Ionization and Recombination in Intense, Short Electric Field Pulses

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We investigate ionization and excitation of H(1s) in the limit of very short electric field pulses, analytically and numerically and both in the limit of small and extremely large peak electric fields. We identify a process of recombination akin to Rabi flopping from the continuum and give an analytic expression for this process after a single-cycle strong-field pulse.

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The development of intense, very short light pulses proceeds apace. Already, advanced laser facilities can achieve pulses with peak electric field $F_0 \sim 1$ a.u. (1 a.u. $= 5.1423 \times 10^9$ V/cm) and pulse lengths $\tau \sim$ 10 fs (1 fs $= 41.341$ a.u.) corresponding to $\sim$1–2 cycles of the electric field at a photon energy $\omega \sim 0.05$ a.u. [1]. On the other hand, high frequency pulses, at the moment of the order of 100 cycles, are produced in DESY from a free electron laser source with frequency around half an atomic unit, intensities 7×10^{13} W/cm^2 (peak electric field $F_0 \sim 4.4 \times 10^{-2}$ a.u.) and total pulse duration of 50 fs [2], as well as from high-harmonic generation with frequency $\omega = 3.3$ a.u. and total duration of 0.65 fs [3]. There is no doubt that much higher peak fields and much shorter pulse lengths with fewer cycles will be produced in the future.

When pulse lengths are so short that only a half or one cycle of the field occurs, the characteristics of ionization and excitation are very different from the very long pulse or continuous wave regime. In the context of ionization from an atomic orbit, "short" implies a pulse length shorter than the characteristic classical orbit time. Almost all treatments, except Refs. [4,5], considering very strong fields ($>$1 a.u.) have been wholly numerical, involving pulses with many cycles and with photon energies comparable to or greater than 0.5 a.u., the ionization energy of hydrogen. Much attention has concentrated on the phenomenon of stabilization as the peak field strength F_0 increases [6]. There are a large number of papers concerning ionization and recombination from Rydberg states (see, for example [7]). Here, we consider not only total ionization probability but also energy and angular distributions for ionization from the *ground state* of the hydrogen atom in half- or one-cycle pulses over a span of field strengths: from the weak, perturbative regime to the very strong, "short pulse approximation" limit, also called impulse or first Magnus approximation [8,9]. The principal results of this work are as follows:

(i) To present perturbation theory analytic results for a sequence of alternating square or sinusoidal pulses and to compare the respective ionization probabilities differential in the electron energy. We also show that ionization and recombination repeat periodically in perturbation theory.

(ii) To show that for a one-cycle pulse, in the strong-field case there is a range of field strengths where ionization caused by the first half-cycle is reversed by the second, i.e., recombination occurs, essentially Rabi flopping involving the continuum. Note that for initial Rydberg states recombination occurs into a large number of neighboring states. We predict that for the initial ground state, recombination occurs predominantly to the ground state, so that one can speak of Rabi flopping.

(iii) To present an analytic formula for the probability of returning to the ground state (recombination) following a second half-cycle, where the first half-cycle causes ionization.

To obtain analytic results we restrict ourselves to the hydrogen atom. Then the high ionization energy implies pulse duration at the moment unachievably short. However, when scaled to other atoms the relevant pulse lengths may soon be available. Atomic units are used throughout.

The probability amplitude for occupation of a continuum state $\Phi_{\mathbf{k}}$ with linear momentum $\mathbf{k}$ at time t after a classical laser field is switched on at a time t_0 is given by

$$a(\mathbf{k}) = \langle \Phi_{\mathbf{k}} | U(t, t_0) | \Phi_i \rangle, \tag{1}$$

where $\Phi_i(t_0)$ is the initial atomic state and $U(t, t_0)$ is the full time-development operator defined by

$$H(t)U(t, t') = i\partial U/\partial t, \tag{2}$$

where in dipole approximation,

$$H(t) = H_0 + \mathbf{r} \cdot \mathbf{F}(t) \equiv H_0 + V(t). \tag{3}$$

Here H_0 is the atomic Hamiltonian and $\mathbf{F}(t)$ describes the linearly polarized electric field. In Eq. (1) we will take $t_0 = 0$ and consider a pulse of finite duration τ, so that for $t > \tau$ the state $\Phi_{\mathbf{k}}$ is a continuum eigenstate of H_0.

The propagator $U(t, t')$ satisfies the equation

$$U(t, t') = U_0(t, t') - i \int_{t'}^{t} U_0(t, t'')V(t'')U(t'', t')dt'', \tag{4}$$

where $U_0(t, t') = \exp[-iH_0(t - t')]$. We will consider two approximations. First, simple first order perturbation theory in which $U(t'', t')$ in (4) is replaced by U_0 to give, from (1)

$$a(\mathbf{k}) = -i \int_0^\tau \langle \Phi_\mathbf{k} | \mathbf{r} \cdot \mathbf{F}(t) | \Phi_i \rangle \exp[i(E - E_i)t] dt \tag{5}$$

with $E = \frac{1}{2}k^2$. Second, appropriate for short pulses, we use the first order Magnus approximation (FMA) [10] to the exact solution (1), i.e.,

$$a(\mathbf{k}) \approx \langle \Phi_\mathbf{k} | \exp(-i\mathbf{q}\cdot\mathbf{r}) | \Phi_i \rangle \tag{6}$$

exact to order τ^3 with

$$\mathbf{q} = \int_0^\tau \mathbf{F}(t) dt \tag{7}$$

appearing as a momentum boost $\mathbf{q}$ provided by the laser field (for peak strength $\mathbf{F}_0$, $\mathbf{q} = \mathbf{F}_0\tau$ for rectangular and $\mathbf{q} = 2\mathbf{F}_0\tau/\pi$ for sine half-cycle pulses). Note that the amplitude (6) is independent of the pulse shape and therefore all pulses are equivalent to a delta-function pulse $\mathbf{F}(t) = \mathbf{q}\delta(t - t_1)$, $t_1 \in (0, \tau)$. The two approximations (5) and (6) are chosen because the conditions of validity [for (5) a weak field of arbitrary duration, for (6) a short pulse of arbitrary strength] are complementary. More importantly, however, and perhaps noteworthy in the field of laser-atom interactions, both approximations admit closed-form analytic solutions without any of the additional approximations (e.g. reduced dimensionality, cutoff potential) which are often made in this field. Similarly, our fully numerical results, based on the discrete variable representation (DVR) [11], are calculated without approximation. It is also interesting that the two approximations have a common region of validity, namely, when the field strength is small and the pulse is short, i.e., $q \ll 1$. Then (6) can be approximated by

$$a(\mathbf{k}) \approx -i\mathbf{q} \cdot \langle \Phi_\mathbf{k} | \mathbf{r} | \Phi_i \rangle. \tag{8}$$

Similarly, when $(E - E_i)\tau \ll 1$, Eq. (5) may be approximated as Eq. (8), i.e., perturbation theory and FMA give identical results. Equation (8), rather then Eq. (6), is called the sudden approximation in Ref. [12] and we will also adopt this notation and reserve the term FMA for the short pulse approximation. Since the transition matrix element is dipole, in both cases, when the initial state is of S symmetry, the final continuum angular distribution will be that of a pure P state. Therefore only when $q \ll 1$ will a unidirectional pulse produce forward-backward symmetrical electron energy distributions.

We consider first of all the perturbation region and the effect of a finite sequence of alternating pulses. For sinusoidal pulses $F(t) = F_0 \sin(\pi t/\tau)$ and $0 < t < n\tau$, of peak strength $\pm F_0$ and duration τ, one can calculate analytically the ionization probability dP/dE from the ground state, integrated over all angles of emission, i.e.,

$$\frac{dP}{dE} = 4\frac{(F_0\pi)^2}{\tau^2} |\langle \Phi_\mathbf{k} | \mathbf{r} | \Phi_1 \rangle|^2 \frac{\sin^2\{[n\tau(E - E_1) - n\pi]/2\}}{[(E - E_1)^2 - (\pi/\tau)^2]^2}, \tag{9}$$

where $E_1 = -0.5$ is the ground-state energy. We have derived a corresponding expression for n alternating rectangular pulses. The energy distributions resulting from 5 full-cycle pulses are shown in Fig. 1. For $\tau = 0.3$ the energy distributions peak strongly at zero energy. In the region near $E = 0$ the energy distribution is independent of pulse shape and shows a broad peak near $E = \pi/\tau = \omega_0$ (the arrow on Fig. 1), corresponding to the frequency of oscillation of the pulses. For longer τ's this peak grows and dominates the emission of zero-energy electrons. Of course this is just the onset of the infinite-pulse behavior, where this resonance peak becomes a δ function. The higher-energy distribution shows peaks at $E_1 + \omega_0 + \omega_0(1 + 2j)/n$, $j \geq 1$. For sinusoidal pulses these peaks decrease smoothly as a function of energy whereas for square pulses they are folded with the Fourier transform of a single half-cycle.

In Fig. 2 the effect of only one, two, or three short alternating pulses is shown, here the numerical DVR calculation and the perturbation theory calculation give identical results. We show the angle-integrated energy distribution for small energies (compared to the scale of Fig. 1). After one-pulse the ionization probability peaks at zero energy with a magnitude of $\sim 10^{-4}$. However, after a second pulse with opposite direction of the electric field almost all ionized electrons have recombined. Continuing the sequence to a third pulse restores the probability almost to the one-pulse value. This feature of perturbation theory with short pulses has not been emphasized to our knowledge, since mostly one concentrates on the long-time Fermi golden rule result. The small residual ioniza-

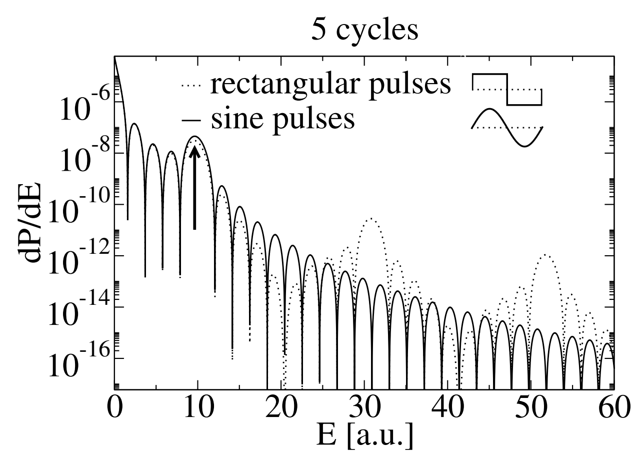

FIG. 1. Energy distribution for $q = 9 \times 10^{-3}$, $\tau = 0.3$. The arrow marks the resonant peak.

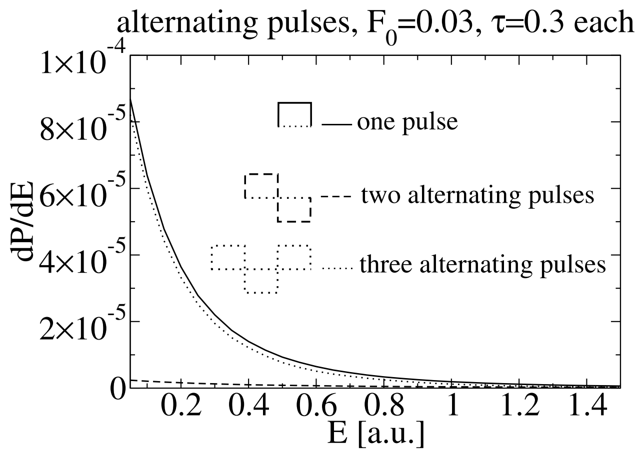

FIG. 2. Angle-integrated energy from the DVR calculation for $F_0 = 0.03$ and $\tau = 0.3$.

tion after two pulses is just due to the high momentum components of the ionized wave packet which leave the interaction region quickly enough to avoid recombination. A further interesting effect in the perturbation regime, as in the strong-field regime [13], is the enormous dependence on the phase of the electric field. If we consider a one-cycle pulse with a $\sin^2$ envelope, i.e., $F(t) = F_0 \sin^2(\pi t/n\tau)\sin(\pi t/\tau + \phi)$ where ϕ is the relative phase of envelope and carrier, one can show that a phase change of $\pi/2$ brings more than 2 orders of magnitude change in the ionization probability near threshold.

Now we switch attention to the strong-field short pulse case, i.e., consider $F_0 \to \infty$ for τ fixed. Up to $q \approx 0.1$ we can use the sudden approximation Eq. (8) to the FMA form of Eq. (6). As has been emphasized in studies of ionization from Rydberg states [8], there is a close connection with fast charged-particle collisions where the first Born approximation also leads to the momentum boost matrix element (6). In that case $\mathbf{q}$ is the momentum transferred from the charged-particle to the atomic electron. For the hydrogen atom these matrix elements are known in closed form [14]. The sudden approximation (8) then corresponds, for collisions, to the dipole limit, where the momentum transfer is vanishingly small.

First of all we have considered a single half-cycle pulse and examined the breakdown of the sudden approximation by comparing dP/dE in both sudden and Magnus approximations. For $q = 0.09$ the two agree perfectly and predict a maximum for $k = 0$ followed by a monotonic decrease for larger k. As q increases beyond ~ 0.1 the sudden approximation breaks down and the FMA result begins to acquire a peak near $k = q$. Here the dipole approximation fails completely and this manifests itself in the angular distribution which is of P symmetry in the dipole approximation but strongly asymmetrically peaked in the field direction for large q. As shown in Eqs. (6) and (7) the FMA is independent of pulse shape and thus the pulse can always be treated as a δ-function pulse with $\mathbf{q} = \int_0^\tau \mathbf{F}(t)dt$. That the FMA is an excellent approximation is illustrated in Fig. 3 for a single pulse with $\tau = 0.3$ and varying q. In Fig. 3 we show both the ionization probability $P_{\rm ion}$ (i.e., dP/dE integrated over E) and the occupation probability P_1 of the initial state. In both cases the FMA of Eq. (6), giving a closed-form analytic result, is in perfect agreement with separate DVR calculations using both sine and rectangular pulses. In Fig. 3 the depletion of the ground state almost mirrors the onset of complete ionization indicating, as we have verified, very small population of excited states.

The above results can be viewed as those for a half-cycle sine pulse. Then one can pose the question of what happens for a full-cycle pulse. In the strict FMA [Eq. (6)] the amplitude of ionization after a full cycle is identically zero, as is also the case in the approximation of Ref. [5]. However, here we modify the FMA by considering a full cycle to be equivalent to two δ pulses separated by a half-cycle time τ. Then, between the two pulses the electron propagates in the nuclear Coulomb field, leading to a nonzero final transition amplitude of the form

$$a_{if} = \langle \Phi_{\mathbf{k}} | \exp(i\mathbf{q}\cdot\mathbf{r}) \exp(-iH_0\tau) \exp(-i\mathbf{q}\cdot\mathbf{r}) | \Phi_i \rangle \tag{10}$$

with an error term $\sim \tau^2 q$. Since for large q (see Fig. 3), the electron occupies the continuum after the first half cycle, we can approximate the result of two pulses by considering only continuum states as intermediate states. Then the probability of population of the ground state after the second δ pulse is

$$P_1^c = \left| \int \langle |\Phi_{\mathbf{k}}| \exp(i\mathbf{q}\mathbf{r}) |\Phi_1\rangle|^2 \exp\left(-i\frac{k^2}{2}\tau\right) d^3\mathbf{k} \right|^2. \tag{11}$$

By considering the analytical form of the boost matrix

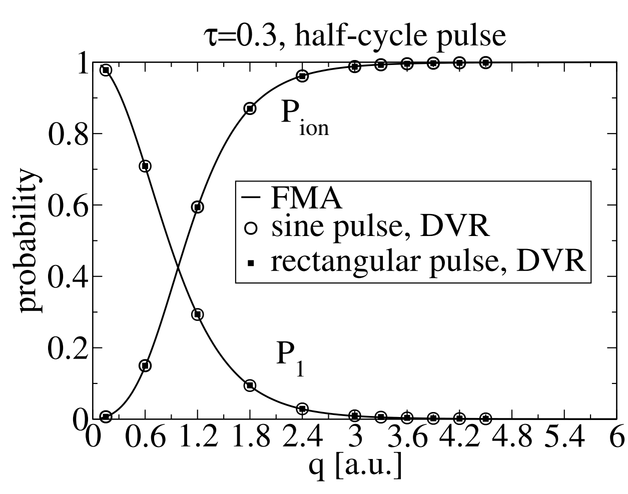

FIG. 3. Probabilities $P_{\rm ion}$ and P_1 for a half-cycle pulse in the FMA, and from the DVR calculation for both sinusoidal and rectangular half-cycle pulses.

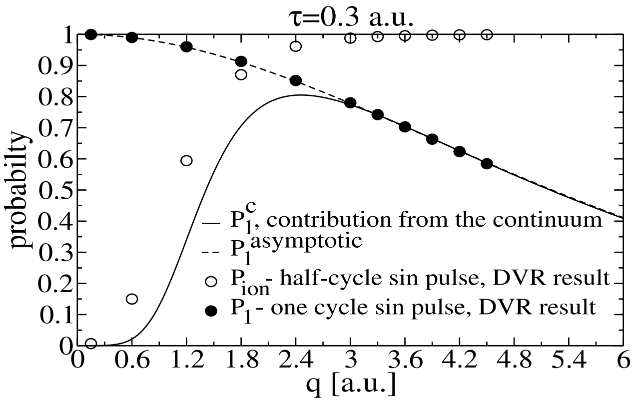

FIG. 4. Comparison between the results obtained by analysis of two δ pulses, and numerical results for half-cycle and one-cycle sine pulses.

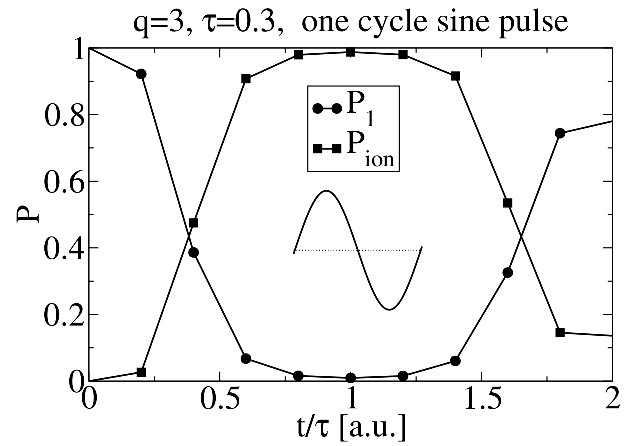

FIG. 5. The time evolution of the probabilities of the population of the ground state (P_1) and the ionization probability ($P_{\rm ion}$) during one-cycle sine pulse for $F_0 = 5\pi$, $\tau = 0.3$.

element in (11) one can show that for asymptotically large q, the ground-state occupation probability assumes the simple analytic form

$$P_1^{\rm asymptotic} = \exp(-2q\tau)\left[1+q\tau+\frac{(q\tau)^2}{3}\right]^2. \qquad (12)$$

The results for a one-cycle sine pulse ($\tau = 0.3$) from the DVR calculation and from the FMA Eq. (11) are shown in Fig. 4. The full circles are the probability P_1 of remaining in the ground state. We show the half-cycle $P_{\rm ion}$ from Fig. 3 (open circles) showing that for $q \geq 2.4$, at half-cycle the electron is in the continuum. Hence, for $q \geq 2.4$, the ground state is first fully depleted and then repopulated on the second half of the pulse. The full curve is precisely the probability for this process calculated according to Eq. (11). Also shown is the asymptotic approximation for P_1 from Eq. (12). For $q \geq 3$ it agrees with P_1 from DVR and FMA. Surprisingly, however, it agrees with the DVR calculation for all q (i.e., all F_0), up to $q = 4.5$, where DVR breaks down. For $q < 3$ this can be viewed as a happy accident arising from the fact that P_1 of Eq. (12) has the correct $q = 0$ value of unity. Nevertheless, even when viewed as only asymptotically justified but empirically everywhere correct, the formula (12) is a remarkably simple result for the population of the ground state after a single-cycle short laser pulse.

Finally, that ionization and recombination really do occur in the two halves of the pulse is shown in Fig. 5 where we plot $P_1(t)$ and $P_{\rm ion}(t)$ for $q = 3$ (the population of excited states never exceeds 10%). On the first half of the pulse there is almost 100% ionization followed by $\sim$80% recombination into the ground state. Note that this process is entirely to be distinguished from "stabilization" of the ground state. Rather, over a small range of q one has a process more akin to "Rabi flopping" between continuum and the ground state. We have compared the analytic formula (12) to numerically converged calculations for a variety of pulse lengths $0 \leq \tau \leq 1$ and found in all cases that the analytic result is in excellent agreement with the numerical one.

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[1] T. Brabec and F. Krausz, Rev. Mod. Phys. **72**, 545 (2000); G. G. Paulus *et al.*, Nature (London) **414**, 182 (2001).
[2] V. Ayvazyan *et al.*, Phys. Rev. Lett. **88**, 104802 (2002).
[3] M. Hentschel *et al.*, Nature (London) **414**, 509 (2001).
[4] L. V. Keldysh, Zh. Eksp. Teor. Fiz. **47**, 1945 (1964) [Sov. Phys. JETP **20**, 1307 (1965)].
[5] G. Duchateau, E. Cormier, and R. Gayet, Eur. Phys. J. D **11**, 191 (2000); Phys. Rev. A **66**, 023412 (2002).
[6] M. Dondera, H. G. Muller, and M. Gavrila, Phys. Rev. A, **65**, 031405(R) (2002).
[7] D. G. Arbó, C. O. Reinhold, and J. Burgdörfer, Phys. Rev. A **69**, 023409 (2004); J. G. Zeibel and R. R. Jones, Phys. Rev. A **68**, 023410 (2003); A. Wetzels *et al.*, Phys. Rev. Lett. **89**, 273003 (2002); C. Wesdorp, F. Robicheaux, and L. D. Noordam, Phys. Rev. Lett. **87**, 083001 (2001).
[8] C. O. Reinhold, M. Melles, H. Shao, and J. Burgdörfer, J. Phys. B **26**, L659 (1993).
[9] P. Krstic and Y. Hahn, Phys. Rev. A **50**, 4629 (1994); A. Bugacov, B. Piraux, M. Pont, and R. Shakeshaft, Phys. Rev. A **51**, 1490 (1995).
[10] W. Magnus, Commun. Pure Appl. Math. **7**, 649 (1954); P. Pechukas and J. C. Light, J. Chem. Phys. **44**, 3897 (1966).
[11] D. Dimitrovski, T. P. Grozdanov, E. A. Solov'ev, and J. S. Briggs, J. Phys. B **36**, 1351 (2003).
[12] L. I. Schiff, *Quantum Mechanics* (McGraw-Hill, New York, 1968), 3rd ed., p. 292.
[13] D. B. Milošević, G. G. Paulus, and W. Becker, Laser Phys. Lett. **1**, 93 (2004).
[14] H. A. Bethe, Ann. Phys. (Leipzig) **5**, 325, (1930); A. R. Holt, J. Phys. B **2**, 1209 (1969); Dž Belkić, J. Phys. B **17**, 3629 (1984).