Efficient adiabatic population transfer by two-photon excitation assisted by a laser-induced Stark shift


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We demonstrate and analyze a novel scheme for complete transfer of atomic or molecular population between two bound states, by means of Stark-chirped rapid adiabatic passage (SCRAP). In this two-laser technique a delayed-pulse laser-induced Stark shift sweeps the transition frequency between two coupled states twice through resonance with the frequency of the population-transferring coupling laser. The delay of the Stark-shifting pulse with respect to the pulse of the coupling-laser Rabi frequency guarantees adiabatic passage of population at one of the two resonances while the evolution is diabatic at the other. The SCA method can give a population-transfer efficiency approaching unity. We discuss the general requirements on the intensity and timing of the pulses that produce the Rabi frequency and, independently, the Stark shift. We particularly stress extension to a double-SCRAP technique, a coherent variant of stimulated emission pumping in the limit of strong saturation. We demonstrate the success of the SCA method with experiments in metastable helium, where a two-photon transition provides the Rabi frequency.

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I. INTRODUCTION

Since the early days of modern atomic, molecular, and optical physics researchers have continually improved techniques that can force an ensemble of particles into a desired quantum state by means of resonant or near-resonant radiation. Some techniques, such as stimulated emission pumping (SEP), off-resonance stimulated-Raman scattering rely mainly on the high spectral energy density of laser radiation, while others, such as π-pulse methods and chirp-induced processes, also exploit its coherent nature for successful completion of the transfer process.

Here we propose and demonstrate a scheme which employs adiabatic passage by chirping a transition frequency and which is particularly promising for pulses of nanosecond duration. This work grew out of previous extensive work on the STIRAP method (stimulated Raman adiabatic passage). STIRAP uses a delayed-pulse sequence of adiabatic interactions to transfer all the population of an initial quantum state to a final one, via radiative coupling through an intermediate state. Briefly stated, adiabatic evolution requires that the pulse area is large (the transition needs to be saturated) and the phase fluctuations must be very small (i.e., a nearly Fourier-transform limited spectral bandwidth).

When the STIRAP method is properly implemented, and when the conditions for adiabatic evolution are met, the intermediate state is never populated. Therefore, this method does not suffer from the often detrimental consequences of spontaneous emission.

Radiation with properties suitable for STIRAP is available in the visible and the near ultraviolet region of the spectrum. However, for many molecules the first electronically excited state, which must provide the coupling between the initial and the final states, can only be reached with radiation in the vacuum ultraviolet (VUV) spectral region. Unfortunately, VUV radiation with adequate power and (narrow) bandwidth is not readily available. It was therefore natural to consider multiphoton versions of STIRAP, where the coupling between the initial and the intermediate states (and possibly also between the intermediate and final states) is induced by a two-photon interaction (a hyper-Raman coupling). Although conditions for successful transfer have been identified and the feasibility of multiphoton STIRAP has been experimentally confirmed, process suffers from unavoidable ac Stark shifts. Their presence prohibits the existence of a dark (population-trapping) state of the dressed Hamiltonian. Such a trapping state correlates to the initial state at early times and to the final target state at late times, and has no population in the intermediate state. When it does not exist, there occurs some transient population in the intermediate state. As a consequence, spontaneous emission is not fully avoided and one of the essential advantages of STIRAP is lost. Therefore, it was an obvious and interesting task to search for alternative methods for efficient population transfer in the presence of substantial ac Stark shifts. The population-transfer method described here—two-photon excitation assisted by a laser-induced Stark shift—is an exten-
sion of well-established chirping techniques (which are based on well-controlled tuning of a two-level system into and out of resonance with a coherent radiation field)\textsuperscript{12–30} to the regime of nanosecond pulses.

Manipulation of the population of quantum states by frequency-chirping techniques has a long history. First developed in the context of nuclear magnetic resonance\textsuperscript{31} its application in the optical region was suggested in the late 1960s.\textsuperscript{32} The process was demonstrated in the 1970s, during the early days of the application of laser radiation in atomic and molecular physics involving one-photon\textsuperscript{33–35} or two-photon\textsuperscript{36–38} transitions. Methods based on frequency sweeping (chirping) are well established for cw lasers (or for pulsed radiation with pulse length exceeding microsecond duration, see, e.g., Refs. 12–14). For continuous (or long-pulse) lasers active electro-optic or acousto-optic modulation techniques provide straightforward means to tune the laser frequency through resonance with the transition frequency of a two-state system. Alternatively, atoms or molecules that cross the axis of a cw laser beam at nearly right angles slightly displaced from the minimum waist of a focused Gaussian beam will experience a frequency chirp due to the curvature of the wave front.\textsuperscript{12–14} It is also straightforward to impose a frequency chirp on picosecond and femtosecond pulses.\textsuperscript{20,39} The large bandwidth of such ultrashort pulses allows spatial dispersion of the spectrum and subsequent time-delayed spatial recombination of the various frequency components.

However, chirp methods are not well developed for pulses of nanosecond duration. Chirping of the laser frequency by active phase modulation, although possible in principle, is inconvenient because it requires driving modulators at gigahertz frequencies. On the other hand, the spectral bandwidth of nanosecond pulses is too small for successful application of the techniques based on spatial dispersion that are well developed for femtosecond pulses. Although there are few, if any, examples of achieving adiabatic passage with nanosecond pulses by chirping the laser frequencies, the possibility of using external electric fields\textsuperscript{33} or laser-induced ac Stark shifts\textsuperscript{34,40} for the manipulation of state population was recognized and demonstrated more than 20 years ago. Transient coherent phenomena\textsuperscript{37}—or even gain\textsuperscript{37}—were observed. A Stark shift induced by a nanosecond laser pulse modifies the transition frequency, and thus offers a natural and straightforward means for inducing adiabatic passage processes. However, such shifts have not yet been exploited to produce efficient population transfer. Unless implemented correctly, the phenomenon of coherent population return (CPR)\textsuperscript{41–43} will drive any transient population in the upper state back to the ground state, thereby diminishing the net population transfer.

In this work we show that efficient population transfer will occur when a pair of laser pulses is appropriately timed: one pulse (usually far off resonance) induces the Stark shift and the other (near resonance) pulse provides two-photon coupling between the states. In order to prevent CPR the two laser pulses must not be simultaneous. As we explain later, the delay between pulses occurs for a different reason than the delay that is essential for STIRAP or the delay usually imposed for SEP.

In Sec. II we explain the basic concept of Stark-chirped rapid adiabatic passage (SCRAP) followed in Sec. III by a discussion of the required conditions on the interaction parameters. A preliminary account of the SCRAP method has been given in Ref. 44. Results from an experimental feasibility study, using metastable helium atoms as a model system is the theme of Sec. IV. Section V mentions important extensions to a “double-SCRAP” scheme. These include a scheme for inducing two sequential adiabatic passage processes with only one Stark-shift pulse. That version of the SCRAP scheme allows efficient and selective population transfer into highly vibrationally excited levels in the electronic ground state of molecules, if the lifetime of the electronically excited state is longer than the laser pulse width. It can be considered as a coherent counterpart of the SEP technique, allowing very efficient population transfer and reducing the detrimental contributions from spontaneous emission.

II. THE SCRAP CONCEPT

In this section we use a model system to demonstrate the conceptually simple, potentially very powerful, and versatile concept combining the convenience of two-photon excitation (a convenient wavelength range, not in the far UV) and the robustness of adiabatic passage (by Stark-induced chirp). The level scheme and radiative couplings, which are the basis of the following discussion, are shown in Fig. 1. Modifications and extensions of that scheme will be discussed later.

A. Summary of rapid adiabatic passage theory

The essence of the SCRAP procedure is the well-established technique of rapid adiabatic passage (RAP),\textsuperscript{4,5} understood most simply from the two-state Hamiltonian

\[
H(t) = \frac{\hbar}{2} \begin{bmatrix} 0 & \Omega(t) \\ \Omega(t) & 2\Delta(t) \end{bmatrix}
\]

where the pulsed Rabi frequency \(\Omega(t)\) connects states \(|\psi\rangle\) and \(|\psi'\rangle\) radiatively, and the variable detuning \(\Delta(t)\) expresses

\[E_0 - \Delta_0 = \hbar \omega_0 - \hbar \Delta(t) = \hbar \omega_p - 2\hbar \omega_p t^2 / \hbar \omega_p t^2,\nonumber\]

where \(\omega_p\) is the frequency of the first pulse, \(\Delta(t)\) is the curvature of the wave front, and \(t\) is the delay between pulses.

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\[\begin{align*}
\text{Fig. 1. Two-photon radiative coupling in a two-state system.} \\
\text{The target state 2 is coupled to the initial state 1 by two photons with frequency \(\omega_p\),} \\
\text{detuned by \(\Delta_0 = \omega_0 - 2\omega_p\) from the Bohr frequency \(\omega_0\). The far off-} \\
\text{resonance radiation of frequency \(\omega_s\) Stark shifts the transition frequency \(\omega_0\).} \\
\end{align*}\]
deviation from resonant excitation. The adjective “rapid” implies that population transfer occurs during a time interval shorter than relaxation times, so that the dynamics is governed by the Schrödinger equation (rather than by a density-matrix equation). Although the details of the time variation in the Hamiltonian elements are established by the choice of the atomic transition and of laser characteristics, for the moment we regard $\Omega(t)$ and $\Delta(t)$ as independently controllable pulses.

When the state vector $\Psi(t)$ initially is a single energy state (say $|1\rangle$) and the Hamiltonian changes sufficiently slowly (adiabatically), then $\Psi(t)$ coincides at all times with one of the two instantaneous eigenstates of the Hamiltonian (1). These eigenstates can be written

$$\Phi_-(t) = \cos \Theta(t)|1\rangle - \sin \Theta(t)|2\rangle,$$

$$\Phi_+(t) = \sin \Theta(t)|1\rangle + \cos \Theta(t)|2\rangle,$$

where the mixing angle $\Theta(t)$ is defined, modulo $2\pi$, by the formula

$$\tan 2\Theta(t) = \Omega(t)/\Delta(t), \quad -\pi \leq 2\Theta \leq 0.$$

Obviously, $\Theta(t)$ is controlled by the ratio of the Rabi frequency and detuning, while the rate of change $\theta(t)$ varies with the time derivative of that ratio.

If during the time interval $-\infty < t < +\infty$ the detuning $\Delta(t)$ sweeps from some very large negative value to some very large positive value (irrespective of whether the laser frequency or the transition frequency is changed), then the angle $\Theta(t)$ rotates clockwise from $0$ to $-\pi/2$ and the adiabatic eigenstate $\Phi_-(t)$ changes from $|1\rangle$ to $|2\rangle$:

$$\Phi_-(\infty) = |1\rangle, \quad \Phi_-(+\infty) = |2\rangle.$$  

Thus such an adiabatic change (chirp) of $\Delta(t)$ will produce complete population transfer (adiabatic passage) from the initially populated state $|1\rangle$ to the initially unpopulated state $|2\rangle$. We note here that adiabatic passage does not depend on the sign of the detuning chirp and it can take place also when $\Delta(t)$ sweeps adiabatically from some very large positive value to some very large negative value; then $\Theta(t)$ rotates counterclockwise from $-\pi/2$ to $0$ and the population is transferred via the eigenstate $\Phi_+(t)$ which changes from $-|1\rangle$ to $|2\rangle$.

In this context slow (adiabatic) variation means that the magnitude of the rate of change in the mixing angle, $|\Theta'(t)|$, must be much smaller than the separation $|E^+(t) - E^-(t)|$=

$$E^{\pm}(t) = \frac{\hbar}{2} \left[ \Delta(t) \pm \sqrt{\Delta^2(t) + 4 \Omega^2(t)} \right].$$

The criterion for adiabatic evolution can be presented as the condition that the adiabaticity function

$$f(t) = \frac{|\Theta(t)|}{\sqrt{\Delta^2(t) + 4 \Omega^2(t)}}$$

is small at all times,

$$f(t) \ll 1.$$  

In practice $f(t) < 0.3$ ensures adiabatic evolution, while $f(t) > 3$ characterizes a diabatic process.

**B. Stark chirping**

1. The Hamiltonian

Extension of the results for the one-photon two-state process to one involving $N$ photons (here $N = 2$ is of interest) is straightforward. The relevant two-photon rotating-wave approximation Hamiltonian can be written as

$$H(t) = \hbar \begin{bmatrix} D_1(t) & \frac{\Omega(t)}{2} \\ \frac{\Omega(t)}{2} & D_2(t) \end{bmatrix}.$$  

The diagonal elements of this Hamiltonian include dynamic Stark shifts $S_1(t)$ and $S_2(t)$ for the two states,

$$D_1(t) = S_1(t),$$

$$D_2(t) = S_2(t) + \Delta_0,$$

where $\Delta_0$ is the static two-photon detuning from the Bohr-frequency $\omega_0 = (E_2 - E_1)/\hbar$. When the two photons come from the same field, as we assume, the detuning is defined by

$$\hbar \Delta_0 = E_2 - E_1 - 2\hbar \omega_p,$$

where $\omega_p$ is the carrier frequency of the pump field. The dynamic Stark shifts are proportional to the product of a pair of electric-field amplitudes [an intensity $I(t)$] and a matrix element of the (frequency dependent) polarizability tensor $\alpha(\omega)$. For radiation linearly polarized (in the $z$ direction), which we assume, and a single pulsed field of intensity $I(t)$ the definition reads

$$S_j(t) = -\frac{1}{2\hbar \epsilon_0} \left[ \langle j|\alpha_{zz}(\omega)|j\rangle I(t), \quad (j=1,2). \right.$$  

When computing dynamic Stark shifts it is important to include the effect of each field upon each transition; the definition (11) must be modified accordingly. Typically the shifts are greater for excited states than for the ground state, because the former are more polarizable.

The Rabi frequency $\Omega(t)$ appearing in Eq. (8) refers to a two-photon interaction; like the dynamic Stark shift, it is proportional to the product of a polarizability matrix element and an intensity,$^5$

$$\Omega(t) = -\frac{1}{2\hbar \epsilon_0} \langle 1|\alpha_{zz}(\omega)|2\rangle I(t).$$

The two-photon Rabi frequency appearing here contrasts with the more common single-photon Rabi frequency, which is the product of a dipole-transition matrix element $\mu$ and an electric-field amplitude $E(t)$ (proportional to the square root of intensity),$^5$

$$\Omega(t) = \mu E(t)/\hbar.$$

2. The eigenvectors

The eigenvectors $\Phi_{\pm}(t)$ of this Hamiltonian are identical to those of the Hamiltonian (1) with the identification

$$\Delta(t) = D_2(t) - D_1(t) = \Delta_0 - S(t),$$

where $S(t) = S_1(t) - S_2(t)$ is the difference between the two Stark shifts and $\Delta_0$ is the static detuning (10). The resonance
condition $\Delta(t)=0$ implies that two photons combine to carry the excitation energy. From Eqs. (5) and (13) the two adiabatic eigenvalues are

$$E_{\pm}(t) = \hbar S(t) \pm \frac{\hbar}{2} \left[ \Delta_0 - S(t) \right] \pm \frac{\hbar}{2} \left[ \Omega^2(t) + \left[ \Delta_0 - S(t) \right]^2 \right].$$

(14)

The essential feature of the transfer process proposed here is to shift the Bohr frequency by using a dynamic Stark shift proportional to the laser intensity. In many experiments strong fixed frequency lasers, operating in the infrared and visible spectral region, are used to pump oscillators or amplifiers and the remainder of this laser is available for use as the “Stark pulse.” As will be explained, it is necessary to use two pulses, whose maxima are offset by a suitable delay; one (pump pulse) which couples the levels and thus controls $\Omega(t)$, and one (Stark pulse) which controls the variation $S(t)$ of the transition frequency. The resulting population transfer we term Stark-chirped rapid adiabatic passage (or SCRAP).

C. Delayed interaction

Here we show examples which demonstrate why an additional interaction with a delayed laser pulse, causing Stark shifts, is needed for complete population transfer.

1. On-resonance case

Because the Stark shift $S(t)$, induced by the Stark-shift laser and/or the pump laser, is pulsed, it starts and ends with a null value, $S(\pm \infty)=0$. Therefore if the laser carrier frequency $\omega_p$ is tuned on two-photon resonance with the Bohr frequency $\omega_0$ in the absence of the Stark shift ($\Delta_0=0$), then the induced Stark shift will take the system away from resonance and therefore excitation will be hindered; see left frame of Fig. 2. The straight dotted horizontal line marks the transition frequency in the absence of any Stark shift of the levels. The other dotted line shows the variation of the transition frequency due to the Stark shift. The solid lines $E^+$ and $E^-$ show the dressed state eigenvalues including the Stark shifts as well as the coupling by the pump laser. This picture is qualitatively the same with or without the Stark pulse.

2. Off-resonance case with coincident pulses

If the pump laser is tuned away from the Bohr frequency ($\Delta_0 \neq 0$) and the static detuning $\Delta_0$ and the Stark shift $S(t)$ have opposite signs, the picture will be qualitatively the same as in the on-resonance case: The Stark shift takes the system further away from resonance as shown in the left frame of Fig. 2. If $\Delta_0$ and $S(t)$ have the same signs, however, as shown in the middle frame of Fig. 2, then a sufficiently large Stark shift will create a pair of degeneracies (level crossings) where the resonance condition $S(t)=\Delta_0$ is temporarily met. One resonance occurs while $S(t)$ is increasing, another one when $S(t)$ is decreasing. The shift caused by the pump laser is usually (much) smaller than the one caused by the Stark-shift laser. In the middle frame of Fig. 2 we show the case when the two pulses coincide.

The behavior of the population depends critically on the strength of the pulsed interaction $\Omega(t)$ at each resonance $\Delta(t) = \Delta_0 - S(t) = 0$. When $\Omega(t)$ is strong at a resonance, the adiabatic curves (see the solid lines) exhibit a strongly avoided crossing; then the system will evolve adiabatically and the atom will undergo complete population transfer. However, for the timing of the pulses shown in the middle frame of Fig. 2 the field is (or the two fields are) of equal strength at both resonances. Therefore population transfer at the first resonance is reversed at the second one (as was the case in the demonstration by Loy $^{33}$). In particular, if the coupling is sufficiently strong at the resonances to assure adiabatic evolution, population transfer will be complete at the first crossing as well as at the second one. If the coupling is weak, there will be no transfer at either crossing. In both cases, the net result at the end is no population transfer at all.

In the case of intermediate pulse strength, there will be partial population transfer at each crossing, resulting in splitting of the wave function at the first crossing and subsequent recombination at the second, which leads to oscillatory behavior of the final populations as a function of the intensity of the lasers or the static detuning $\Delta_0$.

3. Off-resonance case with delayed pulses

From these observations, it is straightforward to arrive at a coupling scheme which results in efficient excitation. The key to one-way transfer is to ensure that the pump field is strong at one crossing but has negligible effect at the other one. We propose to do this by introducing a delay between the pulses, such that the coupling is strong (i.e., population transfer occurs) at only one of the two resonances. This is the essence of the SCRAP technique. The right frame of Fig. 2
shows an example of this case in which the peak value of the pump pulse coincides with the moment of resonance $[\Delta(t) = 0]$ (see the crossing of the dotted lines) and the peak of the Stark pulse occurs later; consequently, there is negligible interaction $\Omega(t)$ at the second resonance. The pump pulse is sufficiently strong to produce an adiabatic passage at the first resonance (see the solid lines), but has negligible effect at the second resonance. The overall result is population transfer between the two atomic states.

Note that the delay between the STIRAP pulses (Stokes before pump) is needed for an entirely different reason: For STIRAP the pulse delay is needed to ensure that the state vector $\Psi(t)$ follows the population-trapping adiabatic state, whereas the delay between the pump and Stark pulses in SCRAP is needed to prevent a second adiabatic passage process. It is also obvious from the above analysis that, in contrast to STIRAP, the pulse ordering in SCRAP is irrelevant and complete population transfer can occur whether the Stark pulse follows or precedes the pump pulse.

II. CONDITIONS FOR SCRAP

A. Analytical estimates

We consider a specific model system in order to identify the conditions needed for SCRAP. We assume that the Rabi frequency of the pump pulse $\Omega(t)$ and the Stark shift $S(t)$ of the transition frequency induced by the second pulse are given by the Gaussian expressions

$$\Omega(t) = \Omega_0 \exp[-t^2/\tau_p^2],$$  \hspace{1cm} (15a)

$$S(t) = S_0 \exp[-(t-\tau)^2/\tau_S^2].$$  \hspace{1cm} (15b)

For a given system $S(t)$ and $\Omega(t)$ are calculated using Eqs. (12) and (11), respectively. The time delay $\tau$ between the two pulses will be assumed positive without loss of generality. For simplicity, we assume that the Stark shift induced by the pump laser is negligibly small compared to $S(t)$ (which is the case in the present experiment); if it is non-negligible but is smaller than $S(t)$, the equations below will be modified quantitatively but the qualitative picture remains unchanged.

1. Existence of level crossings

Obviously, the laser-induced Stark shift $S(t)$ can create level crossings only if its maximum $S_0$ is larger than the static detuning $\Delta_0$ and if $S_0$ and $\Delta_0$ have the same sign,

$$|\Delta_0| < |S_0|,$$  \hspace{1cm} (16a)

$$S_0\Delta_0 > 0.$$  \hspace{1cm} (16b)

Then there are two level crossings, occurring at times $t_1$ and $t_2$, where

$$t_1 = \tau - \tau_S \sqrt{\frac{S_0}{\Delta_0}} \hspace{1cm} t_2 = \tau + \tau_S \sqrt{\frac{S_0}{\Delta_0}}$$  \hspace{1cm} (17)

The idea of SCRAP is to maximize the probability for transition near the first crossing $t_1$ and minimize this probability near the second crossing $t_2$. This means that the evolution should be made adiabatic in a region around $t_1$ and diabatic at $t_2$.

It is important to note that by changing the laser parameters $\tau$, $S_0$, and $\Delta_0$ we can move the crossing points along the time axis. It is natural to make the first crossing occur at $t_1 = 0$ because at that moment the pump pulse reaches its maximum; such a choice would improve adiabaticity and hence maximize the transition probability there. This choice leads to the condition

$$\Delta_0 = S_0 \exp(-\tau^2/\tau_S^2),$$  \hspace{1cm} (18)

which will be assumed in the following analysis; then $t_1 = 0$ and $t_2 = 2\tau$.

2. Adiabatic passage

To achieve transfer efficiency close to unity, one must ensure that the evolution is adiabatic everywhere except at the second crossing $t_2$. This means that the adiabaticity function (6) must be small at any time except $t = t_2$. The coupling between the adiabatic states is given by

$$\Theta(t) = \frac{1}{2} \frac{\Omega(t) [\Delta_0 - S(t)]}{\Omega(t) + S(t) - \Delta_0 - S(t)}$$  \hspace{1cm} (19)

[see Eqs. (3) and (13)] and the eigenenergy splitting is

$$e(t) = \sqrt{\Omega^2(t) + [\Delta_0 - S(t)]^2}.$$  \hspace{1cm} (20)

The condition for adiabatic evolution is

$$n |\Theta(t)| < e(t),$$  \hspace{1cm} (21)

where hereafter $n$ is an appropriately chosen (large) number. Its choice depends on how much nonadiabatic transitions we can allow, the probability for which is $\leq 1/n^2$; hence for $3 \leq n \leq 10$, the transfer efficiency is in the range 91%–99%.

For the pulse shapes (15) and with condition (18) the adiabatic condition (21) applied at the first crossing $t_1$ becomes

$$n\Delta_0 \tau < \Delta_0^2 \tau^2.$$  \hspace{1cm} (22)

This condition imposes an upper limit on the detuning $\Delta_0$ and a lower limit on the Rabi frequency $\Omega_0$. For small-to-moderate pulse delays $\tau$, the fulfillment of condition (22) ensures adiabatic evolution not only at the crossing $t_1$ but also in a certain time interval around it; this is so because the coupling $\Theta(t)$ has its maximum near $t_1$.

For large pulse delay $\tau$, as can readily be verified, the maximum of $\Theta(t)$ near $t_1$ splits into two maxima which move away from the crossing and grow in amplitude as $\tau$ increases. In order to avoid nonadiabatic transitions near these maxima, we must impose the adiabaticity condition (21) there. Particularly significant is the maximum that moves toward early times because the eigenenergy splitting $e(t)$ decreases there (the other maximum, which is situated between $t_1$ and $t_2$, is neutralized by a relatively large eigenenergy splitting). As Eqs. (15) and (19) show, at early times the term involving $\dot{S}(t)$ in Eq. (19) vanishes exponentially and can be neglected in comparison with the term $\Omega(t)/\Omega(t)$ itself. Thus the coupling $\Theta(t)$ is represented by the product of a rapidly (exponentially) changing fraction and a slowly (linearly) changing factor $\Omega(t)/\Omega(t)$.

Hence the
maximum of $\Theta(t)$ will be determined mainly by the maximum of the former fraction which occurs at $\Delta_0 - S(t) = \Omega(t)$. By neglecting the term $S(t)$ [which is small compared to $\Omega(t)$ because of the large delay] we find that $t_{\text{max}} = - T_p \sqrt{\ln(\Omega_0/\Delta_0)}$. Then condition (21), applied at time $t_{\text{max}}$, gives

$$\Delta_0 T_p > n \sqrt{\frac{1}{2} \frac{\Omega_0}{\ln \Delta_0}}. \quad (23)$$

This condition imposes a lower limit on $\Delta_0$ [and via Eq. (18) an upper limit on $\tau$, while the restriction on the Rabi frequency $\Omega_0$ is rather weak.

3. Completeness of the adiabatic passage

To achieve transfer efficiency close to unity, one should also ensure that the ratio $\Omega(t)/\Delta(t)$, which controls the mixing angle $\Theta(t)$ (3), is small at both the initial and final times of the adiabatic passage. At the initial time $t \rightarrow -\infty$ this is always the case because $\Omega(-\infty)/\Delta(-\infty)$ = 0. As a final time of the transition we can take the time $t = \tau$ when $|\Delta(t)|$ is maximum; this leads to the condition

$$n \Omega_0 \exp(-\tau^2/T_p^2) < |S_0 - \Delta_0|. \quad (24)$$

In addition, the transition should be completed by the time $t = \tau$. In the adiabatic limit the transition duration is given approximately by $t_v = \Omega_0/|\Delta(0)|$, a half of this time being before the crossing at $t_1 = 0$ and a half after it. The requirement $t_v < \tau$ leads to the condition

$$\Omega_0 < \frac{2 \Delta_0 \tau^2}{T_p^2}. \quad (25)$$

Both conditions (24) and (25) impose lower limits on $\Delta_0$ and upper limits on $\Omega_0$.

4. Diabatic transition at the second crossing

The condition for diabatic transition at the second crossing $t_2$ requires that

$$|\Theta(t_2)| > n \epsilon(t_2).$$

For the pulse shapes (15) and with condition (18) we have

$$\Delta_0 \tau > n \Omega_0^2 T_p^2 \exp(-8 \tau^2/T_p^2). \quad (26)$$

This condition imposes a lower limit on $\Delta_0$ and an upper limit on $\Omega_0$.

By comparing conditions (22) and (26) one can derive a simple estimate for the minimum time delay:

$$\tau_{\text{min}} = \frac{1}{4} T_p \sqrt{\ln n}. \quad (27)$$

For smaller delays the conditions (22) and (26) cannot be fulfilled simultaneously.

5. Discussion

Conditions (16) and (22)–(26) provide the restrictions on the interaction parameters needed for SCRAP to work. They provide various lower and upper limits for the interaction parameters: the pump Rabi frequency $\Omega_0$, the static detuning $\Delta_0$, the pulse delay $\tau$, the peak Stark shift $S_0$, and the two pulse widths $T_p$ and $T_S$. Hence, when plotted as a function of a given experimental parameter (we plot it against $\Delta_0$), the transfer efficiency is expected to be high within a certain interval and low outside it. The limits of this interval are determined by the most stringent of conditions (22)–(26); which ones these are depends on the values of the other parameters. In the experiment reported in this paper, the experimentally accessible parameters by which the transfer can be controlled are $\tau$, $\Delta_0$, $S_0$, and $\Omega_0$. The pulse widths $T_p$ and $T_S$ are in general not adjustable and they are related by $T_S \geq 1.5 T_P$; it can be shown from the above-derived conditions that the relation $T_S > T_P$ is favorable for SCRAP.

Figure 3 displays the various regimes of adiabatic or diabatic evolution at the two crossings, as controlled by two dimensionless parameters: the ratio $x = \tau/T_S$ of the delay $\tau$ to the width $T_S$ of the Stark-shift pulse, and the (normalized) ratio $y = \Omega_0 T_S / S_0 T_S$ of the peak pump Rabi frequency to the square root of the peak Stark shift. The two curves, $y = \sqrt{3} x e^{-x^2}$ and $y = \sqrt{\frac{1}{2}} x e^{1.5 x^2}$, display Eqs. (22) and (26) with $n = 3$ and $T_S = 1.5 T_P$, where $\Delta_0$ is given by Eq. (18). In order to achieve adiabatic evolution at the first resonance, the Rabi frequency has to be larger than a lower limit, depending on the delay between the pulses and on the peak Stark shift. On the other hand, the Rabi frequency has to be sufficiently small, in order to achieve diabatic evolution at the second crossing (upper curve). These two conditions define a range of Rabi frequencies for which evolution is adiabatic at the first and diabatic at the second crossing (gray). It should be pointed out that the other three SCRAP conditions (23)–(25), not shown in Fig. 3, may further reduce the gray region.

B. Numerical examples

In Fig. 4 we show the time evolution of the Stark shift and the dressed-state eigenenergies for four different values of pulse delay $\tau$ and two values ($\alpha$ and $\beta$) for the pump detuning $\Delta_0$. Figure 4(a) shows the pump pulses. The time dependence of the Stark shift is plotted in the Fig. 4(b). The
vertical lines indicate the maxima of the pump pulse in the four cases (1), (2), (3), and (4). The two horizontal lines indicate the values \( \alpha \) and \( \beta \) of the static detuning \( \Delta_0 \) for which the first crossing with \( S(t) \) (the first resonance) occurs at the maximum of the pump pulses 2 and 3. The four lower frames (c) show the evolution of the diabatic (dashed curves) and adiabatic energies \( E^\pm(t) \) (solid curves) for the different pulse delays and the corresponding set of detunings \( \alpha \) or \( \beta \).

In Fig. 5 we show the population of the target state as a function of the static pump detuning \( \Delta_0 \). The three upper frames refer to the three values of the pulse delay \( \tau \) shown in Fig. 4: (1) large, (3) intermediate, and (2): small. Zero delay is not shown because coherent population return occurs. In each frame, there are three curves which correspond to three different values of pump laser Rabi frequency: small (solid line), medium (dotted line), and large (dashed line). The behavior of the transfer efficiency can be understood on the basis of the analytic estimates derived in Sec. III A.

Figure 4 shows for zero (case 1) or small (case 2) delay, that the pump Rabi frequency is large not only at the first crossing (where it is maximum) but also remains fairly large at the second crossing. Consequently, the condition (26) for diabatic evolution near the second crossing is not valid and the transfer efficiency is very small. Delay (3) is optimal in the sense that the coupling is large around the first resonance but very small around the second. Diabatic evolution near the second crossing is also assured for delay (4) but the delay is too large (for the given detuning) and the condition for adiabatic evolution at the first crossing is only marginally fulfilled.

The upper frame of Fig. 5 [relevant for large delay, see (4) of Fig. 4] shows large transfer efficiency for small detun-
ing. The peak Rabi frequency is obtained early when the Stark shift is still small. With increasing static detuning, the first resonance is met for decreasing pump laser intensity and adiabatic evolution near the resonance is less likely to be achieved. However, with increasing pump laser Rabi frequency conditions for adiabatic evolution are more likely to be established up to larger static detuning $\Delta_0$. Therefore, the transfer efficiency $T_E$ develops, for increasing Rabi frequencies, a plateau as a function of $\Delta_0$.

The second frame of Fig. 5 refers to the delay labeled (3) in Fig. 4. In this case, the transfer efficiency $P_2$ is small for very small static detuning $\Delta_0$ since the pump laser Rabi frequency $\Omega_p$ has not yet reached its maximum when the first resonance occurs. However adiabatic evolution is guaranteed for intermediate values of $\Delta_0$ and intermediate or large intensities ($\Omega_p$). The insensitivity of $P_2$ to the variation of $\Omega_p$ or $\Delta_0$ is a signature of the adiabatic nature of the transfer process. However, if either the detuning or the Rabi frequency $\Omega_p$ are too large then diabatic evolution at the second crossing is no longer assured. As a consequence, a decrease of the $P_2$ with increasing $\Omega_0$ or even an oscillatory behavior with the variation of $\Delta_0$ is found for large detuning.

The third frame of Fig. 5 refers to the small delay, labeled (2) in Fig. 4, where $\Omega_p$ reaches its maximum for detuning $\Delta_0=\beta$. For small detuning we have $P_2\sim 0$ since $\Omega_p$ is too small at the time when the first resonance occurs. For larger detunings (but still $\Delta_0<\beta$) the transfer efficiency increases with increasing $\Omega_p$ since adiabatic evolution near the first resonance is better fulfilled for larger $\Omega_p$ while diabatic evolution is still valid around the second crossing. However, for very large detunings ($\Delta_0>\alpha$), the evolution around the second resonance becomes more adiabatic with increasing $\Omega_p$, and therefore $P_2$ decreases with increasing $\Omega_p$ and $\Delta_0$. When the evolution is neither fully adiabatic nor clearly diabatic at the two crossings, the system will evolve along both paths (the one exclusively relevant in case of diabatic evolution and the one exclusively relevant in case of adiabatic evolution). In this case pronounced oscillations of $P_2$ as a function of $\Delta_0$ will be observed, as seen in this figure for small $\Omega_p$ and large $\Delta_0$.

IV. EXPERIMENTAL DEMONSTRATION

A. Coupling scheme

We chose metastable helium for a demonstration of the feasibility of the SCRAP process. The needed atomic parameters (transition moments, ionization rates, and Stark shifts) can be calculated reliably using a model potential approach.\cite{46, 47}

Figure 6 shows the relevant energy levels of helium. The metastable triplet state $1s\ 2s\,^3S_1$ serves as initial state of the SCRAP process. The target state $1s\ 3s\,^3S_1$ is coupled to the initial state by a two-photon transition induced by the pump laser, operating at 855 nm. The pump laser frequency is detuned several gigahertz from the exact two-photon resonance. The target state decays with a lifetime of 36 ns,\cite{48} which is long compared to the laser pulse durations.

With the laser intensity expressed in watts per squared centimeter, the coupling between initial and target state is described by the two-photon Rabi frequency, given in $s^{-1}$, with $\Omega_0=152.0\ I_P(t)$. The ac Stark shift, which is necessary for the SCRAP process, is induced by a laser pulse at 1064 nm. The infrared radiation is far off resonance from any transition frequency involving the initial or the target state. The Stark shift of the target state with respect to the initial state (i.e., the variation of the transition frequency) induced by the laser pulses is

$$S(t)=153.6\ I_S(t)+23.0\ I_P(t),$$

with the laser intensities expressed in watts per squared centimeter and the Stark shifts in $s^{-1}$. Since we always have $I_S>I_P$, $S(t)$ is clearly dominated by $I_S$.

The population in the target state is probed by photoionization with a laser pulse at 532 nm.

B. Experimental procedure

Figure 7 shows the experimental setup. A supersonic beam of helium is expanded through a pulsed nozzle (General Valve; opening diameter 0.8 mm) at a repetition rate of typically 20 Hz. The stagnation pressure is 1200 mbar. The helium atoms are excited to the metastable states by electron impact in an injection-seeded pulsed gas discharge,\cite{49} being operated between the nozzle orifice and an anode placed 4 mm downstream from the nozzle. A skimmer (diameter 0.8 mm) at a distance of 40 mm from the nozzle collimates the atomic beam and separates the source chamber from the region of laser interaction and detection. Helium ions are detected mass selectively by means of a short time-of-flight segment terminated by a double-thickness microsphere plate (EL Mul Technologies). The output current of the micro-
The pump–pulse laser is derived from a single-mode cw titanium:sapphire laser, operating at 855 nm. The output of that laser is amplified in a pulsed dye amplifier (PDA) pumped by the second harmonic of a single mode pulsed Nd:YAG laser (neodymium dotted yttrium aluminum garnet laser, GCR 4, Quanta Ray). The width of the pump laser pulse at \( \lambda_p = 855 \text{ nm} \) is \( T_p = 3 \text{ ns} \) (half-width at \( 1/e \) of maximum intensity). The wavelength of the titanium:sapphire laser is measured to an accuracy of \( \delta \lambda/\lambda = 2 \times 10^{-6} \) with a Michelson-type wavemeter, using a He–Ne laser, stabilized to an iodine line, as a reference. Pulse energies up to 5.5 mJ are achieved. The laser beam is focused to a diameter of 0.25 mm into the atomic beam, yielding intensities up to 2 GW/cm\(^2\).

The Stark-laser pulse at 1064 nm is provided by the fundamental frequency of the Nd:YAG laser. The pulse width of the infrared radiation is \( T_S = 4.8 \text{ ns} \), with pulse energy up to 500 mJ. The diameter of the Stark laser in the interaction region with the atomic beam is 4 mm, yielding intensities of up to several ten gigawatts per squared centimeter.

The remaining radiation at 532 nm provide the probe laser pulse with a pulse duration of 3.7 ns. The probe laser beam (pulse energy 30 mJ) is focused to a diameter of 0.5 mm, yielding intensities up to 2 GW/cm\(^2\).

C. Experimental results

The data shown in Figs. 8 and 9 demonstrate the validity of the SCRAP concept. The data clearly prove the possibility of complete population transfer from the initial to the target state if detuning, intensities, and pulse delay are appropriately chosen.
between the two pulses. The upper (a) and lower (b) frames for each time delay show the variation of the transfer efficiency for moderate and rather large peak intensities of the Stark-shift laser, respectively. The time delay as well as the Stark shift is shown for each experimental curve.

The peak intensity of the Stark-shift laser was varied by spatial displacement of the Stark-shift laser beam with respect to the pump laser beam. It was difficult to measure reliably the intensity directly within the relevant region because the intensity profile was not smooth. Therefore, we estimated the value of $I_{S}^{(0)}$ by comparison of the experimental results with simulated data (solid lines) for different peak intensities $I_{S}^{(0)}$ and $I_{P}^{(0)}$. The resulting estimates of $I_{S}^{(0)}$ and $I_{P}^{(0)}$ are approximate average values in the relevant region.

The probe laser is delayed by 12 ns with respect to the pump laser in order to avoid any interference between the probe and the SCRAP process. This leads to unavoidable losses ($\approx 28\%$) of population in the target state due to radiative decay. The total loss depends on the time delay between the transfer process and the moment of detection. It is straightforward to correct the experimental data accordingly, i.e., to eliminate the effect of spontaneous decay. Therefore, the numerical simulation of the data (solid lines) does not include spontaneous decay. The absolute transfer efficiency was calibrated by comparison of the population in the excited state with the pump laser tuned to the two-photon resonance and the Stark laser switched on and off. Assuming saturation of the pump transition, 50% of the initial state population is transferred to the target state without Stark shift laser. We confirmed that the pump laser intensities used were sufficiently high to saturate the two-photon transition.

Transfer of population from the $1s2s3S$ to the $1s3s3S_1$ state can, in principle, be observed for pump laser detuning in the range $0 < \Delta_0 < S_0$, where $S_0$ is the maximum Stark shift. The detuning range which allows high transfer efficiency varies, for given Stark laser peak intensity, with the time delay between the pulses and the pump-laser Rabi frequency.

Figures 8(a) and 9(a) show the variation of the transfer efficiency with pump-laser detuning for small peak intensity $I_{S}^{(0)}$ of the Stark-shift laser. Therefore, the maximum Stark shift $S_0$ is small compared to the tuning range of the pump laser shown. Transfer is not possible for $\Delta_0 > S_0$ [see Eq. (16)]. For large time delay between the two pulses, efficient transfer is only achieved for small detuning.

For larger static detuning $\Delta_0$ the Stark-shift-induced resonance occurs only after the pump laser intensity has decreased to a level too small for efficient transfer. The acceptable detuning range increases with decreasing delay $\tau$, see Fig. 9(a). Both Figs. 8(a) and 9(a) show a maximum transfer efficiency of nearly unity when the pump-laser Rabi frequency reaches its maximum at the time of the first resonance ($\Delta_0 = S$). For the relevant figure of merit $n = (\Omega_0 T_S)^2/\Delta_0 \tau$ we find for the optimal detuning $\Delta_0 = S_0 \exp(-\tau/I_3)$ $n = 73$ for the case shown in Fig. 8(a) and $M = 8$ for Fig. 9(a), i.e., in both cases we have $n > 3$, which is sufficient to guarantee adiabatic evolution [see Eq. (22)] near the resonance.

The variation of the transfer efficiency $P_2$ with detuning $\Delta_0$ for much larger maximum Stark shift $S_0$ is shown in Figs. 8(b) and 9(b) for 10.5 and 7 ns delay between the two laser pulses. As observed before, maximum $P_2$ is obtained for small detuning $\Delta_0$ when $\tau$ is large. Here, we find $n = 0.5$ and therefore we expect $T_2 < 1$. A larger pump-laser Rabi frequency $\Omega_{P}^{\text{max}}$ would be required to reach $P_2 = 1$. When the static detuning is increased beyond the value $\Delta_0^{\text{opt}}$ for which maximum $T_2^{\text{max}}$ is achieved, the pump-laser Rabi frequency at resonance is $\Omega_{P} < \Omega_{P}^{\text{max}}$ and $P_2 < P_2^{\text{max}}$ results. Furthermore, for $\Delta_0 > \Delta_0^{\text{opt}}$ the transition frequency is swept through the resonance more rapidly, leading to an even less adiabatic process and a smaller transfer efficiency. The two resonances, one met as the Stark shift $|S|$ increases and the other when it decreases, are separated by more than the pump-laser pulse width. Therefore the transfer efficiency would increase with increasing $\Omega_{P}$ for $\Delta_0 > \Delta_0^{\text{opt}}$ since the evolution would be more adiabatic at the first crossing while remaining diabatic at the second one.

In summary, the experimental data shown in Figs. 8 and 9 nicely confirm the prediction of the SCRAP theory presented in Secs. II and III.

V. DISCUSSION

A. Double SCRAP

An interesting and possibly important extension of what has been discussed in the preceding sections is the application of two sequential SCRAP processes, a first one for the excitation to an electronically excited state and a second one, for instance, for the controlled deexcitation to a target level in the electronic ground state, possibly a highly vibrationally excited level. Figure 10 shows a coupling scheme, the timing of the laser pulses, and the evolution of the dressed state eigenvalues. Here we consider the case when the excitation takes place by a two-photon process and the deexcitation is induced by a one-photon process. A simple description of the states of particle and field starts from atom state 1 in the presence of $n_P$ pump-field photons and $n_S$ Stokes-field photons, as denoted $|1,n_P,n_S\rangle$. Two-photon excitation leads to the state $|2,n_P-2,n_S\rangle$, and subsequent Stokes-field deexcitation leads to the state $|3,n_P-2,n_S+1\rangle$.

As pointed out earlier, strong fixed frequency radiation suitable for Stark shifting the levels is often available and the pulse width is longer than for the visible or ultraviolet radiation generated by frequency conversion from the former. Therefore, if the frequencies of both the pump laser and the Stokes laser are suitably tuned away from their respective resonances, and if the pulses are appropriately timed, then the pump and the Stokes lasers only address one of the two resonances between the dressed states $|1,n_P,n_S\rangle$ and $|3,n_P-2,n_S+1\rangle$. When the pump-laser Rabi frequency reaches its maximum at about the same time as the first resonance of the adiabatic eigenvalues, which correlate asymptotically to states 1 and 2, occurs, then the coupling by this laser is negligibly small near the second resonance between these levels. When the Stokes laser Rabi-frequency reaches its maximum at about the time when the second resonance (of the dressed state eigenvalues which correlate asymptotically to states 2 and 3) is reached,
then the Stokes laser has a negligible affect at the first resonance of these two levels. Therefore, all the population in state 1 is first transferred to the intermediate state, from where it is completely transferred into the final state. Obviously, only one Stark-shifting laser pulse is needed to complete the pump and dump process in this \( \text{SCRAP} \) scheme.

When the Stokes transition is also induced by a two-photon process, and when the Stark shift of the pump and Stokes radiation fields is sufficiently large and has the same sign, then an extra Stark-shift laser is not needed for the transfer process. The reason for that is obvious from Fig. 11 which shows the coupling scheme for the \( \text{SCRAP} \) process, the timing of the laser pulses, and the diagram of dressed-state eigenvalues. In this case, the Stark shift related to the pump laser itself drives the dressed states eigenvalues, which asymptotically relate to the levels 1 and 2, into resonance. Without the Stokes laser, there would be another crossing of the same two dressed-state eigenvalues when the pump laser intensity decreases and the transient population in state 2 would return adiabatically to state 1. However, the Stark shift induced by the Stokes laser prevents the second crossing of these dressed states to occur before the pump-laser Rabi frequency is sufficiently small. Similarly, the pump laser-induced Stark shift drives the eigenvalues of those dressed states which correlate asymptotically to the states 2 and 3 through the resonance before the Stokes laser is sufficiently strong to be effective. It is obvious from Fig. 11 that the pump-laser Rabi frequency cannot be a maximum when the relevant resonance is reached, because the Stark shift is caused by the pump laser itself. Of course, the delay of the two pulses must be, within certain limits, appropriately chosen. For instance, larger delay than that shown in Fig. 11, or larger static detuning, will lead to the process of coherent population return from state 2 back to state 1 before the Stokes laser can carry the population to state 3.

The \( \text{SCRAP} \) process can also be implemented when both the initial and final states are coupled through a one-photon process to the intermediate level by the pump and Stokes laser, respectively. However, when the conditions for adiabatic evolution in such a \( \text{SCRAP} \) process are met, the conditions for successful application of \( \text{STIRAP} \) are also fulfilled. In that case, \( \text{STIRAP} \) often is the better alternative and should be used since, unlike for \( \text{SCRAP} \), the intermediate state is never populated.

**B. Comparison of double \( \text{SCRAP} \), \( \text{STIRAP} \), and \( \text{SEP} \)**

It is interesting to compare briefly the common features and the differences of population transfer by Stark-chirp rapid adiabatic passage (\( \text{SCRAP} \)) implemented in the form of double \( \text{SCRAP} \), stimulated Raman adiabatic passage (\( \text{STIRAP} \)), and stimulated emission pumping (\( \text{SEP} \)). (For a detailed comparison of \( \text{STIRAP} \) and \( \text{SEP} \), see Ref. 50).

1. **Coherence**: \( \text{SEP} \) does not require coherent radiation; it is modeled by rate equations rather than by the time-dependent Schrödinger equation. In fact, a high degree of coherence will lead to Rabi oscillations between the pairs of levels, which would render the overall transfer process sensitive to the intensity (or pulse area) unless there is sufficient

![FIG. 10. Upper diagram: Coupling scheme for the (2+1)-SCRAP process. Lower frame: Intensities of pump \( I_p(t) \), Stark-shift \( I_s(t) \), and Stokes \( I_{\text{Stokes}}(t) \) pulses vs time. Upper frame: Adiabatic eigenvalues (solid lines) and diabatic energies (dotted lines) vs time. The arrow shows course of population flow.](image)

![FIG. 11. Upper diagram: Coupling scheme for the (2+2)-SCRAP process. Lower frame: Intensities of pump \( I_p(t) \) and Stokes \( I_{\text{Stokes}}(t) \) pulses vs time. Upper frame: Adiabatic eigenvalues (solid lines) and diabatic energies (dotted lines) vs time. The arrow shows course of population flow.](image)
pulse-to-pulse fluctuation in pulse energy. Coherence is essential for STIRAP and SCRAP. For those procedures the inverse of the spectral bandwidth of the radiation should be equal to or longer than the interaction time with the system. If that condition is fulfilled, phase fluctuations of the radiation fields during the transfer process are small and will not prevent adiabatic evolution.

b. Delay: In all three techniques there are delays between the various pulses. However, the pulse delay occurs for different reasons. For STIRAP the delay is needed to prepare the system in the trapped state. This requires the system to be in the intermediate level for different reasons. For STIRAP the delay is needed to prevent adiabatic evolution.

c. Two-photon detuning: For best performance of STIRAP the frequency difference of the two laser fields must be tuned exactly to the energy difference of the initial and final states, i.e., the two-photon resonance must be maintained (although some detuning of the individual radiation fields from the one-photon resonance with the intermediate level is tolerable). Neither SEP nor SCRAP require exact two-photon resonance. Unlike for STIRAP (where detuning must be smaller than the two-photon linewidth), frequency tuning within the saturation broadened one-photon linewidth is tolerable for SEP. For SCRAP the maximum Stark shift limits the tolerable detuning.

d. Sensitivity to Doppler shift: The various techniques are applicable not only in experiments with highly collimated particle beams but also in bulk samples, if the efficiency of the processes is not very sensitive to Doppler shift. Because exact two-photon resonance is required for STIRAP, this process is to some extent sensitive to a Doppler shift. This is particularly true when the pump laser and Stokes laser propagate in opposite direction. However, when the two lasers copropagate, the sensitivity to the Doppler shift is reduced. The process becomes insensitive to Doppler shift when the ratio of the two frequencies approaches unity. Both SEP and SCRAP are not very sensitive to Doppler shift. This is true for SEP when saturation broadening is substantial. This is true for SCRAP when the Stark shift induced by the shift laser is large compared to the Doppler width. Particles at different velocities are driven through the resonances at different times.

e. Intermediate-state population: When two-photon resonance is maintained throughout the transfer process induced by STIRAP, there is no transient population in the intermediate level. This leads to a key advantage of the STIRAP method over other schemes because spontaneous emission from that level is prevented. All the population is driven through the intermediate level when SEP or SCRAP is used. Therefore, the transfer process must be completed as quickly as possible, in any case faster than the radiative lifetime of the intermediate level. Since partial overlap of the laser pulses is tolerable for SCRAP but could be problematic for SEP, the often detrimental consequences of spontaneous emission are somewhat reduced for SCRAP.

f. Robustness: The adiabatic passage nature of STIRAP and SCRAP renders these processes, if implemented correctly, insensitive to small variations of the intensity and pulse delay. While SCRAP is also not sensitive to small variations of the laser frequencies, STIRAP suffers from such variations unless they occur synchronously for both lasers and maintain the two-photon resonance. Since SEP is often implemented with the individual transitions driven in the limit of saturation, this process is also not sensitive to small variations of intensity, frequency and pulse delay. The above implies that, unlike processes which are sensitive to the pulse area such as the $\pi$-pulse method, population transfer is efficient over a large area across the laser beams (i.e., not limited to region very near the axis of the laser beams). Furthermore, the population of all members of a group of degenerate levels (for instance, $m$ states) can be transferred, provided the conditions for high transfer efficiency is satisfied for those states with the weakest radiative coupling.

g. Efficiency and selectivity: The most essential properties of any population transfer scheme are its efficiency and selectivity. The transfer efficiency of SEP, implemented with delayed pulses, is typically limited to 25%. A fraction (of the order of 25%) remains in the intermediate state at the end of the dump pulse and decays by spontaneous emission, often also to other levels than the target one. In contrast, the transfer efficiency obtainable with SCRAP and STIRAP approaches unity, which also implies a very high degree of selectivity.

VI. SUMMARY

We have developed a concept for efficient population transfer based on Stark-shift induced rapid adiabatic passage processes which are applicable for lasers with a pulse width in the range of nanoseconds. SCRAP is yet another example for the use of far off-resonant radiations for the manipulation of internal and external degrees of freedom of atoms and molecules. To some extent, this is “an old trick.” However, it is presented and applied in a new form. Although the main body of this manuscript deals with the discussion (and experimental demonstration) of the transfer process between two levels, we anticipate important applications of this scheme in the form of “double SCRAP,” i.e., two sequential adiabatic passage processes are applied to efficiently move population first to an intermediate electronically excited state and then to a target level, which could be a level in another electronic state or in the electronic ground state. With regard to the “pump-dump” configuration, discussed in Sec. V, the SCRAP process can be viewed as a coherent variant of the SEP technique, retaining the flexibility of the latter but substantially increasing its selectivity and efficiency. Because of the widespread use of SEP in spectroscopy and collision dynamics, we anticipate interesting and important applications of SCRAP in these areas as well.
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