Above-threshold ionization of atomic hydrogen via resonant intermediate states

K. J. LaGattuta
Applied Theoretical Physics Division, Los Alamos National Laboratory, Los Alamos, New Mexico 87545
(Received 23 July 1992)

We performed state-of-the-art calculations of the emitted-electron kinetic-energy distribution for atomic hydrogen, initially in its ground state, and interacting with a linearly polarized laser field. The laser had frequencies in the one-photon-resonant ($1s+\omega\rightarrow 2p$), two-photon-ionization regime, and irradiances between 10^{13} and 10^{14} W/cm^2. A direct numerical solution of the time-dependent Schrödinger equation was performed using a standard method, and incorporating "slow" laser turn-on and turn-off pulse envelope switching functions. Structure was observed, in addition to the usual above-threshold-ionization peaks. This was deduced to be an effect of the Rabi coupling between the $1s$ and $2p$ states. The precise form of this structure was found to depend on both the laser detuning and the irradiance, but was unaffected by the turn-on transient.

PACS number(s): 32.80.Rm, 42.50.Hz

INTRODUCTION

Calculations have recently appeared of multiphoton-ionization (MPI) rates, and emitted-electron angular distributions, for ground-state atomic hydrogen, in the one-photon-resonant, two-photon-ionization regime [1]. In this Brief Report we describe related calculations of the emitted-electron kinetic-energy distributions; i.e., the so-called above-threshold-ionization (ATI) spectra.

Many measurements of emitted-electron kinetic-energy spectra produced at high laser irradiances, have been performed, following the initial observation [2]. These experimentally determined spectra have almost always registered the signature feature of intense-field MPI; i.e., multiple peaks, separated by units of the photon energy. Typically, if the laser irradiance is not too high, then the threshold peak is the strongest, while ATI peaks grow weaker with increasing energy. If the irradiance is very high, then the threshold and low-order ATI peaks may be suppressed. A variety of calculations have been able to reproduce qualitatively these experimental results. A comparison between theory and experiment has just appeared [3]. This field has also just undergone a comprehensive review [4].

For systems driven by very short laser pulses, other structures have emerged in the emitted-electron kinetic-energy spectra, both experimentally [5] and theoretically [6]. These have been related to resonances, the positions of which are irradiance dependent. As the laser approaches peak irradiance, excited states of successively lower energy will be lifted into resonance. Then an ionized electron can appear in the continuum with an energy $E_j=E_{\rm th}-I_0(t_j)/(4\omega^2)+n\omega$, where $E_{\rm th}$ is the threshold energy at zero irradiance, ω is the laser frequency, and t_j is the time at which the jth excited state is lifted into resonance, at a given point in the focal volume. For $n=0$, the group of accessible excited states (j values) leads to a family of peaks just below $E_{\rm th}$. This might be called the "threshold family." For $n=1$, a similar family can appear below $E_{\rm th}+\omega$, the "first ATI family"; etc. We assume that at least two photons are required to ionize. (It has been suggested recently that some resonant states ionize very rapidly, so that the time t_j is also essentially the time at which ionization occurs [7]. This should lead to very broad spectral features. Other states ionize more slowly, producing sharper features. In this case, there may even be a large population remaining in the excited state after the pulse has passed [8]. However, the interpretation of these data is still somewhat controversial [9].

In the following, we point out that resonant multiphoton ionization (REMPI) can give rise to structures in the ATI spectrum, in addition to those just described. These are structures due to explicit Rabi coupling between the bound states of the atomic or ionic system driven by the laser. A splitting of the individual peaks in the "ATI family" can occur. Identical structures were described earlier [10], having appeared in calculations performed for $I_0\sim 10^{10}$ W/cm^2, using analytical techniques appropriate to those low irradiances.

RESULTS AND DISCUSSIONS

We report here specific results of numerical calculations aimed at describing the interactions of atomic hydrogen with a linearly polarized pulsed laser, at *moderately high irradiances*. These calculations were performed according to a standard prescription [11], and involved a full solution of the (two-dimensional) time-dependent Schrödinger equation. Since this problem possesses axial symmetry, it is inherently two dimensional. In the following, we use atomic units (a.u.).

The equation to be solved is

$$[H_0+\mathbf{r}\cdot\mathbf{E}(t)-i\partial/\partial t]\Psi(\mathbf{r},t)=0 \tag{1}$$

for $t\geq 0$, subject to the initial condition

$$\Psi(\mathbf{r},0)=\phi_{1s}(\mathbf{r})=2e^{-r}/\sqrt{4\pi}\ , \tag{2}$$

and where H_0 is the Coulomb Hamiltonian

$$H_0=-(1/2)\nabla^2-1/r\ . \tag{3}$$

The laser electric field is

$$\mathbf{E}(t)=\hat{\mathbf{z}}f(t)E_0\sin(\omega t)\ , \tag{4}$$

where $\hat{\mathbf{z}}$ is a unit vector pointing in the z direction. The amplitude function $f(t)$ switches on as a sine-squared function over a period of 100 a.u. of time, is constant at a value of 1 for 1800 a.u., and falls to zero as a sine-squared function over another 100 a.u. of time. The duration of this "pulse" is ~ 50 fsec.

The wave function computed at $t=t_f=2000$ a.u. is then projected onto Coulomb continuum states ϕ_E, and the spectrum of emitted-electron energies determined as

$$dP_E/dE=\left|\int d\mathbf{r}\,\phi_E^*(\mathbf{r})\Psi(\mathbf{r},t_f)\right|^2\ , \tag{5}$$

where P_E is the probability of appearing in the continuum with energy E, and $*$ denotes complex conjugation. The total ionization probability is

$$P_{\text{ion}}=\int dE\,dP_E/dE\ . \tag{6}$$

We consider first the case of exact $1s$-$2p$ resonance, where $\omega=0.375$ a.u., and choose an irradiance $I_0=10^{13}$ W/cm^2, corresponding to a value $E_0=0.0169$ a.u. Under these conditions, the photoionization rate has been determined earlier [1] to be $R_{\text{PI}}=1.5\times10^{12}$/sec, implying a time to ionize of $\sim 3\times10^4$ a.u., approximately 15 times longer than the duration of our simulated pulse. Consequently, the total probability of ionization over the course of this pulse is approximately 0.05.

For this choice of laser parameters, the $1s$ and $2p$ hydrogenic states are closely coupled. They execute Rabi oscillations of period $2\pi/\Omega_R$, where the Rabi frequency is $\Omega_R=r_{1s,2p}E_0/\sqrt{3}=0.013$ a.u. and r_{1s2p} is the dipole matrix element. See Fig. 1, where we plot the analog of Eq. (5) for bound states, while the pulse is on.

The numerical technique allows us to determine the occupation of each continuum partial wave. But, because the irradiance chosen here is not very high, almost all of the ionized flux goes into the very low partial waves. In Fig. 2, we plot dP_E/dE versus E for the sum of all partial waves ($0\le l\le 4$, in this calculation). The threshold peak should appear at an energy of $E=\varepsilon_{1s}+2\omega-I_0/(4\omega^2)$

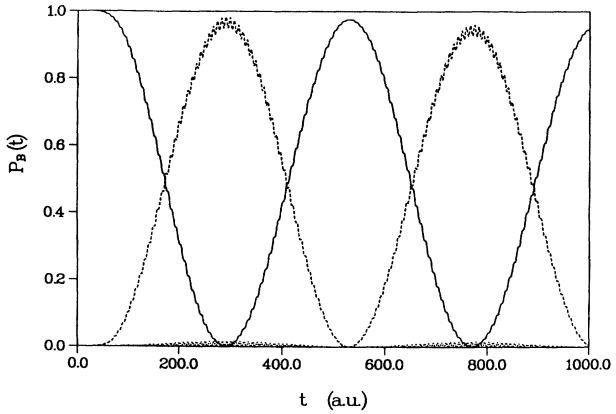

FIG. 1. Probability of being in $1s$ (solid) or $2p$ (dashed) states vs t; $I_0=10^{13}$ W/cm^2 and $\omega=0.375$ a.u.; $2s$ (dotted) probability also appears. Only the first half of the 50-fsec pulse is displayed.

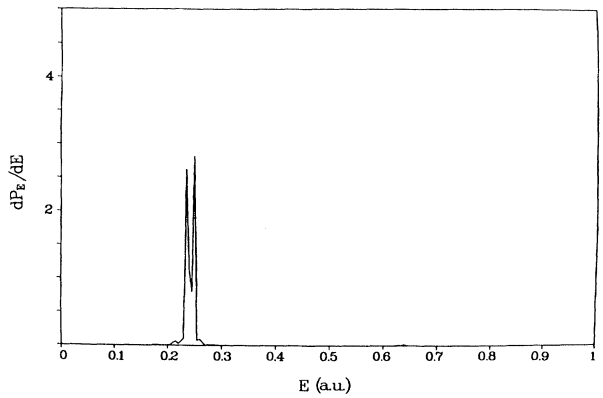

FIG. 2. dP_E/dE vs E; sum of all partial waves; conditions as in Fig. 1.

$=0.250$ a.u., in the even partial waves, where ε_{1s} is the ground-state energy. But, because of the finite mesh size employed in these calculations, there is an offset in the positive-energy spectrum of -0.010 a.u. The expected position of the threshold peak is therefore at $E=0.240$ a.u. In this example, the ponderomotive energy is very small [$I_0/(4\omega^2)\sim 5\times10^{-4}$ a.u.].

In Fig. 2, we see two peaks, of approximately equal magnitude, at energies $E_-=0.235$ a.u. and $E_+=0.249$ a.u. The average of these two values, 0.242 a.u. is, to within the calculational accuracy, at the expected position of the threshold peak. The difference of E_+ and E_- is equal to Ω_R, again to within the calculational accuracy. The first ATI peak is similarly split. See Fig. 3, where we plot dP_E/dE for the $l=1$ partial wave. Note that these splittings are much larger than the peak ponderomotive energy (by a factor of ~ 25).

The splitting of the threshold peak is not difficult to understand. We refer to an argument developed earlier to explain features occurring in strong resonant scattering of radiation by atomic hydrogen [12]. Consider the form of the wave function evolved from the $1s$ state, under conditions of $1s-2p$ resonant drive. Before significant ionization has occurred, this wave function is given, to a good approximation, by

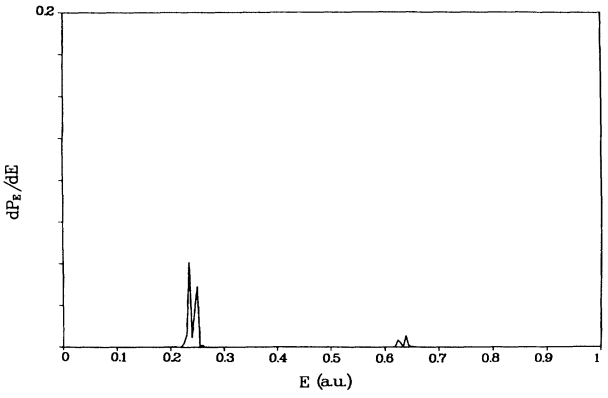

FIG. 3. Same as Fig. 2, but for the $l=1$ partial wave only.

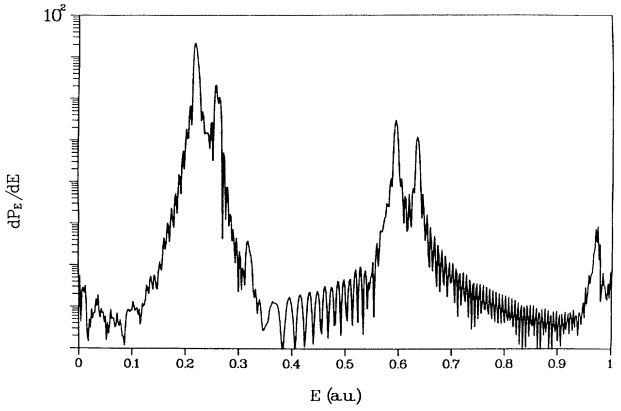

FIG. 4. dP_E/dE vs E for $I_0=10^{14}$ W/cm^2 and $\omega=0.375$ a.u. The sum of all partial waves appears.

$$\Psi(\mathbf{r},t)\sim\phi_{1s}(\mathbf{r})e^{-i\varepsilon_{1s}t}\cos(\Omega_R t/2) +i\phi_{2p}(\mathbf{r})e^{-i\varepsilon_{2p}t}\sin(\Omega_R t/2)\ , \tag{7}$$

ignoring the turn-on transient. Transitions from this state into the continuum are mediated by dipole couplings between the $2p$ component and the energy-conserving continuum state.

Energy conservation demands that these (one-photon) transitions occur at energies $E_\pm=\varepsilon_{2p}+\omega-I_0/(4\omega^2)\pm\Omega_R/2=\varepsilon_{1s}+2\omega-I_0/(4\omega^2)\pm\Omega_R/2$. From the form of the wave function in Eq. (7), the transitions to E_+ and E_- are expected to occur with equal probability. This accords with the results displayed in Fig. 2, which is a manifestation of the Autler-Townes effect [13].

The splitting of the first ATI peak is explained in an analogous fashion. (We note that, if the field has been turned on abruptly, then additional structures would have appeared in the spectrum [4]. The turn-on time of 100 a.u. was selected in our calculations to eliminate the effects of this artifact. Increasing the duration of the turn-on time beyond 100 a.u. does not affect our results.)

The behavior of the ATI spectrum at higher irradiance is not so easily interpreted, even on resonance. In Fig. 4, we plot values analogous to those of Fig. 2, again at

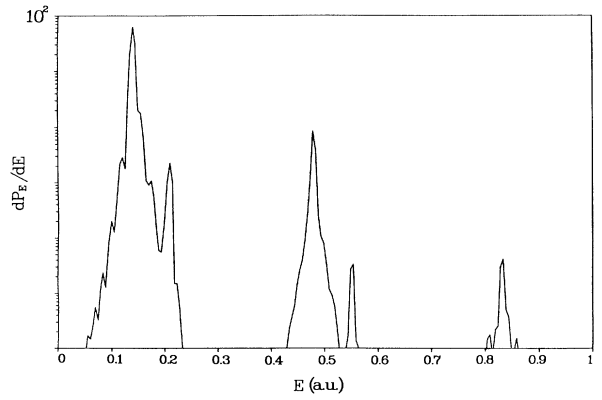

FIG. 5. Same as Fig. 4, but for $\omega=0.331$ a.u.

$\omega=0.375$ a.u., but for $I_0=10^{14}$ W/cm^2. The Rabi frequency is $\Omega_R=0.040$ a.u., still equal to the difference of E_+ and E_-. The average of E_+ and E_- is again equal to the expected threshold energy. (The observed splitting is only ~ 10 times the ponderomotive energy.) An asymmetry between E_+ and E_- is now apparent. This is likely due to a degradation of the Rabi coupled state, with mixing in of higher excited states, as well as the continuum. At this point, we do not understand why the higher-energy component of the doublet is the weaker; however, see the following.

Off resonance, the ATI spectra are even more asymmetric. In Fig. 5, we plot values of dP_E/dE versus E for $\omega=0.331$ a.u. and $I_0=10^{14}$ W/cm^2. Splitting of the threshold and ATI peaks still occurs, but now there is extreme asymmetry, with E_+ appearing at a much lower intensity than E_- (by a factor of ~ 500). The average of E_+ and E_- is now rather far from the expected value of the threshold energy. In fact, $E_-\sim E_{\rm th}$ for this case.

The generalization of Eq. (7) for nonzero detuning is

$$\Psi(\mathbf{r},t)\sim\phi_{1s}(\mathbf{r})e^{-i(\varepsilon_{1s}-\Delta/2)t} \times[\cos(\Omega t'/2)-i(\Delta/\Omega')\sin(\Omega' t/2)] +i(\Omega_R/\Omega')\phi_{2p}(\mathbf{r})e^{-i(\varepsilon_{2p}+\Delta/2)t} \times\sin(\Omega' t/2)\ , \tag{8}$$

where the detuning is $\Delta=\omega-(\varepsilon_{2p}-\varepsilon_{1s})$ and the shifted Rabi frequency is $\Omega'=(\Delta^2+\Omega_R^2)^{1/2}$. Equation (8) implies threshold peaks in the emitted-electron spectrum at

$$E_\pm=\varepsilon_{2p}+\omega-I_0/(4\omega^2)+(\Delta\pm\Omega')/2\ .$$

For the parameters chosen here, this last formula yields $E_+=0.21$ a.u. and $E_-=0.15$ a.u., quite close to the values displayed in Fig. 5. However, Eq. (8) also implies that these peaks should appear with equal intensity.

In fact, the peaks in Fig. 5 could be "explained" in a somewhat simpler fashion. The stronger peak might be ascribed to direct (nonresonant) ionization from the ground state, implying a continuum energy $E=\varepsilon_{1s}+2\omega-I_0/(4\omega^2)=0.15$ a.u. $\approx E_-$. The weaker peak can be thought of as arising from one-photon ionization from the (resonant) $2p$ state, at an energy $E=\varepsilon_{2p}+\omega-I_0/(4\omega^2)=0.21$ a.u. $\approx E_+$. Since the first (excitation) step of the second process is actually quite far off resonance, it might not seem too surprising that its rate is relatively low. Other calculations of photoionization rates for atomic hydrogen, at $I_0=10^{14}$ W/cm^2, have shown that the resonant process disappears into the background of direct two-photon ionization events for such high irradiances [1]. Consequently, our explanation is plausible. However, see Ref. [3], where this subject is considered in more detail.

We have recorded additional spectra at $\omega=0.331$ a.u. and $I_0=10^{13}$ W/cm^2. These show an asymmetry that is less than that seen at 10^{14} W/cm^2, and so are in accord with the preceding argument.

SUMMARY

We have described calculations of the REMPI energy spectra of electrons emitted from ground-state atomic hydrogen, during two-photon ionization, in the $1s$-$2p$ one-photon-resonant regime. The (two-dimensional) time-dependent Schrödinger equation was solved numerically for a simulated laser pulse of duration 50 fsec. The laser was linearly polarized, with irradiances between 10^{13} and 10^{14} W/cm^2.

Structure in the electron spectra was traced to Rabi coupling between the $1s$ and $2p$ hydrogenic states. The positions of the observed peaks was well understood, in every case, on the basis of simple arguments. The relative intensity of these peaks was understood, generally, at a more qualitative level. Previous work in this area [9,14] discovered identical structures, but by means of analytical methods appropriate to much lower irradiances. The present work extends slightly our understanding of structure in ATI spectra into the domain of moderately high irradiances.

[1] K. LaGattuta (unpublished).

[2] P. Agostini, F. Fabre, G. Mainfray, G. Petite, and N. Rahman, Phys. Rev. Lett. **42**, 1127 (1979).

[3] M. Dorr, D. Feldmann, R. Potvliege, H. Rottke, R. Shakeshaft, K. Welge, and B. Wolff-Rottke, J. Phys. B **25**, L275 (1992).

[4] J. Eberly, J. Javanainen, and K. Rzazewski, Phys. Rep. **204**, 331 (1991).

[5] R. Freeman, P. Bucksbaum, H. Milchberg, S. Darack, D. Schumacher, and M. Guesic, Phys. Rev. Lett. **59**, 1092 (1987).

[6] J. Eberly and J. Javanainen, Phys. Rev. Lett. **60**, 1346 (1988).

[7] M. Dorr *et al.*, J. Phys. B **25**, L275 (1992).

[8] M. de Boer and H. Muller, Phys. Rev. Lett. **68**, 2747 (1992).

[9] G. Gibson, R. Freeman, and T. McIlrath, Phys. Rev. Lett. **69**, 1094 (1992).

[10] F. Faisal and J. Moloney, J. Phys. B **4**, 3603 (1981).

[11] K. LaGattuta, Phys. Rev. A **43**, 5157 (1991).

[12] K. LaGattuta, J. Mod. Opt. **39**, 1181 (1992).

[13] S. Autler and C. Townes, Phys. Rev. **100**, 703 (1955).

[14] F. Faisal, *Theory of Multiphoton Processes* (Plenum, New York, 1987), pp. 300–305 and references therein.