Quantum optimal control theory and applications

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Kaj Stenvall, Total control, oil on canvas (2005)
Outline

- Principles of quantum optimal control theory (OCT)

- Examples of OCT in solid-state physics
  - Optimization of entanglement
  - Optimization of local fields
  - Optimization of quantum revival

- OCT for strong-field applications
  - Enhancement of ionization
  - Suppression of ionization
  - Control of high-harmonic generation
Optimal control: Overview

Classical control since 1697

\[
x = \frac{L}{C} \cosh^{-1} \left(1 + \frac{y}{A}\right)
\]

\[
y = A \left( \cosh \frac{Cx}{L} - 1 \right)
\]
Optimal control: Overview

- Classical control since 1697
- Learning-loop exp. since 1960s

Optimal control: Overview

Classical control since 1697

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Quantum OCT since 1980s

Recently: applications in chemistry and condensed matter & solid state physics

Laarmann et al. (2007)  M. Inguscio, LENS  Delft Qubit Project
Quantum optimal control theory (OCT)

Key question: What is the external time-dependent field that drives the system into a predefined goal?

\[ i \frac{d}{dt} |\Psi(t)\rangle = \hat{H} [\epsilon_k(t)] |\Psi(t)\rangle \]

control functions

Usually the control function is an electric field (laser pulse)

\[ \hat{H}(t) = \hat{H}_0 + \epsilon(t) \hat{D} \]

Most commonly the objective is to maximize the transition probability to a target state
Formulation of OCT

Find the extremal points of the functional

\[ J[\Psi, \chi, \epsilon] = J_1[\Psi] + J_2[\epsilon] + J_3[\Psi, \chi, \epsilon] \]

\[ J_1[\Psi] = \langle \Psi(T)|\hat{O}|\Psi(T) \rangle \]

\[ J_2[\epsilon] = -\alpha \left[ \int_0^T dt \, \epsilon^2(t) - E_0 \right] \]

\[ J_3[\Psi, \chi, \epsilon] = -2 \text{Im} \left[ \int_0^T dt \langle \chi(t)|i \frac{d}{dt} - \hat{H}(t)|\Psi(t) \rangle \right] \]

\[ \Rightarrow \text{control equations} \text{ to be solved iteratively (various algorithms)} \]

Control equations

- Forward propagation for \( |\Psi(t)\rangle \)
  \[
i \frac{d}{dt} |\Psi(t)\rangle = \hat{H}(t) |\Psi(t)\rangle, \quad |\Psi(0)\rangle = |\Psi_{\text{initial}}\rangle
\]

- Backward propagation for \( |\chi(t)\rangle \)
  \[
i \frac{d}{dt} |\chi(t)\rangle = \hat{H}(t) |\chi(t)\rangle, \quad |\chi(T)\rangle = \hat{O} |\Psi(T)\rangle
\]

- Solution field:
  \[
  \epsilon(t) = -\frac{1}{\alpha} \text{Im} \left[ \langle \chi(t) | \mu | \Psi(t) \rangle \right] \quad \text{with} \quad \int_0^T dt \epsilon^2(t) = E_0
\]

$B \sim mT$

$I \sim \mu A$
Optimization of entanglement

Two electrons in coupled quantum dots + optical field

Optimized pulses can be complex...

Optimization of *local* fields

\[ U(x, y, t) = g(x, y) f(t) \]

Optimization of quantum revival

OCT in strong-field and ultrafast regime

**What is a “strong” field?**

- One definition: electric fields $\gtrsim 1$ a.u. $\approx 5.14 \times 10^{11}$ V/m corresponding to intensities $\gtrsim 3.51 \times 10^{16}$ W/cm²

- Realization: (i) collision of charged particles, (ii) laser fields
OCT in strong-field and ultrafast regime

What is an “ultrafast” field?
- One definition: pulse lengths below one femtosecond are ultrafast
... or more precisely: for electrons, nuclei, and solid-state devices, ultrafast time scales are as, fs, and ps, respectively
- Current world record: 67 as (Univ. of Central Florida, 2012)

Streaking spectrogram for neon 2p photoelectrons (A. Assion et al., Laser Focus World, (2008)
Femtosecond pulse shaping

Control knobs for NIR, VIS, and VIS-UV, respectively:
- chirp
- CEP
- delay
- energy (beam size)

Synthesized Light Transients
A. Wirth et al.,
Science 334, 195 (2011)
OCT for strong fields: Enhanced ionization

- Target operator: \( \hat{O} = \hat{1} - \sum_{i}^{\text{bound}} |\varphi_i\rangle \langle \varphi_i| \)

- System: \( H^2_+ \) (in 3D)

- Initial pulse:
  \( \omega_0 = 0.114 \) a.u.
  \( I \sim 10^{15} \) W/cm\(^2\)
  \( T = 5.3 \) fs

- OCT constraints:
  - fixed fluence
  - max freq = \( 2\omega_0 \)

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Ionization probabilities:

- no OCT \( \Rightarrow 0.5\% \)
- OCT, cutoff at \( 2\omega_0 \) \( \Rightarrow 20\% \)
- OCT, cutoff at \( 4\omega_0 \) \( \Rightarrow 99\% \)
OCT for strong fields: Suppressed ionization

- Motivation: undesired ionization restricts controlled dissociation or HHG
- Control target: ground-state occupation at the end of the pulse

\[ J_1 = |\langle \Psi(T) | \Psi_{GS} \rangle|^2 \]

Example: 1D hydrogen
Suppressed ionization (1D hydrogen)

IR regime

UV regime

Suppressed ionization in 3D (fixed nuclei)

H$_2^+$

OCT for strong fields: High-harmonic generation

- Optimized pulse
- Desired harmonic, cutoff, or/and yield
HHG spectrum & optimization

\[ \text{yield} = \frac{\left| \mathcal{F}\left[ \frac{d^2}{dt^2} D(t) \right](\omega) \right|^2}{\omega^2} \]

1D model atom

\[ I_{\text{max}} = 10^{13} \text{ W/cm}^2 \]
\[ \lambda = 800 \text{ nm} \]
\[ T \approx 25 \text{ fs} \]
HHG spectrum & optimization

\[
yield = \frac{\left| \mathcal{F} \left[ \frac{d^2}{dt^2} D(t) \right] (\omega) \right|^2}{\omega^2}
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HHG spectrum & optimization

\[
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\]

OCT constraints: fixed fluence, max freq = \( \omega_0 \)
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OCTOPUS code (GPL) (www.tddft.org)