Prevention of decoherence by two femtosecond chirped pulse trains

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We study the vibrational energy relaxation (VER) and collisional dephasing as channels of coherence loss in a vibrational mode that is selectively excited using chirped pulse adiabatic passage in the Raman configuration. Based on the dressed state picture analysis we propose a method to reduce decoherence using femtosecond chirped pulse trains. When applied with a period close to the VER time, the pulse trains allow one to sustain high coherence in the selected vibrational mode. © 2008 Optical Society of America

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A series of femtosecond pulses have been utilized in the frequency comb spectroscopy to control atomic and molecular systems [1–3]. In [1] a coherent train of weak pump-dump pulse pairs is implemented to perform the narrowband Raman transitions using pulse area as a control parameter. A piecewise adiabatic passage is demonstrated in [2], where a series of femtosecond transform-limited pulses are utilized. The experiments on the molecular trace presence and spectroscopic parameters evaluation are described in [3]. A theoretical description of pulse-train excitation of a two-level system has been carried out in [4], revealing conditions for complete population inversion and return.

In this Letter we implement chirped pulse trains to control and reduce decoherence on the time scale close to the incident pulse duration. The problem of decoherence is a challenge for a variety of research fields, from control of chemical reactions [5] to quantum computing [6]. To preserve high coherence in the presence of dephasing caused by vibrational energy relaxation (VER) and fast collisions we propose to use a series of femtosecond pulses that are chirped as described in [7]. The phase difference between the pump and Stokes pulses in each pair is locked. This technique is of vital importance in investigations based on selective excitation of Raman transitions in the presence of decoherence. An example of applications is noninvasive imaging of live tissues with implementation of the coherent anti-Stokes Raman scattering (CARS) microscopy [8,9].

We model molecular vibrational modes by two-level systems (TLSs), Fig. 1. Transition frequencies of the TLSs $\omega_{21,43}$ are chosen to be very close to feature a typically dense vibrational frequency spectrum, e.g., $\omega_{43} - \omega_{21} = \delta \ll \omega_{21}$. Initially only ground states $|1\rangle$ and $|3\rangle$ are evenly populated. Our goal is to generate and sustain high-level coherence in the $|1\rangle$–$|2\rangle$ TLS and zero coherence in the $|3\rangle$–$|4\rangle$ TLS in the presence of decoherence caused by VER and collisional dephasing (CD). Raman transitions are induced by a series of ultrafast pump and Stokes pulses having central frequencies $\omega_{p,s}$. These pulses are described as

\[ E_{p,s}(t) = E_{p,s}^{(0)}(t) \cos(\omega_{p,s}(t-t_0) + \alpha_{p,s}(t-t_0)^2/2), \]

where $E_{p,s}^{(0)}(t) = E_0 \exp(-(t-t_0)^2/2\sigma_{p,s}^2)$ are the pump and Stokes pulse envelopes, $\tau_{p,s} = \tau_0[1 + \alpha_{p,s}^2/\tau_0^2]^{1/2}$ are the chirp-dependent pulse durations, and $\tau_0$ is the duration of a transform-limited pulse [10]. Note that for $\alpha_{p,s} = 0$, $\omega_{p,s} = \omega_{21}$, the temporal ($\alpha$) and spectral ($\alpha'$) chirps are related as $\alpha_{p,s} = \alpha'_{p,s} \tau_0^2/(1 + \alpha_{p,s}^2/\tau_0^2)$.

In [7], the “roof” method was proposed implementing specifically chirped femtosecond pulses to perform the adiabatic passage that results in zero coherence $\rho_{34}$ in the detuned $|3\rangle$–$|4\rangle$ TLS and maximum coherence $\rho_{12}$ in the resonant $|1\rangle$–$|2\rangle$ TLS. Here we analyze VER and CD as factors that cause decoherence in the selectively excited TLSs when characteristic times are close to the pulse duration. A method is then developed to reduce decoherence by means of two coherent pulse trains consisting of chirped pulses pairs as in [7].

The interaction Hamiltonian for a TLS reads $H = \Lambda \hat{\sigma}_z/2 + \Omega_3(t)\hat{\sigma}_x$ [11], where $\hat{\sigma}_x$ and $\hat{\sigma}_z$ are the Pauli matrices, $\Lambda = \delta + (\omega_s - \omega_p)(t-t_0)$ is the effective detuning (for the resonant TLS, $\delta = 0$), $\Omega_3(t) = \mu^2 E_{p}^{(0)}(t) E_{s}^{(0)} \times (t - \tau_0)/\tau_0^2/2$ is the effective Rabi frequency, $\mu^2 = \mu^2/4\hbar^2 \Delta$ is the dipole moment, and $\Delta$ is the detuning from the excited state. Using the Liouville von Neumann equation we derived a set of differential equations for the density matrix that reads

\[ \dot{\rho}_{11} = -2\Omega_3(t) \text{Im} [\rho_{21}] + \gamma_2 \rho_{22}, \quad \dot{\rho}_{22} = 2\Omega_3(t) \text{Im} [\rho_{21}] - \gamma_2 \rho_{22}, \quad \dot{\rho}_{12} = i\Omega_3(t) (\rho_{22} - \rho_{11}) - \gamma_2/2 + \Gamma - i\Lambda) \rho_{12}. \]

The $\gamma_2 = 1/\tau_1$ is the VER rate, and $\Gamma = 1/T_\rho$ is the CD rate. The $\gamma_2$ and $\Gamma$ can be originated from the inelastic and elastic collisions, respectively. We used indexes 1 and 2 to label the states of the resonant TLS, in which case $\delta = 0$. For the detuned TLS, the equations can be obtained by changing the former indexes to 3 and 4, and $\delta$ must be kept nonzero.

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**Fig. 1.** Schematic of the TLSs representing Raman active vibrational modes.

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The values of parameters used in numerical calculations are \( \omega_{21} = 85.35 \text{ THz} \) (2845 cm\(^{-1}\)) and \( \omega_{43} = 88.68 \text{ THz} \) (2956 cm\(^{-1}\)). These are the frequencies of CH\(_2\) and CH molecular groups, which are abundantly present in various biological tissues. The field intensity is \( 28.4 \times 10^{12} \text{ W/cm}^2 \), \( \gamma_0 = 177 \text{ fs} \), and \( \alpha' = 2.8 \times 10^{-4} \text{ cm}^{-2} \), giving \( \tau = 1.58 \text{ ps} \). These parameters are chosen in accordance with [7] to provide the adiabatic passage. The \( \gamma_2 \) and \( \Gamma \) vary from 0 to 0.085 THz, giving the time of VER and CD the flexibility to change from infinity to 11.76 ps. We apply a single pair of pump and Stokes laser pulses chirped as in [7] to induce Raman transitions in the TLSs. In Fig. 2, dotted lines show the dependence of \( \rho_{12} \) on \( \gamma_2/\omega \) for various values of \( \Gamma/\omega_1 \); from \( \Gamma/\omega = 0 \) (the upper curve) to \( \Gamma/\omega = 10^{-3} \) (the lowest curve), solid curves show the dependence of \( \rho_{12} \) on \( \Gamma/\omega \) for analogous values of \( \gamma_2/\omega \). The \( \gamma_2 \) and \( \Gamma \) are given in the units of \( \omega = \omega_{21} \). Given \( \omega_{21} = 84.9 \text{ THz} \), the value \( \gamma_2/\omega = \Gamma/\omega = 10^{-3} \) corresponds to \( \gamma_2 = \Gamma = 0.085 \text{ THz} \). For \( \gamma_2 = 0 \), increasing \( \Gamma \) from 0 to 0.085 THz causes a decrease in \( \rho_{12} \) from the maximum value (0.25) to 0.065. While for \( \Gamma = 0 \), increasing \( \gamma_2 \) causes \( \rho_{12} \) to decrease twice as slow. This demonstrates that the dynamics of induced coherence is very sensitive to the CD, and to a less extent, to the VER.

To understand dynamics in the resonant TLS in the presence of CD and VER, we refer to the time-dependent picture. Figure 3(a) shows coherence and populations as a function of time for two sets of \( \gamma_2/\omega \) and \( \Gamma/\omega \): for \( \gamma_2/\omega = 0 \) and \( \Gamma/\omega = 10^{-3} \) (solid curves) and for \( \gamma_2/\omega = 10^{-3} \) and \( \Gamma/\omega = 0 \) (dashed curves). The CD causes more rapid coherence decay, shown by \( \rho_{12}^{(ad)} \), in comparison to the effect caused by the VER, shown by decay of \( \rho_{12}^{(cd)} \).

Let us consider the case of strong collisions, e.g., \( \gamma_2 = \Gamma = 0.085 \text{ THz} \). In Figs. 3(b) and 3(c) coherence and populations are depicted in two representations: in the field interaction representation as \( \rho_{ij} \) and in the dressed state picture as \( \rho_{ij}^{(d)} \) obtained from the balance equations

\[
\rho_{11}^{(d)} = - (\gamma_2 \sin^2 \Theta + 1/2(\Gamma - \gamma_2/2)\sin^2 2\Theta)\rho_{11}^{(d)} + (\gamma_2 \cos^2 \Theta + 1/2(\Gamma - \gamma_2/2)\sin^2 2\Theta)\rho_{22}^{(d)},
\]

where \( \sin \Theta = \sqrt{1/2(1-R\lambda)}, \cos \Theta = \sqrt{1/2(1+R\lambda)}, \) and \( R = \sqrt{\lambda^2 + 4 \Omega_3(t)^2} \). For the detuned TLS [Fig. 3(b)], \( \rho_{dd} \) (blue online) reaches to a maximum and then falls to zero following the dynamics of the \( \Omega_3(t) \) (dotted curve). The \( \rho_{ii}^{(d)} \) (dashed curve) relaxes to respective \( \rho_{ii} \) (solid curve) asymptotically. In the resonant TLS [Fig. 3(c)], \( \rho_{12} \) (blue online) reaches a maximum at the peak value of the \( \Omega_3(t) \) (dotted curve) and then slowly decays under the influence of strong collisions. The \( \rho_{ii}^{(d)} \) (dashed curve) monotonously changes in time and asymptotically approaches an equilibrium value; the \( \rho_{22} \) returns to the ground state (solid curves). The relation between the \( \rho_{ii} \) and the \( \rho_{ij} \) is \( \rho_{ii}^{(d)} = \cos^2 \Theta \rho_{ii} + \sin^2 \Theta \rho_{ii} - 2 \sin \Theta \cos \Theta \text{Re}[\rho_{12}], \rho_{ii}^{(d)} = \sin^2 \Theta \rho_{ii} + \cos^2 \Theta \rho_{ii} + 2 \sin \Theta \cos \Theta \text{Re}[\rho_{12}] \). Accordingly, at \( t \to \infty \), for the detuned TLS and \( \delta = 0 \), the coefficients are \( \sin^2 \Theta = 0 \) and \( \cos^2 \Theta = 1 \). Under these conditions, the \( \rho_{ii}^{(d)} \) relaxes to the \( \rho_{ii} \) and the \( \rho_{ii}^{(d)} \) relaxes to the \( \rho_{22} \). In the resonant TLS and \( \delta = 0 \), the asymptotic solution is qualitatively different; it gives \( \sin^2 \Theta = \cos^2 \Theta = 1/2 \) and \( \rho_{ii}^{(d)} = 1/2(\rho_{ii} + \rho_{22}) - \text{Re}[\rho_{12}], \rho_{ii}^{(d)} = 1/2(\rho_{11} + \rho_{22}) + \text{Re}[\rho_{12}], \) which means that at \( t \to \infty \), \( \rho_{ii}^{(d)} \) and \( \rho_{ii}^{(d)} \) are equal to \( \rho_{ii}^{(d)} \).

Periodic restoration of population \( \rho_{22} \) of the upper level in the resonant TLS provides the asymptotic solution for the dressed states to be a superposition of populations \( \rho_{11} \) and \( \rho_{22} \) as well as coherence \( \rho_{12} \). This is the essence of the mechanism to counteract decoherence. To do so in the selectively excited TLS, we propose to use two femtosecond coherent pulse trains that are linearly chirped as in [7]. When applied with the train period \( T \) comparable with the VER time, they create and sustain high coherence in the selected TLS.

Figure 4(a) shows the result of applying two sequential pump and Stokes pulse pairs with \( T = 44 \text{ ps} \). The black curve shows the \( \Omega_{12}(t)/\omega \), which is the same for both pump and Stokes pulses. Populations and coherence, calculated in the presence of only VER, are...
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...mum value during the first pulse pair and undergoes decay up to the moment when the second pulse pair arrives and restores population of the upper level as well as coherence. Notably, the second pulse pair does not affect the $\rho_{12}^{\text{cd}}$ because it is not coupled to the population distribution. The restoration of coherence takes place because the $T$ is longer than the VER time. This results in almost complete relaxation of the population to the ground state in the resonant TLS and return of the system into the initial condition. Figures 4(b) and 4(c) show the time-dependent picture of the TLS’s parameters due to the sequence of pump and Stokes pulses having $T=11$ ps, close to the VER and CD. After the first pulse pair, the $\rho_{12}^{\text{er}}$ (dashed curve) decreases to 0.10 due to the VER and, subject to interaction with the subsequent pulses, varies within a 0.10–0.15 region. This is because the $\rho_{22}$ in the TLS does not fully relax to the lower level between pulses. This population distribution determines the initial condition for each subsequent pulse pair, which provides partial adiabatic population transfer to the upper level and preserves coherence within the indicated region. The choice of the $T$ depends on one’s need to either maintain uninterrupted coherence in the desired TLS or to get a maximum coherence periodically. In the detuned TLS, chirped pulse trains suppress the generation of coherence by preserving the population in the ground state. Thus, the proposed method satisfies the goal of selective excitation and creation of coherence in a vibrational mode having known frequency in the presence of decoherence. The method is feasible in the case when the intensity of each pulse in the train is uniformly reduced so that the total energy of the pulse trains is equal to the energy of a single pulse pair applied in Fig. 3, as long as the pulses are strong enough to fulfill the adiabatic passage. The method is robust with regard to the form of the pulse envelope, the value of the linear chirp, and the field intensity.

In summary, we propose a new method to control molecular vibrational excitations and sustain high coherence in a predetermined vibrational mode in the presence of decoherence using the pump and Stokes femtosecond coherent pulse trains having the linear chirp. The pulse trains period must be close to the time of the VER. High coherence is sustained in a selectively excited vibrational mode in the presence of decoherence due to a periodic adiabatic pumping of the population to the upper level. The method can be used for noninvasive imaging of biological structure and femtosecond dynamics studies and as an efficient tool for suppression of the background signal in CARS spectroscopy and microscopy. It demonstrates the possibility of coherent control over spectrally close vibrational modes in nonimpulsive Raman scattering.

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References