Coherent transients mimicked by two-photon coherent control of a three-level system

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We show that two-photon coherent control in a V-shape three-level system projects one-photon coherent transient in a simple two-level system. Higher order chirps of a shaped laser pulse play the roles of time and linear chirp in conventional coherent transients. In a devised scheme of a three-pulse coherent excitation experiment, the phase and amplitude of controlled transition probability is retrieved from a 2D Fourier-transform spectral peak.

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I. INTRODUCTION

Recent advances in ultrafast laser and optical pulse shaping techniques have brought the use of shaped pulses of optical frequency for the manipulation of quantum systems [1-4]. This field, known as quantum control, though being started as a theoretical exercise, has rapidly become an experimental reality in a vast variety of materials extending from atoms and molecules to condensed matter and biological materials [5-9].

One of the simplest way to shape an optical pulse is to chirp, or to make a quadratic spectral phase, i.e.,
\[
\phi(\omega) = \frac{a_2}{2} (\omega - \omega_0)^2, \tag{1}
\]
where \(a_2\) is linear chirp rate and \(\omega_0\) is laser center frequency. Chirped pulses have been used to control molecular vibrational excitation and fragmentation [10, 11], coherent anti-Stokes Raman Scattering microscopy [12], molecular alignments [13], and high harmonic generation [14], to list a few. Of particular relevance in the context of the present paper is the chirped pulse excitation contributions can be easily understood in the frequency domain representation of CTs, given by [13]
\[
c_{21}(\tau) = \frac{\mu_{21}}{\hbar} \left[ i \pi \tilde{E}(\omega_{21}) + \varphi \int_{-\infty}^{\infty} \tilde{E}(\omega) \exp[i(\omega_{21} - \omega)\tau] d\omega \right], \tag{4}
\]
where the finite time integration with a quadratic temporal phase \(\alpha \tau^2\) leads to the transient excited-state population being of a Cornu spiral shape, well known from Fresnel diffraction pattern from a sharp edge [15].

For a short pulse which has broad spectral components, putting chirp on the pulse delays some of those components with respect to others in the time domain, and the instantaneous laser frequency shifts as a function of time. So, from the time when the resonant condition is met, further off-resonant excitation interferes, with the resonant transition, either constructively or destructively, and shows rather an oscillatory transient behavior. The quantum interference between resonant and non-resonant excitation contributions can be easily understood in the frequency domain representation of CTs, given by [13]
\[
\phi(\omega) = a_1(\omega - \omega_0) + \frac{a_2}{2} (\omega - \omega_0)^2 + \frac{a_3}{6} (\omega - \omega_0)^3, \tag{5}
\]
where \(E_0\) is laser center frequency.

We show in a V-shape three-level system the given two-photon coherent control is reduced to the discussed CT in a two-level system, where now \(a_2\) and \(a_3\) play the roles of the time and linear chirp in a regular CT. Furthermore, we apply this coherent control to a three-pulse coherent control scheme for two-dimensional Fourier-transform spectroscopy [22] and measure the amplitude and phase of two-photon inter-excited states transition coefficients.
II. TWO-PHOTON COHERENT CONTROL OF V-SHAPE THREE-LEVEL SYSTEM

We consider a three-level system in V-type configuration, composed of ground state |g⟩ and two excited states |a⟩ and |b⟩, with respective energies \( \hbar \omega_g, \hbar \omega_a, \) and \( \hbar \omega_b \). The excited states are dipole-coupled to the common ground state, with dipole moments \( \mu_{ag} \) and \( \mu_{bg} \), and the transition between the excited states is forbidden (i.e., \( \mu_{ab} = 0 \)). Then, the Hamiltonian is given by

\[
H(t) = H_0 + V(t),
\]

where

\[
H_0 = \sum_i \hbar \omega_i |i\rangle \langle i| \quad \text{and} \quad V(t) = -\sum_{i,j} \mu_{ij} E(t) |i\rangle \langle j| \quad \text{for} \quad i,j \in \{g,a,b\}. \]

With \( T = \exp(-iH_0/\hbar) \), we transform to the interaction picture obtaining the interaction Hamiltonian

\[
H_I(t) = T^\dagger H(t) T + i\hbar \frac{d}{dt} T = \sum_{i,j} V_{ij}(t)e^{i\omega_{ij}t}|i\rangle \langle j|, \quad \text{where} \quad V_{ij}(t) = \langle i|V(t)|j\rangle \quad \text{and} \quad \omega_{ij} = \omega_i - \omega_j.
\]

Then, the transition probability amplitude from state |i⟩ to state |f⟩, defined by \( c_{fi}(t) = \langle f|U_I(t,t_0)|i\rangle \), where \( U_I(t,t_0) \) is the evolution operator given by

\[
U_I(t,t_0) = 1 - \frac{i}{\hbar} \int_{t_0}^{t} H_I(t')U_I(t',t_0)dt',
\]

is obtained by the order of \( V(t) \) as

\[
c^{(0)}_{fi}(t) = \delta_{fi},
\]

\[
c^{(1)}_{fi}(t) = -i \frac{\hbar}{\mu_{fi}} \int_{t_0}^{t} dt' V_{fi}(t') \exp(i\omega_{fi}t'),
\]

\[
c^{(2)}_{fi}(t) = -i \frac{\hbar^2}{\mu_{fi}} \Sigma_{j} \int_{t_0}^{t} dt' \int_{t_0}^{t'} dt'' V_{fj}(t')V_{ji}(t'') \exp(i\omega_{fj}t' + i\omega_{ji}t'').
\]

For an electric field shaped in frequency domain as

\[
\tilde{E}(\omega) = A(\omega) e^{i\phi(\omega)},
\]

where \( A(\omega) \) is spectral amplitude and \( \phi(\omega) \) is spectral phase, the time domain pulse profile is given by

\[
E(t) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} \tilde{E}(\omega) e^{-i\omega t} d\omega,
\]

where the imaginary part of \( E(t) \) is maintained zero all the time by defining \( \tilde{E}(-\omega) = \tilde{E}^*(\omega) \) for \( \omega < 0 \). Then, \( V_{ij}(t) = -\mu_{ij} E(t) \), and the first order (one-photon) transition probability amplitude is simply the corresponding spectral amplitude, given by

\[
c^{(1)}_{fi} = i \frac{\mu_{fi}}{\hbar} \sqrt{2\pi} \tilde{E}(\omega_{fi}),
\]

where we consider the integral limit \([t_0,t] \rightarrow [-\infty, \infty]\) by assuming the pulse duration is considerably shorter than all lifetimes involved.

Now we consider two-photon transition probability amplitude between the excited states |a⟩ and |b⟩. From Eq. (9), the second order transition probability amplitude is obtained as

\[
c^{(2)}_{ba} = i \frac{\mu_{ga} \mu_{bg}}{\hbar^2} \left[ i\pi \tilde{E}^*(\omega_{ag}) \tilde{E}(\omega_{bg}) - \phi \int \tilde{E}^*(\omega) \tilde{E}(\omega_{ba} + \omega) d\omega \right].
\]

For a Gaussian pulse spectrally centered at \( \omega_0 \), i.e., \( A(\omega) = E_0 \exp(-[\omega - \omega_0]^2/\Delta \omega^2) \), with the spectral phase \( \phi(\omega) \) given in Eq. (9). The transition probability amplitude Eq. (13) is simplified to the following form

\[
c^{(2)}_{ba} = i \frac{\mu_{ba}}{\hbar^2} \left[ i\pi \tilde{E}(\omega) - \phi \int_{-\infty}^{\infty} \frac{\tilde{E}(\omega)}{\omega - \omega} d\omega \right],
\]

where \( \tilde{E}(\omega) = E_0^2 \exp(-2(\omega - \omega_0)^2/\Delta \omega^2 + i\omega \phi(\omega)) \).

It is noted that Eq. (14) is of a similar functional form to Eq. (1), the one-photon transition in a two-level system, except for the sign between the resonant and non-resonant contributions.

III. PROJECTION TO ONE-PHOTON TRANSITION IN A TWO-LEVEL SYSTEM

The difference between Eq. (1) and Eq. (14) is resolved by considering the de-excitation. For an electric field of gaussian pulse shape with linear chirp (only), the one-photon transition probability amplitude from |2⟩ to |1⟩ is easily found as

\[
c^{(1)}_{12}(t) = \frac{\mu_{21}}{\hbar} e^{-i(\omega_{21} - \omega_0)t} \left[ i\pi \tilde{E}^*(\omega_{21}) e^{i(\omega_{21} - \omega_0)t} - \phi \int_{-\infty}^{\infty} \frac{\tilde{E}^*(\omega)}{\omega - \omega_{21}} d\omega \right].
\]

As evident from the same structure, the two-photon inter-excited states transition in a V-type system projects one-photon transition (de-excitation) in a simple two-level system. tantalizing part is that, since \( \tilde{E}(\omega) \) has differentiated phase, linear chirp in V-shape system corresponds to time in two-level system, and minus quadratic chirp to linear chirp. Therefore, the obtained solution in Eq. (14), which is the transition probability amplitude \( c^{(2)}_{ba} \) for the two-photon inter-excited state transition in a V-type system, has become formally a one-photon transition probability amplitude, more specifically a de-excitation process, in a two-level system of energies 0 and \( \omega_0 \), induced by the newly defined electric field \( \tilde{E}(\omega) \).

Therefore, if we consider the interaction of the V-shape system with laser shaped pulse of linear and quadratic chirps, then we can achieve duplicated results of coherent transients in a two-level system interacting with a
linearily chirped pulse. With this information, we can derive the same form of Eq. (2) in V-shape system. For this, the electric field, $E(t)$ is the inverse Fourier transformation of the complex conjugate of electric field in frequency domain $\tilde{E}(\omega)$ having $t = \omega_{ba} a_2$ and $a_2 = -\omega_{ba} a_3$. Then, the “CT-like” transition probability amplitude in V-shape system becomes

$$
\alpha^{(2)}_{ba}(a_2, a_3) = -\frac{\hbar o_b}{\hbar^2} E_0^2 \frac{\Delta \omega}{\sqrt{\tau_c/\tau_0}} \exp\left(\frac{i}{2} \tan^{-1} \frac{2a_2}{\tau_0} + i\theta\right) \int_{-\infty}^{\infty} dt' \exp\left[-\frac{t'^2}{\tau_c} - i\left((\omega_{ba} - \omega_0)t' - \tilde{\alpha} t'^2\right)\right],
$$

where $\tilde{\alpha} = \omega_{ba} a_2 + t$, $a_2 = -\omega_{ba} a_3$, $\tau_c = 2\sqrt{2}/\Delta \omega$, $\tau_0 = 2\sqrt{2}/\tau^4$, and $\theta = -(\omega_{ba} - \omega_0)t$, and $\tilde{\alpha} = 2\omega_{ba}/(\tau_0^2 + 4\omega_{ba}^2)$. 

IV. THREE-PULSE EXCITATION SCHEME

The two-photon control in the previous section can be verified by measuring the phase and amplitude of $\alpha^{(2)}_{ba}(a_2, a_3)$. This is achieved by using a three-pulse excitation scheme in a two-dimensional Fourier-transform spectroscopy. The second pulse is the control pulse which induces two-photon inter-excited states transition from ground state, i.e., $|a\rangle$. For this, the atoms need to be excited to $|a\rangle$ by a pre-pulse. In addition, a third pulse is necessary to measure $\alpha^{(2)}_{ba}(a_2, a_3)$. The measurement of the phase and amplitude of $\alpha^{(2)}_{ba}(a_2, a_3)$ via three-pulse coherent excitation is explained in the following.

As our starting point, the quantum system is in the ground state, i.e., $|\psi(t = 0)\rangle = |g\rangle$. By assuming the interaction in the weak field regime (i.e., $V_{ij}(t) \ll \hbar$ for all $t$), we neglect the higher order terms of $V(t)$ and consider the lowest order terms of each transitions. Then, the evolution operator for the first pulse is written in terms of three states $\{|g\rangle, |a\rangle, |b\rangle\}$ as

$$
U_1(\alpha) = \begin{pmatrix}
1 & \alpha^{(1)}_{ag} & \alpha^{(2)}_{ab} \\
\alpha^{(1)}_{ag} & 1 & \alpha^{(2)}_{ab} \\
\alpha^{(1)}_{bg} & \alpha^{(2)}_{ab} & 1
\end{pmatrix},
$$

where $\Delta \omega_{ij} = \omega_{ij} - \omega_0$. Then, after the first pulse, the wave function becomes $|\psi(\tau_1)\rangle = U_1(\beta)|\psi(0+)\rangle$. Likewise, the evolution operator for the second pulse, including the time delay effect, is given by

$$
U_2(\beta) = \begin{pmatrix}
1 & \beta^{(1)}_{ag} e^{-i\Delta \omega_{ag} \tau_1} & \beta^{(1)}_{bg} e^{-i\Delta \omega_{bg} \tau_1} \\
\beta^{(1)}_{ag} e^{i\Delta \omega_{ag} \tau_1} & 1 & \beta^{(2)}_{bg} e^{i(\Delta \omega_{bg} - \Delta \omega_{ag}) \tau_1} \\
\beta^{(1)}_{bg} e^{i\Delta \omega_{bg} \tau_1} & \beta^{(2)}_{bg} e^{-i(\Delta \omega_{bg} - \Delta \omega_{ag}) \tau_1} & 1
\end{pmatrix},
$$

where $\omega^{(1)}_{ij} = \omega_{ij} - \omega_0$. Then, after the second pulse, the wave function becomes $|\psi(\tau_2)\rangle = U_2(\gamma)|\psi(\tau_1)\rangle$. Likewise, the evolution operator for the third pulse, including the effect of the time delay $\tau_2$ relative to the second pulse, is given by

$$
U_3(\gamma) = \begin{pmatrix}
1 & \gamma^{(1)}_{ag} e^{-i\Delta \omega_{ag} (\tau_1 + \tau_2)} & \gamma^{(1)}_{bg} e^{-i\Delta \omega_{bg} (\tau_1 + \tau_2)} \\
\gamma^{(1)}_{ag} e^{i\Delta \omega_{ag} (\tau_1 + \tau_2)} & 1 & \gamma^{(2)}_{bg} e^{i(\Delta \omega_{bg} - \Delta \omega_{ag}) (\tau_1 + \tau_2)} \\
\gamma^{(1)}_{bg} e^{i\Delta \omega_{bg} (\tau_1 + \tau_2)} & \gamma^{(2)}_{bg} e^{-i(\Delta \omega_{bg} - \Delta \omega_{ag}) (\tau_1 + \tau_2)} & 1
\end{pmatrix},
$$

where $\alpha^{(1,2)}$ denote the transition probability amplitudes, respectively defined in Eqs. (3) and (4), for the first pulse. (Likewise, $\beta^{(1,2)}$ and $\gamma^{(1,2)}$ denote the ones for the second and third pulses, in the following.) We note that $\alpha^{(1)}_{ba} = \alpha^{(1)*}_{ab}$ but $\alpha^{(2)}_{ba} \neq \alpha^{(2)*}_{ab}$. Then, the wave function after the first interaction is given by $|\psi(0+)\rangle = |g\rangle + \alpha^{(1)}_{ag} |a\rangle + \alpha^{(1)}_{bg} |b\rangle$. For the second pulse, the time delay $\tau_1$ causes an overall phase shift of $\exp[i(\omega_{ag} - \omega_0)\tau_1]$ to the $V_{ij}(t)\exp(i\omega_{ij})$ term in Eqs. (5) and (6), relative to the ones for $\alpha^{(1,2)}$. Therefore, the first and second order transition probability amplitudes for the second pulse, including the phase shift from the time delay, are obtained, respectively, $\beta^{(1)}_{fg} \exp[i(\omega_{fg} - \omega_0)\tau_1]$ and $\beta^{(2)}_{fg} \exp[i(\omega_{fg} - \omega_0)\tau_1] - i(\omega_{fg} - \omega_0)\tau_1$, where the rotating wave approximation is used for $\beta^{(2)}_{ba}$. Accordingly, the evolution operator for the second pulse, including the time delay effect, is given by
and, after all the three pulsed interactions, the final wave function $|\psi(t_1 + t_2)\rangle$ is obtained as the sum of twenty different terms. By measuring the projection to the $|b\rangle$ state, the probability $P_b = |\langle b|\psi\rangle|^2$ is given by

$$P_b(t_1, t_2) = |\alpha_{ag}^{(1)}|^2 + |\beta_{bg}^{(1)}|^2 + |\gamma_{bg}^{(1)}|^2 + \ldots$$

$$+ |\alpha_{ag}^{(1)}|^{*} |\beta_{bg}^{(2)*}|^{*} |\gamma_{bg}^{(1)}|^{*} e^{i(\Delta\omega_{ag}t_1 + \Delta\omega_{bg}t_2)} + \ldots,$$

where, for example, the term $\alpha_{ag}^{(1)} |\beta_{bg}^{(2)*}|^{*} |\gamma_{bg}^{(1)}|^{*} e^{i(\Delta\omega_{ag}t_1 + \Delta\omega_{bg}t_2)}$ denotes the quantum interference between the two transitions $|g\rangle \rightarrow |a\rangle \rightarrow |b\rangle$ and $|g\rangle \rightarrow |b\rangle$. The coefficient $\alpha_{ag}^{(1)} |\beta_{bg}^{(2)*}|^{*} |\gamma_{bg}^{(1)}|^{*}$ is retrieved from $|\langle b|\psi\rangle|^2$, as the amplitude and phase of the temporarily modulated component with the function $i(\Delta\omega_{ag}t_1 + \Delta\omega_{bg}t_2)$. The modulation $\exp(i\Delta\omega_{ag}t_1)$ and $\exp(i\Delta\omega_{bg}t_2)$ are from the phase evolution that the atoms are respectively in state $|a\rangle$ during $t_1$ and in state $|b\rangle$ during $t_2$. The 2D Fourier-transform spectrum is defined as

$$S(\omega_1, \omega_2) = \int \int P_b(t_1, t_2) e^{-i(\omega_1 t_1 + \omega_2 t_2)} d\tau_1 d\tau_2,$$

which has 49 peaks including a zero frequency peak [22]. The coefficients of the spectral peaks of $S(\omega_1, \omega_2)$ in the first quadrant of the two-dimensional plane are listed in TABLE I. Aside from the constant $\alpha_{ag}^{(1)} |\beta_{bg}^{(1)}|^{*}$, the controlled transition probability amplitude $|\beta_{bg}^{(2)*}|^{*}$ is then retrieved from the peak located at $(\omega_1, \omega_2) = (\Delta\omega_{ag}, \Delta\omega_{bg})$. As a result, the three-pulse coherent control scheme devised for 2D Fourier-transform spectroscopy can be used to measure the two-photon inter-excited states transition coefficients.

| $\omega_1$ \ $\omega_2$ | $\Delta\omega_{ag}$ | $\Delta\omega_{bg}$ | $\Delta\omega_{ag} - \Delta\omega_{bg}$ |
|------------------|-----------------|-----------------|-----------------|
| $\Delta\omega_{bg}$ | $\alpha_{ag}^{(1)} |\beta_{bg}^{(2)*}|^{*} |\gamma_{bg}^{(1)}|^{*}$ | $\alpha_{ag}^{(1)} |\beta_{bg}^{(1)}|^{*} |\gamma_{bg}^{(1)}|^{*}$ | $\alpha_{ag}^{(1)} |\beta_{bg}^{(1)}|^{*} |\gamma_{bg}^{(1)}|^{*}$ |
| $\Delta\omega_{ag}$ | $\alpha_{ag}^{(1)} |\beta_{bg}^{(1)}|^{*} |\gamma_{bg}^{(1)}|^{*}$ | $\alpha_{ag}^{(1)} |\beta_{bg}^{(2)*}|^{*} |\gamma_{bg}^{(1)}|^{*}$ | $\alpha_{ag}^{(1)} |\beta_{bg}^{(2)*}|^{*} |\gamma_{bg}^{(1)}|^{*}$ |

V. CONCLUSION

In summary, we have shown that two-photon coherent control in a $V$-shape three-level system behaves formally like a coherent transient signal in a two-level system, where the roles of time and linear chirp in the latter are duplicated by linear and quadratic chirp rates in the former. For the measurement, a three-pulse excitation scheme is devised, and the phase and amplitude of the controlled transition probability is retrieved from a 2D Fourier-transform spectral peak. It is hoped that this control scheme may underpin coherent control capability on 2D Fourier-transform spectroscopy.

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