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Decoherence control in quantum computing with simple chirped pulses

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Abstract. We show how the use of optimally shaped pulses to guide the time evolution of a system ('coherent control') can be an effective approach towards quantum computation logic. We demonstrate this with selective control of decoherence for a multilevel system with a simple linearly chirped pulse. We use a multiphoton density-matrix approach to explore the effects of ultrafast shaped pulses for two-level systems that do not have a single photon resonance, and show that many multiphoton results are surprisingly similar to the single-photon results. Finally, we choose two specific chirped pulses: one that always generates inversion and the other that always generates self-induced transparency to demonstrate an ensemble CNOT gate.

Keywords. Coherent control; multiphoton excitation; pulse shaping; quantum logic gates.

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1. Introduction

The concept of quantum computer (QC) has attracted considerable attention recently due to its massive parallelism provided by the superposition of the quantum states, which bear no classical analog. Building a quantum computer is a big challenge – the very thing that makes quantum computing so powerful, the coherent superposition of states, also makes it very fragile and difficult to control. Use of optimally shaped pulses to guide the time evolution of a system and thereby control its future would be an effective approach towards quantum computation logic [1–3]. Optical spectroscopy of molecules could potentially become a highly scaleable approach but for the intramolecular relaxation processes, such as intramolecular vibrational relaxation (IVR), which is the most important contributor to decoherence even in isolated molecules. Restriction of IVR by optical schemes [4,5] can be an attractive route towards selective excitation in large molecular systems. Albeit attractive, most of the photon-mediated approaches towards restricting IVR (also called 'photon-locking') use complicated pulse shapes that are not yet demonstrated in the laboratory due to stringent requirements of intensity and precision.

In this paper, however, we show that even simple linearly chirped pulses could restrict IVR in systems at least as complicated as investigated earlier [4,5]. Such results hold even in the extreme case of a two- or multi-photon transition occurring with a chirped pulse, where the lower-order photon processes are non-resonant [6,7], which makes these results even more attractive. We also investigate the case of phase modulated overlapped Gaussian

pulses for two-photon transition. In case of two-level systems, such shaped-pulses result either in population inversion or self-induced transparency.

Exerting selective optical control over molecules in an ensemble is as unique as addressing individual spins in a NMR spectrometer and could essentially lead to *quantum computing in bulk optical systems*. The main aim in 'coherent-control' has been controlling an observable, while in the case of 'quantum computation', individual logic gates would be implemented using the principle of controlling observables. The overall idea, therefore, is to use shaped-optical pulses, which, on interaction with a quantum system, would retain coherence for longer time so that a larger number of logic gates are implementable.

2. Formalism

An isolated two-level system or ensemble without relaxation or inhomogeneities is the simplest model describing a molecular system, which, in fact, often turns out to be a very practical model for most systems interacting with femtosecond laser pulses as the magnitude of the relaxation processes are immensely large compared to the femtosecond interaction time.

Let us apply a linearly polarized laser pulse of the form $E(t) = \varepsilon(t)\,\mathrm{e}^{i[\omega\cdot t+\phi(t)]} = \varepsilon(t)\,\mathrm{e}^{i\left[\omega+\dot{\phi}(t)\right]t}$ to a simple two-level system with $|1\rangle \rightarrow |2\rangle$ transition, where $|1\rangle$ and $|2\rangle$ represent the ground and excited eigenlevels, respectively, of the field-free Hamiltonian (figure 1). The instantaneous amplitude, phase and frequency sweep are given by $\varepsilon(t)$, $\phi(t)$ and $\dot{\phi}(t)$, respectively; ω is the laser carrier frequency or the center frequency for pulsed lasers; $\omega_{\mathrm{R}} = \omega_2 - \omega_1$ is the resonance frequency; $h\omega_1$, $h\omega_2$ are the energies of the ground and excited states; and μ is the transition dipole moment. The Hamiltonian for the simple case of a two-level system under the effect of an applied laser field can be written in the frequency modulated (FM) frame for N-photon transition [7] as $H^{\mathrm{FM}} = \hbar \begin{pmatrix} \Delta + N\dot{\phi}(t) & \Omega \\ \Omega^{*} & 0 \end{pmatrix}$. The time derivative of the instantaneous phase function, $\dot{\phi}(t)$, appears as an additional resonance offset, over and above the time-independent multiphoton detuning $\Delta = \omega_{\mathrm{R}} - N\omega$, while the direction of the field in the orthogonal plane remains fixed. We define the multiphoton Rabi frequencies as complex conjugate pairs: $\Omega(\mathrm{t})=(\mu_{\mathrm{eff}}.\varepsilon(t))^N/\hbar$ and $\Omega^{*}(t) = (\mu^{*}_{\mathrm{eff}}.\varepsilon(t))^N/\hbar$. For the $|1\rangle \rightarrow |2\rangle$ transition, $\omega_{\mathrm{R}} = \omega_2 - \omega_1$ is the single-photon resonance frequency. We have assumed that the transient dipole moments of the individual intermediate virtual states in the multi-photon ladder result in an effective transition dipole moment, μ^N_{eff}, which is a product of the individual N virtual state dipole moments μ_n, (i.e., $\mu^N_{\mathrm{eff}} = \Pi^N_n \mu_n$). This approximation is particularly valid when intermediate virtual levels are non-resonant and as such their multi-photon interaction dynamics can be neglected [6,7]. We use a density matrix approach by integrating

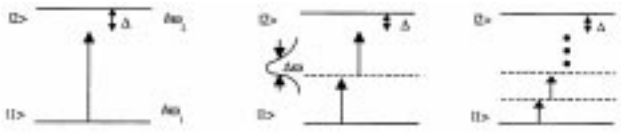

Figure 1. Schematic of single-, two- and multi-photon processes, respectively. Symbols and notations are defined in the text.

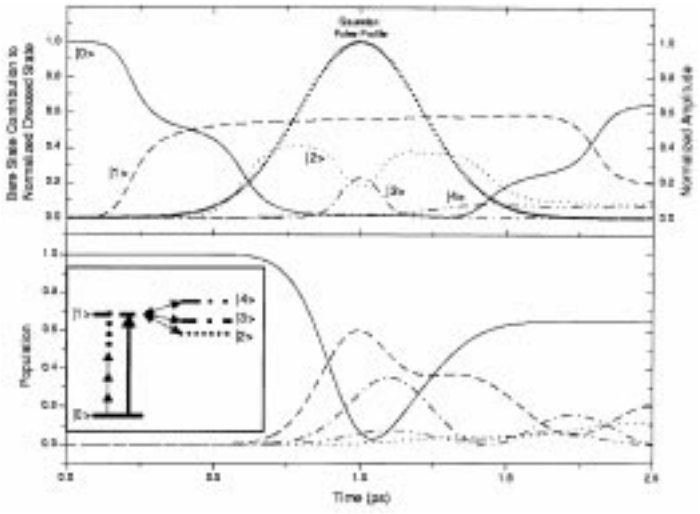

Figure 2. A transform-limited Gaussian pulse interacts with a model anthracene molecule (inset) in a single photon mode or in a multi-photon condition. Inset: Schematic of IVR for anthracene molecule from [5] based on data extracted from experimental measurements in [8].

the Liouville equation $\mathrm{d}\rho(t)/\mathrm{d}t = i/\hbar\left[\rho(t), H^{\mathrm{FM}}(t)\right]$ for a Hamiltonian in the rotating FM frame of reference. $\rho(t)$ is a 2×2 density matrix whose diagonal elements represent populations in the ground and excited states and off-diagonal elements represent coherent superposition of states.

Let us extend the two-level formalism to the multilevel situation involving IVR. In the conventional zeroth order description of intramolecular dynamics, the system can be factored into an excited state that is radiatively coupled to the ground state, and nonradiatively to other bath states that are optically inactive (figure 2 (inset)). These 'dark' states have no radiative transition moment from the ground state as determined by optical selection rules. They can belong to very different vibrational modes in the same electronic state as the 'bright' state, or can belong to different electronic manifolds. These dark states can be coupled to the bright state through anharmonic or vibronic couplings. Energy flows through these couplings and the apparent bright state population disappears. Equivalently, the oscillator strength is distributed among many eigenstates. The general multilevel Hamiltonian in the FM frame for an N-photon transition ($N \geq 1$), expressed in the zero-order basis set is

$$H^{\mathrm{FM}} = \begin{pmatrix} |0\rangle & |1\rangle & |2\rangle & |3\rangle & |4\rangle & \dots \\ 0 & \Omega(t) & 0 & 0 & 0 & \dots \\ \Omega^{*}(t) & \delta_1(t) & V_{12} & V_{13} & V_{14} & \dots \\ 0 & V_{12} & \delta_2(t) & V_{23} & V_{24} & \dots \\ 0 & V_{13} & V_{23} & \delta_3(t) & V_{34} & \dots \\ 0 & V_{14} & V_{24} & V_{34} & \delta_4(t) & \dots \\ \vdots & \vdots & \vdots & \vdots & \vdots & \end{pmatrix}, \tag{1}$$

where $\Omega(t)$ (and its complex conjugate pair, $\Omega^*(t)$) is the transition matrix element expressed in Rabi frequency units, between the ground state $|0\rangle$ and the excited state $|1\rangle$. The background levels $|2\rangle$, $|3\rangle, \ldots$ are coupled to $|1\rangle$ through the matrix elements V_{12}, V_{13}, etc. Both the Rabi frequency $\Omega(t)$ and the detuning frequency $[\delta_{1,2,\ldots} = \Delta_{1,2,\ldots} + N\dot{\phi}(t)]$ are time dependent (the time dependence is completely controlled by the experimenter). In general, the applied field would couple some of the dark states together, or would couple $|1\rangle$ to dark states, and thus, the V_{ij} terms would have both an intramolecular, time independent component and a field-dependent component. As an alternative to eq. (1), the excited states' submatrix containing the bright state $|1\rangle$ and the bath states $|2\rangle$, $|3\rangle, \ldots$ can be diagonalized to give the eigenstate representation containing a set of Δ_i' as diagonal elements and corresponding Ω_i' as off-diagonal elements. Such a representation corresponds closely to what is observed in conventional absorption spectroscopy. As long as the intensity of the field is very low ($|\Omega_i'| \ll \Delta_i'$) the oscillator strength from the ground state (and hence the intensity of the transition, which is proportional to $|\Omega_i'|^2$) is distributed over the eigenstates, and the spectrum mirrors the distribution of the dipole moment. On the other hand, a pulsed excitation creates a coherent superposition of the eigenstates within the pulse bandwidth. Physically, in fact, the presence of the dark states has been key to the loss of selectivity of excitation to a specified bright state.

We explore the effects of a linear sweep in frequency of the laser pulse (i.e., $\dot{\phi}(t) = 2b_2 t$) that can be generated by sweeping from far above resonance to far below resonance (blue to red sweeps), or the opposite. For a sufficiently slow frequency sweep, the irradiated system evolves with the applied sweep and the transitions are 'adiabatic'. If this adiabatic process is faster than the characteristic relaxation time of the system, such a laser pulse leads to a smooth population inversion, i.e., an adiabatic rapid passage (ARP) [8].

We also use the shaped overlapping Gaussian pulses for a two-photon transition that are linearly approaching and going away from resonance which is given by

$$\dot{\phi}(t) = bt, \text{ where } b \text{ changes sign at } t_0. \tag{2}$$

These pulses can also satisfy the ARP conditions. We also extend our calculations to the N-photon case in a simple two-level type of system that supports only an Nth photon transition and show how the phase switches affect the population cycling.

3. Results and discussion

Excitation exactly on resonance results in the population evolution for a simple two-level system without relaxation and creates a complete population inversion when the pulse area (the time integral of the Rabi frequency) equals π. However, the population oscillates between the ground and the excited state as a sine function of the Rabi frequency. These oscillations are not desirable in most cases involving real atoms or molecules. They are washed out by inhomogeneous broadening, the transverse Gaussian profile of the laser, and (in the molecular case) different values of $\mu.\varepsilon$. For a single-photon case, as discussed in [8], frequency modulated pulses can instead produce adiabatic inversion, which avoids these complications. From experimental results on the fluorescence quantum beats in jet-cooled anthracene [9], the respective values (in GHz) of $\Delta_{1,2,\ldots,4}$ are 3.23, 1.7, 7.57 and 3.7; and $V_{12} = -0.28$, $V_{13} = -4.24, V_{14} = -1.86$, $V_{23} = 0.29, V_{24} = 1.82$, $V_{34} = 0.94$. When

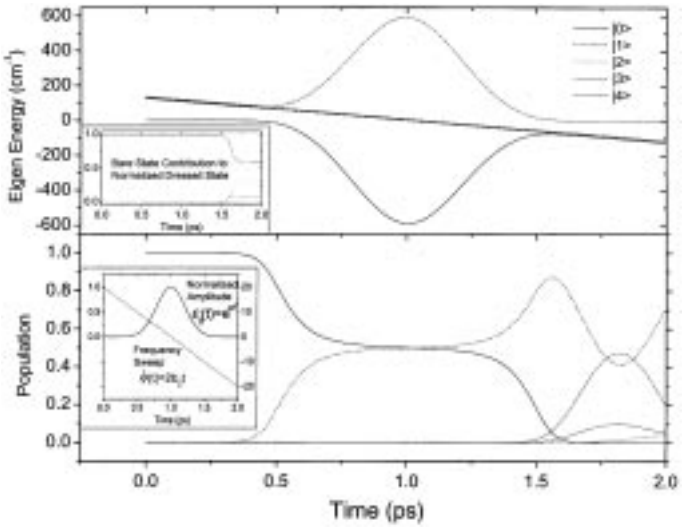

Figure 3. A transform-limited Gaussian pulse interacts with a model anthracene molecule (inset) in a single-photon mode or in a multi-photon condition. Inset: Schematic of IVR for anthracene molecule from [4] based on data extracted from experimental measurements in [9].

these values are incorporated in eq. (1), we obtain the full zero-order Hamiltonian matrix that can simulate the experimental quantum beats (figure 2) upon excitation with a transform-limited Gaussian pulse ($\dot{\phi}(t) = 0$). Since $|0\rangle$ and $|1\rangle$ do not form a closed two-level system, considerable dephasing occurs during the second half of the Gaussian pulse. Thus, in a coupled multilevel system, simple pulses cannot be used to generate sequences of $\pi/2$ and π pulses, as in NMR. The dark states start contributing to the dressed state well before the pulse reaches its peak and results in redistributing the population from the bright state (figure 2).

For a linearly frequency swept (chirped) laser pulse, when the frequency sweep is sufficiently slow such that the irradiated system can evolve with the applied sweep but faster than the characteristic relaxation time of the system, a smooth population inversion occurs with the evolution of the pulse (the well-known ARP). If the frequency sweeps from below resonance to exact resonance with increasing power, and then remains constant, adiabatic *half* passage occurs and photon locking is achieved with no sudden phase shift. However, even under adiabatic *full* passage conditions, figure 3 shows that there is enough slowing down of the E field to result in photon locking over the FWHM of the pulse. These results hold even under certain multi-photon conditions where only an Nth ($N \geq 2$) photon transition is possible [7]. Theoretically, scaling the number of dark states is possible as long as there is finite number of states and there are no physical limitations on Stark shifting.

We next use the overlapping Gaussian pulses (when the overlap is complete it collapses into a single Gaussian) with different phase relationships. Our simulation shows that for shaped overlapping Gaussian pulses the excited state population depends on the form of the frequency sweep. In an isolated two-level system that does not suffer from the population dephasing, the linear swept pulse yield inversion of population, while the overlapping

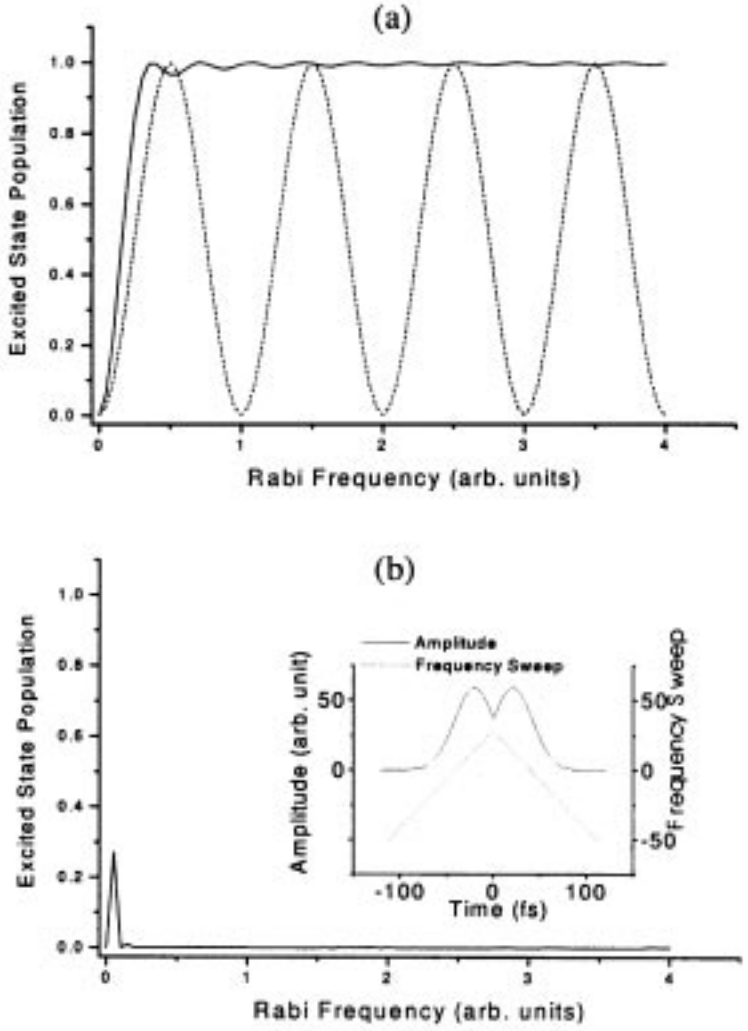

Figure 4. (**a**) Relative excited state population for 2-photon excitation (overlapped Gaussian envelop) as a function of Rabi frequency for a transform-limited (dashed line) vs. a bandwidth equivalent frequency-swept (solid line) pulse. (**b**) However, when the frequency sweep of the second pulse in the two-photon excitation is reversed with respect to the first pulse (see inset), a counter-intuitive self-induced transparency occurs.

Gaussian pulses for a two-photon transition that are linearly approaching and going away from resonance produce self-induced transparency (figure 4). For a smoothly varying linear frequency sweeping to the two-photon resonance and then away from resonance, as given by eq. (2), the results are quite robust to the exact nature of the amplitude profile and intensity. At zero detuning and for small values of Rabi frequency, we have some

population in the excited state. However, when the intensity of applied pulse increases, the excited state population returns to zero. In other words, we are sending shaped pulse into the two-level system but finally there is no excited-state population. Such 'dark pulses' given by eq. (2) are quite insensitive to the changes in the pulse amplitude profile.

For a multilevel system, the induced optical AC Stark-shift by the frequency swept pulse moves the off-resonant coupled levels far from the resonant state leading to an effective decoupling. Under the perfectly adiabatic condition, pulses with their linear sweep approaching and going away from resonance return the system to its unperturbed condition at the end. It is only during the pulse that the Stark-shifting of the dark states are decoupled and IVR restriction is possible in the multi-level situation.

The results are generic and illustrate that the intramolecular dephasing can be kept to a minimum for the duration of the 'locking' period under adiabatic conditions. Since the effect occurs under an adiabatic condition in all these frequency swept pulses, it is insensitive to the inhomogeneity in Rabi frequency. The simulations have been performed with laser pulses with Gaussian as well as hyperbolic-secant intensity profiles over a range of intensities. They show identical results of 'locking' the population in the chosen excited state of a multilevel system, conforming to the adiabatic arguments that there is hardly any effect of the actual envelope profile. Promoting novel chemical reactions during photon locking, or completing several quantum-computing operations can, thus, be accomplished within the pulse before dephasing randomizes the initially prepared state.

In terms of quantum computing, when a sequence of such experimentally achievable chirped-pulses act on a bulk system, and perform a series of quantum logic gates (e.g., AND, NOT etc.), it would essentially constitute 'ensemble quantum computing' [10]. Given the formalism of generalized chirped pulses discussed in this paper, we demonstrate the construction of a CNOT gate as an example of experimentally implementable QC gate involving the entanglement of coherent photon with quantum system. The truth table for this CNOT gate is shown in table 1 for a quantum mechanical ensemble B that can either be in the ground state (0) or excited state (1) interacting with the control pulse A, which provides robust chirped pulse inversion (condition 1) and the self-induced transparency or dark pulse (condition 0). Under the effect of inverting pulse the entanglement results in population inversion, while the effect of the dark pulse is to preserve the original state of the system. Shaped pulses are important for implementing such an ensemble gate since such coupled systems preclude the use of simple pulse area approaches of $\pi/2$ and π sequences for controlling coherence or inducing inversion. Such chirped pulses are also essential in restoring the system to its initial state without being limited by the natural lifetime of the system, which ensures speeding up the subsequent steps.

Table 1. CNOT gate with simple chirped pulses.

Shaped-pulse	A	B	A$\oplus$B
'Inverting' pulse	1	1	0
	1	0	1
'Dark' Pulse	0	1	1
	0	0	0

4. Conclusions

In this paper, we have explored the effects of ultrafast shaped pulses for two-level systems that do not have a single-photon resonance by developing a multi-photon density-matrix approach. We took advantage of the fact that the dynamics of the intermediate virtual states are absent in the femtosecond time-scales, and demonstrated that many multi-photon results can be surprisingly similar to the well-known single-photon results. When we extend the ARP to the multi-photon condition, robust population inversion and dark pulses become possible that are insensitive to the exact profile of the applied electric field. We have shown, therefore, that it is possible to extend the single-photon coherent control ideas to develop femtosecond multi-photon coherent control. The linearly chirped laser pulse approach to a robustly implementable ensemble CNOT gate could provide further impetus for future developments in optical approaches to 'ensemble quantum computing'.

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