Time evolution of two-photon population transfer between the 1*s* and 2*s* states of a hydrogen atom

M. Dörr,[1,2] O. Latinne,[2] and C. J. Joachain[2]
[1]*Max-Born-Institut, D-12474 Berlin, Germany*
[2]*Physique Théorique, Université Libre de Bruxelles, CP 227, B-1050 Bruxelles, Belgium*
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We have solved the time-dependent Schrödinger equation to analyze the time evolution of a hydrogen atom, initially in its ground 1*s* state, interacting with a one- or two-color linearly polarized laser pulse which is tuned close to a two-photon resonance with the 2*s* metastable state. The two-photon transition therefore does not involve an intermediate resonance state. Using a suitable combination of two laser pulses of different frequencies, population can be transferred from the 1*s* state to the 2*s* state via the continuum. In both the single-color and the two-color cases, a maximum amount of 17% of population can be found in the 2*s* state at the end of the pulse. We compare and interpret our results in terms of the time-independent Floquet eigenvalues. The system can also be modeled by an ''essential states'' two-level atom with decay. Both the Floquet and the time-dependent solutions are compared with the model predictions. The model allows systematic optimization studies for population transfer under a wide range of laser pulse parameters. [S1050-2947(97)06504-9]

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I. INTRODUCTION

Pumping an atom from the ground state into an excited state with the help of a laser is usually performed by tuning the laser to resonance and waiting sufficiently long. This process can be understood in terms of a simple two-level model atom [1], in which the laser coupling induces Rabi oscillations of the population between the two states. In principle, population can be transferred to 100% by choosing the pulse length appropriately (''π pulse''). Decay processes can be included in this model, for example the spontaneous decay, which destroys the coherence, but still allows at least 50% population transfer. When laser-induced ionization of the atom becomes important, however, in particular when the ionization rate of the upper level becomes comparable to the Rabi frequency, it is not evident that population can be transferred through Rabi oscillations.

Following the experimental observation [2] of relatively large populations in excited states after the interaction of xenon atoms with a laser pulse, there have been several theoretical time-dependent studies of possible mechanisms of population transfer [3]. The experiment involved a seven-photon resonance from the ground state, the excited state being coupled to the continuum by one photon.

The case considered in the present calculation involves only a two-photon coupling between the ground $(1s)$ and the excited $(2s)$ states, the two photons being either of the same frequency or of different frequencies. We explore different schemes of excitation, varying several of the laser field parameters, namely frequency, intensity, and pulse duration. Atomic units (a.u.) will be used throughout, unless otherwise indicated.

We consider a hydrogen atom, initially in its ground $(1s)$ state, which interacts with a short, strong laser pulse, described classically in the dipole approximation as an electric field $\mathcal{E}(t)=-c^{-1}d\mathbf{A}(t)/dt$, where $\mathbf{A}(t)$ is the vector potential

$$\mathbf{A}(t)=\hat{\boldsymbol{\epsilon}}\overline{A(t)}\cos[\omega(t)t+\phi], \tag{1}$$

with $\hat{\boldsymbol{\epsilon}}$ the polarization vector, $\overline{A(t)}$ the amplitude, and $\omega(t)$ the angular frequency. For sufficiently short laser pulses, this problem can be solved by direct numerical integration of the time-dependent Schrödinger equation [4,5]. Results obtained this way will be presented below and referred to as the ''full solution,'' giving the correct time dependence of the system.

It is useful to interpret the time-dependent results in terms of time-independent eigenstates of the atom in the field, with the help of the Floquet theory [6]. The Floquet calculation takes advantage of the fact that $A(t)$ and $\omega(t)$ are slowly varying functions of t, that is, they are approximately constant over one field period $T=2\pi/\omega$. This yields ''quasistationary'' states describing an ionizing atom in the field at constant $A(t)$ and $\omega(t)$, each state having a ''quasienergy'' with negative imaginary part $\mathrm{Im}(E)=-\Gamma/2$, where Γ is the decay rate of that state. The Floquet approach can be generalized to more than one color [6,7].

As the field intensity or frequency are varied adiabatically, the atomic system follows a particular Floquet eigenstate. If states of the system are resonantly coupled by one or more photon transitions, the single Floquet state approximation may become invalid. If the field parameters vary too rapidly, notably in the vicinity of crossings of Floquet eigenvalue curves, the evolution may no longer be adiabatic and part of the population can be transferred to other Floquet eigenstates. Landau-Zener descriptions of narrow isolated avoided crossings have been successful in describing experimentally observed population transfer [8]. However, many open questions still remain, which are not amenable to the usual Landau-Zener treatment, notably when the crossing is not avoided but true [9], or when the transition time through the crossing is too small for an adiabatic transition. This is the case in the results presented below.

II. METHOD

The time-dependent Schrödinger equation for a hydrogen atom in a laser field described by the vector potential (1) reads

$$i\frac{\partial}{\partial t}\Psi(\mathbf{r},t)=\left[\frac{\mathbf{p}^2}{2}-\frac{1}{r}+\frac{1}{c}\mathbf{p}\cdot\mathbf{A}(t)\right]\Psi(\mathbf{r},t), \tag{2}$$

where the term in $A^2(t)$ has been eliminated by performing a unitary transformation. Spontaneous decay can be neglected if the laser pulse duration considered is much shorter than the spontaneous decay lifetime; this is certainly the case for the $2s$ (metastable) state.

We use standard finite-difference procedures for the solution of this time-dependent equation, working in position space and using a spherical grid with a smooth mask-function absorber at large radius r [4,5,10].

The time-dependent solution thus obtained can be analyzed by projecting the wave function on the unperturbed (field-free) eigenstates $\langle i|$ of the hydrogen atom. Thus we introduce the probabilities

$$P_i=|\langle i|\Psi(t)\rangle|^2. \tag{3}$$

Although the field-free eigenstates are no longer eigenstates when the field is turned on, an analysis in terms of the probabilities P_i can give indications about the dynamics of the system. At the end of the pulse the P_i give the true fractional populations in each of the bound states.

If the intensity and frequency of the laser are assumed to be constant, quasistationary solutions can be calculated with the help of the Floquet theory [6,9,11]. The Floquet eigenstates are frequency- and intensity-dependent and constitute a generalization of the field-free stationary states to the case where the field is not zero.

In the present case, we are considering a near resonance between two discrete states of the atom. Such a system is effectively composed of only two states, which are coupled by the laser, plus one (or more) continuum into which the system can decay. The Hamiltonian for such a two-level atom, in the rotating wave approximation, can be written as a 2×2 matrix [12]

$$H=\begin{pmatrix} E_{1s}+\Delta-i\Gamma_{1s}/2 & \Omega \\ \Omega & E_{2s}-f(\omega)-i\Gamma_{2s}/2 \end{pmatrix}, \tag{4}$$

with the Rabi-frequency Ω proportional to the laser multiphoton coupling matrix element between the two states, Γ the states' ionization widths, the shift of the $1s$ state $\Delta\propto I$, and where ω is the laser frequency and I the intensity. The $2s$ state also experiences a shift in the field that, within the model, we have simply absorbed inside Δ. The presence of the term $f(\omega)$ is due to the fact that the resonance detuning is a function of the frequency (or frequencies). For the one-color two-photon resonance case below, $f(\omega)$ is simply equal to 2ω.

The eigenvalues of the two-level atom complex symmetric Hamiltonian of Eq. (4) can immediately be obtained and compared to the eigenvalues from a full Floquet calculation. The parameters in the model can be adjusted to reproduce the full Floquet results as closely as possible. Such a comparison is discussed in Fig. 2 below. Generally, the agreement is good as long as only two bound states are coupled by the field. The global effects of the other bound states can be incorporated by appropriate (and generally nonperturbative) parameters and field dependence of the Γ and Δ. If good modeling of the atom is achieved in this way, the two-level Hamiltonian can be used in a time-dependent calculation, solving

$$i\frac{\partial}{\partial t}|\psi(t)\rangle=H(t)|\psi(t)\rangle, \tag{5}$$

where $|\psi(t)\rangle$ is a column vector with two components and where $H(t)$ incorporates the explicit dependence on the time-varying field parameters. The 2×2 model solution can then again be compared to the full solution of the time-dependent Schrödinger equation (2).

III. RESULTS

A. Single-color two-photon resonance

The $1s$-$2s$ two-photon resonance in atomic hydrogen in a linearly polarized single-color laser field has been extensively used in studies of the zero-field energy difference of these two states [13]. state Usually, field-induced resonances produce avoided crossings in the dressed states' energies as a function of a parameter (for example, the laser intensity or frequency). In such two-level atoms, when ionization is neglected, Rabi oscillations are visible when projecting the time-dependent wave function onto the field-free eigenstates. These oscillations are damped by the ionization out of the bound states. When the ionization rate becomes larger than the Rabi frequency, no more oscillations will be visible [12].

In the present case, a laser field whose frequency is close to the two-photon $1s$-$2s$ resonance frequency will also ionize the excited $2s$ state by a single photon. The two-photon coupling matrix element between the $1s$ and the $2s$ states, $D^{(2)}_{2s,1s}$, turns out to be of almost the same magnitude as the square of the matrix element between the $2s$ state and the continuum, $|D^{(1)}_{\epsilon,2s}|2$, where ϵ denotes the (p) continuum state at energy $\epsilon=E_{2s}+(E_{2s}-E_{1s})/2$.

For this case, the parameters in the effective 2×2 Hamiltonian matrix (4) are, at low intensity,

$$\Omega\propto D^{(2)}_{2s,1s}\times I, \tag{6}$$

$$\Gamma_{2s}\propto|D^{(1)}_{\epsilon,2s}|^2\times I, \tag{7}$$

and

$$f(\omega)=2\omega. \tag{8}$$

Figure 1 shows the imaginary and real parts of the two eigenvalues of the Hamiltonian (4) for a fixed frequency $\omega=0.2$ a.u. *versus* intensity. A small field-induced off-resonant two-photon ionization width Γ_{1s} of the ground state has been incorporated in Eq. (4)—the perturbative half-widths in the absence of coupling (setting $\Omega=0$) are indicated by the dashed lines in the upper part of the figure. In the lower part, the dashed lines indicate the perturbative con-

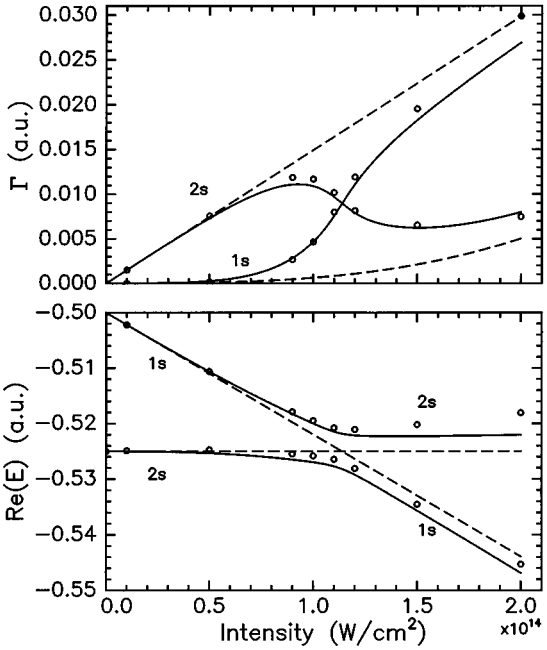

FIG. 1. Quasistationary Floquet eigenvalues for a fixed frequency $\omega=0.2$ a.u., as a function of intensity. Upper, width $\Gamma=-2\,\mathrm{Im}(E)$; lower, real part. Solid line, model (4); circles, full Floquet calculation [11]; dashed, perturbative result without resonance.

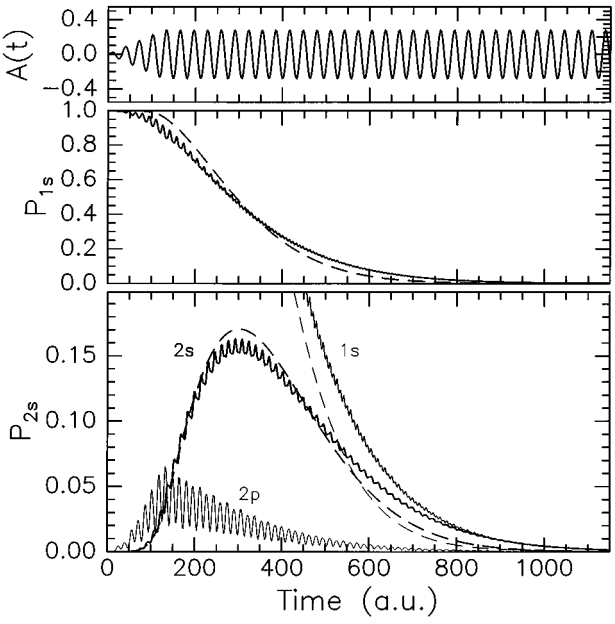

FIG. 2. Top part, vector potential $A(t)$. Frequency $\omega=0.2$ a.u., linear ramp turn-on from $t_0=0$ to $t_1=126$ a.u., constant thereafter, field strength $\mathcal{E}_0=0.056$ a.u. ($I=1.1\times10^{14}$ W/cm2). Lower parts, time evolution of the projection onto the unperturbed $1s$ and $2s$ eigenstates. Solid line, full solution [5]; dashed line, model (4). The thin lines in the lower (P_{2s}) plot are a repetition of the P_{1s} results above. The thin line labeled $2p$ gives the population of the $2p$ state (in the full solution).

tinuation of the uncoupled shifts and one should note the deviation from these lines at high intensity. This deviation is due to the fact that as a function of intensity the coupling energy grows as rapidly as the detuning and thus the deviation from the perturbative uncoupled (dashed) result is roughly constant. The main point of the figure, however, is to exhibit the excellent quantitative agreement between the full Floquet solution [11] and the model (4). In fact, the parameters of the model have been taken from the full Floquet calculation rather than from a perturbative calculation.

Since both Ω and Γ_{2s} in Eq. (4) are proportional to the field intensity, there is a scaling invariance of the system with intensity: for a particular frequency, the intensity at which the crossing occurs is given by the (intensity-dependent) shift $\Delta(I)$. At sufficiently low intensities, $\Delta(I)=\Delta\times I$. By rescaling the time and the detuning $\delta=E_{1s}+\Delta(I)+2\omega-E_{2s}$, the behavior (e.g., as a function of frequency ω) is the same, at all intensities for which the model (4) is applicable. It breaks down at intensities higher than about 10^{14} W/cm^2 as is apparent from Fig. 1. Therefore, the time dependence discussed below applies at any lower laser intensity, even in the very weak perturbative limit. This scaling applies in fact to any two-photon resonance, where the upper state is connected to the continuum by a single photon. The explicit time dependence, however, depends on the magnitude of the two matrix elements, $D^{(2)}_{2,1}$ and $|D^{(1)}_{\epsilon,2}|^2$, relative to each other and to the photon energy.

The gap at the avoided crossing of the real parts has the same magnitude as -2 times the imaginary parts at the crossing. This indicates that the coupling between the states is just as large as the damping due to the ionization width of the upper state. Therefore, the system should behave as a critically damped oscillator. This behavior is indeed exhibited in Fig. 2. The upper part shows the field's vector potential, $A(t)$, which is ramped on rapidly and then kept constant, corresponding to the intensity $I=1.1\times10^{14}$ W/cm^2, just at the resonance for $\omega=0.2$ a.u. In the lower parts, the projection of the time-dependent wave function onto the field free eigenstate $2s$ is shown. The thicker solid line is from a full numerical solution of the Schrödinger equation for the hydrogen atom [5], while the dashed line is the solution to the time-dependent Schrödinger equation using the Hamiltonian (4). The agreement between the two is again very good. The behavior at times larger than 500 a.u., however, is qualitatively different for the full solution as compared to the two-level model. The P_i of the full solution converge to the same near-exponential decay curve, while the model results cross and show different and in part nonsingle-exponential behavior. In fact, for a resonance case one should not expect single-exponential decay and the full solutions in Fig. 2 can be fitted very well by $P_i(t)=P_{i0}t\exp(-\alpha_i t)$ up to $t=1600$ a.u., where our numerical precision starts to run out. We obtain $\alpha_{1s}=0.0083$ a.u. and $\alpha_{2s}=0.0078$ a.u., which is of the order of but not identical to the Floquet widths at the crossing. In the lower part the thin lines give the results for the $1s$ and for the $2p$ populations, respectively. The $2p$ state is never actually populated but it contributes to the ''field dressed'' $1s$ state.

The main point of this second figure is that only a maximum of about 15% of population is ever found in the $2s$ excited state. This amount of real population transfer can indeed be obtained, by employing a finite laser pulse. It is evident from Fig. 2 that the pulse duration must be of the order of 300 a.u. In Fig. 3 we show the results for a trapezoi-

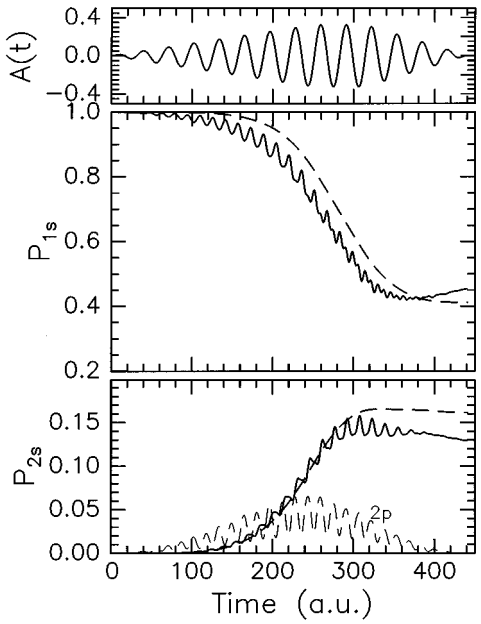

FIG. 3. Same as for Fig. 2, real population transfer with a short pulse. Frequency $\omega=0.2$ a.u., trapezoidal pulse from $t_0=0$ to $t_1=251$ to $t_2=314$ to $t_4=440$ a.u., peak field strength $\mathcal{E}_0=0.065$ a.u. ($I=1.5\times10^{14}$ W/cm^2).

dal pulse, where the field strength is increased linearly from t_0 to t_1, then held constant from t_1 to t_2, and finally turned off linearly from t_2 to t_3. The same amount can be obtained with other pulse shapes (e.g., a $\sin^2$ or a sech^2 pulse) showing that the population transfer is robust with respect to the pulse shape (as long as it is reasonably smooth).

As said above, this result is also robust under variation of the crossing intensity and frequency: the time after which a maximum of population will be transferred into the excited state depends on the ionization rate at the crossing, but the maximum amount will always be around 15%. Conversely, if the atom experiences a laser pulse of fixed duration τ a maximum $2s$ signal will be seen after the pulse for a particular frequency ω_X, which is approximately given by

$$\omega_X \approx \frac{E_{2s}-E_{1s}}{2} - \frac{\pi\overline{\Delta}}{D^{(2)}_{2s,1s}\tau}. \tag{9}$$

This condition follows since for ω_X the crossing is at an intensity of $I_X=(E_{2s}-E_{1s}-2\omega_X)/\Delta$, and the duration τ must be approximately equal to π/Ω, where Ω is the energy width of the gap at the crossing, with $\Omega=D^{(2)}_{2s,1s}\times I_X$. For an optimal population transfer with a short pulse, in order to reduce ionization losses, the peak intensity I_0 must be chosen somewhat larger than I_X (roughly a factor 1.3), in order to make the pulse as short as possible, as seen in Fig. 2. Since the shift is no longer linear at higher intensity, the condition (9) is valid only for I_0 not too large, as mentioned above. If a two-photon transition between a bound state and a member of a Rydberg series of higher-lying excited states is considered, a particular peak intensity will induce optimal population transfer only to a few specific Rydberg states, for a fixed τ and ω.

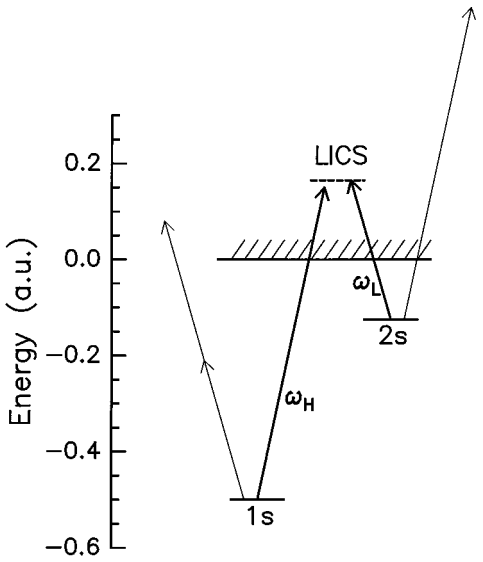

FIG. 4. Schematic energy diagram for laser-induced continuum structure involving a two-color coupling of the two states, via the continuum.

The question now arises whether more efficient population transfer is possible using the intensity variation together with a frequency variation (chirp) of the laser pulse. The avoided crossing seen in Fig. 2 might be used by choosing a path in intensity-frequency parameter space which leads to an adiabatic transfer of population from the $1s$ to the $2s$ state. The problem with this hypothetical adiabatic variation is that an adiabatic variation through the avoided crossing must occur during a time which is longer than the inverse of the Rabi splitting at the crossing. But since this time is of the same order as the lifetime of the states at the crossing (see Fig. 2, bottom part), the system will be ionized during the slow passage through the crossing. Thus, in Fig. 3, the optimal pulse duration is only 450 a.u., which is roughly equal to π/Ω. Due to the intensity scaling mentioned above, the situation will not change by choosing a lower intensity and corresponding different frequencies. Systematic optimization studies using the model (4) also demonstrate that the additional freedom of a frequency chirp does not lead to larger population transfer.

Also, a combination of two laser pulses does not yield a higher $2s$ population, as has been verified by systematic studies on the two-level model, varying the pulse durations, peak intensities, and temporal offsets (or overlaps). It is interesting to note, however, that after the first pulse has passed, a second pulse can be applied which coherently *depopulates* the $2s$ level completely, actually pumping back some population into the $1s$ level.

B. Laser-induced continuum structure

Many authors have considered the ''Λ-configuration,'' depicted in Fig. 4, in which two bound states 1 and 2 of an atom are coupled via the continuum, using two lasers of angular frequencies ω_L and ω_H [14,15]. Usually the ω_H laser (the probe) is kept weak so that the ω_L laser effectively induces continuum structure (LICS) analogous to an autoionizing resonance. This resonance gives rise to a marked inter-

ference structure in the ionization rate of the ground state. At the heart of this interference lies a degeneracy of two *complex* Floquet eigenvalues, at a particular value of the two frequencies and intensities. Similar degeneracy points have been discussed in [16,17]. This opens the fascinating prospect of observing a geometric phase [18] by encircling such a degeneracy point by a suitable variation of the laser parameters. The main problem comes from the fact that the atom is decaying in the field, and most of the population is ionized when the path is completed. In the present case we are interested in optimal population transfer and this is only effected by a nonadiabatic transition. In the single-color case discussed in Sec. III A above, for exactly the same reason the optimal population transfer was also given by a nonadiabatic transition.

Figure 5 shows the complex Floquet quasienergies for atomic hydrogen in two laser fields with $\omega_L=0.3$ a.u. fixed, $I_L=2\times10^{14}$ W/cm^2, ω_H as indicated on the curves, and I_H varying from 0 to 3×10^{14} W/cm^2 along the four curves on the left and to 1×10^{15} W/cm^2 along the four curves on the right. Comparison with the results of [17] shows that the simple LICS model is approximately applicable here. In Eq. (4), the width Γ_{2s} is now given by the sum of the two colors' ionization probabilities for the upper state, Δ includes the shifts of both states (in the absence of resonance), and Γ_{1s} is principally due to the ionization width of the $1s$ state by the laser field ω_H alone. The two-photon coupling of the $1s$ and the $2s$ states now occurs via the continuum and thus Ω is now complex and proportional to $(q-i)D^2_{2s,1s}$, where $D^2_{2s,1s}$ is equal to $\sqrt{\Gamma_{1sH}\Gamma_{2sL}}$. The quantities Γ_{1sH} and Γ_{2sL} are, respectively, the one-photon ionization rate of the $1s$ state by frequency ω_H alone and of the $2s$ state by ω_L alone. The Fano parameter $q=0.35$ can be obtained from a Fano-profile fit to the Floquet rate of the $1s$ state versus ω_H, at low I_H. As discussed in [17], the resulting complex-symmetric Hamiltonian matrix (4) has pairs of two degenerate eigenvalues at two different complex energies, corresponding to two different particular sets of field parameters. The real parts of the two degeneracies lie asymmetrically about $\delta=0$, where the detuning is defined as $\delta=E^{(0)}_{2s}-E^{(0)}_{1s}+\omega_L-\omega_H$. The energy separation between the two degeneracy points is of the order of Γ_{2s}. In order to follow a quasienergy trajectory, one must vary the parameters (ω_H and I_H) slowly enough, while in order not to lose all population due to ionization along the way the parameter variation must be fast enough. The first condition implies two relations: (i) the pulse duration τ of the lasers must be long enough for the frequency ω_H to be defined within $\Delta\omega_H=|d_1-d_2|\approx\Gamma_e$, thus $T_H/\tau<\Delta\omega_H/\omega_H\Leftrightarrow\tau>2\pi/\Gamma_e$ (noting $T_H=2\pi/\omega_H$), and (ii) the passage should be slow on the time scale set by the two-color Rabi coupling frequency Ω_R between the two states g and e. Estimating $\Omega_R\approx\Gamma_e$ yields the relation $\tau>1/\Gamma_e$. The second condition, on the other hand, implies roughly $\tau<1/\Gamma_e$. Evidently, these relations are incompatible, but only marginally so.

C. Optimal two-color population transfer

We note that the present two-color transfer mechanism uses noncommensurable frequencies and thus does not rely on a fixed phase relation between the two colors. However, the general ideas of coherent control are applicable. Wishing to gain maximal output of the cost function, given as the final population in the excited state after the laser pulses, one must find the optimal laser parameters within a defined parameter variation space. Computation of the cost function through solution of the full time-dependent Schrödinger equation would be too expensive in a multidimensional parameter space. We have therefore used the two-level model and applied a numerical multidimensional minimization procedure to find the optimal laser pulse parameters in order to transfer a maximum amount of population into the $2s$ state at the end of the pulses. We have allowed parameter variations of the peak intensities of both pulses, the frequency of the higher frequency pulse and the pulse shapes, keeping the lower frequency fixed at $\omega_L=0.3$. We have also allowed for a chirp in the higher frequency pulse, the optimal transfer requiring no chirp, however.

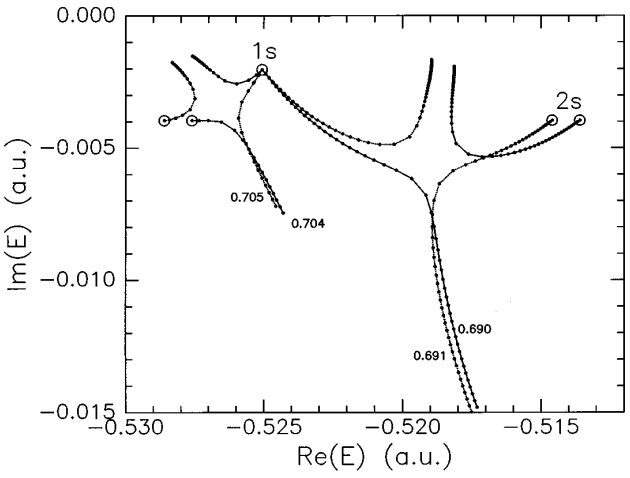

FIG. 5. Floquet quasienergy trajectories in the complex energy plane for two-photon ionization of atomic hydrogen in the $1s$ or $2s$ state. Frequency $\omega_L=0.3$ a.u. fixed, intensity $I_L=2\times10^{14}$ W/cm^2 fixed, ω_H indicated in a.u., and I_H varying along the curves from 0 to 3×10^{14} W/cm^2 (left curves) or to 1×10^{15} W/cm^2 (right curves). The dots denote steps in intensity of 2×10^{13} W/cm^2.

In Fig. 6 we show the optimal transfer result, in which a final population of 16% is obtained in the $2s$ state, while the $1s$ state is almost empty at the end of the pulse. The result is robust, that is, it is not too sensitive to the exact numerical values of the parameters of the laser pulses. The reason for the near-complete depopulation of the $1s$ state is simply that its ionization rate through the higher-frequency field alone is much larger than the ionization rate of the $2s$ state by the high-frequency field. Our result shows that the population transfer through the continuum in a real system, including the incoherent decay channels, need not be as small as noted in [15]. The pulse parameters must of course be optimized within a model including all interactions, as we have done.

It must be stressed that the present optimal pulse parameters can only be obtained with the help of the adapted model, since a minimization must be performed in a multidimensional parameter space for the two laser pulses, which would require prohibitively large computer time when using the full time-dependent solution.

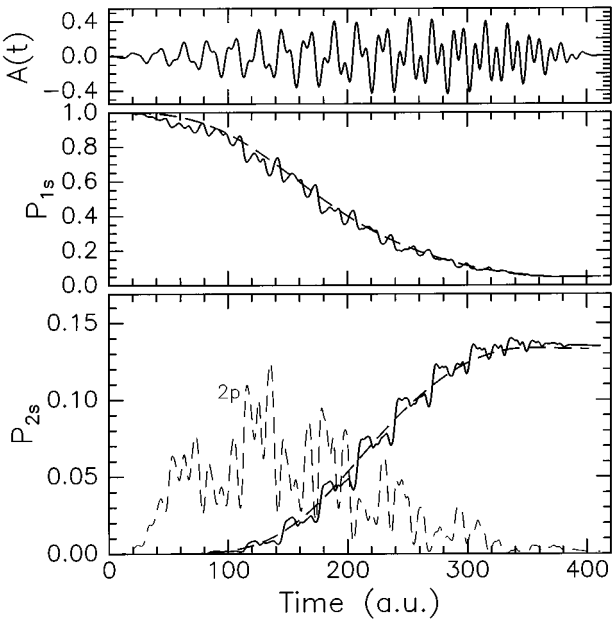

FIG. 6. Same as for Fig. 3, but for a combination of two laser pulses of different frequency. Lower frequency $\omega_L = 0.3$ a.u., trapezoidal pulse from $t_0 = 12$ to $t_1 = 290$ to $t_2 = 340$ to $t_3 = 400$ a.u., peak field strength $\mathcal{E}_{0L} = 0.0755$ a.u. Higher frequency $\omega_H = 0.697$ a.u., sin-pulse from 0 to 410 a.u., peak field strength $\mathcal{E}_{0H} = 0.174$ a.u. These values give an optimal population transfer.

IV. CONCLUSIONS

When a hydrogen atom, initially in its $1s$ ground state, is subject to a laser pulse whose frequency is close to the two-photon resonance with the metastable $2s$ excited state, the system behaves like an overdamped oscillator. Only a maximum of 16% of population can be transferred into the excited state. It is not possible to follow quasienergy curves adiabatically for the hydrogenic two-photon $1s$-$2s$ coupling since the damping (ionization) is comparable to the coupling (Rabi frequency). It is never possible to achieve an inversion, that is, a population larger in the excited than in the ground state, for this case. Chirping the frequency of the laser pulse does not lead to more effective population transfer in the present case.

For a two-color coupling of the two states, via the continuum, effective population transfer is again possible, in fact to about the same amount. This can be achieved by an optimized choice of the laser pulse parameters. In this case, the ground state is severely depopulated, since its ionization rate by the high-frequency laser alone is much larger than the one of the $2s$ state. Since the energy difference of the degeneracies is comparable to the ionization rate, in the two-color case again it is not possible to follow quasienergy curves adiabatically and it is consequently not possible to resolve the two complex energy degeneracy points.

We have shown that for resonant couplings, even via the continuum, a two-by-two effective Hamiltonian reproduces to satisfactory accuracy the solution of the full Schrödinger equation, both fully time-dependent and within the Floquet approach. It is important in order to obtain accurate modeling to adjust the model parameters to the full nonperturbative Floquet calculation; if perturbative couplings are used, the agreement is usually not good. The adjusted effective Hamiltonian has been used in a numerical minimization calculation, which yields optimal laser pulse parameters. In the present case, optimization has been performed on the population in the excited state at the end of the laser pulses.

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