Population trapping in the excited states using vacuum-induced coherence and adiabatic process

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Abstract
We theoretically investigate how population can be trapped in the closely spaced excited levels in presence of vacuum-induced coherence (VIC). We employ delayed pulses to transfer population from a meta-stable state to the excited states. Subsequently, spontaneous emission from these excited states builds coherence between them. This coherence can be probed by using chirping, which leads to the decoupling of the excited states from the ground state thereby ensuring population transfer via delayed pulses. Our results indicate that the existence of VIC leads to the generation of a mixed state in the excited state manifold, where trapping of the population occurs even in the presence of large decay. This trapping may be realized in molecular systems and can be interpreted as a sensitive probe of VIC. We present suitable numerical analysis to support our results.

Keywords: vacuum-induced coherence, adiabatic transfer, delayed pulses, chirping, purity

1. Introduction

Quantum coherence [1] in atomic/molecular systems is the backbone of coherent control of optical properties of the medium. Such coherence arises from the interaction between a coherent source of light and the medium. Beside this, incoherent process such as spontaneous emission also leads to coherence between the excited states in the medium. One of the pronounced coherence phenomena called, vacuum-induced coherence (VIC), arises due to interference between different pathways of spontaneous emission from the closely spaced excited states to a single ground state [2]. In last decades, many atomic systems are studied for VIC to show fascinating applications [3–10]. However, the occurrence of VIC requires a stringent condition that the dipole moments of participating transition should be non-orthogonal [2, 11]. The unavailability of this condition in atomic systems restricts their applications. A possible realization of VIC has been suggested in ions [12, 13], quantum dots [14, 15], Mössbauer nuclei [16], quantum emitters placed in vicinity of localized surface plasmons [17], cavity quantum electrodynamics [18] and microwave superconducting cavity [19]. In spite of these proposals, the clear signatures of VIC in atomic systems are still difficult to achieve. However, contrary to atoms or ions, molecules are the favorable candidates for the inspection of VIC. In fact, VIC arises naturally in molecules due to availability of similar excited states belonging to same molecular electronic state thereby satisfying the requirement of non-orthogonality. Using this feature, the existence of VIC is reported in the excited ro-vibrational states of an atom-molecule system by means of ultracold photoassociative techniques [20]. The presence of such VIC in ultracold molecules can also be detected and estimated using magneto-optical rotation [21]. In another proposal, VIC is appreciated in terms of the existence of long-lived quasi-stationary coherence in a V-type molecular system which is excited incoherently [22].
Population trapping in excited states requires suitable initial preparation of the system. One possible way of transferring population into the trapped state could be to transfer it from the ground state by using a ‘+’ pulse. However, this would also create a coherence between the excited states and ground state. To avoid creation of such coherence, we employ the stimulated Raman adiabatic passage (STIRAP). Originally STIRAP was introduced as an efficient way to transfer the population between the dipole forbidden levels in multilevel systems. Using this technique, Shore et al. showed that one can transfer population to a desired energy level, without populating the intermediate levels. The STIRAP has been extensively studied for numerous applications ranging from quantum optics to quantum chemistry. Besides the atomic and molecular systems, STIRAP has also been employed in superconducting circuits based on Josephson effect. Recently, the combination of STIRAP with short nonadiabatic Rabi pulses is utilized in preparing a qutrit system in any desired state.

In this paper, we show how to adiabatically transfer the population to closely spaced excited states, exhibiting VIC, from a third excited state, thanks to STIRAP. The third excited state can be chosen as a meta-stable state. Contrary to usual STIRAP, which involves two or more ground states, we rather employ this technique to transfer population between a manifold of excited states. Once initially prepared, the subsequent spontaneous emission from the excited states to common ground state creates VIC. It should be borne in mind that such VIC is shown to lead to a coherent superposition of the excited states in a V-type configuration during photoassociation of cold molecules. Moreover, under suitable conditions, the superposition state of the excited states can become a dark state (immune to the relaxation) where initially transferred population gets trapped.

In this paper, we propose an alternative approach using STIRAP to create such a superposition state (albeit partially pure) at steady state, in presence of dissipative channels. We specifically show that it is possible to trap population in the closely spaced excited levels coherently, with the aid of VIC and using the technique STIRAP. We analyze our result by investigating the purity of the state thus prepared.

The paper is organized as follows: in section 2 we describe the theoretical model with the dynamical evolution including VIC. In section 3 we present the numerical analysis by considering different cases. We conclude the paper in section 4.

2. Model

The key requirement for VIC is the availability of non-orthogonal transitions which can arise in diatomic molecules due to the coupling of rotation of the molecular axis with the molecular electronic angular momentum. In real molecules, transitions to the same final rotational states but different vibrational states satisfy the conditions of parallel transition dipole moments. We consider a molecular system whose ground state can be represented as \( |0\rangle = |v_0, J = 0, M_J = 0; \Lambda = 0, \Sigma = 1; +\rangle \), where \( v_0 \) is the vibrational level, \( J \) is the rotational level, \( \Lambda \) is the axial component of electronic orbital quantum number \( L \), \( M_L(M_J, \Sigma) \) being the projection of \( L \) (\( J, \Sigma \)) on the internuclear axis and ‘+’ represents the parity of ground state. In the same manner, we choose a set of excited states \( |1\rangle = |v_1, J = 1, M_J = +1; \Lambda = 0, \Sigma = 0; -\rangle \) and \( |2\rangle = |v_2, J = 1, M_J = +1; \Lambda = 0, S = 0, \Sigma = 0; -\rangle \), where \( v_i \) \((i = 1, 2)\) represents the vibrational quantum numbers of the excited states and ‘-’ is the parity of the excited states. Both of these states belong to \( \Sigma \) state. Note that \( \Lambda = 0 \) for the ground and excited states are chosen for \( L = 0 \) and \( L = 1 \), respectively. Now, in order to have small spacing between the excited states, the vibrational levels should be chosen close to the dissociation threshold. Recent development in the production of Feshbach molecules provides a platform to choose such appropriate excited vibrational levels suitable for VIC by transitions from a molecular state in the triplet ground-state potential. For instance, \( \text{Cs}_2 \) molecule at ultracold temperature can be used to access highly excited vibrational levels from triplet ground state. Further, we should mention that the two transitions considered here occur between the same rotational and electronic states. Therefore, the electronic part of the dipole matrix elements will be the same thereby making the transition dipole moments parallel to each other.

We consider a four-level configuration as shown in figure 1, in which the ground state \( |0\rangle \) is coupled to the closely spaced excited levels \( |1\rangle \) and \( |2\rangle \) by an electromagnetic field \( \vec{E}_1 \). Another excited state \( |3\rangle \) is coupled to the ground state \( |0\rangle \) via an electromagnetic field \( \vec{E}_2 \). These electromagnetic fields are represented as

\[
\begin{align*}
\vec{E}_1(t) &= \hat{x}_1(t)e^{-i\omega_1t} + \text{c.c.}, \\
\vec{E}_2(t) &= \hat{x}_2(t)e^{-i\omega_2t} + \text{c.c.},
\end{align*}
\]

where, \( \hat{x}_1(t) \) and \( \hat{x}_2(t) \) are the time-dependent amplitudes of the electromagnetic fields and their polarization are perpendicular to each other.

![Figure 1](image-url)
to each other. The time-dependent phases \( \phi_{1,2}(t) \) denote frequency-sweep or chirping.

The Hamiltonian of this system under the dipole approximation can be written as

\[
\hat{H} = \hbar \omega_1 |1\rangle \langle 1| + \omega_2 |2\rangle \langle 2| + \omega_3 |3\rangle \langle 3| - i [\hat{d}^{\dagger}|0\rangle \langle 1| + \hat{d}^{\dagger}|2\rangle \langle 1| + \text{H.c.}) \hat{E}_1 - i [\hat{d}^{\dagger}|3\rangle \langle 0| + \text{H.c.}) \hat{E}_2.
\]

(2)

Here, zero of the energy is defined at the level \( |0\rangle \) and \( \hbar \omega_{1,2,3} \) is the energy difference between \( |\alpha\rangle \) and \( |\beta\rangle \). The dynamical evolution of the system can be described by the Markovian master equation:

\[
\dot{\rho} = -i/\hbar [\hat{H}, \rho] + \frac{1}{2} \sum_{i=1}^{3} \gamma_0 (2A_{0i} \rho A_{i0} - A_{i0}^\dagger \rho - \rho A_{i0}) + \frac{1}{2} \gamma_{12} \sum_{i,j=1,i\neq j}^{2} (2A_{ij} \rho A_{ji} - A_{ji}^\dagger \rho - \rho A_{ji}).
\]

(3)

Here, \( \gamma_{ij} \) is the decay rate from the level \( |j\rangle \) to \( |i\rangle \). The term \( \gamma_{12} = \sqrt{\gamma_0} \) is the decay rate of the coherence between level \( |f\rangle \) and \( |i\rangle \), and \( \gamma_{coll} \) is the collisional decay rate. Further, \( \Delta_1(t) = \omega_{g1} + \phi_g(t) - \omega_{10} \), and \( \Delta_2(t) = \omega_{g2} + \phi_g(t) - \omega_{30} \) are the detuning between the frequency of the field and the transition frequency, where \( \phi_g(t) = -\chi_f G(t) \). The transformation used to drive the density matrix elements in (4) are as follows: \( \rho_{01} = \rho_{01}\text{e}^{-i\omega_{10}t}, \rho_{02} = \rho_{02}\text{e}^{-i\omega_{30}t}, \rho_{03} = \rho_{03}\text{e}^{-i\omega_{12}t} \), while the rest of the elements remain the same. Also, the above density matrix elements satisfy the conditions \( \rho_{00} = \rho_{11} = \rho_{22} = \rho_{33} = 1 \) and \( \rho_{ij} = \rho_{ji}^\dagger \).

3. Numerical analysis

As discussed in the Introduction, we choose to prepare the molecule in a third excited state \( |3\rangle \), which can be chosen to be a meta-stable state. One can apply pulses in suitable sequence such that the population is transferred (albeit partially) to the closely spaced excited levels. We consider Gaussian profile for the time-dependence of these pulses

\[
G_{1}(t) = G_{01}\exp(-t^2/\tau^2),
\]

\[
G_{2}(t) = G_{02}\exp(-t^2/\tau^2),
\]

(5)

where, \( \tau_0 \) is the pulse delay, \( \tau \) in the pulse width and \( G_{01} \) and \( G_{02} \) are the maximum amplitudes of corresponding pulses. We consider Gaussian profile for the time-dependence of these pulses

3.1. Case I: Without chirping

Next, we solve (4) using (5). We first consider that the interaction of the fields with the system at exact resonance \( (\Delta_0 = 0) \) without chirping. Also, we consider a situation where maximum VIC exists in the system (i.e. \( \theta = 0 \)). We display the evolution of the population of the different levels and the pulse shape of Rabi frequencies \( G_{1} \) and \( G_{2} \) in figure 2.

Clearly, almost 50% population is transferred to the closely spaced excited states and gets trapped at steady state. The transferred population gets confined in excited state manifold that refers to a coherent (albeit partially) superposition state of excited states arising due to VIC.

To investigate further, we show the variation of real and imaginary part of coherence \( \rho_{10}, \rho_{20}, \rho_{31} \) and \( \rho_{32} \) with respect to time in figure 3.

We found that the magnitudes of these coherences are non-zero that means that the density matrix of the system does not get factorized into the basis \( \{|1\rangle, |2\rangle\} \). This further refers

4 Here, \( \theta = 0 \) is considered as both the excited rotational levels are chosen to be same. If they happen to be different then a situation where \( \theta = 0 \) might be considered. See supplementary of [22].
Figure 2. Population variation of level $|0\rangle$ (Black line), $|1\rangle$ (red line), $|2\rangle$ (blue line) and $|3\rangle$ (magenta line) with respect to time, where angle between transition dipole $\vec{d}_{01}$ and $\vec{d}_{20}$ is $\theta = 0$, $G_{01} = 0.9\gamma$, $G_{02} = 0.3\gamma$, pulse width $\tau = 4/\gamma$, pulse delay time $t_0 = 2.5\tau$, and spontaneous decay rates $\gamma_{01} = 5.8\gamma$, $\gamma_{02} = 2.2\gamma$ and $\gamma_{03} = 0.1\gamma$. The inset show the pulse shape of Rabi frequency $G_1$ (Red dash-dotted line) and $G_2$ (Blue dotted line).

Figure 3. Variation of real and imaginary part of coherence $\tilde{p}_{10}$ (black line), $\tilde{p}_{20}$ (blue line), $\tilde{p}_{31}$ (red line) and $\tilde{p}_{32}$ (green line) with respect to time at exact resonance($\Delta t = 0$), here solid (dashed) lines represent the real (imaginary) part of coherence and other common parameters are same as in figure 2. The inset show the pulse shape of Rabi frequency $G_1$ (Red dashed-dotted line) and $G_2$ (blue dotted line).

Figure 4. Variation of real and imaginary part of coherence $\tilde{p}_{10}$ (black line), $\tilde{p}_{20}$ (blue line), $\tilde{p}_{31}$ (red line) and $\tilde{p}_{32}$ (green line) with respect to time for $\Delta t = 0$ at $\chi_1 = 0.3$ and $\chi_2 = 0.2$, here solid (dashed) lines represent the real (imaginary) part of coherence. Other common parameters are same as figure 3.

We display the real and imaginary parts of the coherences $\tilde{p}_{10}$, $\tilde{p}_{20}$, $\tilde{p}_{31}$ and $\tilde{p}_{32}$ in figure 4 for $\Delta t \neq 0$.

We find that for suitable choice of $\chi_i$, the real and imaginary parts of these coherences become zero at the steady state. Therefore, the excited states are no longer coupled with the ground state and the meta-stable state. This means that population gets transferred from the meta-stable to the excited states via delayed pulses.

So far, we discussed about how the population gets transferred to the closely spaced excited states. Now, we will discuss, why this population gets confined at excited states for longer time? To find the answer of this question, we calculate coherence $\tilde{p}_{21}$ as discussed next.

The time-dependent real and imaginary part of coherence $\tilde{p}_{21}$ are shown in figure 5. Clearly, the magnitude of this coherence has non-zero value. Since the transition dipole moments $\vec{d}_{01}$ and $\vec{d}_{20}$ are parallel to each other, the cross coupling term $\gamma_{12}$ exhibits non-zero value. This means that the spontaneous emission channels from the excited states create a cross-talk via the vacuum of electromagnetic field (though the direct transition between these two states is dipole forbidden) and this interaction is responsible for VIC between excited states. It is interesting to see that the VIC causes to trap the population in the excited states, even in the presence of large spontaneous emission rate. This trapping of population in the closely spaced excited levels may be considered as a probe of VIC. It is to be noted that in this paper, the only coherence that exists is the VIC whereas the other coherence terms vanish even in the presence of the field. This feature makes our system novel as compared to the other works on VIC [1, 20].

The application of STIRAP technique between the excited states makes our model further interesting.

To understand the role of VIC, we continue to investigate the purity $\text{Tr}(\rho^2)$ of the state thus prepared. Here $\rho$ designates the density matrix block in the $\{1\}, \{2\}$ subspace only at the steady state. We show in figure 6 how the purity changes with $\theta$. As $\theta$ increases to $\pi/2$, the VIC ceases to exit and the coherence in the excited states also vanishes. clearly, the VIC to a coherent signature of the ground state in the final density matrix. We ask the following question next: Is it possible to trap the population maximally exclusively in the closely spaced excited levels, assisted by VIC? In the following, we find an affirmative answer.

3.2. Case II: With chirping

To eliminate coherence between the excited states and the ground state, we next employ chirped pulses as follows:

$$\Delta_1(t) = \chi_1 \text{tanh}(t),$$
$$\Delta_2(t) = \chi_2 \text{tanh}(t - t_0).$$

\begin{equation}
\end{equation}
plays very crucial role to maintain the coherence in the state, thus prepared.

It is clear that in the presence of VIC (corresponding to \( \theta = 0 \)), the purity becomes maximum and zero for \( \theta \gtrsim 0.3 \). Note that in the present case, the system is prepared in a mixed state at long times.

4. Conclusion

In conclusion, we have analyzed the creation of a dark state in the closely spaced excited states in molecular systems. By means of STIRAP, the population is adiabatically transferred from a meta-stable state to the excited states and gets trapped therein at steady state due to the existence of VIC. Further, the chirped detuning in our analysis specified that the transfer of population occurs solely due to pulse. We verified the effect of VIC and the pulse through numerical analysis of time evolution and purity. Apart from its fundamental importance \([1, 39, 40]\), our prototype for population trapping in excited states with the aid of adiabatic pulses in the presence of VIC in molecular systems, can be extended to study decoherence in quantum information processing \([41]\), the investigation of the role of noise-induced coherence \([22]\), in quantum heat engines \([42]\), and biological processes \([43]\).

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