Stimulated Raman adiabatic passage in a two-state system

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We present a method, using adiabatic passage, for creating a maximally coherent superposition in a two-state atom. The method exploits the analogy between the two-state Bloch equations and the three-state stimulated Raman adiabatic passage (STIRAP) equations, and uses two sequential but overlapping pulsed fields, a driving pump pulse and a delayed but overlapped detuning pulse. Like STIRAP, this method is robustly insensitive to small changes in pulse properties.

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I. INTRODUCTION

Many possibilities exist for coherent manipulation of the state vector describing a two-state system by pulsed radiation. These include resonant excitation (for which exact analytic solutions exist for the relevant time-dependent Schrödinger equation) and rapid adiabatic passage produced by a frequency sweep (chirp) across the resonance frequency \([\omega_L, \omega_R]\). Particularly well known, and experimentally demonstrated, are implementations of these two techniques that will transfer all population from one quantum state to the other. During recent years interest has shifted to manipulations that create a coherent superposition of two quantum states—processes that complete only a partial transfer of population, with well-defined phase. It is this class of state-vector motions that we consider here.

With the development of lasers as tools for producing coherent excitation, attention turned to multiphoton processes that we consider here.

II. SCHröDINGER EQUATION AND BLOCH EQUATION

A. The Bloch equation for a two-state system

Coherent excitation of a two-state atom, whose unperturbed energies are \(E_1\) and \(E_2\), is governed by the time-dependent Schrödinger equation. In the rotating-wave approximation (RWA) this equation, for RWA probability amplitudes \(C_1(t)\) and \(C_2(t)\), reads

\[
\frac{d}{dt} \begin{bmatrix} C_1 \\ C_2 \end{bmatrix} = \begin{bmatrix} 1 & 0 & \Omega \\ 2 & \Omega & 2\Delta \end{bmatrix} \begin{bmatrix} C_1 \\ C_2 \end{bmatrix}.
\]

The time-dependent Rabi frequency \(\Omega(t)\), which couples the two states, quantifies the interaction energy and is proportional to the envelope \(\tilde{E}(t)\) of the pulsed laser electric field

\[
\mathbf{E}(t) = e\tilde{E}(t)\cos(\omega_0 t)
\]

and the transition dipole moment

\[
\mathbf{d}_{12} = -\mathbf{d}_{12} \cdot e\tilde{E}(t)/\hbar.\]

The generally time-dependent detuning

\[
\Delta(t) = \omega_0(t) - \omega_L(t)
\]

is the offset between the Bohr transition frequency \(\omega_0(t) = (E_2 - E_1)/\hbar\) and the laser carrier frequency \(\omega_L(t)\), either of which may vary with time.

As has long been known [4], this equation for complex-valued probability amplitudes can be recast as three coupled equations for real-valued variables. The resulting optical Bloch equation for a coherently driven two-state system, in RWA reads [1,2,4]

\[
\frac{d}{dt} \begin{bmatrix} u \\ v \\ w \end{bmatrix} = \begin{bmatrix} 0 & -\Delta & 0 \\ \Delta & 0 & -\Omega \\ 0 & \Omega & 0 \end{bmatrix} \begin{bmatrix} u \\ v \\ w \end{bmatrix}.
\]

The three real-valued time-dependent quantities \(u, v,\) and \(w\) define the coordinates of the Bloch vector, moving in an essential-states treatment. The remaining amplitude and frequency controls are to be incorporated in the mathematical model.

This paper is organized as follows. In Sec. II we describe the analogy between the two-state Bloch equation and the three-state Schrödinger equation. In Sec. III we describe the basic properties of 2S-STIRAP by exploiting the knowledge of the usual 3S-STIRAP. In Sec. IV we discuss possible experimental implementations and in Sec. V we summarize the conclusions.

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TABLE I. Correspondence between two- and three-state systems.

<table>
<thead>
<tr>
<th>Two-state system</th>
<th>Three-state system</th>
</tr>
</thead>
<tbody>
<tr>
<td>$w$</td>
<td>$C_1$</td>
</tr>
<tr>
<td>$v$</td>
<td>$-iC_2$</td>
</tr>
<tr>
<td>$u$</td>
<td>$-C_3$</td>
</tr>
<tr>
<td>$w(-\infty)=-1$</td>
<td>$C_1(-\infty)=-1$</td>
</tr>
<tr>
<td>$u(+\infty)=-1$</td>
<td>$C_2(+\infty)=1$</td>
</tr>
<tr>
<td>$2\Delta$</td>
<td>$\Omega_S$</td>
</tr>
<tr>
<td>$2\Omega$</td>
<td>$\Omega_P$</td>
</tr>
<tr>
<td>Dephasing</td>
<td>Irreversible loss from 2 and 3</td>
</tr>
<tr>
<td>Irreversible loss from 2</td>
<td>All population in the system</td>
</tr>
<tr>
<td>Pure state</td>
<td>$</td>
</tr>
<tr>
<td>$u^2+v^2+w^2=1$</td>
<td>Not all population in system</td>
</tr>
<tr>
<td>Mixed state</td>
<td>$</td>
</tr>
<tr>
<td>$w^2+v^2+w^2&lt;1$</td>
<td>Depleted initial state ($C_0=0$)</td>
</tr>
<tr>
<td>Maximal coherence ($w=0$)</td>
<td></td>
</tr>
</tbody>
</table>

This system involves three unperturbed energies $E_1$, $E_2$, and $E_3$, and two carrier frequencies, $\Omega_P$ and $\Omega_S$, that of a pump field $\omega_P$ and that of a Stokes field $\omega_S$. We require that both laser fields are on exact resonance with the respective transitions: $|E_2-E_1|=\hbar \omega_P$ and $|E_3-E_2|=\hbar \omega_S$. Then all diagonal elements of the RWA Hamiltonian vanish, as seen in Eq. (3). The Rabi frequencies $\Omega_p(t)$ and $\Omega_S(t)$ quantify the interaction energies for the pump and Stokes transitions, respectively.

After replacing the probability amplitudes $C_2$ and $C_3$ by the amplitudes $\tilde{C}_2=-iC_2$ and $\tilde{C}_3=-C_3$, and exchanging the places of $C_1$ and $\tilde{C}_3$, the Schrödinger equation (3) takes the form

$$\frac{d}{dt} \begin{bmatrix} C_1 \\ C_2 \\ C_3 \end{bmatrix} = \frac{1}{2} \begin{bmatrix} 0 & \Omega_P & 0 \\ \Omega_P & 0 & \Omega_S \\ 0 & \Omega_S & 0 \end{bmatrix} \begin{bmatrix} C_1 \\ C_2 \\ C_3 \end{bmatrix}. \quad (3)$$

This equation is completely equivalent to the 2S Bloch equation (2) after making the identifications summarized in Table I and described below.

C. Correspondence between the two-state Bloch vector and the three-state state vector

Most notably, the Bloch vector components $u$, $v$, and $w$ correspond to the probability amplitudes $\tilde{C}_3$, $\tilde{C}_2$, and $C_1$, as illustrated in Fig. 1. The detuning $\Delta(t)$ corresponds to the Stokes pulse $\Omega_S(t)$ (apart from a factor of 2) and the Rabi frequency $\Omega(t)$ to the pump pulse $\Omega_P(t)$.

The initial condition for the 2S system describing population in the lower state, $w(-\infty)=-1$, corresponds to $C_1(-\infty)=-1$ in the 3S system. The STIRAP procedure then uses adiabatic passage, with the Stokes pulse before the pump pulse, to move the population into state 3, meaning $|C_3|=1$. Hence STIRAP, run in the 2S system, will create a state with $u=1$, $v=w=0$.
States for which \( u^2+v^2=1 \) and \( w=0 \) are states of maximal coherence in the 2S system. In the 3S system these correspond to a depleted initial state, \( C_1=0 \). Hence a process that depletes the initial state in the 3S system, e.g., STIRAP, will create a maximally coherent superposition state in the 2S system.

Dephasing in the 2S system corresponds to irreversible population losses from states 2 and 3 in the 3S system. There are no analogs in the 2S system for the 3S system processes of spontaneous emission, irreversible loss from state 2, single-photon or two-photon detunings. Likewise, spontaneous emission in the 2S system has no analog in the 3S system.

A pure (mixed) state in the 2S system corresponds to having all (not all) population in the 3S system. A completely incoherent state in the 2S system \( (u=v=w=0) \) corresponds to no population at all in the 3S system.

III. TWO-STATE STIRAP

A. Dark state and population transfer

The dark state, which is the population transfer vehicle in STIRAP, is defined as the zero-eigenvalue eigenstate of the RWA Hamiltonian in Eq. (3),

\[
|d(t)\rangle = \cos \theta(t)|1\rangle - \sin \theta(t)|3\rangle,
\]

where \( \theta(t)=\arctan[\Omega_p(t)/\Omega_2(t)] \). When the Stokes pulse precedes the pump pulse, we have

\[
\theta(-\infty) = 0, \quad \theta(\infty) = \pi/2,
\]

and hence the dark state has the limits \( |d(-\infty)\rangle=|1\rangle \) and \( |d(\infty)\rangle=|3\rangle \), thereby providing an adiabatic link between states \( |1\rangle \) and \( |3\rangle \). Hence, after starting in state \( |1\rangle \), the 3S system will end in state \( |3\rangle \), with unit probability in the adiabatic limit.

The dark “state” in the 2S system is not really a quantum state of the system, but a sum of the inversion \( w(t) \) and the coherence \( u(t) \),

\[
d(t) = w(t)\cos \theta(t) + u(t)\sin \theta(t),
\]

with \( \theta(t)=\arctan[\Omega(t)/\Delta(t)] \). When the detuning pulse \( \Delta(t) \) precedes the pump pulse \( \Omega(t) \) the mixing angle \( \theta(t) \) has the same asymptotic values (6) as in STIRAP; hence \( d(-\infty)=w(-\infty) \) and \( d(\infty)=u(\infty) \). Thus we know that, in a 2S system, we can move the Bloch vector from alignment with \( w \) to alignment with \( u \) by applying first a detuning pulse and then an excitation pulse, while maintaining adiabatic conditions. The result will be that the Bloch vector moves on the Bloch sphere from the south pole to the equator, aligned with \( u \), i.e., the system ends in a coherent superposition of the two states.

Because the adiabatic passage is robust, this procedure is robust: it depends only weakly on the overlap of the two pulses and the peak values of \( \Delta(t) \) and \( \Omega(t) \), and only requires that the areas be large.

\( u > 0.9 \)

FIG. 2. (Color online) The numerically calculated coherence \( u \) resulting from a two-state STIRAP versus the peak values of the detuning \( \Delta_0 \) and the Rabi frequency \( \Omega_0 \), for Gaussian shapes \( \Delta(t) = \Delta_0 e^{-[t+\tau/2]^2/T^2} \) and \( \Omega(t) = \Omega_0 e^{-[t-\tau/2]^2/T^2} \), with delay \( \tau=1.5T \). The contour lines, labeled with the corresponding values of \( u \), display the gradual increase of the coherence as \( \Delta_0 \) and \( \Omega_0 \) grow. The dashed curve is the circle \( \Delta_0^2 + \Omega_0^2 = (15/T)^2 \).

B. Adiabatic condition

The adiabatic condition is another example of what one can learn from 3S STIRAP. The local adiabatic condition reads [3]

\[
|\dot{\theta}(t)| \ll \bar{\Omega}(t),
\]

where \( \bar{\Omega}(t)=\sqrt{\Omega_p^2(t)+\Omega_2^2(t)} \) is the rms Rabi frequency. The global adiabatic condition is obtained after integration of Eq. (8),

\[
\frac{\pi}{2} \ll \int_{-\infty}^{\infty} \bar{\Omega}(t)dt,
\]

which is interpreted as a condition for large pulse area.

For pulsed interactions it is known that there is an optimum delay for which the adiabatic regime is most easily reached. For example, for Gaussian shapes the optimum delay is about \( \tau\approx 1.5T \), where \( T \) is the pulse width [3]. Similar features must hold for two-state STIRAP too, upon the identification \( \bar{\Omega}(t)=\sqrt{\Delta^2(t)+\Omega^2(t)} \).

It is also known that in STIRAP the energetically most efficient choice (associated with minimal rms pulse area) is when the peak Rabi frequencies of the pump and Stokes pulses are nearly equal. A stronger pump or Stokes, beyond this balance, improves adiabaticity only marginally. The same property must hold for 2S STIRAP as well.

Figure 2 shows the coherence \( u \) obtained after 2S-STIRAP process, plotted as a function of the peak values of the detuning \( \Delta_0 \) and the Rabi frequency \( \Omega_0 \). The coherence approaches unity as \( \Delta_0 \) and \( \Omega_0 \) increase. The figure is nearly symmetric with respect to the line \( \Delta_0=\Omega_0 \). The dashed curve shows the circle \( \Delta_0^2 + \Omega_0^2 = (15/T)^2 \), which nearly touches the contour line \( u=0.9 \) around \( \Delta_0=\Omega_0 \). Thus this figure demonstrates that, indeed, nearly equal values \( \Delta_0 \) and \( \Omega_0 \) provide
C. Counterintuitive vs intuitive sequence

As we discussed above, and as is well known, 3S STIRAP populates state $|3\rangle$ with unit probability in the adiabatic limit; hence it also depletes the initial state $|1\rangle$ completely [5]. However, instead of transferring the population to state $|3\rangle$, it creates an oscillating superposition of states $|2\rangle$ and $|3\rangle$ [5]. In 2S STIRAP this means that the detuning-pump sequence (counterintuitive) will create a state with $|u| = 1$, $v = w = 0$, while the pump-detuning (intuitive) sequence will create a state with $|u|^2 + |v|^2 = 1$, $w = 0$. In both cases an equal superposition of states $|1\rangle$ and $|2\rangle$ is created ($w = 0$), with $v = 0$ in the former case and $v \neq 0$ in the latter.

IV. IMPLEMENTATIONS

For the implementation of 2S STIRAP the main challenge is to create a pulse-shaped detuning $\Delta(t)$ vanishing at early and late times; this pulse-shaped detuning must be delayed in time with respect to the pulse-shaped Rabi frequency of the excitation pulse. Such pulse-shaped detuning can be designed by at least three techniques.

One can use a computer-controlled pulse shaper to engineer the desired time dependences of $\Omega(t)$ and $\Delta(t)$ for a pico- or femtosecond pulse [6]; in this case the time variation of the detuning derives from the variation of the frequency of the driving pulse field.

Alternatively, one can use the dynamic Stark shift induced by a strong off-resonant Stark pulse to modify the energies of the two states [7]. In this case the detuning chirp derives from the Stark-controlled variation of the Bohr transition frequency.

Finally, one can use a pulsed magnetic field to modify the Bohr transition frequency. In this case one needs to choose magnetic sublevels with different magnetic quantum numbers, for example $(J = 0, m = 0)$–$(J = 1, m = 1)$, and circularly polarized laser pulse.

V. CONCLUSIONS

In this paper we have exploited an intriguing analogy between STIRAP and chirped two-state excitation. This situation is a rare example when one can learn something about a simpler system (the two-state system) by using the knowledge about a more complex system (the three-state system). This occurs because STIRAP has been very well studied in the past 15 years.

The variation of the detuning is actually a pulse, not a monotonic chirp, as in chirped adiabatic passage. We have suggested several experimental implementations.

In a two-state system, the described STIRAP analog operates between the population inversion $w$ and the real part of the coherence $u$; 2S STIRAP transfers inversion into coherence, with the same efficiency and robustness as in 3S STIRAP.

The described technique has the potential to not only be a curious and intriguing example of isomorphism between quantum systems of different dimensions (or different realizations of the same mathematical group) but also a useful, efficient and robust experimental technique for creating maximally coherent superposition states, for example, for applications in quantum information [8].

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