PHYSICAL REVIEW A VOLUME 60, NUMBER 6 DECEMBER 1999

Source of metastable H(2*s*) atoms using the Stark chirped rapid-adiabatic-passage technique

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(Received 13 August 1999)

We propose the use of Stark chirped rapid-adiabatic-passage method, a technique in which the energy of a target state is swept through resonance by a slowly varying dynamic Stark shift to induce complete population transfer from the ground 1*s* state to the metastable 2*s* state of the hydrogen atom. Parasitic ionization processes are strongly reduced by using a two-color excitation scheme. Our detailed numerical calculations show that under judicious choice of pulse parameters, up to 98% of the population can be found in the 2*s* state at the end of the process. [S1050-2947(99)51112-8]

PACS number(s): 42.50.Md

Control of population transfer between specific quantum states has been a major goal in atomic and molecular physics during the last three decades. The recent availability of efficient schemes for selective population transfer, such as the stimulated Raman adiabatic passage (STIRAP) technique, have opened new directions in collision dynamics and spectroscopy [1]. One important aim of selective population transfer is to provide pure and dense sources of excited atoms. The preparation of hydrogen atoms in their metastable 2*s* state is of particular interest due to the long lifetime (provided the electric field is small) and high energy of this state. Furthermore, hydrogen lends itself to quantitatively accurate theoretical analysis. However, application of STIRAP to the transfer of population from the 1*s* to the 2*s* state of hydrogen requires coupling to a higher electronic state. The required wavelength to access this state must be less than 120 nm. In this range, radiation with suitable intensity and coherence is not currently available. A prerequisite for the construction of a pure H(2*s*) source is the development of more efficient population transfer schemes, as current optical population transfer methods are limited, by photoionization, to an efficiency of less than 20% [2].

Recently we have developed a population transfer scheme called ''Stark chirped rapid adiabatic passage'' (SCRAP) [3]. This technique builds on the well-known method of rapid adiabatic passage [4–8] in which a one- or two-photon transition between two discrete-energy states is adiabatically swept through resonance, thereby inducing a nearly complete population inversion. Our method utilizes one laser pulse (the ''pump''), tuned slightly away from resonance with the two-photon transition between the states, and a second pulse (the ''Stark'') that sweeps through the resonance by inducing a dynamic Stark shift. The result is similar to the population inversion reported by Loy [8], who used adiabatic quasistatic pulses of ~5-μs duration, to induce dc Stark shifts. However, he induced two sequential population inversions per pulse (an excitation for the leading edge and deexcitation for the trailing edge of each pulse), with the final result that no net population transfer took place. In SCRAP a time delay between the ''Stark'' pulse and the ''pump'' pulse ensures that only one transfer process occurs and that the entire population is in the excited state when the process is over.

Commercially available pulsed lasers enable the completion of the inversion process within a few nanoseconds with the SCRAP method. Shaping quasistatic pulses of this duration is a much more difficult task. It may also be argued that femtosecond chirping techniques could be used to sweep the resonance. However, the bandwidth of ns pulses is far too small to apply traditional short-pulse methods [9].

The SCRAP method was demonstrated successfully for the 2*s*-3*s* transition in metastable helium [3]. In this Rapid Communication, we present a theoretical study of the SCRAP scheme, as applied to produce metastable H(2*s*) atoms by coherent population inversion from ground-state ensembles. The excitation scheme is modified from our experiment in He* to a two-color pump pulse in order to avoid unwanted ionization from the 2*s* state. We study the inversion efficiency as a function of the relative intensities of the two pump pulses and their detuning from two-photon resonance, and show that by appropriate choice of parameters, nearly complete population inversion is obtained.

The SCRAP technique, as previously implemented [3], proceeds by a one-color, two-photon excitation from the ground state with a fixed small detuning $\Delta_0 \equiv \omega_a - 2\omega$, where ω is the pump laser frequency and $\omega_a \equiv (E_{2s} - E_{1s})/\hbar$ is the atomic resonance frequency. The transition is then swept through resonance by the Stark pulse, which induces a time-dependent dynamic Stark shift, thereby scanning the interlevel spacing (mainly by shifting the 2*s* level). The intensity of the Stark pulse should be sufficiently strong to modify the atomic frequency by more than Δ_0 in order to sweep through resonance. The situation is depicted by dashed lines in Fig. 1. If the Stark pulse is coincident with the pump, as with Loy [8], the resonance is swept through twice [see Fig.

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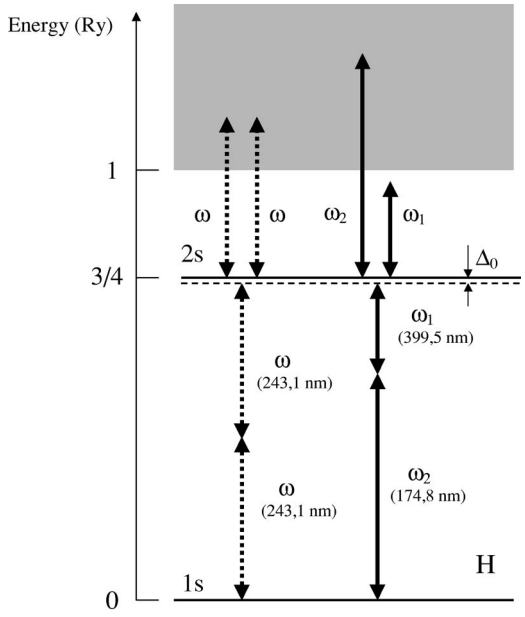

FIG. 1. Two-photon excitation schemes for SCRAP in hydrogen: dashed arrows, one-color; solid arrows, two-color. The detuning Δ_0 is grossly exaggerated.

2(a)]: first by the leading edge of the pulse and next by its trailing edge. As a result, the entire population remains in the $1s$ state when the pulses are over. To avoid this double population transfer, a time delay is introduced between the Stark and pump pulses [see Fig. 2(b)], rendering the second crossing ineffective.

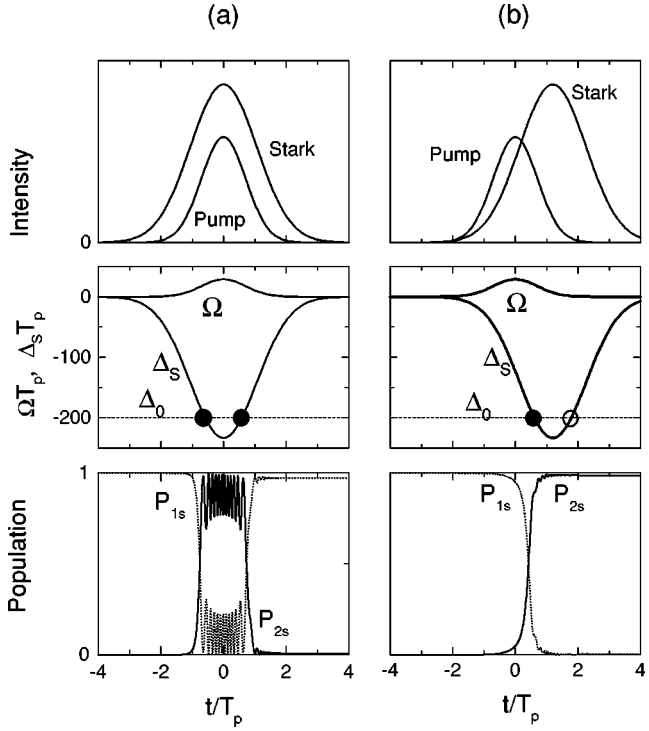

FIG. 2. Comparison of coincident-pulse scheme (a) and SCRAP scheme (b) for the hydrogen $1s$-$2s$ transition. Shown are the Stark and pump laser pulses (upper trace); the two-photon transition rate $\Omega(t)$, the Stark-free detuning Δ_0, and the Stark shift $\Delta_S(t)$ (middle trace); and the populations of the two levels (lower trace).

In brief, the two-level Hamiltonian for the system is

$$H(t)=\frac{\hbar}{2}\begin{pmatrix} 0 & \Omega(t) \\ \Omega(t) & 2\Delta(t) \end{pmatrix}. \tag{1}$$

In the above, $\Omega(t)$ is the two-photon transition rate and $\Delta(t)$ is the Stark-shifted detuning

$$\Delta(t)=\Delta_0-\Delta_S(t), \tag{2}$$

where $\Delta_S(t)$ is the relative Stark shift, i.e., the time-dependent difference between the Stark shifts of the two levels. The two-level Schrödinger equation with the Hamiltonian of Eq. (1) has two adiabatic solutions,

$$\Psi_-(t)=\cos\Theta(t)|1s\rangle-\sin\Theta(t)|2s\rangle, \tag{3a}$$

$$\Psi_+(t)=\sin\Theta(t)|1s\rangle+\cos\Theta(t)|2s\rangle, \tag{3b}$$

given in terms of a mixing angle $\Theta(t)$ defined, modulo 2π, as

$$\tan 2\Theta(t)=\Omega(t)/2\Delta(t). \tag{4}$$

The corresponding adiabatic eigenvalues are

$$E_\pm=\frac{\hbar}{2}[\Delta(t)\pm\sqrt{\Omega(t)^2+\Delta(t)^2}]. \tag{5}$$

On adiabatically sweeping $\Delta(t)$, via $\Delta_S(t)$, from a large negative value through the resonance to a large positive value, the adiabatic eigenstate $\Psi_-(t)$ evolves smoothly from the initially populated state $|1s\rangle$ to the target state $|2s\rangle$, with the final result that population is completely transferred to the target state. To maintain adiabaticity, the time derivative of the mixing angle $\Theta(t)$ must at all times be much smaller in magnitude than the spacing between the adiabatic energies of Eq. (5). This requirement leads to the adiabaticity condition,

$$|\dot{\Theta}(t)|\ll\sqrt{\Omega(t)^2+\Delta(t)^2}, \tag{6}$$

which must be maintained at all times. Equation (6) is easily satisfied when $\Delta(t)$ is large. However, on resonance, $\Delta(t)=0$, adiabatic evolution requires a sufficiently strong coupling $\Omega(t)$. We have previously shown [3] that for time delayed, Gaussian shaped pulses,

$$\Omega(t)=\Omega_0\exp\left(\frac{-t^2}{T_p^2}\right),\quad \Delta_s(t)=S_0\exp\left(\frac{-(t-\tau)^2}{T_s^2}\right), \tag{7}$$

the condition set by Eq. (6)) translates to the following requirements for the experimentally controllable laser parameters Ω_0, T_p, Δ_0, S_0, T_S, and τ,

$$\exp\left(\frac{-8\tau^2}{T_p^2}\right)\ll\frac{\Delta_0}{\Omega_0^2T_S}\sqrt{\ln\frac{S_0}{\Delta_0}}\ll 1. \tag{8}$$

The inequalities (8) imply that it is preferable to work with large pump intensities and small detuning. However, for large pump intensities, ionization rates are increased and population is lost. Therefore, it is essential to obtain optimal intensities for the pump pulses.

We have simulated the SCRAP process on the $1s$-$2s$ hydrogen transition. A potential difficulty in using SCRAP for

TABLE I. Theoretical values for simulation. Here $\hbar S_{ij}$ and Γ_{ij} are the dynamic Stark shift of the energy of state $i=1(1s)$ or $2(2s)$ and ionization rate of this state produced by laser j; Ω is the effective Rabi frequency for the two-photon transition from state $1s$ to state $2s$; I_1, I_2, and I_3 are the intensity of uv, vuv, and Stark IR lasers expressed in W/cm^2. The ionization rates, Stark shifts and Rabi frequency are expressed in s^{-1}.

Ω	$11.49\sqrt{I_1 I_2}$		
Γ_{11}	0	S_{11}	$-1.44I_1$
Γ_{12}	0	S_{12}	$-2.30I_2$
Γ_{13}	0	S_{13}	$-1.34I_3$
Γ_{21}	0	S_{21}	$-1.44I_1$
Γ_{22}	$2.56\times I_2$	S_{22}	$4.65I_2$
Γ_{23}	0	S_{23}	$-53.01I_3$

this transition is that there may be significant losses due to ionization of the H($2s$) atoms by the pump photons (whose energy is of the order 3/8 Ry, greater than the $2s$ ionization energy $E_{ion}^{(2s)}=1/4$ Ry). To overcome this difficulty, we propose a two-color excitation scheme, as depicted by solid arrows in Fig. 1. Two coincident excitation pulses are used; with carrier frequencies $\omega_1<E_{ion}^{(2s)}/\hbar$ and $\omega_2=\omega_a-\Delta_0-\omega_1$, and intensities I_1 and I_2. Because ω_1 is too small to induce a single-photon transition from $2s$ into the continuum, it is possible to minimize ionization by increasing I_1, while concomitantly decreasing I_2, thereby maintaining a fixed two-photon transition rate, which is proportional to $\sqrt{I_1 I_2}$. The wavelengths of the excitation lasers were taken to be 399.4 nm (25034.4 cm^{-1}, uv) and 174.75 nm (57223.8 cm^{-1}, vuv) for ω_1 and ω_2, respectively. This choice nullifies the dynamical Stark shift induced by the strong uv pulse, which would make it difficult, if not impossible, to guarantee adiabatic evolution. We have previously shown [3] that in order to maintain adiabaticity, this shift should be no greater than the two-photon transition rate Ω. Because $d\Delta_S/d\nu_1$ at $\nu_1=25\,034.4$ cm^{-1} is $0.0426I_1$ cm/s and Ω is $11.49\sqrt{I_1 I_2}$ s^{-1} (where I_1 and I_2 are in units of W/cm^2), the frequency of the uv laser should be kept within an interval of $\Omega/(d\Delta_S/d\nu_1)=269.7\sqrt{I_2/I_1}$ cm^{-1} around ν_1. Therefore, for the characteristic intensity ratios of $I_2/I_1\sim10^{-4}$ used in our calculations, the required uv frequency is $25\,034.4\pm2.7$ cm^{-1}. The wavelength of the Stark laser was 1064 nm.

The radiation needed for the uv laser pulse at 399.4 nm can be provided by frequency-doubling the output of a tunable laser (e.g., a titanium sapphire laser or an optical parametric oscillator). The vuv pulse at 174.75 nm can be generated by resonant frequency-difference mixing of two strong laser fields in a rare-gas or metal vapor [14].

The possibility of ionization by two ω_1 photons at high I_1 was also considered. Calculated two-photon ionization rates from the hydrogenic $2s$ level [10] at the relevant wavelengths ($\sim$400 nm) and intensities ($\sim10^9$ W/cm^2) are of the order of 10^5 s^{-1}, amounting to negligible losses of order 0.01% for a ns pulse. Two-photon transition rates, dynamic Stark shifts and ionization rates were calculated as described in Ref. [11] and are given in Table I. We have simulated the time evolution of the SCRAP process as a function of the pump and Stark pulse fluences and of the Stark-free detuning. The width of the Stark-shift laser pulse was taken to be about 1.5 times the width of the pump pulses. This is a realistic assumption, since often the latter radiation is obtained by frequency up-conversion from the infrared radiation, and the pulse width is shortened by the conversion process.

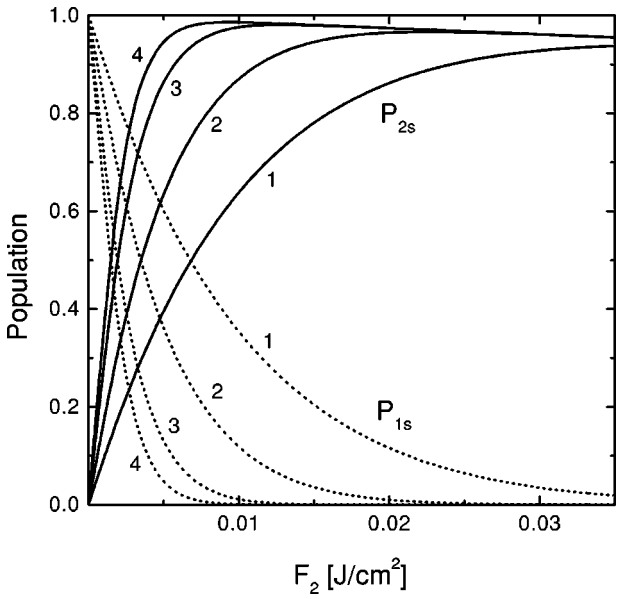

FIG. 3. Final populations in the $1s$ and the $2s$ states as a function of vuv fluence, at 25 J/cm^2 (1), 50 J/cm^2 (2), 100 J/cm^2 (3), and 150 J/cm^2 (4) uv fluence. The Stark pulse fluence is 1 J/cm^2 and its duration is $T_S=1.5T_p$. The time delay between the pulses is $\tau=1.2T_p$ and the Stark-free detuning is $\Delta_0=-15/T_p$.

The results of a time-delayed SCRAP scheme in which the pump pulses precede the Stark pulse, are plotted in Fig. 2(b). The resonance is swept through twice by the Stark pulse but, due to the time-delayed pulse sequence, the second crossing takes place when the pump pulse is essentially over and is therefore ineffective. As a result, population is being transferred adiabatically from the ground state to the excited state. Ionization is minimized by the two-color pumping scheme to less than 2%, resulting in a total transfer efficiency of 98%.

While ionization losses are generally low, they become more significant on increasing the pump pulse intensities. It is particularly important to maintain a low intensity of the vuv (I_2), laser which is capable of one-photon ionization from the $2s$ state. On the other hand, the adiabaticity condition of Eq. (8) implies that strong pump intensities are required to obtain efficient population transfer. These contradicting requirements suggest that there is an effective optimal pump intensity for which the inversion yield is maximized. In Fig. 3 we plot the final populations of the two states, as a function of the vuv fluence $F_2=\sqrt{\pi}I_2T_p$, at four different values of the uv fluence $F_1=\sqrt{\pi}I_1T_p$. As expected, the $2s$ population increases with increasing the vuv fluence until a maximum is reached. Any further increase in this fluence would only enhance the one-photon ionization rate and reduce efficiency. Weaker I_2 intensities are required in order to obtain the same efficiency for increasingly stronger I_1. This is simply due to the fact that the population distributions are dependent only on the overall rate Ω that is proportional to $\sqrt{I_1 I_2}$.

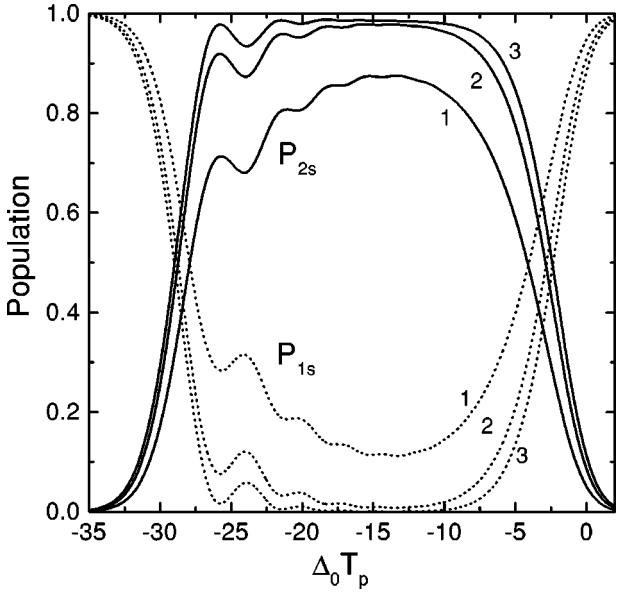

FIG. 4. Calculated two-photon line shapes for three values of uv-pulse fluence: 50 J/cm^2 (1), 100 J/cm^2 (2), and 150 J/cm^2 (3). The vuv-pulse fluence is 0.01 J/cm^2 and the IR (Stark) pulse fluence is 1 J/cm^2. The Stark pulse duration is $T_S = 1.5T_p$ and the time delay is $\tau = 1.2T_p$.

It is also of interest to study the effect of the Stark-free detuning Δ_0 on the efficiency of SCRAP. As depicted in Fig. 4, at low values of Δ_0 the coupling $\Omega(t)$ at the crossing point is not sufficiently strong to induce population transfer. This may be viewed as a violation of condition (8) due to the high value of $\sqrt{\ln(S_0/\Delta_0)}$. On the other hand, when Δ_0 approaches S_0, the two crossing points get closer, the resonance is swept through twice at comparable pump-pulse intensities, and the efficiency decreases and oscillates with respect to Δ_0. Finally, when Δ_0 is larger than S_0, the Stark pulse is too weak to sweep through the resonance and no population transfer takes place. The two-photon bandwidth increases for increasingly larger pump intensities.

The SCRAP method, as applied in this article for the $1s$-$2s$ transition in hydrogen, is an efficient mechanism for population inversion. In fact, the entire ensemble of atoms can be converted from the ground state ($1s$) to the metastable state ($2s$). Possible applications include the production of a much needed, intense Lyman-α source [15] with a near unity conversion efficiency (compared with 10^{-4} conversion efficiencies of frequency-mixing techniques in rare- or metal-gas vapors [15]). The two-color excitation scheme may also serve for probing hydrogenic Bose-Einstein condensates [12] by two-photon spectroscopy [13]. The total detection efficiency of the currently used scheme [16], limited to $\sim 10^{-5}$, may be greatly improved.

To conclude, we have shown that large populations of metastable hydrogen in the $2s$ state may be obtained from ground-state ensembles, via the SCRAP technique. We propose a two-color pump scheme that keeps ionization losses low while maintaining a high overall rate. Optimal population transfer efficiencies of 98% are predicted for pulse intensities that are sufficiently high to maintain adiabaticity throughout the process but not so large that substantial ionization occurs.

This work was supported by the German-Israeli foundation for scientific research and by the Deutsche Forschungsgemeinschaft. B.W.S. thanks the Alexander von Humboldt Foundation for financial support; his work is supported in part under the auspices of the U.S. Department of Energy at the Lawrence Livermore National Laboratory under Contract No. W-7405-Eng-48. We would like to thank S. Steuerwald and T. Rickes for valuable discussions.

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