Coherent Control of Multiphoton Excitations: From Weak-Field to Intermediate-Field Regime

Zohar Amitay
Lev Chuntonov
Andrey Gandman
Leonid Rybak

Schulich Faculty of Chemistry
Technion – Israel Institute of Technology
Haifa, Israel
Coherent Decoupling of coupled multiphoton excitations

**Intermediate-field regime:**
(extend the frequency-domain analysis beyond the weak-field regime)
A family of pulse shapes optimal for enhanced two-photon absorption

Photo-induced coherent information processing ("problem-specific"): Anti-Symmetry check

("the other side of the coin")
Optical Coherent Control of Quantum Systems

Manipulation of constructive / destructive interferences between pathways to enhance / attenuate transition probability from initial state to final state.

physical quantum system + light + interaction = pathway

optical field : \{ frequency, amplitude, phase, polarization \}

The present work :
Coherent phase control using (a priori) shaped femtosecond pulses
Ultrashort Pulse

Bandwidth = 145 cm\(^{-1}\) (~10 nm)

Duration = 100 fs

Frequency
\[ \Delta \omega \Delta \tau \geq 0.88 \, \pi \]

Time

Broad Bandwidth
+ Coherence
+ All freq. in phase
  "Mode Locking"

Short Pulse
"Transform Limited"
“Pulse Shaping”

every frequency has its own phase!

The Control Knobs

\[ E(\omega) = |E(\omega)| \exp \left[ i \Phi(\omega) \right] \]
Experimental Excitation Scheme - Na atom

[ pulse: ~5 nm bandwidth @ 780 nm, TL duration ~ 180 fs ]
[ ensemble of Na atoms @ static cell, 550 K ]
(I) Non-Resonant Two-Photon Absorption

\[ A_{4s}^{(2)} \propto \int_{-\infty}^{\infty} E(\omega)E(\omega_{4s,3s} - \omega) \, d\omega \]

\[ P_{4s} = \left| A_{4s}^{(2)} \right|^2 \]

(2nd-order time-dep. perturbation theory)

D. Meshulach and Y. Silberberg,
Nature 396, 239 (1998); PRA 60, 1287 (1999)
(II) Resonance-Mediated (2+1) Three-Photon Absorption

\[ A_{7p}^{(3)} \propto i\pi E(\omega_{7p,4s})A_{4s,3s}^{(2)} + \]

on-resonance with 4s \( \propto A_{4s}^{\text{final}} \)

\[ + P.V \int_{-\infty}^{\infty} A_{4s,3s}^{(2)}(\omega_{4s,3s} - \delta) E(\omega_{7p,4s} + \delta) \] \( d\delta \)

near-resonance with 4s

where \[ A^{(2)}(\Omega) = \int_{-\infty}^{\infty} E(\omega)E(\Omega - \omega) d\omega \]

\[ P_{7p} = |A_{7p}^{(3)}|^2 \] (3rd-order time-dep. perturbation theory)
Resonance-Mediated (2+1) Three-Photon Absorption

- **on-resonant pathways**
  - 7p
  - 4s
  - 3s
  - 781.2 nm

- **near-resonant pathways**
  - 7p
  - 4s
  - 3s
  - ≠ 781.2 nm
  - δ

**weak-field regime**
Enhancement of the near-resonant component

\(\pi\)-step: only Intra-group interferences

7p population (3\(\gamma\) absorption)
4s population (2\(\gamma\) absorption)

exp. and calc.
Branching Ratio Phase Control by “Coherent Decoupling”

The basic idea:
Any phase-pattern addition that is anti-symmetric around $\omega_{4s,3s}/2$ (777 nm) will not change $A_{4s}$ but will change $A_{7p}$!

\[ \Delta \Phi(\omega) = -\Delta \Phi\left(\omega_{4s,3s} - \omega\right) \]

\[
A_{4s}^{(2)} \propto \int_{-\infty}^{\infty} E(\omega)E(\omega_{4s,3s} - \omega) \, d\omega =
\]

\[
= \int_{-\infty}^{\infty} |E(\omega)||E(\omega_{4s,3s} - \omega)| \exp\{i \left[ \Phi(\omega) + \Phi(\omega_{4s,3s} - \omega) \right] \} \, d\omega
\]

\[
A_{7p}^{(3)} \propto i\pi E\left(\omega_{7p,4s}\right) A_{4s}^{(2)} + P.V \int_{-\infty}^{\infty} \frac{A_{4s}^{(2)}(\omega_{4s,3s} - \delta)E(\omega_{7p,4s} + \delta)}{\delta} \, d\delta
\]

where \[ A^{(2)}(\Omega) = \int_{-\infty}^{\infty} E(\omega)E(\Omega - \omega) \, d\omega \]
Anti-symmetric phase-pattern addition that changes only the three-photon absorption.

Basic phase-pattern that controls the two-photon absorption.
two-photon dark pulse pattern (π-step) + variable anti-symmetric phase addition

Coherent Decoupling – Two-photon Dark Pulses

The two-photon absorption is zero while the three-photon absorption can be tuned to be between 0 and ~25% of the TL case!

Calculated map of 7p population (relative to TL case)

The anti-sym phase addition

\( \lambda_{\text{asym}} \)

\( \Phi_{\text{asym}} \)

\( \Delta \)
The anti-sym phase addition

The two-photon absorption is zero while the three-photon absorption can be tuned to be between 0 and ~25% of the TL case!
**Coherent Decoupling – additional examples**

\[ \Phi_{\text{asym}} = \pi \]

![Graphs showing population changes with step position for 4S and 7P levels, highlighting the antisymmetric \( \pi \) step position with a red circle labeled (3.2).](image)
Coherent Decoupling - Summary

- 7p population (3γ absorption)
- 4s population (2γ absorption)
normalized to TL signal

Coherent Decoupling - Summary

7p population (3γ absorption)
4s population (2γ absorption)
"The other side of the coin": Photo-Induced Coherent Information Processing by a Quantum System

Using the outcome of the photo-induced dynamics to characterize an unknown driving pulse of optically encoded information (huge config. space). ~inversion

problem-specific global characterization
The implemented computational task: **Efficient Anti-Symmetry Check**

**Input**  
**UNKNOWN Optically Encoded Numerical Function**  
*f(x)* values are encoded as spectral phases ("x=0": 777 nm)

| Processing | 4s population :  
(2γ absorption) | TL Signal | TL Signal | not TL signal |
|-----------|----------------|-----------|-----------|---------------|
| Processing | 7p population :  
(3γ absorption) | TL Signal | not TL signal | not TL signal |
| Output    | Constant function | Anti-symmetric function | “Random” |

\[ f(x) = C \]
\[ f(x) = -f(-x) \]

**Computational Complexity of O(1) !!!**  
(independent of function’s size)

**Classical computation: Complexity of O(N)**
Efficient Anti-Symmetry Check of Optically Encoded Function

Experimental Results

Total of 20,000 functions
10,000 anti-symmetric

Experimental false identification – below 7%
(anti-sym as const / const as anti-sym)
Higher pulse intensities – Intermediate-field Regime
(beyond lowest-order perturbative regime)

Frequency-domain analysis & design !!!

Two-photon absorption
Increased Pulse Intensity – 4s population – Exp. Results & Exact TDSE Calc.

- **8 µJ/pulse**
  - $I_{TL-peak} = 2.4 \times 10^{10}$ W/cm²
  - $I_{effective} = 0.5 \times 10^{10}$ W/cm²

- **12 µJ/pulse**
  - $I_{TL-peak} = 3.6 \times 10^{10}$ W/cm²
  - $I_{effective} = 1.5 \times 10^{10}$ W/cm²

- **23 µJ/pulse**
  - $I_{TL-peak} = 6.9 \times 10^{10}$ W/cm²
  - $I_{effective} = 2.3 \times 10^{10}$ W/cm²

- **30 µJ/pulse**
  - $I_{TL-peak} = 8.9 \times 10^{10}$ W/cm²
  - $I_{effective} = 3.5 \times 10^{10}$ W/cm²

- **47 µJ/pulse**
  - $I_{TL-peak} = 1.4 \times 10^{11}$ W/cm²
  - $I_{effective} = 5.3 \times 10^{10}$ W/cm²

[ spatial averaging; enhancement@777nm, 780nm; dip@781.2nm; dark pulses ]
Beyond Lowest-Order Perturbative Regime

FREQUENCY DOMAIN PICTURE

Two-photon absorption

3s  4s  7p

4s Resonance-Mediated $4\gamma = \{2\gamma + \text{Resonant Raman}\}$

on-resonant with 4s
near-resonant with 4s
4s_Resonance-Mediated_4γ = \{2γ + Non-Resonant Raman\}

on-resonant, near-resonant with 4s
\[ A_{4s}^{(2)} \propto |E_0|^2 \int_{-\infty}^{\infty} E_N(\omega) E_N(\omega_{4s,3s} - \omega) d\omega \]

\[ A_{4s}^{(4)} = |E_0|^4 \left[ A_{4s}^{(2+nR)} + A_{4s}^{(2+rR)} \right] \]

\[ A_{4s}^{(2+nR)} \propto i\pi A^{(2)}(\omega_{4s,3s}) A^{(nR)}(0) + \phi \int_{-\infty}^{\infty} d\delta \frac{1}{\delta} A^{(2)}(\omega_{4s,3s} - \delta) A^{(nR)}(\delta) \]

\[ A_{4s}^{(2+rR)} \propto i\pi A^{(2)}(\omega_{4s,3s}) A^{(rR)}(0, \omega_{7p,4s}) + \phi \int_{-\infty}^{\infty} d\delta \frac{1}{\delta} A^{(2)}(\omega_{4s,3s} - \delta) A^{(rR)}(\delta, \omega_{7p,4s}) \]

where

\[ A^{(2)}(\Omega) \equiv \int_{-\infty}^{\infty} E_N(\omega) E_N(\Omega - \omega) d\omega \]

\[ A^{(nR)}(\Delta\Omega) \equiv \int_{-\infty}^{\infty} E_N(\omega) E_N^*(\omega - \Delta\Omega) d\omega \]

\[ A^{(rR)}(\Delta\Omega, \Omega_r) \equiv i\pi E_N(\Omega_r + \Delta\Omega) E_N^*(\Omega_r) - \phi \int_{-\infty}^{\infty} d\delta' \frac{1}{\delta'} E_N(\Omega_r + \Delta\Omega - \delta') E_N^*(\Omega_r - \delta') \]
The 2nd & 4th order perturbation theory description is valid up to $I_{\text{TL-peak}} \sim 5 \times 10^{10} \text{ W/cm}^2$.
Higher attenuation of the 2\textsuperscript{nd}-order term by the 4\textsuperscript{th}-order term for a TL pulse as compared to a pulse of π-step@777 nm !!!

Here – red-detuned pulses: 780 vs. 777 nm
For blue-detuned pulses: attenuation!

\begin{itemize}
\item 2\textsuperscript{nd}-order & 4\textsuperscript{th}-order terms are of opposite signs!
\end{itemize}
Intermediate Fields - Special Family of Pulses Shapes

Anti-Symmetric Phase Patterns

\[ \Phi\left(\frac{\omega_{4s,3s}}{2} - \Omega\right) = -\Phi\left(\frac{\omega_{4s,3s}}{2} + \Omega\right) \]

Calculations: 2nd + 4th Order Pert. Theory

Increased TL Intensity (higher 4th-order contribution)

Each hist: 5000 random anti-symmetric patterns

Phase patterns anti-symmetric around \( \omega_{4s,3s}/2 \) (777 nm):

Weak-field regime (2nd-order term) \( \rightarrow \) Two-photon absorption equal to the TL pulse.

Intermediate-field regime (2nd- & 4th-order terms) \( \rightarrow \) Exceed the absorption induced by the TL pulse !!!
Intermediate Fields - Special Family of Pulses Shapes

**Anti-Symmetric Phase Patterns**

\[
\Phi \left( \frac{\omega_{4s,3s}}{2} - \Omega \right) = -\Phi \left( \frac{\omega_{4s,3s}}{2} + \Omega \right)
\]

Experimental Results

**Increased Pulse Energy**
(higher 4\(^{\text{th}}\)-order contribution)

Each hist: 5000 **random** anti-symmetric patterns

**for Photo-Induced Information Processing:**
Discriminates between constant and anti-symmetric functions without the three-photon (7p) channel.
Intermediate-Field Regime

Best Anti-Symmetric Phase Pattern

\[ I_{\text{TL-peak}} = 5 \times 10^{10} \text{ W/cm}^2 \]

\[ P_{4s} = 5\% \]

\[ (S_{\text{TL-pulse}} = 0.7\pi) \]

Enhancement factor over TL = 4.9 : \( P_{4s} = 25\% \)

Enhancement factor over TL = 7.0 : \( P_{4s} = 35\% \)
\[ \text{on-resonant with 4s} \]

\[ \text{near-resonant with 4s} \]

\[ 4s \_ \text{Resonance-Mediated} \_ 4\gamma = \{2\gamma + \text{Resonant Raman}\} \]
Intermediate-Field Regime - Freq.-Domain Analysis:
Pulse shapes that enhance the two-photon absorption

Weak-Field Regime:
Coherent Decoupling of coupled multiphoton excitations

Information Processing aspects
THE END