science

to bridge the gap between basic concepts and complexity of physical platforms

in molecular physics

to explore fundamental limits of what can / cannot be controlled

we can energetically separate population in degenerate states left-handed from right-handed molecules how to adapt these concepts when scaling up system size?
**summary**

**quantum optimal control is a versatile tool**

*in qu info science*

**to bridge the gap between**

**basic concepts and complexity**

**of physical platforms**

*in molecular physics*

**to explore fundamental limits**

**of what can / cannot be controlled**

---

**we can determine**

- practically useful protocols for various platforms
- fundamental performance bounds
**Summary**

Quantum optimal control is a versatile tool in quantum information science to bridge the gap between basic concepts and complexity of physical platforms.

In molecular physics, we can explore fundamental limits of what can / cannot be controlled.

We can energetically separate population in degenerate states.

We can determine:

- Practically useful protocols for various platforms
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---

**Diagram:**

- Diagram showing quantum states and population dynamics.
- Diagram illustrating the relationship between control fields and molecular states.
Quantum optimal control is a versatile tool in quantum information science to bridge the gap between basic concepts and complexity of physical platforms.

In molecular physics, we can explore fundamental limits of what can / cannot be controlled.

We can determine:
- Practically useful protocols for various platforms
- Fundamental performance bounds

How to adapt these concepts when scaling up system size?

We can energetically separate:
- Population in degenerate states
- Left-handed from right-handed molecules
acknowledgements

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C. Pérez, M. Schnell (DESY)

Karl Horn  Sabrina Patsch
Monika Leibscher
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