Electromagnetically induced transparency with quantum interferometry

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This talk has been divided into parts

- **Quantum Interferometry of Wavepackets (QIW)** and how it is *controlled*.

- **Electromagnetically Induced Transparency (EIT)** and how it is *generated*.
Quantum Interferometry of Wavepackets:

Quantum interferometry of wavepackets is an emerging technique and it has a number of potential applications:

- Initially this technique has proved a success in the field of coherent control of atomic and molecular dynamics.

- With the advent of ultrashort lasers huge advancement in this field has occurred leading to precise control in wavepacket shaping and populating selective excited states.

- Utilizing its potentiality in coherent control, in the last decade there has been lot of progress in quantum computation and quantum information processing.


**What is quantum interferometry of wavepackets:**

- In this method, *two wavepackets* which are generated by *two delayed phase-locked pulses* are *allowed to interfere coherently* in the excited state of an atom or molecule.
- This *resulting wavepacket* assumes different *shape* and *amplitude* due to constructive or destructive interference of two wavepackets.
- The *shaping* of the interfering wavepacket is *controlled* in such a way that it *oscillates in phase with wavefunctions* of some *selective levels* in the excited state leading to *enhancement of excitation* in those selective levels.
- Simultaneously, it oscillates out of phase with wavefunctions of other levels leading to *suppression of absorption* in those levels.
- *Control over quantum interference* of two wavepackets can be achieved by controlling the *phase difference*, *delay*, *pulse duration* and *carrier frequency* of pulses.
For quantum interferometry of vibrational wavepackets ultrashort pulses are used.

An ultrashort laser pulse can access many vibrational eigenstates simultaneously in the molecule.

Wavepacket interferometry makes the use of ultrashort pulses to generate vibrational wavepackets on an excited state.

The vibrational wavepacket generated by a pulse is a superposition of a set of vibrational levels covered by the bandwidth of the pulse.

Procedure for quantum interferometry by control-probe method:

First wavepacket is excited from the ground state by the first ultrashort pulse.

First wavepacket is propagated freely on the excited state till the time (a delay) a second wavepacket is excited by the second ultrashort pulse.

The delayed second wavepacket excited by the probe pulse interferes with the first leading to interference of transition amplitudes to different vibrational levels excited by the carrier frequency and the other frequency components of the pulses.

The wavepacket designed from the interference of two excited wavepackets oscillates in phase with a set of selective vibrational levels odd(even) of the excited state and the oscillation is out of phase with the other set of vibrational levels even(odd) covered by the bandwidth of the pulses leading to enhancement and damping of overlap with these vibrational levels, respectively.
Quantum Interferometry of two vibrational wavepackets in molecules:

- The wavepacket generated by the control pulse is controlled or probed by the interference of the second wavepacket generated by another similar pulse delayed from the first.

- The nature and the degree of interference between the wavepackets depend on the delay and relative phase between the pulses and on the characteristics of the pulses such as temporal width and the carrier frequency.

- Designing of wavepacket due to interference of two wavepackets in the excited state. Thus enhancement and damping of population in the selective vibrational levels in the excited state.

Selective Vibrational excitation in NaH molecule
For evolution of wavepacket time dependent Schrodinger equation has been solved by Fourier Grid Hamiltonian method (one dimensional)

❖ Excitation by first pulse of duration $T$:

First wavepacket on the $A^1\Sigma^+$ is given by

$$i/\hbar \chi_{e1}(R,T) = \int_0^T dt_1 \exp\{-iH_e(T-t_1)/\hbar\} [D_{ge}(R)\varepsilon(t_1)] \exp\{-iH_g t_1 / \hbar\} \chi_{gv}(R,0)$$

$D_{ge}(R) \rightarrow$ Dipole transition moment for transition from ground state to $A^1\Sigma^+$ state.

$\varepsilon(t) \rightarrow$ Electric field amplitude of the pulse at time $t$

$\varepsilon(t) = g(t) \exp [i(\omega t + \theta)]$

$$g(t) = \frac{1}{\sqrt{2\pi\sigma}} e^{-t^2/2\sigma^2}$$

$H_e, H_g \rightarrow$ Hamiltonian for excited and ground states.

$\chi_{gv}(R,0) \rightarrow$ wavefunction of Ground vibrational level in $X^1\Sigma^+$ state

❖ The First wavepacket is propagated freely on the excited state for time delay $t'$
**Excitation by second pulse:**

The second pulse of duration $T'$ generates another wavepacket $\chi_{e2}$ on the $A^{1}\Sigma^{+}$ state of the same form as $\chi_{e1}$.

- The resultant wave packet on the $A^{1}\Sigma^{+}$ state is the sum of two wavepackets given by

\[ \chi_{e}(R,T + t' + T') = \chi_{e1}(R,T + t' + T') + \chi_{e2}(R,T') \]

- It is allowed to propagate on this state for a long time $t$ much greater than the vibrational time period of the resonant vibrational level of the $A^{1}\Sigma^{+}$ state.

- At each value of time autocorrelation function $F(t)$ is calculated, given by

\[ F(t) = \langle \chi_{e}(t_{0}) | \chi_{e}(t) \rangle \]

- Excitation Cross-section can be obtained from the Fourier transform of the autocorrelation function as

\[ \sigma = \{\pi \nu / 3c\varepsilon_{0}\hbar\} \int_{-\infty}^{\infty} \exp(iEt / \hbar)F(t)dt \]
Autocorrelation fn. for two 20 fs pulses at $\tau=5476$ a.u.
What is electromagnetically induced transparency (EIT)?

Electromagnetically induced transparency (EIT) is a process of creation of a narrow Transparency Window in an otherwise opaque medium using quantum interference.

- **EIT by interference of absorption amplitudes in a multilevel system:**

  - Frequency domain analysis

- **EIT by quantum interferometry (QI) of two vibrational wavepackets:**

  - Time domain analysis

In both the cases end result is EIT

EIT of probe light in a three level system is achieved by inducing destructive interference of absorption channels from the ground state to two dressed states obtained by coupling the excited state to another level of the system by a strong coupling laser.
Electromagnetically Induced Transparency (EIT) and how it is generated by quantum interference of wavepackets.

Quantum interference of wavepackets is controlled to excite either a set of even vibrational levels or a set of odd vibrational levels by controlling the phase difference and delay between two ultrashort pulses.

Electromagnetically Induced Transparency (EIT) window is generated between two absorption peaks at two consecutive even or two consecutive odd vibrational levels by suppressing absorption in odd/even vibrational level in between two consecutive absorption peaks.

Complete lossless transmission of ultrashort pulses is possible if the spectral bandwidth of the pulse (longer duration) is smaller than the energy difference between two consecutive odd or between two consecutive even vibrational levels of the molecule.

For pulses with spectral bandwidth greater (shorter duration) than the width of the transparency windows only band of frequencies are transmitted. Complete or partial transmission of pulse also depends on the carrier frequency.

Since the spacing between vibrational levels in molecules is large compared to spacing between hyperfine levels in atoms, EIT windows as large as tens of terahertz (∼23 × 10^{12} \text{ Hz}) can be achieved in the present method.
RESULTS

Excitation spectrum by single 4 fs pulse

Bound-bound spectrum for excitation from ground $X^1\Sigma^+$ state to the excited $A^1\Sigma^+$ state with single 4 fs pulse of Carrier frequency 25310 cm$^{-1}$ resonant with $v=10$ level of $A^1\Sigma^+$ state
Dependence of absorption to selective vibrational levels on phase difference of 4fs pump and control pulses:

**Bound-bound spectrum for** $\tau_{\text{control}} = 1.5 T_{\text{vib.}}, \phi = 0$

Absorption in odd vibrational levels is damped.

**Bound-bound spectrum for** $\tau_{\text{control}} = 1.5 T_{\text{vib.}}, \phi = \pi/2$

Absorption in even vibrational levels is damped.

**Bound-bound spectrum for** $\tau_{\text{control}} = 1.5 T_{\text{vib.}}, \phi = \pi$

Absorption in odd vibrational levels is damped.

**Bound-bound spectrum for** $\tau_{\text{control}} = 1.5 T_{\text{vib.}}, \phi = 3\pi/2$

Absorption in odd vibrational levels is damped.
Two-pulse spectrum for 20 fs pulses

Figure 1: Excitation cross section for excitation from \( X^1\Sigma^+ \) to \( A^1\Sigma^+ \) states of NaH molecule by two similar pulses (Control and Probe) with delay \( \tau = 5476 \) a.u., phase difference \( \phi = \pi/2 \) and carrier frequency = 25310 cm\(^{-1}\).
Comparison of spectra for single pulse and delayed phase locked two-pulse absorption

Delay = 5476 a.u., $\phi = \pi/2$

Pulse Duration = 20 fs

Carrier frequency = 25310 cm$^{-1}$

Generation of Electromagnetically Induced Transparency Window (EIT)

Transparency window between two absorption peaks at $v=9$ and $v=11$ levels is generated by suppressing absorption at $v=10$ level for 20 fs pulse of carrier frequency 25310 cm$^{-1}$. For single pulse excitation, absorption occurs at all vibrational levels $v=9$ to 11.
Interfering Wavepacket is controlled to oscillate in phase with \( V=9 \) and \( V=11 \) levels; and oscillation is out of phase with \( V=10 \) level.

Nature of oscillation is independent of pulse duration.

Carrier frequency 25310 cm\(^{-1}\) resonant with \( V=10 \) level.

Comparison of Wavepacket Oscillation with Vibrational levels.
Overlap of wavpacket with vibrational levels

- Overlap of $v=9$ and $v=11$ levels of $A^1\Sigma^+$ with $v=0$ of $X^1\Sigma^+$ is enhanced.
- Overlap of $v=10$ level of $A^1\Sigma^+$ with $v=0$ of $X^1\Sigma^+$ is damped.
Absorption peaks differ with carrier frequency of delayed phase-locked pulses (20 fs)

\[ \phi = \pi/2, \text{ delay} = 5476 \text{ a.u.} \]
Dependence of phase of oscillation of wavepackets on carrier frequency

\[ 24929 \text{ cm}^{-1} \]

\[ 25689 \text{ cm}^{-1} \]
Dependence of spectrum on carrier frequency of phase-locked delayed 20fs pulses

- For \( \text{Delay} \approx 5476 \text{ a.u.} \) \( \phi=\pi/2 \) only odd vibrational levels \((v=9 \text{ & 11})\) of \( A^1\Sigma^+ \) state are excited for all carrier frequencies from 24929 cm\(^{-1}\) to 25689 cm\(^{-1}\).

- Height of absorption peaks depends on carrier frequency.

- Absorption at \( v=10 \) level is suppressed for all the carrier frequencies.

- Energy difference of two absorption peaks at \( v=9 \) and \( v=11 \) levels is 760 cm\(^{-1}\).
Population code for different carrier frequency of 20 fs pulse

Twin pulse spectrum for different carrier frequency

Transparency Window of width 760 cm\(^{-1}\) remains unchanged for different carrier frequencies.

A transparency window of width sufficiently large for absorptionless transmission of 20 fs control pulse of carrier frequency 25310 cm\(^{-1}\) is achieved.
Two pulse spectrum for different pulse durations and phase difference $\Phi=3\pi/2$

**Cross Section for $\tau_{\text{control}} = 1.5 \tau_{\text{vib.}}, \phi=3\pi/2$**

**Carrier frequency=25310 cm$^{-1}$**

Overlap at $\tau=0$ for $\tau_{\text{control}} = 1.5 T_{\text{vib.}}, \phi=3\pi/2$
Comparison of spectra for single pulse and delayed phase locked two-pulse absorption for pulses with duration 10fs and 20fs

Dealy = 5504 a.u.; $\phi = 3\pi/2$
Carrier frequency = 25310 cm$^{-1}$

- For two delayed phase-locked pulses absorption occurs only at even vibrational levels.
- Two Electromagnetically Induced Transparency (EIT) Windows are generated between (i) $v=8 \& 10$ and (ii) $v=10 \& 12$
- For single pulse excitation absorption occurs at all the vibrational levels within the FWHM of pulse.
Comparison of Wavepacket Oscillation with Vibrational levels

\[ \tau_{\text{control}} = 1.5 \ T_{\text{vib.}}, \ \phi = \frac{3\pi}{2} \]

carrier frequency 25310 cm\(^{-1}\)
resonant with v=10 level

- Wavepacket oscillates in phase with v=10 level and
- out of phase with v=9 and 11 levels

\[ |\psi|^2 (R) \]

10 fs

\[ R \ (\text{a.u.}) \]

20 fs
For Dealy $1.5 T_{vib}; \phi=3\pi/2$ only even vibrational levels of $A^1\Sigma^+$ state are excited.

Absorption spectrum for pulses with carrier frequencies varying from 24549 cm\(^{-1}\) to 26068 cm\(^{-1}\) shows absorption only at even levels $v=8, 10 \& 12$.

Absorption at odd vibrational levels $v=9 \& 11$ is completely suppressed.

Height of absorption peaks depends on the carrier frequency.

Energy Gap (i) $v=8$ to $10=761$ cm\(^{-1}\), (ii) $v=10$ to $12= 758$ cm\(^{-1}\)
Generation of Electromagnetically Induced Transparency (EIT) Window

For $\phi=\pi/2$, delay = 5476 a.u.,
- Energy difference between two absorption peaks at $v=9$ & $11$ is 760 cm$^{-1}$ > FWHM of 20 fs pulse
- Single mode lossless transparency window for 20 fs Pulse of carrier frequency $=25310$ cm$^{-1}$

For $\phi=3\pi/2$, delay=5504 a.u.
- Double mode lossless transparency windows for 20 fs Pulses of carrier frequencies $24929$ cm$^{-1}$ (resonant with $v=9$ level) and $25689$ cm$^{-1}$ (resonant with $v=11$ Level).
- Width of the transparency Windows are 761 cm$^{-1}$ and 758 cm$^{-1}$ respectively > FWHM of 20 fs pulse
Transparency of pulses depends on pulse durations

- Odd vibrational levels of $A^1\Sigma^+$ state are excited for pulses of different durations 10 fs, 20 fs and 30 fs.
- Width of transparency window $>$ FWHM of 20 & 30 fs pulses.
- Lossless transmission of 20fs and 30fs pulses of carrier frequency $25310\text{ cm}^{-1}$
- Width of transparency window $<$ FWHM of 10 fs pulse.
- Transmission of Bands of frequencies of 10 fs pulse.
Lossless transmission of band of frequencies of 10 fs pulses through windows generated in NaH molecule

- For $\phi=\pi/2$, delay = 5476 a.u., three windows are generated between absorption peaks at $v=7$ & $9$; $v=9$ & $11$ and $v=11$ & $13$

- For $\phi=3\pi/2$, delay = 5504 a.u., two windows are generated between absorption peaks at $v=8$ & $10$ and $v=10$ & $12$,

- Three bands of frequencies of 10 fs pulse: (i) $24\ 171$ cm$^{-1}$ and $24\ 929$ cm$^{-1}$, (ii) $24\ 929$ cm$^{-1}$ and $25\ 689$ cm$^{-1}$ and (iii) $25\ 689$ cm$^{-1}$ and $26\ 444$ cm$^{-1}$ for pulses with three different carrier frequencies (i) $24\ 549$ cm$^{-1}$, (ii) $25\ 310$ cm$^{-1}$, and (iii) $26\ 068$ cm$^{-1}$ transmitted

- Two bands of frequencies between (i) $24\ 549$ cm$^{-1}$ and $25\ 310$ cm$^{-1}$ and (ii) $25\ 310$ cm$^{-1}$ and $26\ 444$ cm$^{-1}$ for pulses with two carrier frequencies $24\ 929$ cm$^{-1}$ and $25\ 689$ cm$^{-1}$

Triple mode and double mode transmission of band of frequencies of 10 fs pulses
CONCLUSION:

- Demonstrated that by controlling the phase difference and the delay between two femtosecond pulses, single- and multi-mode transmission of pulses of different carrier frequencies can be achieved by controlling the Quantum Interference of Vibrational wavepackets in diatomic molecules.

- The widths of the transparency windows depend on the spacing between different vibrational levels in the excited state. Hence, depending on the temporal width (spectral bandwidth) of the pulses, complete or partial transmission of pulses occurs without loss.

References:

Thank You for your Attention