Stabilization of Atomic Hydrogen in Superintense, High-Frequency Laser Fields of Circular Polarization

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We present the first quantitative results for the ionization of hydrogen at relatively high frequencies, from low intensities to superintensities in excess of the atomic unit $I_0=3.51\times10^{16}$ W/cm^2. We find that at superintensities the atom has a tendency to stabilize against multiphoton ionization, even at low frequencies. We show, however, the existence of very short lifetimes for the ground state, at intensities just below those needed for stabilization.

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Physical quantities characterizing the interaction of atoms with purely monochromatic radiation may be derived from a Floquet analysis of the Schrödinger equation of the problem. By applying this analysis to the "space-translated" version of the equation, Gavrila and Kaminski have developed an iterative approximation scheme, valid at sufficiently high frequencies ω.[1] In the high-frequency *limit* the theory predicts stabilization of the atom against decay by multiphoton ionization (MPI), i.e., the existence of stationary, albeit distorted, atomic states.[2] At finite, though sufficiently large ω, the theory allows for MPI.[3] So far only the distortion of low-lying states of H has been studied accurately for linear polarization.[4] We have meanwhile applied the theory to the ionization of H and are presenting here the first (nonperturbative) results to be obtained at relatively high frequencies for the decay rates and lifetimes. Besides, this is the first calculation allowing us to follow continuously the evolution of the lifetime from low intensities to superintensities, which have now become available experimentally. (Our results are subject to relativity and retardation corrections, which may become sizable under these circumstances.) At lower frequencies, where a full Floquet analysis is needed, Potvliege and Shakeshaft have also calculated lifetimes,[5] but only up to several times 10^{13} W/cm^2.

The values of the lifetimes play an essential role in assessing the possibility of exposing the (neutral) atom to superintense fields (for a discussion of this issue, see Lambropoulos, Ref. 6). As these are produced in the form of very short, picosecond or subpicosecond, laser pulses, in order that the atom can survive to feel the peak intensity in the pulse, its lifetime should exceed the rise time of the latter at all intensities passed. Our results answer the question of the atomic survival in the realm of high frequencies. Note that we may still apply the stationary Floquet approach to this (nonstationary) problem, provided that the pulse rise time is not exceedingly short,[7] by allowing I to become a slowly varying function of time.

We shall be interested here in the case of circular polarization.[8] The corresponding classical vector potential can be taken as

$$\mathbf{A}=-a[\mathbf{e}_1\sin(\omega t)\pm\mathbf{e}_2\cos(\omega t)]\,, \tag{1}$$

where the unit vectors $\mathbf{e}_1,\mathbf{e}_2$ and the unit vector $\boldsymbol{\nu}$ of the direction of propagation are assumed to form a right-handed system. The upper (lower) sign in Eq. (1) stands for right (left) circular polarization.

The independent parameters appearing in a natural way in the high-frequency theory are the frequency ω and a characteristic length α_0, related to ω and the intensity I in the case of circular polarization by $\alpha_0=(I/2)^{1/2}\omega^{-2}$ a.u.[9,10] In fact, it was shown that in the high-frequency limit the energies E of the levels are eigenvalues of the Schrödinger equation

$$[\tfrac{1}{2}\mathbf{P}^2+V_0(\alpha_0,\mathbf{r})]\psi_0=E\psi_0\,, \tag{2}$$

where $V_0(\alpha_0,\mathbf{r})$ is the "dressed potential" corresponding to the original potential $V(\mathbf{r})$, and $\psi_0(\mathbf{r})$ is the corresponding normalized eigenfunction. Both E and ψ_0 depend only on α_0, and not (explicitly) on ω. The n-photon ionization amplitude of the state ψ_0 is given by

$$f_n=-(1/2\pi)\langle\psi_{\mathbf{k}_n}^{(-)}|V_n|\psi_0\rangle\quad(n\geq1)\,, \tag{3}$$

where $\psi_{\mathbf{k}_n}^{(-)}$ is the continuum solution of Eq. (2) behaving asymptotically like a plane wave of momentum $\mathbf{k}_n$ and amplitude 1, plus incoming spherical waves. Energy conservation requires that $k_n^2/2=E_0+n\omega$. The dressed potential V_0 entering Eq. (2) and the V_n $(n\geq1)$ appearing in Eq. (3) are given by

$$V_n(\alpha_0,\mathbf{r})=\frac{1}{2\pi}\int_0^{2\pi}e^{in\phi}V(\mathbf{r}+\alpha_0(\mathbf{e}_1\cos\phi\mp\mathbf{e}_2\sin\phi))d\phi\quad(n\geq0)\,. \tag{4}$$

The angular-dependent n-photon ionization rate was shown to be[2]

$$\frac{d\Gamma_n}{d\Omega}=k_n|f_n|^2\,. \tag{5}$$

It was argued that[1,2] the high-frequency interaction theory, limited to Eqs. (2) and (3), is a good approximation within the exact Floquet approach provided that

$$\omega \gg |E_0(\alpha_0)|\,, \tag{6}$$

where $|E_0(\alpha_0)|$ is the binding energy of the ground state of the *atom in the field,* and in which α_0 can take any finite value.

We note that Eq. (2), although not Eq. (3), was obtained earlier by other methods by Henneberger,[11] and by Gersten and Mittleman.[11]

In order to evaluate the transition amplitudes from Eq. (3) we have calculated *accurate eigenvalues and eigenfunctions for the ground state.*[12] The dressed Coulomb potential V_0 for circular polarization, obtained by inserting $V(r) = -1/r$ into Eq. (4), is in fact the electrostatic potential generated by spreading uniformly the proton charge on the circle of radius α_0, centered at the origin of the $(\mathbf{e}_1,\mathbf{e}_2)$ plane. V_0 is obviously axially symmetric around $\boldsymbol{\nu}$ and has even parity with respect to the origin. A graphical representation of V_0 was given in Ref. 13, Fig. 1.

The ground state of Eq. (2) has axial symmetry around $\boldsymbol{\nu}$ and even parity.[12,13] The solution of Eq. (2) thus reduces to a two-dimensional problem, in a plane passing through the axis of symmetry. We have solved it by applying accurate (finite-element and finite-difference) grid methods.

The results of this calculation are given in Table I as a function of α_0. We list the values of the binding energy $|E_0(\alpha_0)|$, and of the normalized eigenfunction $\psi_0(\alpha_0,\mathbf{r})$ on the circle of charges, denoted by $[\psi_0]_c$, which we shall need subsequently. As α_0 increases we are faced with a dramatic decrease of the binding energy, similar to that for the linearly polarized case.[4] This has a favorable influence on the possibility of having the validity condition Eq. (6) satisfied at large α_0 (intensity), because then smaller ω are allowed.

TABLE I. α_0 dependence of the ground-state energy of hydrogen $E_0(\alpha_0)$, and of the wave function on the "circle of charges" $[\psi_0]_c$ (all in a.u.).

α_0	$E_0(\alpha_0)$	$[\psi_0]_c$
0.0	−0.5000	0.564
0.2	−0.4813	0.422
0.3	−0.4651	0.366
0.5	−0.4303	0.284
1	−0.3538	0.172
2	−0.2582	0.0882
5	−0.1442	0.0312
10	−0.0847	0.0139
20	−0.04743	0.00644
50	−0.02142	0.00269
70	−0.01597	0.00196
100	−0.01170	0.00140
200	−0.006372	0.000715
500	−0.002831	0.000289

The limiting case of large α_0 was studied analytically exactly in great detail in Ref. 13. Our numerical eigenvalues at large α_0 (e.g., for $\alpha_0 = 100,500$) are in excellent agreement with the analytic formula presented there (Ref. 13, p. 5668). From Ref. 13 we can also derive the large-α_0 expression for $[\psi_0]_c$:

$$[\psi_0(\alpha_0)]_c \cong 0.147/\alpha_0\,. \tag{7}$$

This is in agreement with the numerical results of Table I for $\alpha_0 \geq 100$.

In the following we shall first give simple *analytic expressions for the n-photon decay rates.* As the MPI amplitudes are anyway valid only at high ω, i.e., when the energy of the final state $E_n = k_n^2/2$ is large with respect to E_0 [see Eq. (6)], it follows that we can apply the Born approximation to the final state, i.e., replace $\psi_{\mathbf{k}_n}^{(-)}$ by a plane wave. We thus obtain for the n-photon angular decay rates, to dominant order in $(1/\omega)$,

$$\frac{d\Gamma_n}{d\Omega} = 4k_n^{-3} J_n^2(\alpha_0 \mathbf{k}_n \sin\theta)[\psi_0]_c^2\,, \tag{8}$$

where $[\psi_0]_c$ is the value of the ground-state wave function on the circle of charges (see Table I), and the argument of the Bessel function J_n contains θ, the angle between $\mathbf{k}_n$ and $\boldsymbol{\nu}$. The angle integrated rates can be written as

$$\Gamma_n \cong \frac{8\pi}{\alpha_0 k_n^4}[\psi_0]_c^2 \int_0^{2\alpha_0 k_n} J_{2n}(\xi)\,d\xi\,. \tag{9}$$

We can give simple estimates for the Γ_n in some limiting cases. At sufficiently low I, specific for the application of conventional lowest-order perturbation theory (LOPT), we have $\alpha_0 \ll 1$ [see Eq. (2)]. This means that the radius α_0 of the ring of charges is small in comparison to the a.u., and hence $[\psi_0]_c$ can be replaced by the value of the unperturbed wave function at the origin, $\pi^{-1/2}$. Moreover, no matter how large ω or k_n, we also have $\alpha_0 k_n \ll 1$, and we can replace the Bessel function in Eq. (9) by the first term of its series expansion to obtain

$$\Gamma_n \cong \frac{16}{k_n^3}\frac{(\alpha_0 k_n)^{2n}}{(2n+1)!}\,. \tag{10}$$

This agrees with the high-frequency limit of LOPT ionization cross sections.[14] The Γ_n have the well-known I^n dependence, and differ by orders of magnitude from each other. Consequently, we have for the total rate Γ: $\Gamma \cong \Gamma_1$.

We now consider the case of superintense fields, i.e., the extreme nonperturbative limit $\alpha_0 \gg 1$ and $\alpha_0 k_n \gg 1$. [Note that then the basic high-frequency condition Eq. (6) is automatically fulfilled.] By replacing the upper limit of the integral in Eq. (9) by ∞, the integral reduces to 1. Further, by also taking into account Eq. (7), we get the dominant $1/\omega$ behavior

$$\Gamma_n \cong \frac{0.136}{\alpha_0^3 \omega^2}\frac{1}{n^2}\,. \tag{11}$$

In contrast to the LOPT result, all decay rates Γ_n are now of the same order of magnitude, and vary as $1/n^2$. Since the sum over $1/n^2$ yields $\zeta(2)=\pi^2/6$, the total decay rate becomes

$$\Gamma \cong 0.223/\alpha_0^3\omega^2 = 0.631\omega^4/I^{3/2}. \quad (12)$$

Note that the $n \geq 2$ contributions to Eq. (12) represent a sizable amount, nearly 40% of the total.

We shall now comment on the qualitative dependence of Γ on ω and I, as emerging from the limiting forms Eqs. (10)–(12). The $\Gamma \cong \Gamma_1$ of LOPT, Eq. (10), has the well-known $\omega^{-9/2}$ steep decrease with ω.[14] The Γ of the extreme nonperturbative limit Eq. (12) yields the anticipated result[2] that, at fixed α_0, Γ vanishes when ω increases. When expressed in terms of ω and I, however, Eq. (12) yields the surprising result that in this radiation regime *at fixed I the total rate* Γ *increases with* ω. [For any given I, by allowing ω to grow indefinitely, α_0 will decrease in the process and ends up being smaller than 1, so that the appropriate formula becomes then the LOPT Eq. (10).] Besides, *at fixed* ω, Γ *decreases with* I, in contrast to the LOPT result. (From the standpoint of the high-frequency theory there is no limitation of the increase of I, as long as α_0 stays finite.) Therefore, at sufficiently large I, i.e., in the regime of validity of Eq. (12), the atom has the tendency to stabilize against decay of MPI.

Our results for the lifetime, based on Eq. (9), are given in Fig. 1 as a function or I, at various ω. We have considered there some lower ω ($\omega=0.125$–0.5) for which at small α_0 or I the high-frequency condition Eq. (6) is violated (e.g., more than one photon is needed to ionize). The lifetime for these cases has to be obtained from other theories (e.g., LOPT or Ref. 5). Nevertheless, even for these ω, at sufficiently large α_0 or I, after one-photon ionization has become possible, Eq. (6) ends up being satisfied (see Table I), so that our theory becomes applicable. When this begins to occur, the lifetimes are quite short, in the femtosecond range. They then increase with I towards large values, in agreement with Eq. (12).

For the larger values of ω in Fig. 1 ($\omega=1$–8, for which no intense lasers are available at the present time) one-photon ionization is always possible, even though Eq. (6) may not be properly satisfied occasionally. For these ω we can follow the lifetime from small intensities to superintensities. As apparent, at small I the lifetime begins by decreasing with I [in agreement with the LOPT Eq. (10)], whereas at large I it increases [as predicted by Eq. (12)]. The large-I lifetimes are indeed remarkably long (picosecond to nanosecond). However, our $\omega \geq 1$ curves show a new occurrence: *The lifetime passes a minimum* for an I which increases with ω. The minimum lies in the intensity range where the transition from the LOPT Eq. (10) to the large-I Eq. (12) takes place. For the ω considered, its value is below 1 fs (it does increase slowly with ω).[15]

The Floquet lifetimes given in Fig. 1 were calculated

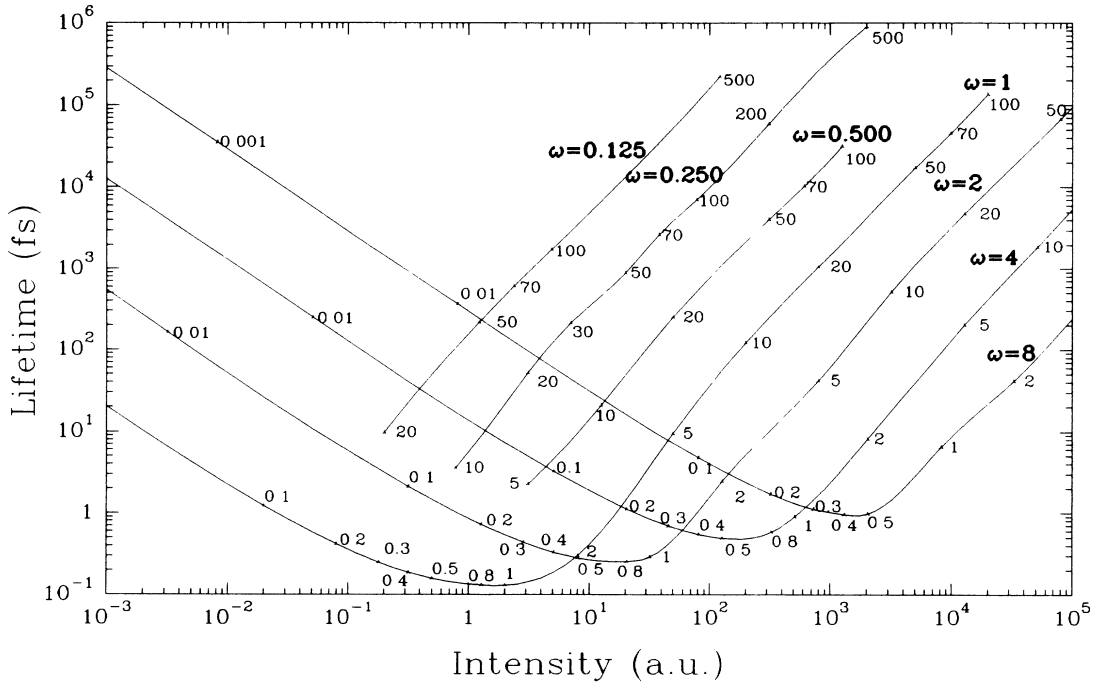

FIG. 1. Lifetime (in femtoseconds) of atomic hydrogen as a function of intensity (in a.u., $I_0=3.51\times10^{16}$ W/cm^2), for various ω (in a.u.). The numbers adjacent to the points give the corresponding value of α_0 (in a.u.). The curves for $\omega \leq 0.5$ have not been extended to lower intensities because our condition Eq. (6) is no longer satisfied.

for the ideal case in which the atom undergoes stationary decay in the field, ignoring the problem of how it was brought into this situation. This applies in particular to the regime of stabilization we have identified above. On the other hand, in actual experiments with present superintense laser pulses, the rise time is not shorter than 50 fs. When such a pulse is applied to the H atom, as the intensity increases gradually and enters the realm of Fig. 1, its lifetime will end up becoming shorter than this duration. The atom will therefore not survive up to the intensities needed to enter into the stabilization regime, at none of the frequencies we have considered: The dip in the lifetimes acts like a "death valley." True x-ray frequencies would be needed in order to make the atom in the ground state survive up to intensities in excess of 1 a.u. at the aforementioned pulse duration. The prospects of generating such superintense x rays have become palpable recently.[16] On the other hand, other schemes can be envisaged to expose the atom to superintensities (to be discussed elsewhere; see also Ref. 8).

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[7]The pulse rise time should be long in comparison to the light period, and also long enough so that no "sudden" transient effects can occur.

[8]The case of linear polarization will be discussed in detail elsewhere [see also M. Pont, Ph.D. thesis, University of Amsterdam, 1990 (unpublished)].

[9]We are using atomic units except as noted. We define the a.u. of averaged intensity, $I_0=3.51\times10^{16}$ W/cm^2, as that corresponding to the a.u. of electric-field strength (linear polarization).

[10]Note that the α_0 defined for circular polarization differs by $2^{1/2}$ from the one we have been using for linear polarization in Ref. 3 [see also W. C. Henneberger, Phys. Rev. Lett. **21**, 838 (1968); J. I. Gersten and M. H. Mittleman, J. Phys. B **9**, 2561 (1976)].

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[14]M. Gavrila and J. Z. Kaminski (unpublished). For $n=1$ compare with the Born approximation photoeffect cross section $d\sigma_1/d\Omega$ [e.g., see H. Bethe and E. Salpeter, *Quantum Mechanics of One- and Two-Electron Systems* (Springer-Verlag, Berlin, 1957), Eq. (70.6)], by taking into account that $d\Gamma_1/d\Omega=N\,d\sigma_1/d\Omega$, where N is the photon flux.

[15]In the case of $\omega=1$, the minimum lifetime (0.13 fs) falls below the light period (0.15 fs), which represents an extreme case of physical inadequacy of the Floquet approach.

[16]R. R. Freeman, "Multiphoton Effects in Soft X Rays and Laser Plasmas," in Proceedings of the International Conference on Multiphoton Processes, Paris, 1990 (to be published).