PHYSICAL REVIEW A VOLUME 52, NUMBER 5 NOVEMBER 1995

Time evolution of a hydrogen atom in a strong, ultrashort, high-frequency laser pulse

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(Received 23 May 1995)

We have solved the time-dependent Schrödinger equation for a hydrogen atom, initially in its ground state, subject to a strong, ultrashort laser pulse of high frequency, $\omega = 2.0$ a.u. We compare and interpret our results in terms of the time-independent Floquet eigenvalues. Even for an ultrashort pulse of 3 cycles half-width, the Floquet adiabatic picture is a very good approximation. We discuss the physical reasons and also the visible deviations from the single-resonance-state approximation.

PACS number(s): 32.80.Rm

The most direct theoretical investigation of the interaction of a quantum system with a strong laser field in the nonrelativistic domain consists in solving numerically the time-dependent Schrödinger equation. This approach has been applied to a wide variety of one-electron atomic systems, ranging from one-dimensional model "atoms" [1,2], to single-active-electron approximations, to multielectron systems [3], comprising of course the hydrogen atom [4–6].

In the present work, 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. The time dependent Schrödinger equation reads [we use atomic units (a.u.) throughout, unless otherwise indicated]

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

We recall that within the dipole approximation the term in $A^2(t)$ can be eliminated from Eq. (1) by performing a unitary transformation on the wave function. The dipole approximation is well justified for the present case [6].

We employ standard procedures for the solution of this time-dependent equation, working in configuration space and using a finite-difference approximation for the momentum operator $\mathbf{p} = -i\nabla$ [3,6]. Furthermore, we take the direction $\hat{\mathbf{z}}$ of $\mathbf{A}(t) = \hat{\mathbf{z}}A(t)$ fixed, implying that all fields are linearly polarized along the $\hat{\mathbf{z}}$ direction. As a result, Eq. (1) exhibits cylindrical symmetry about the $\hat{\mathbf{z}}$ axis, reducing the problem to two spatial dimensions.

We compute the wave function on a radial grid, comprising 66 points in the polar angle θ and up to 16 500 equidistant radial points between $r=0$ and $r=165$. We have checked convergence in all of these values. In order to avoid reflections from the outside border of our grid, we set the outer part of the wave function to zero (periodically, once every field cycle) by multiplying with a continuous mask function which goes from 1 to 0 linearly between $r=55$ and $r=110$.

The time-dependent solution thus obtained can be analyzed in terms of the unperturbed (field-free) eigenfunctions of the hydrogen atom. The field-free ground state energy we obtain using our grid is $\bar{E}_{1s} = -0.499\,800$. For the next few bound states the energies are $\bar{E}_{2s} = -0.124\,987$, $\bar{E}_{2p} = -0.125\,252$, $\bar{E}_{3s} = -0.055\,553$, $\bar{E}_{3p} = -0.055\,628$, $\bar{E}_{3d} = -0.055\,463$. These values are all within less than 0.1% of the exact values $-1/2n^2$. The corresponding eigenstates are of course no longer eigenstates when the field is turned on. Nevertheless, an analysis in terms of the probabilities

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

where $\langle i|$ represents the ith field-free bound eigenstate of atomic hydrogen, can give some indications about the shape of the full time-dependent wave function $\Psi(\mathbf{r},t)$. At the end of the pulse the P_i give the true populations in the bound states.

The appropriate "time-independent" eigenstates for the atom in the field can be obtained within the Floquet theory [7]. This yields "quasistationary" states describing an ionizing atom in the field, 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 calculation for the present single color case assumes a fixed intensity and frequency of the laser, i.e., a monochromatic field. As the field intensity or frequency are varied, the atomic system follows adiabatically a particular Floquet eigenstate. If states of the system are resonantly coupled by one or more photon transition, the single Floquet state approximation may become invalid. We do not consider this question in the present work, deferring a detailed study of adiabaticity for the case of several near-resonantly coupled states to a future publication. In this paper, we select a frequency that is so high that no single- or multiphoton resonances between discrete states of atomic hydrogen are present. Even when no crossing is present, it is interesting to inquire what a minimum pulse duration should be for the Floquet approach to be useful.

Floquet eigenvalues (E_α) and eigenvectors (Ψ_α) can be obtained directly from a time-dependent calculation by diagonalizing the one-cycle time evolution operator

$$\Psi(t+T) = \sum_\alpha e^{-iE_\alpha T}\Psi_\alpha\langle\Psi_\alpha|\Psi(t)\rangle \tag{3}$$

$$= U(t+T,t)\Psi(t), \tag{4}$$

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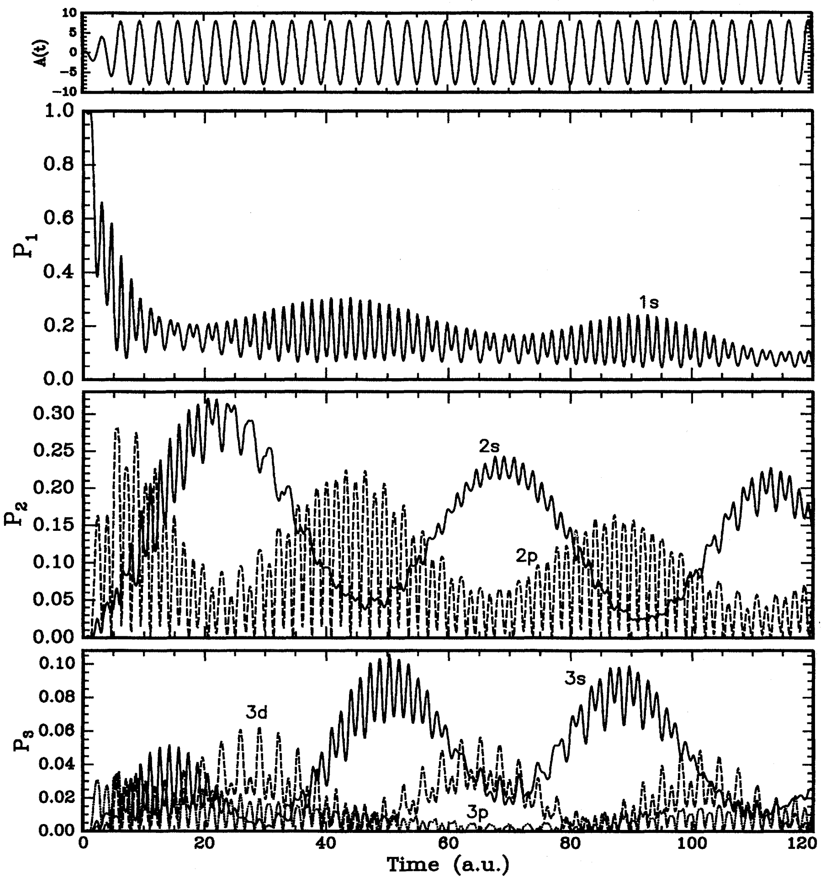

FIG. 1. Time evolution of the projection of the wave function onto the zero-field eigenstates of the hydrogen atom : 1*s*; 2*s*, and 2*p*; 3*s*, 3*p*, and 3*d*. Top: $A(t)$, magnitude of the vector potential, linearly ramped on over two cycles, then held constant.

where $T=2\pi/\omega$, and ω is the constant angular frequency of the laser field. This method has been used for analyzing the time-dependence for one-dimensional models [1,8].

We also remark that the applicability of the Floquet approximation in the absence of resonances has been tested and compared successfully with experiment in the case of multiphoton ionization of the negative hydrogen ion [9,10].

Even in the presence of resonances, the single Floquet state approximation can be very successful at describing certain overall features of the time-dependent case. Indeed, for hydrogen atoms subject to a 0.5 psec optical-frequency laser pulse, the resonance peaks in the electron spectrum have been reproduced to very good accuracy by a Floquet calculation [11]. The reason for the agreement in this case lies in the fact that all crossings with the excited states have a small gap and that the lifetimes of the excited states are of the order of the optical period, much shorter than the ground state lifetime.

In the present calculation, we consider an angular frequency $\omega=2.0$ a.u., well above the field-free atomic hydrogen ionization energy of 0.5 a.u. This frequency is in the adiabatic stabilization regime [12]. Furthermore it is high enough to allow a nonrelativistic treatment for the intensities considered here.

In Fig. 1 we show our time-dependent results, obtained by numerically solving Eq. (1) for a ramped-constant field

$$A(t)=A_0\, f(t)\, \cos\omega t,$$

$$f(t)=[\Theta(t)\; t-\Theta(t-\tau)\; (t-\tau)]/\tau,$$

$$\Theta(t)=\begin{cases}1, & t>0\\ 0, & \text{otherwise}\ .\end{cases} \tag{5}$$

The turn-on is effected very rapidly, over two cycles only, $\tau=2T=2\pi$. The peak field amplitude is $\mathcal{E}_0=16$ a.u., corresponding to an intensity of $I=9\times10^{18}$ W/cm^2. We observe that excited state population is visible mainly in the $2p$ state. This should be interpreted not as "real" population but rather as describing the polarization of the $1s$ "Floquet" state. In fact, we see oscillations between the $1s$ and the $2p$ populations with the laser frequency, indicating that both states are strongly mixed, and the corresponding Floquet solution therefore contains a mixture of both field-free states besides important admixtures of other states, including field-free continuum states. We have followed the time evolution of the wave function for a long duration. Several features appear at this large intensity. In particular, there is a slow oscillation visible in the $2s$ probability, with a frequency of about 0.14 a.u. The same periodic oscillation is also visible in the envelope of the $1s$ probability. This corresponds to an interference between the coherently populated $1s$ and $2s$ Floquet states. The transfer of population to the Floquet state which has predominantly "$2s$" character does not occur through a

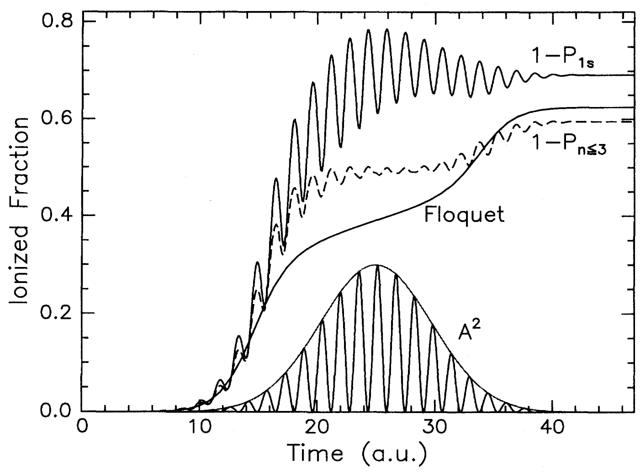

FIG. 2. Time evolution of the ionization probability in an ultrashort laser pulse of frequency 2 a.u., with Gaussian envelope, for which the quantity $|A(t)|^2$ is indicated, in arbitrary units. Floquet: time average of the quasistationary Floquet results. $1-P_{1s}$: projection onto the unperturbed $1s$ eigenstate. $1-P_{n\leq 3}$: projection onto the lowest eigenstates up to and including the $n=3$ manifold.

level crossing, since the frequency is too large. It occurs at the beginning of the pulse because the short pulse has a significant frequency spread, since it has been ramped on rapidly. We have performed Floquet calculations [13] with the result that the real parts of the Floquet energies are $E_{1s}=-0.226$ a.u. and $E_{2s}=-0.084$ a.u. Thus the energy difference is 0.14 a.u. and corresponds exactly to the beat frequency observed. The decay rates of the two states are quite different, $\Gamma_{1s}=0.006$ a.u. and $\Gamma_{2s}=0.0005$ a.u. This also agrees with an estimate of the decay rate obtained from a fit to the maxima in the oscillations.

In Fig. 2 we show our time-dependent results for an ultrashort laser pulse with Gaussian envelope

$$A(t)=A_0 e^{-(t-t_0)^2/\tau^2}\cos\omega t \qquad (6)$$

for $\omega=2.0$ a.u. and $\tau=9.43$ a.u., $\mathcal{E}_0=10$ a.u. (corresponding to a peak intensity of $I_0=3.5\times 10^{18}$ W/cm^2). The line labeled "A^2" shows $A^2(t)$ in arbitrary units. Thus the pulse comprises only about 10 cycles of the field. Similar pulses have been considered in [5]. The smooth line in the figure gives the Floquet adiabatic ionization probability

$$P_F(t)=1-\exp\left[-\int_0^t \Gamma[I(t')]dt'\right]. \qquad (7)$$

It is important to note that at the end of the pulse the Floquet result $P_F=0.625$ is quite close to the time-dependent result $1-P_{1s}=0.692$ (wavy line). The agreement becomes even better when one computes the true ionization probability at the end of the pulse from the time-dependent calculation, namely $P_{\text{ion}}=1-\Sigma_i P_i$, where i runs over all bound states. In the figure we show $P_{n\leq 3}=1-\Sigma_i P_i$ (with $i=$ $1s$, $2s$, $2p$, $3s$, $3p$, $3d$) as the wavy dashed line; its final value is 0.595. The main excited state population at the end of the pulse is $P_{2s}=0.090$, all others being much smaller ($P_{3s}=0.005$). The excited states are populated due to the frequency spread of the pulse, which has a power spectrum

$$P(\omega)\propto\exp[-4\tau^2(\omega-2)^2], \qquad (8)$$

whose half-width is $\sqrt{\ln 2}/\tau=0.09$ a.u. That $1-P_{1s}$ and P_F disagree during the pulse and that the cycle-average of $1-P_{1s}$ peaks at the peak of the pulse and then decreases, seemingly "pulling probability back into the $1s$ state" is not surprising, since, as stated above, P_{1s} should not be interpreted as the probability of population in the ground state during the pulse. The stairlike inflections in P_F at strong fields are a result of the reduction of Γ_{1s} at large intensity and this behavior is also visible in the dashed curve (for a discussion of this adiabatic stabilization, cf. [12]).

The width Γ from the Floquet calculation gives the probability to leave the quasibound state and reach the continuum. Coherent excitation processes to other states that are energetically allowed for the short pulse are not taken into account in a Floquet calculation. It is therefore reasonable that the Floquet results lie between the $1-P_{1s}$ and the $1-\Sigma_i P_i$ results.

This work has been supported by the EC through the HCM Contracts Nos. ERBCHRXCT 920013, 940470 and ERBCHBGCT 930419, 940552 and by the Institut Interuniversitaire des Sciences Nucléaires (Belgium). M.D. was supported by a Habilitations-Stipendium of the Deutsche Forschungsgemeinschaft.

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