Two-photon excitation of the metastable 2s state of hydrogen assisted by laser-induced chirped Stark shifts and continuum structure

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Traditional two-photon excitation suffers from two difficulties that prevent complete population transfer: dynamic Stark shifts and multiphoton ionization. We describe a technique for overcoming these limitations by using a second field to control dynamic Stark shifts via Stark-chirped rapid adiabatic passage (SCRAP) and simultaneously suppress undesired photoionization via laser-induced continuum structure (LICS). We illustrate this proposed procedure by numerical simulations of two-photon excitation of the metastable 2s state of hydrogen, showing that efficient excitation can be expected when the two laser pulses are properly crafted.

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I. INTRODUCTION
The long lifetime (0.12 s) and high internal excitation energy (10 eV) of the metastable 2s state of hydrogen has long attracted interest of experimenters, while the simple internal structure continues to hold interest of theoreticians. This metastable state is of interest for use in precision spectroscopy of transitions starting from the 2s state and for improved accuracy in determining fundamental constants, e.g., the Lamb shift and the Rydberg constant [1,2]. Precision measurements of hyperfine splitting in 2s are desirable for the information they give on a combination of proton structure and QED [3].

The metastable state 2s is nearly degenerate with the 2p states, and so the excitation is readily quenched, with production of Lyman-alpha radiation, either by collisions or by weak electric fields that mix the two states [4]. It may therefore be useful as a source of Lyman-alpha radiation for high-resolution spectroscopy [5]. The high internal energy of excited hydrogen atoms makes these attractive for detecting single atoms by electron multipliers [1].

The theoretical simplicity of atomic hydrogen makes this a desirable species for studying basic collision phenomena, and so these states have been the subject of atomic beam studies of collision phenomena—e.g., collision-induced ionization, reactive scattering, and molecular dissociation processes. Theoretical understanding and modeling of such experiments using hydrogen are much simpler than for multi electron atoms, for the quantum-mechanical properties of the hydrogen atom can be analytically treated. Nevertheless, accurate description of the collision cross sections between H(2s) and H(1s) remains a challenge for theory [1]. A source of H(2s) would be of use for verifying theoretical predictions of cross sections for dissociative ionization and double excitation transfer in H(2s)−H(2s) collisions [6] and to verify calculations of long-range H(2s)−H(2s) interaction potential [5]. The density dependence of the frequency of the two-photon 1s−2s transition is an important diagnostic in Bose-Einstein condensation [7].

Thus there has long been a need for techniques that prepare sources of metastable hydrogen. The most common technique has been to pass a beam of hydrogen molecules through an electric discharge, where electron collisions induce dissociation and excitation [2,3]. Charge exchange between protons and vapor has also been used [8]. More recently optical pumping techniques have been used, with laser excitation of the 1s−3p transition using Lyman-beta radiation followed by spontaneous radiative cascade from the 2s state [2]. A technique using Stark-chirped rapid adiabatic passage (SCRAP), suggested earlier [9–12], is an important element in the present proposal. We here suggest significant modification of that technique for efficient excitation to the 2s state from the ground 1s state of hydrogen. The procedure involves a sequence of overlapping pulses of two frequencies which together overcome some of the limitations of the previous proposal.

To place the present proposal into context Fig. 1 shows linkage patterns for three schemes of laser excitation: (a) straightforward resonant two-photon excitation (inevitably accompanied by photoionization) produced by the single pulse P; (b) the SCRAP procedure using three laser pulses P1, P2, and S proposed earlier [9]; and (c) the present proposal using two laser pulses, P and S.

We note that other multifrequency proposals for similar population transfer or inversion have been proposed. One proposal combined steady strong two-photon excitation with a second steady linkage to an autoionizing resonance in the photoionization continuum [13]. A recent proposal, aimed at producing population transfer in sodium dimers, uses three delayed pulses to produce rapid adiabatic passage while suppressing photodissociation through laser-induced continuum structure LICS [14]. The use of a two-pulse sequence, in the presence of a third “dressing” field, has been proposed as a means of producing coherent control of dissociation (or photoionization) products during adiabatic passage, i.e., suppression of one continuum while enhancing another [15].
II. DESCRIPTION OF THE SUGGESTED TECHNIQUE

The preparation of metastable states of atoms, in particular H(2s), by radiative excitation through a two-photon transition, though highly desirable as a practical tool, suffers from two well-known problems. First, the presence of a field of sufficient strength to produce appreciable population transfer will inevitably cause other two-photon processes. In particular, laser-induced Stark shifts are, like the desired two-photon transition, directly proportional to laser intensity. These time-dependent shifts make it difficult to establish and maintain the needed two-photon resonance conditions.

Second, because the metastable state is typically highly excited, the absorption of a third laser photon may readily ionize the target metastable state [see Fig. 1(a)] thereby diminishing the resulting population transfer.

Each of these individual handicaps has been separately overcome. The unavoidable dynamic Stark shifts that accompany any conventional two-photon process can be counteracted by use of a second field, nonresonant, that produces a carefully timed sweep of adiabatic energies. This procedure, demonstrated experimentally and described theoretically, has been termed Stark-chirped rapid adiabatic passage (SCRAP) [9–12]. The photoionization loss that will inevitably accompany any laser field of sufficiently short wavelength has been suppressed with the aid of laser-induced continuum structure (or “pseudoautoionizing state”), a technique that can, in principle create a narrow transparency window in the variation of the photoionization cross section versus frequency.

The following subsections summarize these two individual mechanisms, SCRAP and LICS. We propose to combine them, using only two pulsed fields, as described in subsequent sections.

A. The SCRAP technique

Our proposed implementation of the SCRAP method is based on the coordinated interaction of the atom with two suitably delayed pulsed laser fields, termed P and S. The S laser, typically an infrared laser, induces Stark shifts of the 1s and 2s levels, while the P-laser field drives a two-photon transition between the 1s and 2s states. The P laser is slightly detuned from the two photon resonance; the detuning is of the order of, but less than, the maximum Stark shift induced by the S laser.

The Stark-shifting S laser drives the 1s–2s transition frequency twice through resonance with the two photons of the P laser: first during the rise of the S pulse and second when the S-pulse intensity decreases. For optimum performance of the SCRAP technique the P-laser pulse should have a slightly shorter pulse duration than the S laser pulse and should be delayed so as to reach maximum intensity at a time when the two-photon resonance condition is met. The result of the combined action of the two pulses is a Landau-Zener-type adiabatic passage whereby the atom undergoes a transition from the 1s to the 2s state. Adiabatic passage occurs only at one of the resonances, since the P-laser intensity is too small (ideally zero) at the other resonance. Figure 2 shows the operation of this general idea applied to the 1s–2s transition.

In the previous implementation of the SCRAP technique [9], schematically indicated in Fig. 1(b), the excitation step used two concurrent pulses (P₁ and P₂) of different frequencies in order to reduce detrimental photoionization of the 2s state: a strong field of frequency too low (wavelength λP₁ = 399.5 nm) to produce photoionization loss, and a weak field whose frequency was sufficiently high (wavelength λP₂ = 174.8 nm) that photoionization was into the continuum well above threshold, where the photoionization cross section is small. With such an approach the desired two-photon transition is driven mainly by the strong low-frequency field, while photoionization occurs only by the weak high-frequency field. Simulations have predicted good population
B. Ionization suppression by LICS

The mechanism for suppression of photoionization at a preselected frequency is based on creating a resonance structure in the photoionization continuum such that there occurs, at the chosen frequency, destructive interference between various ionization pathways—an effect similar to that seen in autoionization and described by a Fano profile [16]. This laser-induced continuum structure (LICS) has been discussed for 30 years [17] and has been demonstrated in many experiments [18,19]. Numerous theoretical papers have presented the complete theory for the effect [20,21] and it has been extensively reviewed [22]. (For more recent references, see Ref. [14].) Under favorable conditions it can substantially suppress the photoionization that would otherwise be produced by the excitation laser [19].

The possibility of using LICS to enhance two-photon excitation of the 2s state in hydrogen has been considered by Dörr et al. [23] who simulated the use of two laser pulses of different frequencies to transfer population from the 1s state to the 2s state via the continuum. They predicted that, at most, 17% of the population could be found in the 2s state at the end of the pulse. In addition to the low efficiency of this proposed approach, it would require an inconvenient laser wavelength shorter than 90 nm.

C. Combining SCRAP with LICS: A three-level model

We here describe how to implement a combination of SCRAP and LICS using only two laser fields. We demonstrate, using numerical simulation, that this is a practical scheme for efficient preparation of the metastable 2s state of hydrogen, via single-frequency two-photon adiabatic transition from the 1s ground state.

Figure 1(c) shows an idealized level scheme of the hydrogen atom. A two-photon transition, involving a pulsed laser field $P$ of constant carrier frequency $\omega_P$ (and wavelength near 243 nm) links the ground state $g$ of energy $E_g$ (the 1s state for hydrogen) with a metastable excited state $e$ of energy $E_c$ (the 2s state for hydrogen) via a two-photon transition.

The $P$ field also photoionizes the bound state $b$, into a continuum at energy $E_e = E_b + \hbar \omega_p$, with the rate $\Gamma_{bp}(t)$. As required for SCRAP, we intentionally introduce an initial frequency mismatch, parametrized by the detuning $\Delta_P$ defined by the equation

$$\hbar \Delta_P = 2 \hbar \omega_P - (E_c - E_b).$$

(1)

We introduce a second pulsed laser field $S$ of carrier frequency $\omega_S$, to connect the continuum at energy $E_c$, as reached by a three-photon process of the $P$ field, to a particular bound state $b$, of energy $E_b$ (the 5s state for hydrogen). The needed wavelength is around 552 nm. The detuning from the resonance for this transition is determined through the equation

$$\hbar \Delta_{PS} = \hbar (\omega_S - \omega_P) - (E_c - E_b).$$

(2)

The $S$ field frequency is insufficient to photoionize state $e$ in a single step, but it can produce two-photon ionization, with the rate $\Gamma_{e2b}(t)$. The $P$ and $S$ fields can each photoionize state $b$, with rates $\Gamma_{bp}(t)$ and $\Gamma_{bs}(t)$, respectively.

Laser pulses with the needed wavelengths and intensities are readily obtained from commercial laser systems. Frequency doubling of pulsed ns lasers conveniently provides 1–2 mJ at a wavelength of 243 nm. Intensity of 2 GW/cm$^2$, as required for the $P$ field, is obtained by focusing this laser to a 100-μm-diameter spot. The $S$ laser, at wavelength 552 nm, requires no frequency conversion. The needed wavelengths and intensities are readily obtained from commercial laser systems.

D. The Hamiltonian

As has been shown in theoretical treatments of LICS [20] the continuum states can be adiabatically eliminated to produce a three-state system in which there is no reference to the continuum. The state vector then has the expansion

$$\Psi(t) = \exp[-i \zeta_c(t)]C_c(t)\psi_c + \exp[-i \zeta_b(t)]C_b(t)\psi_b + \exp[-i \zeta_p(t)]C_p(t)\psi_p,$$

(3)

where $\psi_{n}(n=g,e,b)$ are the bare quantum states in the absence of laser fields and the $\zeta_n(t)$ are time-dependent phases chosen to enable the rotating-wave approximation. The resulting Schrödinger equation reads, in matrix form, $(d/dt)C(t) = -i \mathcal{W}(t)C(t)$, where the effective Hamiltonian is (see the Appendix)

$$\mathcal{W}(t) = \frac{1}{2} \begin{pmatrix}
2S_p(t) + 2\Delta_p & \Omega_p(t) & 0 \\
\Omega_p(t) & 2S_e(t) - i\Gamma_e(t) - i\gamma_e & -(i + q)\Omega_{sp}(t) \\
0 & -(i + q)\Omega_{sp}(t) & 2S_b(t) + 2\Delta_{PS} - i\Gamma_b(t) - i\gamma_b
\end{pmatrix}. $$

(4)

The detunings $\Delta_P$ and $\Delta_{PS}$ are defined by Eqs. (1) and (2).

The other quantities appearing here have the following meaning.

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The quantity \( S_n(t) \) is the dynamic Stark shift of state \( n \), expressible as the sum of a contribution from each of the two fields,

\[
S_n(t) = S_{np}(t) + S_{ns}(t), \quad n = e, g, b.
\]  

Each contribution is proportional to the related time-dependent laser intensity and to a time-independent but frequency-dependent polarizability.

The photoionization rates from levels \( e \) and \( b \) are sums of contributions from the two fields,

\[
\Gamma_{e}(t) = \Gamma_{ep}(t) + \Gamma_{eS}(t),
\]

\[
\Gamma_{b}(t) = \Gamma_{bp}(t) + \Gamma_{bS}(t).
\]  

Here each single-photon rate is proportional to a laser intensity while the two-photon ionization rate \( \Gamma_{eS} \) is proportional to the square of the \( S \)-laser intensity.

The excited states \( e \) and \( b \) can, in principle, decay by spontaneous emission at rates \( \gamma_e \) and \( \gamma_b \) respectively. However, in our discussion state \( e \) is metastable (lifetime 0.12 s), and so its spontaneous emission decay is negligible. Furthermore, the spontaneous emission rate of state \( b \) (lifetime 352 ns) is much smaller than the photoionization rate, so it too can be neglected in the present modeling.

The strength of the coupling between states \( g \) and \( e \) is parametrized by the two-photon Rabi frequency \( \Omega_p(t) \). This is proportional to the intensity of the \( P \) laser.

The Hamiltonian of Eq. (4) models the coupling between states \( e \) and \( b \) through two contributions: by a two-step transition via resonant continuum states [the term \( i \Delta_sp(t) \)], and by Raman-type transitions through nonresonant intermediate discrete and continuum states [the term \( q \Omega_sp(t) \)]. The quantity \( \Omega_sp(t) = \sqrt{\Gamma_{ep}(t)\Gamma_{bS}(t)} \) is a term of the effective Rabi frequency and the quantity \( q \) is the Fano parameter; its value may be calculated from wave functions for bound and continuum states, see Eq. (A12) of the Appendix. The two-photon detuning \( \Delta_pS \) is relevant for the Raman coupling between states \( b \) and \( e \) resulting from absorption of a photon from the \( P \) field and emission of a photon into the \( S \) field.

We use the usual nonrelativistic Pauli approximation for the wave functions of the hydrogen atom so all needed atomic parameters (dipole moments, polarizabilities, and photoionization cross sections) can be calculated from analytic expressions with arbitrary numerical accuracy, as described in Ref. [24]. Table I presents the values needed for the numerical simulations reported here.

### III. NUMERICAL SIMULATION

Two criteria are useful for quantifying the success of any excitation scheme involving probability losses: One is the fraction of the initial population that is transferred to the target state \( e \) [i.e., the probability \( P_e(\infty) \)], often termed the excitation efficiency. The other is the fraction of the final population that is found in this state, i.e., the final population inversion after the pulses cease. The two criteria have different uses. For example, even though a substantial fraction of the atoms photoionize, it may still be possible to achieve complete population inversion of the atoms remaining un-ionized at the end of the pulse sequence. Our aim is to exhibit, using solutions of the time-dependent Schrödinger equation to construct probabilities \( P_n(t) = |C_n(t)|^2 \), conditions of laser intensity and frequency detuning that will maximize either the final population \( P_e(\infty) \) in state \( e \)—the population transfer—or the final population inversion between states \( e \) and \( g \). We quantify this inversion by the fraction

\[
\eta = \frac{P_e(\infty) - P_g(\infty)}{P_e(\infty) + P_g(\infty)}. \tag{8}
\]

For the simulations we take the \( P \) and \( S \) pulses to be Gaussian functions,

\[
I_P(t) = I_P \max \exp(-t^2/t_P^2),
\]

\[
I_S(t) = I_S \max \exp(- (t-t_S)^2/t_S^2) \tag{9}
\]

with durations \( t_P \) and \( t_S \), offset in time by the delay \( t_S \), and with peak intensities \( I_P \max \) and \( I_S \max \).

#### A. Optimal conditions: Theory

To ensure effective and robust population transfer to the state \( e \), the timings of the laser pulses and their static detunings have to be chosen to fulfill both the SCRAP criteria for complete population transfer [9–12] and the LICS criteria for efficient ionization suppression.

The SCRAP technique involves adiabatic evolution of the state vector. In turn, this requires that the temporal area \( A_P \) of the \( P \) pulse must be sufficiently large,

\[
A_P = \int_{-\infty}^{\infty} dt \Omega_p(t) \gg \pi. \tag{10}
\]

In order that LICS produce effective ionization suppression the \( S \) field must be large and almost constant when the \( P \) pulse acts significantly, during the time interval \([−\tau_P, τ_P]\). This means that the \( P \) pulse must be substantially shorter than the \( S \) pulse, \( \tau_P \ll τ_S \). Adiabatic passage requires a monotonic change of the Stark shifts (a chirp) during the interval \([−\tau_P, τ_P]\). The two-photon resonance condition must hold at some moment during this interval. Therefore the time delay \( t_S \) has to be larger than \( τ_P \). Although the SCRAP mechanism can succeed with either pulse delayed, in our implementation

**TABLE I. Theoretical values used in the numerical simulations reported here. All values (other than \( q \)) are in ns\(^{-1}\) for intensities \( I_P \) and \( I_S \) in GW/cm\(^2\).**

<table>
<thead>
<tr>
<th>( \Gamma_{eP} )</th>
<th>( S_{gP} )</th>
<th>( −1.7I_P )</th>
<th>( \Gamma_{eS} )</th>
<th>( S_{gS} )</th>
<th>( −1.4I_S )</th>
<th>( \Gamma_{bP} )</th>
<th>( S_{eP} )</th>
<th>( 8.8I_P )</th>
<th>( \Gamma_{bS} )</th>
<th>( S_{eS} )</th>
<th>( 39.5I_S )</th>
<th>( \Gamma_{eS} )</th>
<th>( 0.000 , 47I_S^2 )</th>
<th>( S_{bP} )</th>
<th>( 8.45I_P )</th>
<th>( \Omega_P )</th>
<th>( 4.63I_P )</th>
<th>( 44.1I_S )</th>
<th>( q )</th>
<th>( −0.083 )</th>
</tr>
</thead>
</table>
the $P$ pulse must be delayed with respect to $S$ pulse to minimize the two-photon ionization loss. The detuning of the $P$-laser frequency from the two-photon resonance due to Stark shifts changes roughly linearly with time, at a rate approximately $\Delta_{\text{Stark}}/2\tau_p$ where

$$\Delta_{\text{Stark}} = S_e(\tau_p) - S_g(\tau_p) - [S_e(-\tau_p) - S_g(-\tau_p)].$$

(11)

For SCRAP to succeed this rate cannot be too large, meaning

$$\Omega_P(0)^2 \gg \frac{\Delta_{\text{Stark}}}{\tau_p}$$

(12)

and the amplitude of changes cannot be too small, meaning

$$\Omega_P(0) \ll \Delta_{\text{Stark}}.$$ 

(13)

When these conditions are not satisfied the SCRAP process fails to produce effective population transfer. We estimate the optimal value of the $P$-field detuning from two-photon resonance by requiring that exact two-photon ionization occurs at the maximum of the $P$ field, i.e., at $t=0$. This choice produces the condition

$$\Delta_{\text{P opt}} = S_e(0) - S_g(0).$$

(14)

Our simulation studies, discussed below, show that this simple expression satisfactorily describes the optimal condition for maximizing population transfer and inversion. The metastable state undergoes population loss due to photoionization by the $P$ field. However, this one-photon ionization can be suppressed, via the LICS mechanism, by proper choice of detuning, $\Delta_{\text{SP opt}}=\Delta_{\text{SP}}$. When all fields are constant the choice

$$\Delta_{\text{SP opt}} = S_e(0) - S_g(0) - \frac{1}{2} \Gamma_{\text{opt}}(0) q$$

(15)

completely suppresses single-photon ionization. Our simulation studies show that this expression provides a satisfactory condition for maximizing population transfer with pulsed excitation.

From Eqs. (10) and (12) one might conclude that larger intensities provide more effective population transfer. However, two-photon ionization is present for the entire duration of the pulse. Therefore large intensities produce large two-photon ionization losses, and hence smaller population transfer, although the results become less sensitive to small changes in pulse properties.

**B. Optimal conditions: Simulation**

To demonstrate the success of our proposed method we integrated the time-dependent Schrödinger equation to obtain the $2s$ population at the conclusion of pulses, $P_2(\infty)$, for a variety of conditions. This section presents some of those results.

Figure 3 shows a contour plot of $P_2(\infty)$, the final population in state $e$, as a function of the two peak intensities $I_{P_{\text{max}}}$ and $I_{S_{\text{max}}}$. For these and the following calculations the pulse durations $\tau_s$, $\tau_p$ and pulse delay $t_d$ are

$$\tau_s = 10 \text{ ns}, \quad \tau_p = 1 \text{ ns}, \quad t_d = -3 \text{ ns}.$$ 

(16)

For each choice of $I_{P_{\text{max}}}$ and $I_{S_{\text{max}}}$ the detunings are the optimal ones defined by Eqs. (14) and (15).

Figure 4 shows a contour plot of the inversion $\eta$ as a function of the two peak intensities $I_{P_{\text{max}}}$ and $I_{S_{\text{max}}}$. The pulse duration and pulse delay are the same as in Fig. 3; again the detunings were optimized for each choice of intensity. Two local maxima are evident in Fig. 4, labeled $A$ and $B$. These regions are also identified in Fig. 3, although there the region $B$ has no local maximum.

The local maxima of region $A$ in the two figures occurs for pulse areas of about $1.6 \pi$. Because such a value does not satisfy the criterion (10), the SCRAP mechanism does not apply in this region. The absolute maximum of the final population of level $e$ (see Fig. 3), approaches 86% for the particular choice $I_{P_{\text{max}}}=0.6 \text{ GW/cm}^2$ and $I_{S_{\text{max}}}=2.4 \text{ GW/cm}^2$ (region $A$). For these intensities the population inversion see Fig. 4) is $\eta=0.993$.

Region $B$ is associated with larger pulse areas than region $A$. In Fig. 4 the local maximum of region $B$ occurs for the $P$-pulse area of $5.3 \pi$, a value consistent with applicability of

![Figure 3](image.png)

**FIG. 3.** Contour plot of the final excited-state population, $P_2(\infty)$, for different peak intensities $I_{P_{\text{max}}}$ and $I_{S_{\text{max}}}$ of the $P$ and $S$ pulses. The pulse parameters are $t_d=-3 \text{ ns}$, $\tau_p=1 \text{ ns}$, and $t_d=10 \text{ ns}$. For each pair of intensities the detunings $\Delta_P$ and $\Delta_{SP}$ are chosen to be the optimal values defined by Eqs. (14) and (15).

![Figure 4](image.png)

**FIG. 4.** Contour plot of the population inversion $\eta$ for different intensities of $P$ and $S$ pulses. Pulse parameters are the same as in Fig. 3.
the variation of the population transfer

The intensities (chosen to maximize population transfer in region A of Fig. 3), are \( I_{P,\text{max}} = 0.6 \text{ GW/cm}^2 \), \( I_{S,\text{max}} = 2.4 \text{ GW/cm}^2 \). The other parameters are those of Fig. 3.

the SCRAP procedure. Thus the letters A and B in Figs. 3 and 4 denote regions in which conditions for SCRAP are violated and obeyed, respectively. The association of regions A and B with the failure or success of SCRAP are confirmed by an examination of the time evolution of the state vector viewed in an adiabatic basis, as shown below. The contours in region A, where SCRAP is not effective, are more closely spaced than those of region B, where SCRAP contributes to population transfer. This indicates that population transfer is less sensitive to the variations of intensities when the SCRAP mechanism acts, i.e., the SCRAP mechanism contributes to robustness of population transfer.

There is a large range of intensities in the SCRAP region B of Fig. 4 where the inversion exceeds 90%. In the corresponding B region of Fig. 3 the population transfer is greater than 60%. The inversion approaches 99.998% for \( I_{P,\text{max}} = 1.96 \text{ GW/cm}^2 \) and \( I_{S,\text{max}} = 9.1 \text{ GW/cm}^2 \). The final population of level \( e \) in this case is 64%.

The contour plots of Figs. 5 and 6 confirm the suitability of the optimal detuning, Eqs. (14) and (15). Figure 5 shows the variation of the population transfer (to state \( e \)) with detunings \( \Delta_P \) and \( \Delta_{SP} \), for intensities that maximize the population transfer, as expressed by Eq. (14). Figure 6 shows the variation of the inversion \( \eta \) with these detunings, for intensities that maximize the inversion, as expressed by Eq. (15).

Table II gives the values of the intensities that maximize inversion in Fig. 4. This table also gives the optimal detunings calculated from Eqs. (14) and (15) for these intensities. The numerical values for the detunings which maximize population transfer in Fig. 5 and inversion in Fig. 6 are very close to the calculated values. The difference is less than 1 ns\(^{-1}\).

### C. Doppler effect

Examination of Fig. 5, which corresponds to regions A of Figs. 3 and 4 where SCRAP fails, or of Fig. 6, which corresponds to regions B where SCRAP succeeds, shows that the range of detunings which allow good population transfer via SCRAP is substantially larger than the range of detunings which are effective without SCRAP. These observations also lead to an estimate of the influence of Doppler broadening on the efficiency of population transfer. For a hydrogen atom moving with the velocity \( v \) the two-photon detuning \( \Delta_P \) has to be replaced by the effective detuning \( \Delta_P = \Delta_P - (4\pi/\lambda_P)\nu \) and \( \Delta_{PS} \) by \( \Delta_{PS} = \Delta_{PS} + (2\pi/\lambda_P - 2\pi/\lambda_S)\nu \). Then from the contour plots on Figs. 5 and 6 we conclude that for the SCRAP region the velocity interval of effective population of the level \( e \) (larger than 50%) is about \( \Delta v \approx 400 \text{ m/s} \) (meaning a bandwidth of 1.7 GHz for the \( P \) laser) whereas for region A, where SCRAP fails, it is much smaller, \( \Delta v \approx 100 \text{ m/s} \) (meaning a \( P \)-field bandwidth of about 0.4 GHz).

### IV. UNDERLYING POPULATION DYNAMICS

#### A. Adiabatic states

It is instructive to examine in some detail the time dependence of the populations in various levels to understand the concerted operation of the two mechanisms, SCRAP and LICs, in the excitation process. For this purpose we introduce three adiabatic states \( \Phi_{\nu}(t) \) and the associated eigenvalues \( \epsilon_{\nu}(t) \), defined as instantaneous solutions to the time-dependent Schrödinger equation

\[
W(t)\Phi_{\nu}(t) = \epsilon_{\nu}(t)\Phi_{\nu}(t). \tag{17}
\]

These adiabatic states coincide with bare states in the absence of the laser fields, and so we label them using indices indicative of this connection before the arrival of the pulses. Using such states we write the statevector as the expansion

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**TABLE II. Parameters for maximizing inversion for region A and region B.**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Region A</th>
<th>Region B</th>
</tr>
</thead>
<tbody>
<tr>
<td>( I_{P,\text{max}} )</td>
<td>0.6 GW/cm(^2)</td>
<td>1.96 GW/cm(^2)</td>
</tr>
<tr>
<td>( I_{S,\text{max}} )</td>
<td>2.4 GW/cm(^2)</td>
<td>9.1 GW/cm(^2)</td>
</tr>
<tr>
<td>( \Delta_P = \Delta_{P,\text{opt}} )</td>
<td>96 ns(^{-1})</td>
<td>360.7 ns(^{-1})</td>
</tr>
<tr>
<td>( \Delta_{SP} = \Delta_{SP,\text{opt}} )</td>
<td>( -9.2 ) ns(^{-1})</td>
<td>( -35.2 ) ns(^{-1})</td>
</tr>
</tbody>
</table>
Because the Hamiltonian is not Hermitian, the eigenvalues

\( F \)

finding the system in the adiabatic state

\( F \)

probability to find an atom in

\( d \)

c if the state vector

\( d \)

and 4

example we consider the SCRAP regime
deduced from examining the adiabatic eigenvalues. As an

II.

maximum inversion. The pulse parameters are given in Table

II (region B).

\( \Psi(t) = B_g(t)\Phi_g(t) + B_e(t)\Phi_e(t) + B_b(t)\Phi_b(t). \) (18)

Because the Hamiltonian is not Hermitian, the eigenvalues are
complex valued. Furthermore, the eigenvectors are not

orthogonal and therefore \( |B_g(t)|^2 \) is not the probability of

finding the system in the adiabatic state \( \Phi_g(t) \) at time \( t \): even

if the state vector \( \Psi(t) \) coincides with \( \Phi_g(t) \), there is some

probability to find an atom in \( \Phi_g(t) \) or \( \Phi_b(t) \). This fact does

not affect the mathematics we use here.

B. SCRAP regime, region B

We first illustrate the dynamics during the interaction as
deduced from examining the adiabatic eigenvalues. As an

example we consider the SCRAP regime (region B of Figs. 3

and 4), choosing parameters of laser pulses that produce maximum inversion. The pulse parameters are given in Table

II.

Figure 7(a) shows the time dependence of the intensities

of the \( P \) pulse (solid line) and the \( a \) pulse (dashed line). These pulses induce time variation in the real and imaginary parts of the adiabatic eigenvalues \( e_n(t) \), as shown in Figs.

7(b) and 7(c), respectively. Two of the adiabatic states, \( \Phi_g(t) \) and \( \Phi_e(t) \), undergo slow probability loss (of at most

0.12 \( \text{ns}^{-1} \)), while \( \Phi_b(t) \) undergoes rapid probability loss (the

loss rate is about 35 \( \text{ns}^{-1} \)). This difference in loss rates is a

manifestation of the coherent population trapping effect that

underlies LICS: the coupling of two discrete states through

the continuum leads to the formation of “dark” and “bright”
eigenstates with very different ionization losses. In an ideal

case, when condition (15) is fulfilled for all times, the

probability of ionization of the dark state decreases to zero. The

existence of this dark state underlies the possibility for ion-

ization suppression by LICS and population transfer through

the continuum [18,20]. In our case one more discrete state is

involved in the interaction. As a result we have two eigen-

states with small ionization loss and one with large loss.

The adiabatic state \( \Phi_g(t) \), which undergoes relatively slow

probability loss, is connected with the ground state \( \psi_g \) for

early times and with the excited state \( \psi_e \) for late times [see

Fig. 7(f)]. Thus if the time evolution is adiabatic there are no

transitions between adiabatic states and an atom which was

initially in the ground state will be found in the excited state

after the interaction—the SCRAP process is active. The

population transfer is not complete because of the complex

nature of the eigenvalues of the effective Hamiltonian. In the

adiabatic limit the population \( |B_g(t)|^2 \) of the adiabatic state

\( \Phi_g(t) \) is given by

\[ B_g(t) = B^{(\text{adiab})}_g(t) = \exp \left[ -i \int_{-\infty}^{t} e_g(t')dt' \right] B_g(-\infty). \] (19)

The consequent maximum excitation probability is

\[ P_{e \text{ max}} = \exp \left[ -2 \int_{-\infty}^{\infty} \text{Im} e_g(t)dt \right]. \] (20)

The conditions for adiabatic evolution can be met more easily,

i.e., with lower laser power, when the pulses are long, but

the maximum transfer becomes smaller with an increase of

the time interval during which \( \text{Im} e_g(t) \) provides the domi-

nant contribution to the exponent of Eq. (20). Hence an in-

crease of the population transfer comes at the cost of de-

creasing robustness.

Figure 8(a) shows the time dependence of the adiabatic state

populations as measured by \( |B_g(t)|^2 \) and \( |B_e(t)|^2 \). The

values of \( |B_g(t)|^2 \) are all too small to be visible here (less than

10\(^{-4}\)). The values of \( |B_e(t)|^2 \) is very close to the ideal case of

adiabatic following [short dash curve, calculated using Eq.

(19)], whereas \( |B_e(t)|^2 \) is very small (less than \( 4 \times 10^{-2} \)). This

means that the system can be represented very well by the

adiabatic state \( \Phi_g(t) \).

The decrease of the population transfer efficiency for in-

creasing pulse amplitudes, when SCRAP becomes dominant,

results from an increase of the imaginary part of the corre-

sponding eigenvalue. There are two reasons for this increase.

First, two-photon ionization by the \( S \) field increases. Second,
loss due to imperfect LICS. Transfer, but this is accompanied by an increase of population.

crease of laser intensities leads to more robust population.

intensities are far from these optimal values. Thus an in-
tween the actual and the ideal adiabatic following when the
simulations show much more pronounced differences be-
have been chosen to maximize population transfer. Our
states primarily during the

One can see here that transitions occur among adiabatic
states during the

pulse but for a lossy system affected by dynamic Stark
fields with large intensity we increase the range of values of
possible only for static fields and detunings. Using pulsed
fields with high intensity we increase the range of values of
the Stark shifts, and thereby increase the ionization rate.

C. Regime where SCRAP fails, region A

Figure 8(b) illustrates the regime in which SCRAP fails.
This figure shows the time dependence of $|B_2(t)|^2$ and $|B_3(t)|^2$ for the intensities and detunings given in Table II (region A). One can see here that transitions occur among adiabatic states primarily during the $P$ pulse.

At the conclusion of the $P$-field interaction there is little
difference between the computed populations and the ideal
adiabatic following of $|B_2(t)|^2$, because the pulse intensities
have been chosen to maximize population transfer. Our
simulations show much more pronounced differences be-
tween the actual and the ideal adiabatic following when the
intensities are far from these optimal values. Thus an in-
crease of laser intensities leads to more robust population
transfer, but this is accompanied by an increase of population
loss due to imperfect LICS.

D. Robustness

For small intensities the time evolution is not adiabatic.
Nevertheless, it is possible to find some intensities which
maximize the population transfer (this is an analog of a $\pi$
pulse but for a lossy system affected by dynamic Stark

FIG. 9. The final population of the excited state, $P_e(\infty)$, as a
function of the intensity of the $S$ laser in units of the optimal $S$-laser
intensity for fixed $P$-laser intensity and fixed detunings as given in
Table II. The full line is region $A$, where SCRAP fails, dashed line is
region $B$, where SCRAP succeeds.

shifts). The difference in robustness for the regions where
SCRAP succeeds and where it fails is evident in Figs. 3 and
4. There we observe a narrow maximum for efficient popu-
lation transfer in region $A$ and a broader dependence in re-
region $B$. The difference in robustness is evident also in Figs. 5
and 6: the interval of detunings where appreciable population
transfer exists is substantially different for the two regions.

It is important to note that in the calculations shown in
Figs. 3 and 4 the detunings were adjusted to their optimal
value for each pulse. In any experiment the pulse intensities
fluctuate from pulse to pulse, so it is useful to see the influ-
ence of these fluctuations on the robustness of the population
transfer. For both regions $A$ and $B$ a change of intensities
with fixed static detunings will, because of the incremental
Stark shifts, take the atom away from the condition for two-
photon resonance. Therefore the intensity fluctuations
decrease the population transfer. Figure 9 shows the population
of the excited state as a function of the intensity of the $S$
laser, in units of the optimal $S$-laser intensity, for fixed
$P$-laser intensity and fixed detunings of both lasers, as given
in Table II. We see that the population transfer efficiency is
quite sensitive to fluctuations of the laser intensity. The de-
sired stability of the laser pulse energy is about $5\%$. Note that
the robustness with regard to intensity fluctuations is larger
for the SCRAP region than for the region where SCRAP
fails.

V. CONCLUSIONS

We have shown by numerical simulation studies that
population transfer from the ground state of hydrogen to the
metastable $2s$ state can be as high as $86\%$, and that popu-
lar inversion between the $2s$ and $1s$ can be as high as $99\%$,
although the two achievements do not occur for the same
pulse conditions. The successful population transfer results
from the concerted action of the mechanism of Stark-chirped
rapid adiabatic passage (SCRAP) for population transfer and
the suitable implementation of laser-induced continuum
structure (LICS) for the suppression of detrimental photoionization processes. Because SCRAP requires a suitably crafted Stark shift but the creation of pronounced LICS suffers from Stark shifts, the laser parameters (peak intensities, pulse delay and detuning) must be chosen carefully. If that is done, the optimal contributions of SCRAP and LICS to the transfer process can be realized with only two laser pulses and with intensities readily available from commercial laser systems.

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APPENDIX: THE LICS EFFECTIVE HAMILTONIAN

We consider an atom interacting with two laser fields: a P laser with amplitude \( e_p(t) \), intensity \( I_p(t) \), and carrier frequency \( \omega_p \), and an S laser with amplitude \( e_S(t) \), intensity \( I_S(t) \), and carrier frequency \( \omega_S \). The total electric field is

\[
E(t) = \text{Re}[e_S e_S^*(t) \exp(-i\omega_S t) + e_p e_p^*(t) \exp(-i\omega_p t)] ,
\]

(A1)

where \( e_j \) is a unit vector prescribing the polarization of field \( j \). We express the state vector \( |\Psi(t)\rangle \) of the atom as a combination of three particular bound states \( n = g, e, b \), with energies \( E_g, E_e, E_b \), together with a sum over all other bound states and an integral over continuum states, using the Dirac picture:

\[
|\Psi(t)\rangle = \sum_{n=g,e,b} C_n(t) \exp(-iE_nt/\hbar)|n\rangle \quad \text{(A2)}
\]

The theoretical ionization rates \( \Gamma_n(t) \) and Stark shifts \( S_n(t) \) appearing here are obtainable by summing partial rates and shifts:

\[
\Gamma_n(t) = \sum_{k,a} \Gamma_n^{(k)}(t), \quad S_n(t) = \sum_a S_{na}(t) \quad \text{(A8)}
\]

where \( \Gamma_n^{(k)}(t) \) is the ionization rate from state \( n \) to continuum \( k \) caused by laser \( \alpha \),

\[
\Gamma_n^{(k)}(t) = \frac{\pi}{2\hbar} |E_n(t)|^2 \sum_{\alpha,k} |\langle n|e_\alpha \cdot \mathbf{d}|E = \hbar\omega_\alpha - E_n,k\rangle|^2 ,
\]

(A9)

and \( \hbar S_{na}(t) \) is the dynamic Stark shift of the energy of state \( n \) produced by laser \( \alpha \):

\[
\hbar S_{na}(t) = -\frac{1}{4} |E_n(t)|^2 \mathbf{e}_\alpha \cdot [\mathbf{Q}_n(\omega_\alpha) + \mathbf{Q}_n(-\omega_\alpha)] \cdot \mathbf{e}_\alpha^* .
\]

(A10)

The quantity

\[
\mathbf{Q}_n(\omega) = \frac{\sum_k \langle m|\mathbf{d}|k\rangle \langle k|\mathbf{d}|n\rangle}{\sum_k \langle m|\mathbf{d}|k\rangle \langle k|\mathbf{d}|n\rangle}.
\]

(A5)

where \( \mathbf{d} \) is the dipole-moment operator. The continuum integral includes the resonance energy \( E = E_m + \hbar\omega \), whose singularity is dealt with by breaking the integration into a nonsingular principal value part (denoted by \( \mathcal{P} \)) and a resonant part (yielding the transition rate of a Fermi golden rule),

\[
\text{Re} \mathbf{Q}_{nn}(\omega) = \sum_k \langle m|\mathbf{d}|k\rangle \langle k|\mathbf{d}|n\rangle + \mathcal{P} \int dE \sum_k \langle m|\mathbf{d}|E_k\rangle \langle E_k|\mathbf{d}|n\rangle ,
\]

(A6)

The adiabatic elimination yields a three-state time-evolution equation, which may be written [22]
\[ \Omega_{SP}(t) = \sqrt{\gamma_{sp}^{(q)}(t)\Gamma_{Q}} \]  
\hspace{1cm} (A11)

is an effective Rabi frequency for the two-step transition from the state \( e \) to the state \( b \) with intermediate population of the continuum states \( k \) with energy \( E \approx \hbar\omega_k + E_e = \hbar\omega_S + E_p \). The Fano \( q \) parameter appearing here can be evaluated from the expression [21,22]

\[ q \Omega_{SP}(t) = \frac{1}{2} |\xi_p(t)\xi_S(t)| \mathbf{e}_p \cdot \left[ Q_{sb}(\omega_P) + Q_{sb}(-\omega_S) \right] \mathbf{e}_S, \]  
\hspace{1cm} (A12)

The two-photon detuning \( \Delta_{PS} \) is given by

\[ \hbar\Delta_{PS} = \hbar(\omega_P - \omega_S) - (E_p - E_e). \]  
\hspace{1cm} (A13)

Equation (A7) is usable for any two excited states; hence there appears a constant loss rate \( \gamma_n \) for each state, expressing the rate at which spontaneous emission removes population from state \( n \).

The effective Hamiltonian implied by Eq. (A7) requires dynamic Stark shifts \( S_n(t) \), photoionization rates \( \Gamma_n(t) \), spontaneous emission rates \( \gamma_n \), an effective Rabi frequency \( \Omega_{SP}(t) \), and the Fano parameter \( q \). In principle, all of these quantities can be calculated if the atomic wave functions are known, as they are for hydrogen.