Quantum Control with Quantum Light

Controlling Non-Adiabaticity in Molecules

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July 10, 2019
Overview

1. Introduction
2. Coherent control with quantum light – basic idea
3. Results for LiF
4. Summary
5. Outlook: Molecules and atoms in cavities
Introduction
Molecules In Cavities

- Confined light modes
- Strong coupling
- Modification of potential energy surfaces
- Applications in photo chemistry
- Quantum Optimal Control?

Morigi et al. PRL 99, 073001 (2007)
Schlawin et al. NJP, 19, 013009 (2017)
Brumer, Shapiro, "Quantum Control of Molecular Processes" (2012)
Quantizing the Photon Mode
From Control with Classical Light to Control with Quantum Light

Classical light:
- Single mode
- Dipole interaction
- \( V_c = -\mu(R)E_0 \cos(\omega_L t + \phi) \)

Quantum light:
- Single cavity mode
- Dipole interaction
- Fock states

\[
H_c = \hbar \omega_c \left( a^\dagger a + \frac{1}{2} \right) \\
V_c = \varepsilon_c \mu(R) \left( a^\dagger + a \right) \left( \sigma^\dagger + \sigma \right)
\]

Kowalewski et al., PNAS, 114, 3278 (2017)
To Diagonalize or not to Diagonalize?
Dressed States vs. Bare States

- **Bare molecular states:**
  - Fock states
  - Only position dependent couplings $g(R)$
  - PES less intuitive?

- **Photon displacement coords:**
  - Arbitrary photon states
  - Beyond RWA ($a\dagger\sigma\dagger + a\sigma$)

- **Dressed states:**
  - Avoided crossings
  - Derivative couplings

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Nonadiabatic couplings in the curve crossing region: \( f = \langle \phi_k | \partial_q \phi_l \rangle \)

Adiabatic curves, avoided crossings:
→ Localized couplings, intuitive picture from QC

Detuning, gradient difference, derivative of tr. dipole

\[
\hat{H}_{kl} = \hat{T} + \delta_{kl} \hat{V}_{kl} + \sum_i \frac{1}{m_i} \left( f^{(i)}_{kl} \frac{\partial}{\partial q_i} + \frac{1}{2} h^{(i)}_{kl} \right)
\]


Field anihilation operator:

\[ a = \sqrt{\frac{\omega_c}{2\hbar}} \left( \hat{\mathbf{x}} + \frac{i}{\omega_c} \hat{\mathbf{p}} \right) \]

In displacement coordinates:

\[ H_c = \frac{1}{2} \frac{d^2}{d\mathbf{x}^2} + \frac{1}{2} \omega_c^2 \hat{\mathbf{x}}^2 \]

\[ V_c = g \sqrt{2\hbar \omega_c} \hat{\mathbf{x}} \left( \hat{\sigma}^\dagger + \hat{\sigma} \right) \]

\( \mathbf{x} \): dimensionless coordinate.

Kowalewski et al. JPCL, 7, 2050 (2016)
Flick et al., JCTC, 13, 1616 (2017)
Pulse shaping, classical light:
- Frequency domain
- Amplitude
- Phase
- (Polarization)

Quantum light:
- Super pos. Fock states
- Different photon numbers
- Amplitude, phase
- Non-classical states

\[ E(t) = \sum_n E_n \cos(\omega_n t + \phi_n) \]
Pulse shaping, classical light:

- Frequency domain
- Amplitude
- Phase
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Quantum light:

- Super pos. Fock states
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→ Idea: start with single mode

\[ E(t) = \sum_n E_n \cos(\omega_n t + \phi_n) \]
Quantum Light and Molecules
Combining the photon mode with vibrational degrees of freedom

Coherent states:

- Two electronic states
- Nuclear coordinate
- Photon coordinate
- Linear coupling
- \( V_c = g \sqrt{2\hbar \omega_c} \hat{x} (\hat{\sigma}^\dagger + \hat{\sigma}) \)
Quantum Light and Molecules
Combining the photon mode with vibrational degrees of freedom

Coherent states:

Control avoided crossing:

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Squeezed vacuum state:

- Two electronic states
- Nuclear coordinate
- Photon coordinate
- Linear coupling
- \( V_c = g \sqrt{2\hbar \omega_c} \hat{x} (\hat{\sigma}^\dagger + \hat{\sigma}) \)
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Combining the photon mode with vibrational degrees of freedom

Squeezed vacuum state:

- Two electronic states
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- \( V_c = g \sqrt{2 \hbar \omega_c} \hat{x} (\hat{\sigma}^\dagger + \hat{\sigma}) \)

control coupling via shape of photon wave packet
Test Case: Lithium Fluoride
Controlling population transfer at the avoided crossing

- Pump-Pulse launches dynamics in $\Sigma_2$
- Cavity mode resonant at avoided crossing
- Different initial states of photon mode
- Fock, squeezed vacuum, squeezed coherent

Numerical Approach
Wave Packet Dynamics

- Photon coordinate displacement coordinates $x$
- Nuclear coordinate $R$
- Intrinsic avoided crossing
- MCDTH for WP dynamics

$$H_{kl} = \delta_{kl} \left( -\frac{\hbar^2}{2m} \frac{\partial^2}{\partial R^2} + \hat{V}_k(R) - \frac{\hbar^2}{2} \frac{\partial^2}{\partial x^2} + \frac{1}{2} \omega_c^2 \hat{x}^2 \right)$$

$$+ (1 - \delta_{kl}) g(R) \sqrt{2\hbar\omega_c} \hat{x}$$

$$+ (1 - \delta_{kl}) \frac{1}{2m} \left( 2f_{kl}(R) \frac{\partial}{\partial R} + \frac{\partial}{\partial R} f_{kl}(R) \right)$$
Coherent States

The classical analogy?

Control parameter: initial displacement phase (≡ carrier phase)
fixed initial displacement parameter.

Coherent state phase

Coherent

Squeezed

0.0 0.5 1.0 1.5 2.0
initial phase,

0.0 0.5 1.0 1.5 2.0
initial squeezing phase,

r=0.5
r=1
r=2
r=3

a)
b)
What’s the Closest Correspondence to a Classical State?
Fock state or coherent state?

Let’s have look at a two level atom coupled to a cavity:

**Fock state**

**Coherent state**
Squeezed Vacuum States
Quantum Light – No classical analogy

Control parameter: initial squeezing phase
fixed squeezing parameter.

![Graph showing final population versus initial squeezing phase](attachment:image.png)

Coherent

Squeezed

Initial phase,

Coherent

Squeezed

Reaction coordinate [Å]

Photon displacement coordinate

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Squeezed Coherent States

Control landscapes

Squeezing phase: $\Theta$

Coherent state phase: $\varphi$

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Quantum Control with Quantum Light
Squeezing phase: $\Theta$
Coherent state phase: $\varphi$
Summary:
- Quantized field modes and molecular quantum dynamics
- Non-classical states of light steer population
- Single cavity mode
- Only suppression of population has been observed
- Control beyond phase/amplitude control of laser pulses

Outlook:
- Multiple modes (laser pulse analogy)
- Molecular ensembles
- Investigate different control scenarios
- Creation of initial states?
Acknowledgment

Thank you for your attention

Thanks to:

- András Csehi
- Gábor J. Halász
- Ágnes Vibók
- Eric Davidson

Funding:

- Stockholm University
- The Swedish Research Council
Stockholm 2021?
Conference Announcement

- Nordita: Coherent Control with Modified Vacuum Fields
- Summer school
- Conference & Workshop
- Planned for ≈August 2021
- We need to get funding first!

→ If you are interested send an email to markus.kowalewski@fysik.su.se
Subject ”Cavities in Stockholm 2021”