Laser Phase Modulation Approaches towards Ensemble Quantum Computing

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Abstract

Selective control of decoherence is demonstrated for a multilevel system by generalizing the instantaneous phase of any chirped pulse as individual terms of a Taylor series expansion. In the case of a simple two-level system, all odd terms in the series lead to population inversion while the even terms lead to self-induced transparency. These results also hold for multiphoton transitions that do not have any lower-order photon resonance or any intermediate virtual state dynamics within the laser pulse-width. Such results form the basis of a robustly implementable CNOT gate.
Minimizing decoherence is an important challenge towards the realization of quantum computing (QC). While Nuclear Magnetic Resonance (NMR) spectroscopy of molecules in liquid phase is an important approach to QC [1] and has long decoherence times, it suffers in terms of scalability to larger number of qubits [2]. Alternative approaches for QC are, therefore, being sought [3]. Optical spectroscopy of molecules and nanoparticles could potentially become a highly scalable 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. Controlling IVR is also important for “coherent-control” due to its connection to “bond-selective” chemistry [4]. The prospect of QC can lead to a further resurgence of activity in this area. Restriction of IVR by optical schemes [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”) have used complicated pulse shapes that are yet to be demonstrated in the laboratory due to stringent requirements of intensity and precision.

In this letter, we use simple chirped pulses, which, by contrast, have been produced routinely at very high intensities and at various different wavelengths for many applications, including selective excitation of molecules in coherent control. Such selective optical control over molecules in an ensemble is as unique as addressing individual spins in a NMR spectrometer and lead to quantum computing in bulk optical systems. The main aim in coherent control has been controlling an observable, while in the case of QC 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.

We show that even simple linearly chirped pulses could restrict IVR in systems at least as complicated as investigated earlier [5]. We prove that higher-order chirps that distort the linear chirps experimentally, in fact, result in better performance in restricting IVR. In case of two-level systems, such shaped-pulses result either in population inversion or self-induced transparency. Similar results hold even in the extreme case of a two- or multiphoton transition occurring with a chirped pulse, where the lower-order photon processes are non-resonant [6,7]. This makes these results more attractive, since the intensity of the laser fields needed to reach adiabatic limits often leads to multiphoton processes.

We apply a linearly polarized laser pulse of the form $E(t) = \varepsilon(t) e^{i[\omega t + \phi(t)]} = \varepsilon(t) e^{i[\omega + \dot{\phi}(t)t]}$ 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. We have $\varepsilon(t)$, $\phi(t)$ and $\dot{\phi}(t)$ as the instantaneous amplitude, phase and frequency-sweep, respectively; and $\omega$ is the laser carrier frequency or the center frequency for pulsed lasers. If we expand the instantaneous phase function of $E(t)$ as a Taylor series with constants $b_n$, we have

$$\phi(t) = b_0 + b_1 t + b_2 t^2 + b_3 t^3 + b_4 t^4 + b_5 t^5 + ...$$
$$\dot{\phi}(t) = b_1 + 2b_2 t + 3b_3 t^2 + 4b_4 t^3 + 5b_5 t^4 + ...$$
$$\ddot{\phi}(t) = \sum_{n=1}^{\infty} n b_n t^{(n-1)} \tag{1}$$

To our knowledge this is the first formulation of a general expression for the continuously frequency modulated (“chirped”) pulses. Establishing this generalization enables us to treat
all possible chirped pulse cases by exploring the effects of each of the terms in Eqn. (1) initially for a simple two-level system and then extend it to the multilevel situation for a model five-level system of anthracene molecule, which has been previously investigated with complicated shaped-pulses [6,7]. We use a density matrix approach by integrating the Liouville equation \( \frac{d\rho(t)}{dt} = i\hbar \left[ H, \rho(t) \right] \) for a Hamiltonian in the rotating Frequency Modulated (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. The Hamiltonian for the simple case of a two-level system under the effect of an applied laser field can be written in the FM frame for N-photon transition as,

\[
H_{\text{FM}} = \hbar \begin{pmatrix}
\Delta + N\dot{\phi}(t) & \Omega \\
\Omega^* & 0
\end{pmatrix}.
\]

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 \( \Delta = \omega_R - N\omega_i \), while the direction of the field in the orthogonal plane remains fixed. We define the multiphoton Rabi Frequencies as complex conjugate pairs: \( \Omega(t) = (\mu_{\text{eff}}/\varepsilon(t))^N/h \) and \( \Omega^*(t) = (\mu_{\text{eff}}^*\varepsilon(t))^N/h \). For the \( |1\rangle \rightarrow |2\rangle \) transition, \( \omega_R = \omega_2 - \omega_1 \) is the single-photon resonance frequency. We have assumed that the transient dipole moment of the individual intermediate virtual states in the multiphoton ladder result in an effective transition dipole moment, \( \mu_{\text{eff}}^N \), which is a product of the individual N virtual state dipole moments \( \mu_n \), (i.e., \( \mu_{\text{eff}}^N = \Pi_n \mu_n \)). This approximation is particularly valid when intermediate virtual levels are non-resonant and as such their multiphoton interaction dynamics can be neglected [8,9].

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 (Fig. 1 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:

\[
H_{\text{FM}} = \begin{pmatrix}
|0\rangle & |1\rangle & |2\rangle & |3\rangle & |4\rangle & \ldots \\
0 & \Omega(t) & 0 & 0 & 0 & \ldots \\
\Omega^*(t) & \delta_1(t) & V_{12} & V_{13} & V_{14} & \ldots \\
0 & V_{12} & \delta_2(t) & V_{23} & V_{24} & \ldots \\
0 & V_{13} & V_{23} & \delta_3(t) & V_{34} & \ldots \\
0 & V_{14} & V_{24} & V_{34} & \delta_4(t) & \ldots \\
\vdots & \vdots & \vdots & \vdots & \vdots & \ddots
\end{pmatrix}
\]

(2)

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 \( V_{12}, V_{23}, \) etc. Both the Rabi frequency \( \Omega(t) \) and the detuning frequency \( \delta_1,2,3,\ldots = \Delta_1,2,3,\ldots + N\dot{\phi}(t) \) 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 Eqn. (2), the excited states’ submatrix containing the bright state $|1\rangle$ and the bath states $|2\rangle$, $|3\rangle$, $|4\rangle$, ..., can be diagonalized to give the eigenstate representation containing a set of $\Delta'_i$ as diagonal elements and corresponding $\Omega'_i$ 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.

From experimental results on the fluorescence quantum beats in jet-cooled anthracene $[8]$, 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 these values are incorporated in Eqn. (2), we obtain the full zero-order Hamiltonian matrix that can simulate the experimental quantum beats (Fig. 1) 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 $\pi$ 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 ($|1\rangle$) into the dark states (Fig. 1).

A linear sweep in frequency of the laser pulse (i.e., $\dot{\phi}(t) = 2b_2t$) can be generated by sweeping from far above resonance to far below resonance (blue to red sweeps), or its 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) $[9,10]$. 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, Fig. 2 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 multiphoton conditions where only an $N^{th}$ ($N \geq 2$) photon transition is possible $[4]$. 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.

The Quadratic Chirp, i.e., $\dot{\phi}(t) = 3b_3t^2$, is the most efficient in decoupling the bright and dark states as long as the Stark shifting of these states prevail at the peak of the pulse. As the pulse is turned off, the system smoothly returns to its original unperturbed condition (Fig. 3). This would be a very practical approach of controlling the coupling of the states with realistic pulse shapes. The cubic term, i.e., $\dot{\phi}(t) = 4b_4t^3$ behaves more like the linear term (Fig. 4). It also decouples the bright and dark states as long as the Stark shifting of these states prevail at the peak of the pulse. However, the oscillatory nature of the “photon-locking” shows that the higher-order terms in the Taylor series involve more rapid changes
and fails to achieve perfect adiabatic conditions. As the pulse is turned off, it attempts to invert the bright state population, which quickly dephases, analogous to the linear chirp case. Thus, in an isolated two-level system that does not suffer from the population dephasing, the linear, cubic, and all the higher odd-order terms of the Taylor series (Eqn. (1)) yield inversion of population, while the even-order terms produce self-induced transparency.

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 the even terms in the Taylor series return the system to its unperturbed condition at the end. In fact, all higher-order odd terms behave in one identical fashion and the even terms behave in another identical fashion. 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. In the present calculations, we have used equal values to $b_n$ in Eqn. (1), to bring out the effects of the higher-order terms in the series. In practice, since Eqn. (1) represents a convergent series, only lower-order terms are more important, and since all higher-order terms produce the same qualitative results as the lower-order terms, one needs to consider only up to the quadratic term.

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” \[1\]. Given the formalism of generalized chirped pulses discussed in this letter, 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 $\pi$ sequences for controlling coherence or inducing inversion.

This straightforward laser phase modulation approach towards a robustly implementable ensemble CNOT gate will hopefully form the basis for future developments towards “ensemble quantum computing” with optical schemes.
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FIGURES

FIG. 1. A transform-limited Gaussian pulse interacts with a model Anthracene molecule (inset) in a single photon mode or in a multiphoton condition. (Inset) Schematic of IVR for Anthracene molecule from Ref. [5] based on data extracted from experimental measurements in Ref. [8].

FIG. 2. A linearly swept Gaussian pulse (bottom inset, where $b_2=10\text{cm}^{-2}$) can generate “photon-locking”. The evolution of the dressed state character is unchanged while locking occurs (top inset) but as the pulse is turned off, the eigen-energy curves cross and the bright state population quickly dephases.

FIG. 3. The Quadratic Chirped Gaussian pulse (bottom inset, where $b_3=10\text{cm}^{-2}$) is the most efficient in decoupling the bright and dark states during the pulse. The eigen-energy curves and the corresponding evolution of the dressed state character (top inset) shows that the entire process is highly adiabatic.

FIG. 4. Effect of a Cubic Chirped Gaussian pulse (bottom inset, where $b_4=10\text{cm}^{-2}$) is similar to the linearly swept pulse (Fig.2), although the evidence of population oscillation indicates that this chirp is not as adiabatic as the linear chirp. The eigen-energy curves cross towards the end of the pulse (top inset) and the bright state population gets redistributed.
TABLE I. CNOT Gate with Simple Chirped Pulses.

| Shaped-Pulse      | A  | B  | A⊕B |
|-------------------|----|----|-----|
| “Inverting” Pulse | 1  | 1  | 0   |
|                   | 1  | 0  | 1   |
| “Dark” Pulse      | 0  | 1  | 1   |
|                   | 0  | 0  | 0   |
\[ \phi(t) = 3b_t^2 \]
