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 —



Juffmann et al., Nature Nanotech. 7, 297 (2012)



Juffmann et al., Nature Nanotech. 7, 297 (2012)

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

current applications

(laser) control of atoms & molecules

NMR

quantum information

current applications

NMR

(laser) control of atoms & molecules

quantum information





courtesy: R. de Vivie-Riedle

current applications

(laser) control of atoms & molecules

NMR

quantum information









courtesy: R. de Vivie-Riedle

courtesy: S. Glaser

current applications

NMR

(laser) control of atoms & molecules





quantum information









courtesy: F. Jelezko

courtesy: R. de Vivie-Riedle

courtesy: S. Glaser

Glaser et al., Eur. Phys. J. D 69, 279 (2015)

define the objective :

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

as a functional of the external field ϵ

Glaser et al., Eur. Phys. J. D 69, 279 (2015)

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include additional constraints:

$$J = J_T + \int_0^T J_t(\boldsymbol{\epsilon}, \boldsymbol{\varphi}) \, \mathrm{d}t$$

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optimize *J* by demanding:

$$\partial_{\boldsymbol{\epsilon}} J = \boldsymbol{\hat{0}} \qquad \partial_{\varphi(t)} J = \boldsymbol{\hat{0}} \qquad \partial_{\boldsymbol{\epsilon}}^{2} J > \boldsymbol{\hat{0}} \\ |\varphi(t)\rangle = \boldsymbol{\hat{U}}(t, 0; \boldsymbol{\epsilon}) |\varphi_{\text{ini}}\rangle \qquad \boldsymbol{\hat{H}} = \boldsymbol{\hat{H}}_{0} + \boldsymbol{\epsilon}(t) \boldsymbol{\hat{H}}_{1}$$

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

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include additional constraints:

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optimize J by demanding:

$$\partial_{\boldsymbol{\epsilon}} J = 0$$
 $\partial_{\varphi(t)} J = 0$ $\partial_{\boldsymbol{\epsilon}}^2 J > 0$
 $|\varphi(t)\rangle = \hat{\mathbf{U}}(t, 0; \boldsymbol{\epsilon}) |\varphi_{\text{ini}}\rangle$ $\hat{\mathbf{H}} = \hat{\mathbf{H}}_0 + \boldsymbol{\epsilon}(t) \hat{\mathbf{H}}_1$

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 $|n, (l), m_l^{max}\rangle$

a first control problem fast & accurate preparation of circular state



control problem

fast & accurate preparation of circular state



-50 -49 -48 -47 -3 -2 -1 0 1 2 3 47 48 49 50

• super long lifetime

 \sim qu sensing \sim cavity QED

 \land qu interfaces \land qu computing

	A LE LEVE, GARAGE
LETTER	on a Rydberg atom
A sensitive electrometer based in a Schrödinger-cat state	nan warten kannanel. Wechs terme ^r h

why?

- super long lifetime
- \land qu sensing \land cavity QED
- \sim qu interfaces \sim qu computing





why?

- super long lifetime
- \sim qu sensing \sim cavity QED

 \sim qu interfaces \curvearrowright qu computing





n = 51

n = 50



n = 51

n = 50

after laser excitation, use RF drive: π -pulse



problem for speed up: larger amplitudes drive unwanted transitions to lower m_ℓ



preparation of circular state optimization: fast & accurate state transfer



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



optimized pulse in experiment

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

improved calibration & simplified pulse



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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superposition of $|m_l = 1\rangle$ and $|m_l = m_l^{max}\rangle$ no known protocol! but we can "train Schrödinger's cat" 25 $S_{
m RF}({{\rm mV}\over{
m cm}})$ simulation experiment $-25 \\ 1.0$ 97% -3 population ---- 50 ---- 51 19 0.50.060 80 1201401602040 100t (ns)

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 $\iff \partial_t \rho = 0$

dissipation as friend, not foe
driven dissipative evolution

— quantum reservoir engineering —

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

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 $\iff \partial_t \rho = 0$

dissipation as friend, not foe



Boulder experiment

Lin, Gaebler, Reiter, Tan, Bowler, Sorensen, Leibfried, Wineland, Nature 504, 415 (2013)



Boulder experiment

Lin, Gaebler, Reiter, Tan, Bowler, Sorensen, Leibfried, Wineland, Nature 504, 415 (2013)



plus laser cooling on Mg⁺



population flow for entanglement generation



population flow for entanglement generation

→ laser cooling

-vvv→ repump

 \longleftrightarrow microwave

 \longleftrightarrow sideband







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entanglement generation

Lin, Gaebler, Reiter, Tan, Bowler, Sorensen, Leibfried, Wineland, Nature 504, 415 (2013)

all fields continuously on : $\mathcal{F} = 0.75$



with optimized parameters

















comparison two sidebands vs original scheme



comparison two sidebands vs original scheme



ultimately attainable error



ultimately attainable error



ultimately attainable error



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?

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)-(-)-Thalidomid

(R)-(+)-Thalidomid

chirality in pharmacology



(S)-(-)-Thalidomid



(R)-(+)-Thalidomid

(S)-ibuprofen

can we distinguish left/right-handed molecules with el. fields?

can we distinguish left/right-handed molecules with el. fields?

yes! with three-wave mixing microwave spectroscopy



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



can we distinguish left/right-handed molecules with el. fields?

yes! with three-wave mixing microwave spectroscopy







This works for randomly oriented molecules and requires only electric-dipole transitions !

enantiomer-selective excitation ?



enantiomers have identical spectra

except for parity violation-related energy shifts

enantiomer-selective excitation !


enantiomer-selective excitation !





enantiomer-selective excitation !



- two excitation pathways from $|0\rangle$ to $|2\rangle$
- constructive vs destructive interference

enantiomer-selective excitation !



- two excitation pathways from $|0\rangle$ to $|2\rangle$
- constructive vs destructive interference

VOLUME 87, NUMBER 18 PHYSICAL REVIEW LETTERS 29 OctoBer 2001

Cyclic Population Transfer in Quantum Systems with Broken Symmetry

Petr Král and Moshe Shapiro

enantiomer-selective excitation

and it doesn't even matter how exactly you do it

Leibscher, Giesen, CPK, J Chem Phys 151, 014302 (2019)



but you do need three mutually orthogonal polarization directions

enantiomer-selective excitation

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Leibscher, Giesen, CPK, J Chem Phys 151, 014302 (2019)



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Leibscher, Giesen, CPK, J Chem Phys 151, 014302 (2019)



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



what's the problem ?

M-degeneracy

temperature

Initial states: 101



what's the problem ?

M-degeneracy

Initial states: 101





Thermal distribution 1K

all states are initially populated





loss due to forbidden transition

problem of temperature



problem of temperature



\sim use thermally unpopulated states !

e.g. vibrationally excited states

Leibscher, Giesen, CPK, J Chem Phys 151, 014302 (2019)





- Different Rabi-frequencies
- Forbidden transitions
- Multiple transitions

$$(J'', K'', M''|D^1_{MK}|J', K', M')$$

$$= \sqrt{2J'' + 1}\sqrt{2J' + 1}(-1)^{M'' + K''} \\ \times \left(\begin{array}{ccc} J' & 1 & J'' \\ M' & M & -M'' \end{array} \right) \left(\begin{array}{ccc} J' & 1 & J'' \\ K' & K & -K'' \end{array} \right)$$

Transition matrix elements depend on M





- Different Rabi-frequencies
- Forbidden transitions
- Multiple transitions

 $\Delta J=0$

Transitions between states with M=0 are forbidden



- Different Rabi-frequencies
- Forbidden transitions
- Multiple transitions

x- and y- polarization ($\Delta M = \pm 1$)



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

Leibscher, Pozzoli, Perez, Schnell, Sigalotti, Boscain, CPK, arXiv:2010.09296



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but a solution exists !

controllability

Molecular Hamiltonian

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

Lie-algebra

 $H_0, H_1, \dots H_K$

 $[H_0, H_i], [H_i, H_j]$

 $\left[H_0, \left[H_i, H_j\right]\right], \left[H_k, \left[H_i, H_j\right]\right]$

Interaction Hamiltonian

$$H_{int} = \sum_{i=1}^{K} u_i(t) H_i$$
 control fields

number of linear independent operators = dimension of Lie-algebra

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

Domenico D'Alessandro, Introduction to quantum control and dynamics, Chapman & Hall/CRC (2008)

enantiomer-selective controllability

Leibscher, Pozzoli, Perez, Schnell, Sigalotti, Boscain, CPK, arXiv:2010.09296

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



$$H_0^{chiral} = \begin{pmatrix} H_0 & 0\\ 0 & H_0 \end{pmatrix} \qquad H_{int}^{chiral} = \begin{pmatrix} H_{int}^{(K)} & 0\\ 0 & H_{int}^{(S)} \end{pmatrix}$$

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

Leibscher, Pozzoli, Perez, Schnell, Sigalotti, Boscain, CPK, arXiv:2010.09296

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



$$H_0^{chiral} = \begin{pmatrix} H_0 & 0\\ 0 & H_0 \end{pmatrix} \qquad H_{int}^{chiral} = \begin{pmatrix} H_{int}^{(R)} & 0\\ 0 & H_{int}^{(S)} \end{pmatrix}$$

Without conversion between enantiomers:

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enantiomer-selective controllability

Leibscher, Pozzoli, Perez, Schnell, Sigalotti, Boscain, CPK, arXiv:2010.09296

- 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)



how many fields are needed?

Leibscher, Pozzoli, Perez, Schnell, Sigalotti, Boscain, CPK, arXiv:2010.09296

single enantiomer controllability

enantiomer-selective controllability



four

five

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



Leibscher, Pozzoli, Perez, Schnell, Sigalotti, Boscain, CPK, arXiv:2010.09296

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



pulse design — step 2: separate *R*/*S*



Leibscher, Pozzoli, Perez, Schnell, Sigalotti, Boscain, CPK, arXiv:2010.09296

pulse design — step 2: separate *R*/*S*



a much simpler solution

Leibscher, Pozzoli, Perez, Schnell, Sigalotti, Boscain, CPK, arXiv:2010.09296



a much simpler solution

Leibscher, Pozzoli, Perez, Schnell, Sigalotti, Boscain, CPK, arXiv:2010.09296



feasible with current microwave technology

a much simpler solution



feasible with current microwave technology

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

 8.34×10^2

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

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
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we can energetically separate

- population in degenerate states
- left-handed from righthanded molecules

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in molecular physics to explore fundamental limits of what can / cannot be controlled



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how to adapt these concepts when scaling up system size ?

acknowledgements



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E. Pozzoli, M. Sigalotti, U. Boscain (INRIA) C. Pérez, M. Schnell (DESY)



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