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Creation of coherent superpositions using Stark-chirped rapid adiabatic passage

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Abstract

We show that the technique of Stark-chirped rapid adiabatic passage (SCRAP), hitherto used for complete population transfer between two quantum states, offers a simple and robust method for creating coherent superpositions of states. SCRAP uses two laser pulses: a strong far off-resonant pulse modifies the transition frequency by inducing ac Stark shifts in the energies of the two states, and an appropriately offset in time, near-resonant and moderately strong pump pulse drives the population between the states via one of the induced diabatic level crossings. The populations in the created superposition are controlled by the detuning of the pump laser from the transition frequency and are insensitive to variations in the intensities of the pump and Stark lasers, as long as these are sufficiently large to allow adiabatic evolution. © 2002 Elsevier Science B.V. All rights reserved.

1. Introduction

The ability to prepare atoms and molecules in well-defined coherent superpositions of quantum states is crucial to various fields of contemporary quantum physics. A quantum system in a coherent superposition of states can lead to qualitatively different phenomena, compared to a system ini-

tially in a single state. Examples can be found in coherent control of chemical reactions [1], quantum information [2], atom optics [3], light propagation through matter [4], lasing without inversion [5], quantum optics [6], high harmonic generation [7], and others.

As is well understood, any technique that can completely transfer population between two quantum states can, in principle, be used to create coherent superpositions: one need only halt the transfer process before its completion. For example, the excited-state population of a two-state atom, coherently excited by constant-intensity resonant radiation, undergoes periodic variation at the Rabi frequency Ω [8]. By choosing the pulse duration T such that the pulse area ΩT is an odd

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multiple of π , all population is placed into the excited state (odd- π pulses). Alternatively, a superposition that equally balances the two states is created when the pulse area is adjusted to $\pi/2$. However, this technique is not robust: variations in pulse area and detuning from resonance, e.g. due to intensity and phase fluctuations or Doppler shifts, can lead to considerable changes in the populations.

A popular technique for inducing complete and robust population transfer between three states in a Raman-type linkage is stimulated Raman adiabatic passage (STIRAP) [9]. STIRAP uses two delayed, but partly overlapping pulses – pump and Stokes – with the Stokes pulse applied first (in the counterintuitive order). Because this technique is based on adiabatic evolution, it is insensitive to pulse areas, and it is therefore robust, so long as the pulse areas are sufficiently large to make the evolution adiabatic and two-photon resonance between the initial state and the final state is maintained. It has been suggested [10] and demonstrated [11,12] that STIRAP can be used to produce a coherent superposition of a ground state and a metastable state or of two ground Zeeman sublevels. Then it is necessary to ensure that, after some moment during the evolution, the two pulses maintain a fixed constant ratio; this ratio determines the composition of the created superposition. With pulsed lasers, however, maintaining an exact ratio may be difficult because of frequency and intensity fluctuations.

Coherent superpositions of states can be created in a robust fashion by an interesting variation of STIRAP, tripod-STIRAP [13]. It supplements the three states in STIRAP with a fourth state, coupled to the intermediate state by another, control laser. Tripod-STIRAP allows the creation of coherent superpositions of two or three states, which can be controlled by the ordering of the three pulses (pump, Stokes, and control), the time delays between them, and the intensity of the control pulse.

Recently, we have proposed [14], and experimentally demonstrated [15], an alternative technique of STIRAP, named Stark-chirped rapid adiabatic passage (SCRAP), which achieves complete population transfer between two bound

states. SCRAP uses a nearly-resonant pump laser pulse, which drives the population between a ground state ψ_1 and an excited state ψ_2 , and an intense far-off-resonance Stark laser pulse, which modifies the transition frequency by Stark shifting the energies of the two states. Thus by detuning the pump laser carrier frequency from resonance with the Bohr transition frequency one can create two separated diabatic level crossings. If the pump pulse is sufficiently intense and is applied at one of these crossings, it will produce complete adiabatic passage of population from ψ_1 to ψ_2 around this crossing. If the pump pulse width is sufficiently small, its intensity will be negligible at the other crossing and the population will remain in state ψ_2 (i.e., there the evolution will be diabatic). Thus the net result of the interaction will be complete population transfer to the excited state ψ_2 . Indeed, if properly implemented, the efficiency of SCRAP is close to unity [15].

The main advantage of SCRAP over STIRAP-based techniques is that SCRAP can be used with multiphoton transitions. Such transitions are always accompanied by laser-induced ac Stark shifts that modify the transition frequencies. For STIRAP, such time-dependent shifts are detrimental because they destroy the two-photon resonance, which is crucial for the population transfer. In SCRAP, the Stark shifts are less harmful because, as a level-crossing technique, it does not require maintaining a resonance condition. Moreover, SCRAP takes advantage of the Stark shifts, induced by the off-resonance laser, to create the level crossings needed for population transfer.

In the present paper we show that for a certain range of pump laser detunings Δ_0 , the SCRAP technique will produce a coherent superposition of states ψ_1 and ψ_2 – it will operate as *half*-SCRAP. The composition of the created superposition is controlled by Δ_0 and is robust against variations in the other interaction parameters.

This paper is organized as follows. In Section 2, we provide some basic definitions and equations describing coherent excitation. In Section 3 we discuss in detail the diabatic–adiabatic mechanism of SCRAP and half-SCRAP. In Section 4 we show that the superpositions created by half-

SCRAP are robust against variations in the interaction parameters. In Section 5 we present an analytically solvable model that illustrates half-SCRAP. Finally, in Section 6 we summarize the results.

2. Background

2.1. Basic definitions and equations

We assume that the two-state system is prepared initially, at time t_i , in state ψ_1 , and we wish to create at the final time t_f , using half-SCRAP, a coherent superposition of states ψ_1 and ψ_2 ,

$$\Psi(t_f) = \psi_1 \rightarrow \Psi(t_f) = \sqrt{1-p}\psi_1 + \sqrt{p}e^{-i\varphi}\psi_2, \quad (1)$$

except for an unimportant common phase factor. Here p is the transition probability from ψ_1 to ψ_2 and φ is the relative phase of the two components.

Because we wish to create a *coherent* superposition, we assume that during the excitation there are no incoherent processes, such as spontaneous emission and collisional relaxation. Then the system is described by its state vector $\Psi(t)$, which can be expanded in the basis of the unperturbed atomic states ψ_1 and ψ_2 as

$$\Psi(t) = C_1(t)e^{-i\zeta_1(t)}\psi_1 + C_2(t)e^{-i\zeta_2(t)}\psi_2. \quad (2)$$

Here $C_1(t)$ and $C_2(t)$ are the probability amplitudes of states ψ_1 and ψ_2 , and the respective populations are $P_k(t) = |C_k(t)|^2$, ($k = 1, 2$). For the sake of later convenience we define the arbitrary phases $\zeta_1(t)$ and $\zeta_2(t)$ as

$$\zeta_1(t) = E_1 t / \hbar + \int_{t_i}^t S_1(t') dt', \quad (3a)$$

$$\zeta_2(t) = \zeta_1(t) + \omega_L t + \phi_L, \quad (3b)$$

where E_1 and E_2 are the unperturbed energies of states ψ_1 and ψ_2 , $S_1(t)$ and $S_2(t)$ are their Stark shifts, and ω_L and ϕ_L are the carrier frequency and the phase (both assumed constant) of the pump laser pulse.

The evolution of the two-state system is governed by the Schrödinger equation,

$$i\hbar \frac{d}{dt} \mathbf{C}(t) = \mathbf{H}(t) \mathbf{C}(t), \quad (4)$$

where $\mathbf{C}(t) = [C_1(t), C_2(t)]^T$. In the rotating-wave approximation and for the phase choice (3a) and (3b), the Hamiltonian is given by [8]

$$\mathbf{H}(t) = \hbar \begin{bmatrix} 0 & \frac{1}{2}\Omega(t) \\ \frac{1}{2}\Omega(t) & \Delta(t) \end{bmatrix}. \quad (5)$$

The coupling between the states is quantified by the Rabi frequency $\Omega(t)$, whose time dependence derives from that of the pump-laser electric-field amplitude $\mathcal{E}(t)$. For single-photon electric-dipole transitions, $\Omega(t)$ is proportional to $\mathcal{E}(t)$. For two-photon transitions, $\Omega(t)$ is proportional to $\mathcal{E}^2(t)$, i.e. to the laser intensity.

The detuning $\Delta(t)$ is the offset of the laser carrier frequency from the Stark-shifted transition frequency,

$$\Delta(t) = \omega_A + S_2(t) - S_1(t) - \omega_L = \Delta_0 - S(t), \quad (6)$$

where $\omega_A = (E_2 - E_1)/\hbar$ is the Bohr transition frequency, $\Delta_0 = \omega_A - \omega_L$ is the static detuning of the pump pulse, and $S(t) = S_1(t) - S_2(t)$ is the difference of the Stark shifts of states ψ_1 and ψ_2 .

In SCRAP the transition frequency is modified by an intense far-off-resonance laser pulse, which induces Stark shifts in the energies of the two states, but it does not affect the system otherwise. The pump pulse itself induces Stark shifts too. The Stark shifts induced by each laser are proportional to the corresponding laser intensity, and the total Stark shifts are sums of these,

$$S_1(t) = S_1^P f_P(t) + S_1^S f_S(t), \quad (7a)$$

$$S_2(t) = S_2^P f_P(t) + S_2^S f_S(t). \quad (7b)$$

Here the dimensionless functions $f_P(t)$ and $f_S(t)$ are the envelopes of the pump and Stark laser intensities, while S_k^a ($k = 1, 2$; $a = P, S$) are the maximum Stark shifts. Because the Stark shifts of the ground and excited states are different (usually $|S_1(t)| \ll |S_2(t)|$), the detuning (6) will be also Stark shifted.

The Stark shifts induced by the pump laser can be significant for multiphoton transitions but are negligible for single-photon transitions. We will assume for simplicity that these shifts can be neglected; then

$$S(t) = S_1(t) - S_2(t) \approx S_0 f_S(t), \quad (8)$$

with $S_0 = S_1^S - S_2^S$.

2.2. Adiabatic basis

The analysis of SCRAP and half-SCRAP is facilitated by introducing the adiabatic states $\Phi_{\pm}(t)$ – the instantaneous eigenstates of the Hamiltonian $H(t)$,

$$\Phi_{-}(t) = \psi_1 \cos \vartheta(t) - \psi_2 \sin \vartheta(t), \quad (9a)$$

$$\Phi_{+}(t) = \psi_1 \sin \vartheta(t) + \psi_2 \cos \vartheta(t), \quad (9b)$$

where the mixing angle $\vartheta(t)$ is defined (modulo $\pi/2$) as

$$\vartheta(t) = \frac{1}{2} \arctan \frac{\Omega(t)}{\Delta(t)}. \quad (10)$$

The eigenvalues of $H(t)$ are the *adiabatic energies*,

$$\hbar \varepsilon_{\pm}(t) = \frac{\hbar}{2} \left[\Delta(t) \pm \sqrt{\Omega^2(t) + \Delta^2(t)} \right], \quad (11)$$

as contrasted with the *diabatic energies*, which are the diagonal elements of $H(t)$: 0 and $\hbar \Delta(t)$.

The amplitudes of the diabatic and adiabatic states are linked via an orthogonal rotation,

$$\mathbf{C}(t) = \mathbf{R}[\vartheta(t)] \mathbf{A}(t), \quad (12)$$

where $\mathbf{A}(t) = [A_{-}(t), A_{+}(t)]^T$ and

$$\mathbf{R}(\vartheta) = \begin{bmatrix} \cos \vartheta & \sin \vartheta \\ -\sin \vartheta & \cos \vartheta \end{bmatrix}. \quad (13)$$

The Schrödinger equation in the adiabatic basis reads

$$i\hbar \frac{d}{dt} \mathbf{A}(t) = \mathbf{H}_A(t) \mathbf{A}(t), \quad (14)$$

with $\mathbf{H}_A = \mathbf{R}^{-1} \mathbf{H} \mathbf{R} - i\hbar \mathbf{R}^{-1} \dot{\mathbf{R}}$, or explicitly,

$$\mathbf{H}_A(t) = \hbar \begin{bmatrix} \varepsilon_{-}(t) & -i\dot{\vartheta}(t) \\ i\dot{\vartheta}(t) & \varepsilon_{+}(t) \end{bmatrix}. \quad (15)$$

2.3. Adiabatic and diabatic evolutions

Adiabatic evolution occurs when the coupling $\dot{\vartheta}(t)$ between the adiabatic states is negligible compared to the separation of the adiabatic energies,

$$|\dot{\vartheta}(t)| \ll \varepsilon(t), \quad (16)$$

where

$$\varepsilon(t) = \varepsilon_{+}(t) - \varepsilon_{-}(t) = \sqrt{\Omega^2(t) + \Delta^2(t)}. \quad (17)$$

Then the transitions between the adiabatic states $\Phi_{-}(t)$ and $\Phi_{+}(t)$ are negligible and their populations are conserved. In particular, if the state vector $\Psi(t)$ is associated initially with a single adiabatic state, it will remain associated with this adiabatic state at all subsequent times. However, the populations of the diabatic states ψ_1 and ψ_2 may change because the composition of the adiabatic states $\Phi_{-}(t)$ and $\Phi_{+}(t)$ may change.

The concept of adiabatic evolution is particularly useful for explaining level-crossing transitions, when the diabatic energies 0 and $\hbar \Delta(t)$ cross, i.e. when the detuning $\Delta(t)$ crosses resonance. As evident from Eq. (17), at any point t_c of diabatic crossing [$\Delta(t_c) = 0$], the adiabatic energies $\hbar \varepsilon_{\pm}(t)$ have an avoided crossing if the Rabi frequency $\Omega(t)$ is nonzero, because $\varepsilon(t_c) = \Omega(t_c)$. Because a level crossing reverses the asymptotic behaviors of the adiabatic states, each adiabatic state is identified with a different diabatic state before and after the crossing. Hence if the evolution is adiabatic, complete population transfer will occur between states ψ_1 and ψ_2 .

Diabatic evolution occurs when the coupling $\dot{\vartheta}$ between the adiabatic states $\Phi_{-}(t)$ and $\Phi_{+}(t)$ is much larger than the adiabatic energy splitting,

$$|\dot{\vartheta}(t)| \gg \varepsilon(t); \quad (18)$$

Then the state vector follows the same diabatic state and the populations of the diabatic states ψ_1 and ψ_2 are conserved, while those of the adiabatic states $\Phi_{-}(t)$ and $\Phi_{+}(t)$ may change. In the most trivial case, diabatic evolution takes place when there is no radiative coupling between the diabatic states, i.e. when $\Omega(t) \approx 0$.

3. The SCRAP technique

3.1. SCRAP setup

The mechanism of SCRAP can be understood by viewing a plot of the diabatic and adiabatic

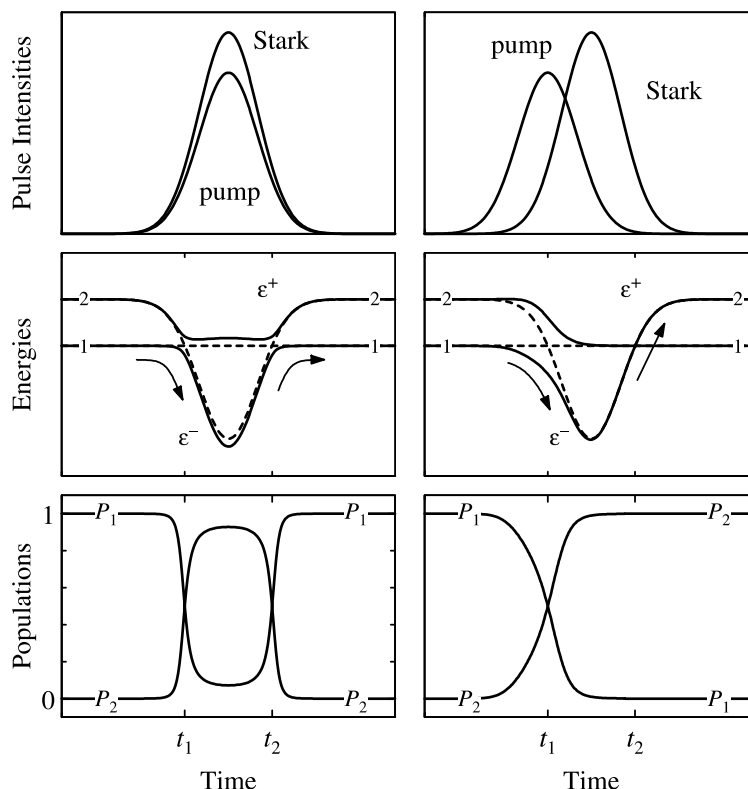


Fig. 1. Time evolution of the laser pulses (top frames), the level energies (middle frames), and the populations (bottom frames) in a two-state system driven by simultaneous (left frames) and offset in time (right frames) pump and Stark pulses.

energies against time, as shown in Fig. 1. As time progresses, the Stark pulse shifts the diabatic energy of state ψ_2 [i.e., the detuning $\Delta(t)$]: first in one direction as the pulse intensity increases, and then, as the pulse subsides, in the other direction. By choosing the static detuning Δ_0 appropriately, we can make the two diabatic energies cross twice – during the rise and during the fall of the Stark pulse. For definiteness we assume that $S_0 > 0$; then for level crossings to occur, Δ_0 must be in the range $0 < \Delta_0 < S_0$.

If the timing of the pulses is such that the pump pulse is sufficiently strong at both crossings, e.g. if the pump and Stark pulses coincide in time, then adiabatic evolution will occur at *both* crossings (Fig. 1, left frames). Such a pulse timing would drive the population first from ψ_1 to ψ_2 , and then back to ψ_1 . The success of SCRAP comes from using delayed pulses, such that the pump Rabi frequency is appreciable at only *one* of the cross-

ings. For definiteness we here assume that the pump pulse is present only during the rising portion of the Stark pulse (Fig. 1, right frames). Then the system, starting from state ψ_1 initially, will evolve adiabatically through the first crossing at time t_1 , following the adiabatic state $\Phi_-(t)$, and thus will make a transition to state ψ_2 . At the second crossing at time t_2 there is almost no pump laser field present and hence the system will evolve diabatically, i.e. it will follow the diabatic state ψ_2 in which it is prior to this crossing. The net result of this adiabatic–diabatic scenario will be complete population transfer from ψ_1 to ψ_2 .

3.2. SCRAP zones

The dependence of the transition probability on the static detuning Δ_0 exhibits *five distinct regions*, which are shown in Fig. 2 and separated by vertical lines.

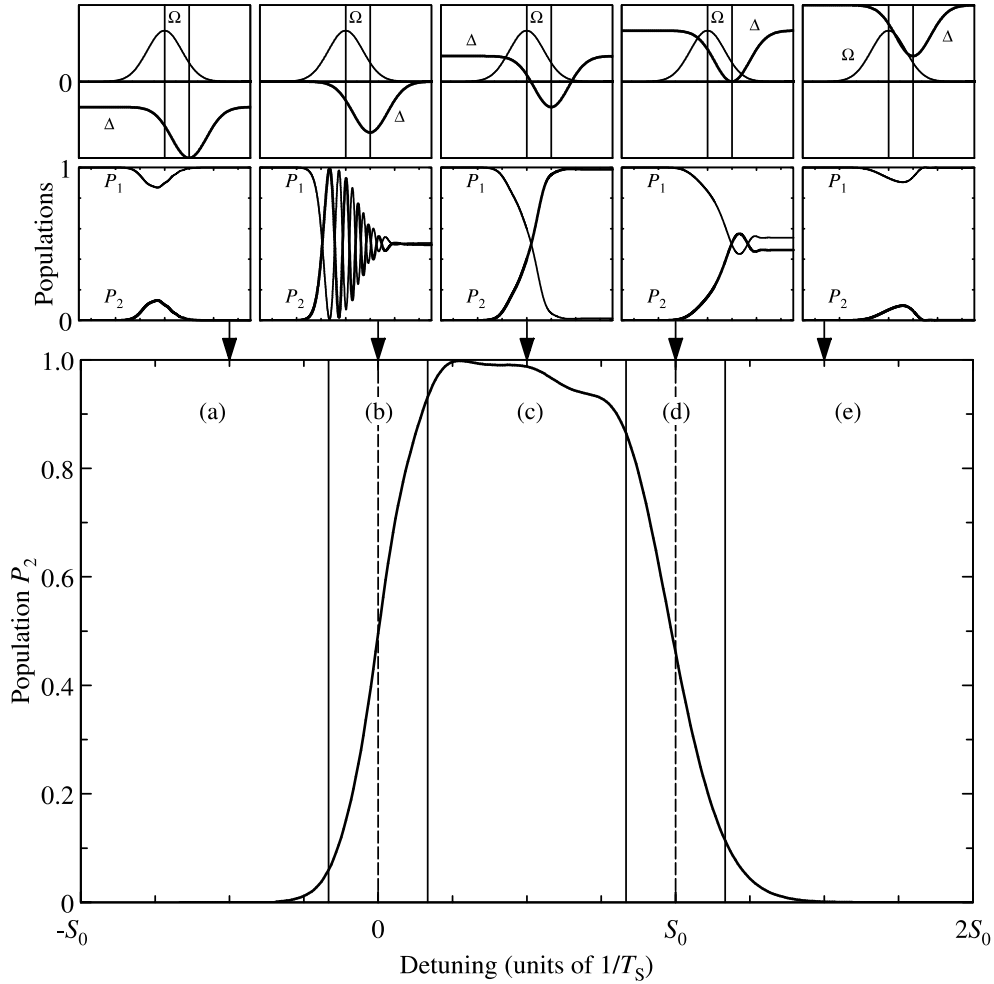


Fig. 2. Numerically calculated excited-state population versus the static detuning Δ_0 . The small frames on the top show the time dependences of the Rabi frequency $\Omega(t)$ and the Stark-shifted detuning $\Delta(t) = \Delta_0 - S(t)$ for five values of Δ_0 , shown by arrows. The small frames in the middle row show the time evolutions of the populations in these cases. We have assumed Gaussian shapes for the pump and Stark pulses, $\Omega(t) = \Omega_0 e^{-t^2/T_P^2}$ and $S(t) = S_0 e^{-(t-\tau)^2/T_S^2}$, with $\Omega_0 = 10T_S^{-1}$, $S_0 = 15T_S^{-1}$, and $\tau = T_P = T_S$.

(a) *Large negative static detuning* ($\Delta_0 < 0$). In this case there are no level crossings between the diabatic energies, because the effective detuning is always negative, $\Delta(t) = \Delta_0 - S(t) < 0$. The adiabatic energies are separated by a large gap, which prevents nonadiabatic transitions, i.e. the evolution is adiabatic. The system starts and ends in the same diabatic state ψ_1 (although some transient population may visit state ψ_2) and almost no population is left in ψ_2 at the end: $P_1 \approx 1$, $P_2 \approx 0$. This is an example of *coherent population return* [16].

(b) *Small static detuning* ($\Delta_0 \approx 0$). Near $\Delta_0 = 0$, the transfer efficiency rises from nearly zero to nearly unity. In this region, the atom ends up in a superposition of ψ_1 and ψ_2 . In particular, as we will show below, on resonance ($\Delta_0 = 0$) half of the population is transferred to ψ_2 if the evolution is adiabatic, $P_1 \approx P_2 \approx 1/2$.

(c) *SCRAP region* ($0 \lesssim \Delta_0 \lesssim S_0$). For such detunings, there are two level crossings. The evolution is adiabatic around the crossing where the pump pulse is applied and diabatic at the other crossing. Consequently, nearly complete popula-

tion transfer to ψ_2 occurs, $P_1 \approx 0$, $P_2 \approx 1$. The plateau in Fig. 2 displays the SCRAP region.

(d) *End of SCRAP region* ($\Delta_0 \approx S_0$). Near $\Delta_0 \approx S_0$, the transfer efficiency drops from nearly unity to nearly zero, which signals the end of the SCRAP plateau. In this region, as in the region around $\Delta_0 = 0$, the atom ends up in a superposition of ψ_1 and ψ_2 .

(e) *Large positive static detuning* ($\Delta_0 \gtrsim S_0$). In this case, as for large negative Δ_0 , there are no diabatic level crossings, because the effective detuning is always positive, $\Delta(t) = \Delta_0 - S(t) > 0$. The large gap between the adiabatic energies leads to adiabatic evolution and negligible population transfer, $P_1 \approx 1$, $P_2 \approx 0$.

Obviously, two detuning regions in Fig. 2 are candidates for half-SCRAP: the regions around $\Delta_0 = 0$ and $\Delta_0 = S_0$. However, the population transfer in the latter depends on the Stark shift S_0 and is therefore vulnerable to variations in the Stark laser intensity. In contrast, the region around $\Delta_0 = 0$ is immune to such fluctuations.

Fig. 3(a) displays the population P_2 of state ψ_2 plotted as a function of the static detuning Δ_0 for four different values of the maximum Stark shift S_0 (for the same maximum pump Rabi frequency Ω_0). Fig. 3(b) shows plots of the population P_2 versus Δ_0 for four different values of the maximum pump Rabi frequency Ω_0 (for the same maximum Stark shift S_0). As both figures demonstrate, in the region around $\Delta_0 = S_0$ the populations are sensitive to the laser parameters, whereas in the region around $\Delta_0 = 0$ the populations are quite robust against small-to-moderate variations in Ω_0 and S_0 . Therefore, we select the region around $\Delta_0 = 0$ for achieving half-SCRAP, and we explain below the robustness of the population transfer in this region.

4. Half-SCRAP

4.1. Half-SCRAP for zero detuning ($\Delta_0 = 0$)

Unlike SCRAP, in half-SCRAP the two opposite pulse sequences – pump-Stark and Stark-pump – lead to different results; we will consider them separately.

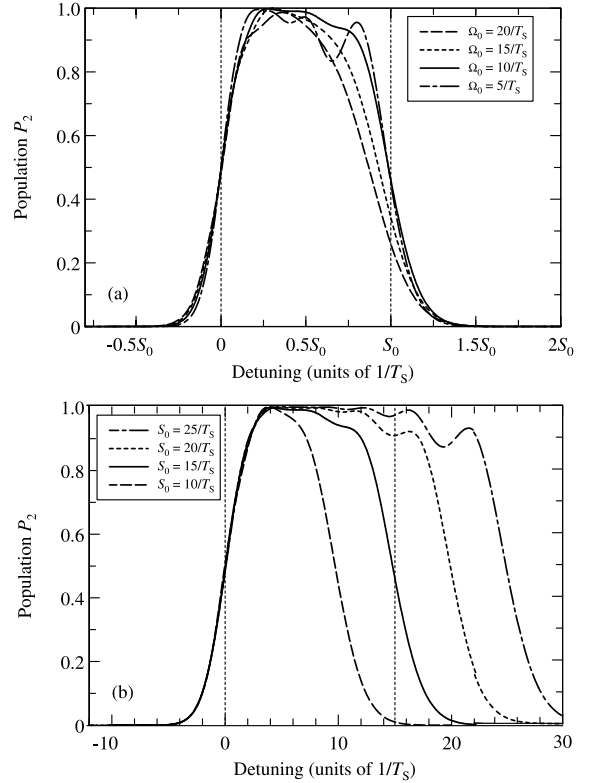


Fig. 3. Numerically calculated population of state ψ_2 versus the static detuning Δ_0 for Gaussian shapes of the pump and Stark pulses, with $\tau = T_p = T_S$; (a) four different peak pump Rabi frequencies Ω_0 for the same peak Stark shift $S_0 = 15T_S^{-1}$, (b) four different peak Stark shift values S_0 for the same peak pump Rabi frequency $\Omega_0 = 10T_S^{-1}$.

4.1.1. Pump pulse before Stark pulse

For $\Delta_0 = 0$, we have $\Delta(t) = -S(t)$. Hence, when the pump pulse precedes the Stark pulse,

$$-\infty \xleftarrow{t_i \leftarrow t} \frac{\Omega(t)}{\Delta(t)} \xrightarrow{t \leftarrow t_f} 0. \quad (19)$$

Then $\vartheta(t_i) = -\pi/4$, $\vartheta(t_f) = 0$, and the adiabatic states have the following asymptotics:

$$\frac{1}{\sqrt{2}}(\psi_1 + \psi_2) \xleftarrow{t_i \leftarrow t} \Phi_-(t) \xrightarrow{t \leftarrow t_f} \psi_1, \quad (20a)$$

$$\frac{1}{\sqrt{2}}(\psi_2 - \psi_1) \xleftarrow{t_i \leftarrow t} \Phi_+(t) \xrightarrow{t \leftarrow t_f} \psi_2. \quad (20b)$$

Thus, the adiabatic states are equally populated initially. If the system evolves adiabatically, their populations will not change and after the

interaction, the system will end up in an equal superposition of ψ_1 and ψ_2 ,

$$\Psi(t_f) = \frac{1}{\sqrt{2}}(\psi_1 + e^{-i\varphi_{PS}}\psi_2), \quad (21)$$

except an unimportant common phase, with

$$\varphi_{PS} = \omega_L(t_f - t_i) + \phi_L + \int_{t_i}^{t_f} \varepsilon(t) dt + \frac{\pi}{2}. \quad (22)$$

The robustness of the populations in the created superposition (21) against variations in the interaction parameters derives from the robustness of adiabatic passage. The superposition phase φ_{PS} , however, depends on these parameters.

4.1.2. Stark pulse before pump pulse

In the opposite pulse order, when the Stark pulse precedes the pump pulse, we have

$$0 \xleftarrow{t_i \leftarrow t} \frac{\Omega(t)}{\Delta(t)} \xrightarrow{t \rightarrow t_f} -\infty. \quad (23)$$

Then $\vartheta(t_i) = 0$, $\vartheta(t_f) = -\pi/4$, and

$$\psi_1 \xleftarrow{t_i \leftarrow t} \Phi_-(t) \xrightarrow{t \rightarrow t_f} \frac{1}{\sqrt{2}}(\psi_1 + \psi_2), \quad (24a)$$

$$\psi_2 \xleftarrow{t_i \leftarrow t} \Phi_+(t) \xrightarrow{t \rightarrow t_f} \frac{1}{\sqrt{2}}(\psi_2 - \psi_1). \quad (24b)$$

Hence state $\Phi_-(t)$ is the only adiabatic state populated initially, and in the adiabatic limit, no population will escape to the other adiabatic state $\Phi_+(t)$. Thus the system will be driven from state ψ_1 into the superposition

$$\Psi(t_f) = \frac{1}{\sqrt{2}}(\psi_1 + \psi_2 e^{-i\varphi_{SP}}) \quad (25)$$

except an unimportant common phase, with

$$\varphi_{SP} = \omega_L(t_f - t_i) + \phi_L. \quad (26)$$

The comparison of Eqs. (21) and (25) shows that, unlike full SCRAP, in half-SCRAP the pulse order makes a difference: the created superpositions have the same populations, but different relative phases between ψ_1 and ψ_2 . This difference occurs because for the pump-Stark order, the population flows through both adiabatic states $\Phi_-(t)$ and $\Phi_+(t)$, while for the Stark-pump order, only the adiabatic state $\Phi_-(t)$ is populated. Consequently, there is an additional dynamic phase in the former case.

4.1.3. Relative phase of the superposition

As evident from Eqs. (22) and (26), the relative phase between the components of the created superposition depends on the pump laser phase ϕ_L and changes in time after the preparation of the superposition. In some applications it may be important to have a constant phase. This is possible only if $E_2 = E_1$, which requires a two-photon Raman-type interaction between degenerate states. Because Raman pulses can be generated from one laser, and the coupling paths can be distinguished by polarization, the static detuning can be set automatically to zero ($\Delta_0 = 0$), and the relative laser phase can be set to zero or some fixed value.

4.1.4. Effect of Stark shifts induced by the pump pulse

So far we have ignored the Stark shifts induced by the pump pulse, $S_1^P f_P(t)$ and $S_2^P f_P(t)$. This is well justified when states ψ_1 and ψ_2 are connected by a single-photon transition because then the Rabi frequency is usually much larger than these Stark shifts. For a two-photon transition, however, these Stark shifts have to be accounted for, even if they are much smaller than the ones induced by the Stark laser, because they are comparable to the Rabi frequency. The most significant implication of their inclusion is that for $\Delta_0 = 0$, the populations in the created superposition are not equal, but are

$$\Psi(t_f) = \psi_1 \cos \Theta + \psi_2 e^{-i\varphi} \sin \Theta, \quad (27)$$

where $\Theta = \frac{1}{2} \arctan[\Omega_0 / (S_2^P - S_1^P)]$. Here φ denotes φ_{PS} for the pump-Stark sequence and φ_{SP} for the Stark-pump one. The pump-induced Stark shifts lead also to minor shifts in the times of the crossings.

4.2. Half-SCRAP for small detuning ($\Delta_0 \lesssim T_P^{-1}$)

For nonzero static detuning Δ_0 , we have $\vartheta(t_i) = \vartheta(t_f) = 0$; hence, at early and late times each adiabatic state tends to the same diabatic state, $\Phi_-(t_i) = \Phi_-(t_f) = \psi_1$ and $\Phi_+(t_i) = \Phi_+(t_f) = \psi_2$. Then the partial population transfer seen for $|\Delta_0| \lesssim T_P^{-1}$ in Figs. 2 and 3 can only result from transitions between the adiabatic states. The probability for such transitions depends on the nonadiabatic

coupling $\dot{\vartheta}(t)$ and the splitting $\varepsilon(t)$ between the adiabatic energies,

$$\dot{\vartheta}(t) = \frac{1}{2} \frac{\dot{\Omega}(t)[\Delta_0 - S(t)] + \Omega(t)\dot{S}(t)}{\Omega^2(t) + [\Delta_0 - S(t)]^2}, \quad (28a)$$

$$\varepsilon(t) = \sqrt{\Omega^2(t) + [\Delta_0 - S(t)]^2}. \quad (28b)$$

Nonadiabatic transitions are most likely to occur in the region around the maximum of the function

$$f(t) = \frac{|\dot{\vartheta}(t)|}{\varepsilon(t)}. \quad (29)$$

It can be shown that for the detuning range $|\Delta_0| \lesssim T_p^{-1}$ the maximum of $f(t)$ occurs in the outer wing of the pump pulse, away from the Stark pulse. Hence we can neglect the Stark shift $S(t)$ in Eqs. (28a) and (28b). Considered as a function of Δ_0 , $f(t)$ has its maximum at

$$\Omega(t_0) = |\Delta_0|\sqrt{2}. \quad (30)$$

Because the peak pump Rabi frequency is assumed large we have $\Omega_0 \gg T_p^{-1} \gtrsim |\Delta_0|$. Hence Eq. (30) is satisfied at times when $\Omega(t_0) \ll \Omega_0$, i.e. at several pulse widths from the center of the pump pulse; there the Stark shift $S(t)$ is negligibly weak, which justifies its neglect above.

By replacing Eq. (30) into Eqs. (28a) and (28b) we find

$$\dot{\vartheta}(t_0) = \frac{1}{3\sqrt{2}} \frac{\dot{\Omega}(t_0)}{\Omega(t_0)}, \quad \varepsilon = |\Delta_0|\sqrt{3}. \quad (31)$$

Therefore, in the nonadiabatic region, the nonadiabatic coupling $\dot{\vartheta}(t)$ does not depend on the peak Rabi frequency Ω_0 , and the eigenenergy splitting $\varepsilon(t)$ depends only on the detuning Δ_0 . Hence the probability for nonadiabatic transitions $\mathcal{P}$, and consequently, the composition of the superposition created by half-SCRAP, are primarily determined by the value of the static detuning Δ_0 and do not depend on the other interaction parameters.

It is easy to see that there is a symmetry against the sign of Δ_0 : when Δ_0 reverses its sign the populations in the superposition are reversed. In order to show this, we first note that the transition

probability in the adiabatic basis $\mathcal{P}$ does not depend on the sign of Δ_0 but only on $|\Delta_0|$. However, the transition probability in the diabatic basis p depends on the sign of Δ_0 . Indeed, because for $\Delta_0 > 0$ there is a level crossing, the transition probability in the adiabatic basis $\mathcal{P}$ is equal to the probability of no transition in the diabatic basis $1 - p$, i.e. the diabatic populations are $P_1 = \mathcal{P}$, $P_2 = 1 - \mathcal{P}$. In contrast, for $\Delta_0 < 0$ there is no level crossing, and the transition probabilities in the two bases are equal, $p = \mathcal{P}$; hence $P_1 = 1 - \mathcal{P}$, $P_2 = \mathcal{P}$.

In reality, because nonadiabatic transitions occur also away from the maximum of $f(t)$, the populations in the superposition depend (slightly) also on the pump Rabi frequency Ω_0 and the Stark shift S_0 . Indeed, small variations of the populations with Ω_0 and S_0 can be seen in Fig. 3 for small Δ_0 .

5. Analytic model

A simple, but realistic illustration of half-SCRAP is provided by the analytically solvable Demkov–Kunike (DK) model [17],

$$\Omega(t) = \Omega_0 \operatorname{sech}(t/T), \quad (32a)$$

$$\Delta(t) = \Delta_0 - B[\tanh(t/T) + 1], \quad (32b)$$

where the parameters Ω_0 , Δ_0 , B and T are assumed positive. The hyperbolic-secant time dependence of $\Omega(t)$ is close to Gaussian, which is used to model laser pulses in typical experiments. The behavior of the DK detuning (32b) is slightly different from the SCRAP detuning (6), which approaches the static detuning Δ_0 at both early and late times. In the DK model, $\Delta(t)$ starts from Δ_0 at $t \rightarrow -\infty$ and reaches monotonically the value $\Delta_0 - 2B$ as $t \rightarrow +\infty$. Hence the DK detuning (32b) behaves as the SCRAP detuning (6) from early to intermediate times, i.e. during the action of the pump pulse. Its divergence from a realistic SCRAP detuning at late times is not very significant because then the pump pulse vanishes and the interaction ceases. Hence the DK model (32a) and (32b) describes reasonably well a half-SCRAP pulse sequence, with the pump pulse acting first. Note that the DK parameter B is related to the maximum Stark shift S_0 .

The transition probability for the DK model is [17]

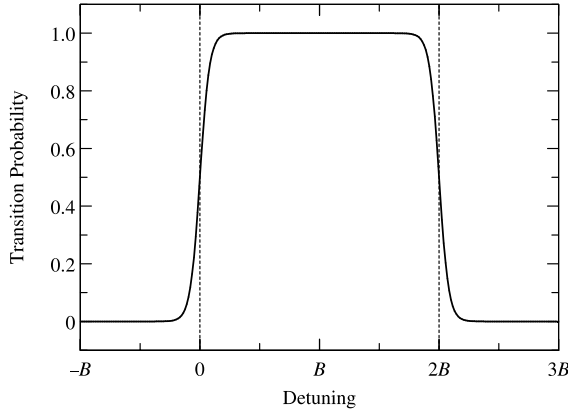


Fig. 4. The transition probability (33) for the DK model (32a) and (32b) versus the static detuning Δ_0 for $BT = 8$ and $\Omega_0 T = 10$.

$$P = \frac{\cosh(\pi TB) - \cos(\pi T \sqrt{\Omega_0^2 - B^2})}{\cosh(\pi TB) + \cosh(\pi T(B - \Delta_0))}. \quad (33)$$

It is plotted in Fig. 4 versus the static detuning Δ_0 . The observed profile is similar to those in Figs. 2 and 3.

For sufficiently large B ($BT \gtrsim 1$) and for sufficiently intense pump pulse [$(\Omega_0 T)^2 \gtrsim BT$], Eq. (33) reduces to

$$P \approx \frac{1}{1 + e^{-\pi \Delta_0 T}}. \quad (34)$$

Hence, the transition probability is determined primarily by the static detuning Δ_0 and depends very weakly on the other parameters, in agreement with our general analysis in Section 4.2. On resonance ($\Delta_0 = 0$), $P_1 \approx P_2 \approx 1/2$. Off resonance, the composition of the created superposition is controlled by Δ_0 ; for example, a superposition with populations P_1 and P_2 is created by static detuning

$$\Delta_0 \approx \frac{1}{\pi T} \ln(P_2/P_1). \quad (35)$$

Thus the change of sign in Δ_0 causes swap of P_1 and P_2 .

6. Conclusions

We have shown that the SCRAP technique, hitherto exploited for complete and robust popu-

lation transfer between two states, can be used to create coherent superpositions of the two states. SCRAP uses an intense off-resonant laser pulse (Stark pulse) to Stark shift the Bohr transition frequency and create a pair of diabatic level crossings. A moderately strong near-resonant, but slightly detuned, pump pulse, applied at one of these crossings, induces adiabatic population transfer. By taking the pump pulse width sufficiently small one can ensure that the pump pulse has little or no effect at the other crossing and so the evolution is diabatic there. This adiabatic–diabatic scenario achieves complete population transfer between the two states.

In the present paper we have shown that when the carrier frequency of the pump pulse is on resonance with the atomic Bohr frequency, an equal superposition of the two states is created. Off resonance, the created superposition has unequal contributions from the two states, which are determined mainly by the pump detuning Δ_0 . It is particularly significant that the superposition is insensitive to small-to-moderate changes of the intensities of the pump and Stark pulses. Half-SCRAP is robust also against variations in the carrier frequency of the Stark pulse and the delay between the two pulses.

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References

- [1] P. Brumer, M. Shapiro, *Annu. Rev. Phys. Chem.* 43 (1992) 257.
- [2] C.P. Williams, S.H. Clearwater, *Explorations in Quantum Computing*, Springer, Berlin, 1997;
D. Bouwmeester, A.K. Ekert, A. Zeilinger, *The Physics of Quantum Information: Quantum Cryptography, Quantum Teleportation, Quantum Computation*, Springer, Berlin, 2000.

- [3] C.S. Adams, M. Sigel, J. Mlynek, *Phys. Rep.* 240 (1994) 143.
- [4] S.E. Harris, *Phys. Today* 50 (1997) 36;
J.P. Marangos, *J. Mod. Opt.* 45 (1998) 471.
- [5] J. Mompart, R. Corbalán, *J. Opt. B* 2 (2000) R7.
- [6] A.S. Parkins, P. Marte, P. Zoller, H.J. Kimble, *Phys. Rev. Lett.* 71 (1993) 3095;
A.S. Parkins, P. Marte, P. Zoller, O. Carnal, H.J. Kimble, *Phys. Rev. A* 51 (1995) 1578;
A.S. Parkins, H.J. Kimble, *J. Opt. B* 1 (1999) 496.
- [7] F.I. Gauthey, C.H. Keitel, P.L. Knight, A. Maquet, *Phys. Rev. A* 52 (1995) 525;
J.B. Watson, A. Sanpera, X. Chen, K. Burnett, *Phys. Rev. A* 53 (1996) R1962.
- [8] B.W. Shore, *The Theory of Coherent Atomic Excitation*, Wiley, New York, 1990.
- [9] K. Bergmann, H. Theuer, B.W. Shore, *Rev. Mod. Phys.* 70 (1998) 1003;
N.V. Vitanov, T. Halfmann, B.W. Shore, K. Bergmann, *Ann. Rev. Phys. Chem.* 52 (2001) 763;
N.V. Vitanov, M. Fleischhauer, B.W. Shore, K. Bergmann, *Adv. At. Mol. Opt. Phys.* 46 (2001) 55.
- [10] P. Marte, P. Zoller, J.L. Hall, *Phys. Rev. A* 44 (1991) R4118;
M. Weitz, B.C. Young, S. Chu, *Phys. Rev. A* 50 (1994) 2438;
L.S. Goldner, C. Gerz, R.J.C. Spreeuw, S.L. Rolston, C.I. Westbrook, W.D. Phillips, P. Marte, P. Zoller, *Phys. Rev. Lett.* 72 (1994) 997;
Quantum Opt. 6 (1994) 387;
J. Lawall, M. Prentiss, *Phys. Rev. Lett.* 72 (1994) 993;
N.V. Vitanov, K.-A. Suominen, B.W. Shore, *J. Phys. B* 32 (1999) 4535.
- [11] M. Weitz, B.C. Young, S. Chu, *Phys. Rev. Lett.* 73 (1994) 2563.
- [12] P.D. Featonby, G.S. Summy, J.L. Martin, H. Wu, K.P. Zetie, C.J. Foot, K. Burnett, *Phys. Rev. A* 53 (1996) 373;
P.D. Featonby, G.S. Summy, C.L. Webb, R.M. Godun, M.K. Oberthaler, A.C. Wilson, C.J. Foot, K. Burnett, *Phys. Rev. Lett.* 81 (1998) 495.
- [13] R. Unanyan, M. Fleischhauer, B.W. Shore, K. Bergmann, *Opt. Commun.* 155 (1998) 144;
H. Theuer, R.G. Unanyan, C. Habscheid, K. Klein, K. Bergmann, *Opt. Express* 4 (1999) 77.
- [14] L.P. Yatsenko, B.W. Shore, T. Halfmann, K. Bergmann, A. Vardi, *Phys. Rev. A* 60 (1999) R4237.
- [15] T. Rickes, L.P. Yatsenko, S. Steuerwald, T. Halfmann, B.W. Shore, N.V. Vitanov, K. Bergmann, *J. Chem. Phys.* 113 (2000) 534.
- [16] N.V. Vitanov, *J. Phys. B* 28 (1995) L19;
N.V. Vitanov, P.L. Knight, *J. Phys. B* 28 (1995) 1905;
A. Kuhn, S. Steuerwald, K. Bergmann, *Eur. Phys. J. D* 1 (1998) 57.
- [17] Y.N. Demkov, M. Kunike, *Vestn. Leningr. Univ. Fiz. Khim.* 16 (1969) 39;
K.-A. Suominen, B.M. Garraway, *Phys. Rev. A* 45 (1992) 374.