Adiabatic stabilization of excited states of H in an intense linearly polarized laser field

R. M. Potvliege and Philip H. G. Smith

Physics Department, University of Durham, Durham DH1 3LE, England

(Received 24 March 1993)

The lifetime of atomic hydrogen exposed to an intense "high-frequency" linearly polarized laser field has been calculated, in a Sturmian-Floquet approach, for the case where the atom is initially in an excited state with $m = 3$, 4, 5, or 6, $l=m$, $m+1$, or $m+2$, and $n \leq 8$, for several wavelengths between 455 and 1064 nm. In this range of parameters, the intensity at which the lifetime of the atom reaches a minimum value is given in good approximation by a simple "empirical" law. The lifetime is longer than that predicted by previous approximate Floquet calculations, and its minimum occurs at a smaller intensity. Results for higher-lying Rydberg states are also presented.

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

In general, an atom exposed to a weak laser field decays more and more rapidly as the intensity increases, with occasional rises and falls of the ionization rate that occur when the initial state is brought in resonance with other states. However, when exposed to an intense laser field, an atom may, for various reasons, become stable against ionization [1]. This Rapid Communication is concerned with "adiabatic stabilization," a remarkable nonperturbative effect whereby the rate of multiphoton ionization of an atom in a *constant* intense laser field is a decreasing function of the intensity. ("Diabatic stabilization," in contrast, is induced by the temporal variation of the intensity during the passage of a laser pulse.) Adiabatic stabilization has not been demonstrated experimentally so far, but several Floquet calculations [2–5] and time-dependent calculations [6, 7] have established that this effect should occur in intense high-frequency fields, for ionization from the ground state (dressed by the field) as well as for ionization from excited states; here, "high frequency" means that the photon energy is larger than the ionization potential of the initial unperturbed state. It is unlikely that adiabatic stabilization of the ground state of hydrogen or of the ground state of a rare gas could ever be observed in an experiment, since this would involve using superintense sub-fs vacuum ultraviolet (vuv) laser pulses. However, it has been suspected for a long time that the ionization rate of highly excited states, in particular of circular states, should remain small at any intensity and that such an atom could survive the rising edge of a realistic laser pulse until the onset of stabilization. This prediction was verified in a time-dependent calculation by Pont and Shakeshaft [6], whose results were confirmed by a Sturmian-Floquet calculation [4] and by the high-frequency Floquet calculation of Vos and Gavrila [5].

The Sturmian-Floquet method [8] consists of finding an appropriate "stationary" solution of the time-dependent Schrödinger equation, that is, a solution of the form $|\Psi(t)\rangle = \exp(-iEt/\hbar)|\mathcal{F}(t)\rangle$, where $|\mathcal{F}(t)\rangle$ is periodic in time with the same period as the laser field. The calculation is performed in the dipole approximation, in the $\mathbf{p} \cdot \mathbf{A}$ gauge, on a discrete basis of complex radial Sturmian functions and spherical harmonics; the method is formally equivalent to the complex dilatation method. The quasienergy E is complex: $E = E_i + \Delta - i\Gamma/2$. E_i is the eigenenergy of the unperturbed initial state, and Δ is the Stark shift. The total ionization rate is simply $\Gamma/\hbar$, and the half-life of the state is $\hbar \ln 2/\Gamma$. To the extent that the atom remains in a single Floquet state as the intensity increases adiabatically from 0 to the peak intensity of the pulse, the ionization yield obtained in this way is numerically exact (for atomic hydrogen). A "stationary" wave function is also sought in the high-frequency Floquet method adopted by Vos and Gavrila, but the calculations are performed in the Kramers-Henneberger frame, some approximations being made in the coupling with the field and, what is a greater cause of concern, in the ionization amplitude (i.e., the Coulomb interaction is neglected in the final state, the outgoing photoelectron being represented by an undistorted plane wave) [5, 9].

We have used the Sturmian-Floquet method to recalculate the lifetime of the $7i$ ($m = 5$) state of hydrogen studied by Vos and Gavrila (the magnetic quantum number m is defined with respect to the polarization direction). The two sets of results are shown in Fig. 1. They are in qualitative agreement, although in our more accurate calculation the onset of stabilization occurs at lower intensity and the lifetime of the atom is longer—at 1064 nm wavelength, $\hbar/\Gamma$ is longer than 8 ps at any intensity [10, 11]. The Sturmian-Floquet results are supported by results of time-dependent calculations [12] and are in excellent agreement with those calculated independently by Buchleitner and Delande [13]. They can also be reproduced by summing to high order the Rayleigh-Schrödinger perturbation series, as will be described elsewhere.

Sturmian-Floquet results and first-order perturbative results are presented in Table I for various other initial states with nonzero magnetic quantum number and for a few frequencies such that $\hbar\omega/10 \lesssim |E_i| \lesssim \hbar\omega/2$. Note that the field couples the initial state only to states with the same value of m. Hence, the circular states (with $n = m+1$) do not interact with states belonging to lower Rydberg manifolds or to the same manifold, which simplifies their dynamics considerably. The rate of photoionization from Rydberg levels decreases with the electronic density in the vicinity of the nucleus when the angular momentum of the initial state increases—or to a lesser extent when its principal quantum number n ($n \gg l$)
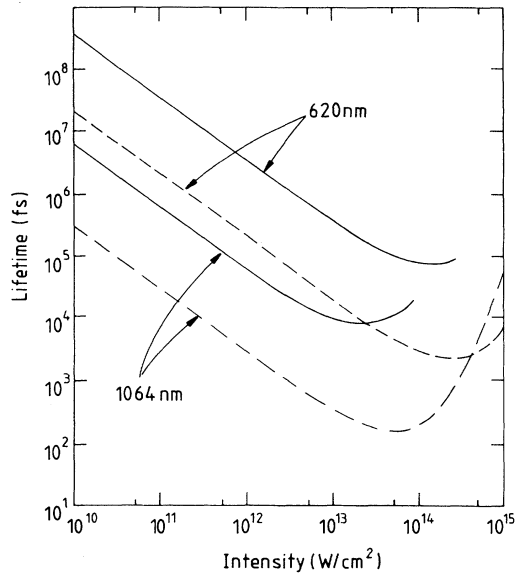
FIG. 1.   Lifetime $(\hbar/\Gamma)$ of a hydrogen atom that is initially in the state $n = 7$, $l = 6$, $m = 5$, vs the intensity of the incident laser field, for two different wavelengths. Solid curves, present Sturmian-Floquet results; dashed curves, high-frequency Floquet results [5].

increases; this accounts for the disparities between the lifetimes of the different states at low intensity. The induced width Γ of the states considered here is always much narrower than the distance to the dressed states belonging to the nearest manifold.

The main results presented in Table I are the intensities $I_{\min}$ at which the lifetime of the atom reaches its minimum value, and the corresponding minimum half-lives $\tau_{\min}$. Clearly $I_{\min}$ increases with the frequency, with the magnetic quantum number, and (for a given m) with the orbital angular momentum quantum number l. However, it only weakly depends on the principal quantum number. Comparing the results for the shortest wavelengths suggests a simple scaling law for $I_{\min}$: it appears that the minimum of the lifetime occurs at an intensity which is given in good approximation by

$$I_{\rm sc} = (\omega/\omega_0)^3 I_0\, m\, (l + 1 - m)! \tag{1a}$$

$$= \frac{\mu c \hbar}{8\pi} \frac{4\pi\epsilon_0}{e^2}\, \omega^3\, m\, (l + 1 - m)!, \tag{1b}$$

where ω is the angular frequency of the field, ω_0 is the atomic unit of angular frequency, $I_0 = 3.51 \times 10^{16}$ W/cm^2, and μ is the mass of the electron. Values of $I_{\rm sc}$ are given in Table I for the sake of comparison. The "empirical" scaling law is verified for 0.04 a.u. $\leq \omega \leq 0.1$ a.u., with $m = 4$, 5, or 6, $l = m$, $m+1$, or (with some disagree-

TABLE I.   Half-life $(\hbar \ln 2/\Gamma)$ of various excited states of atomic hydrogen, which have, initially, principal quantum number n, orbital angular momentum quantum number l, and magnetic quantum number m, for several values of the wavelength of the incident laser beam. $\tau_{\rm p}(a)$, half-life calculated in first-order perturbation theory at 1×10^{10} W/cm^2; $I_{\rm sc}$, intensity (in W/cm^2) where the lifetime is predicted to have a minimum by the empirical scaling law; $I_{\min}$, intensity (in W/cm^2) where the lifetime reaches a minimum value; $\tau_{\min}$, minimum of the half-life calculated in Sturmian-Floquet approach; $\tau_{\rm p}(b)$, half-life calculated in first-order perturbation theory at $I_{\min}$. The numbers in brackets represent powers of 10. The entries corresponding to the results shown in Fig. 1 are indicated by arrows.

$l-m$	m	n	$\tau_{\rm p}(a)$	$I_{\rm sc}$	$I_{\min}$	$\tau_{\min}$	$\tau_{\rm p}(b)$	$l-m$	m	n	$\tau_{\rm p}(a)$	$I_{\rm sc}$	$I_{\min}$	$\tau_{\min}$	$\tau_{\rm p}(b)$
Wavelength: 455.6 nm ($\omega = 0.1$ a.u.)								
0	4	5	6.94 ns	1.40[14]	1.54[14]	1.37 ps	451 fs	0	4	8	378 ps	2.21[13]	2.17[13]	527 fs	174 fs
0	6	7	5.38 μs	2.11[14]	2.55[14]	637 ps	211 ps	1	4	8	1.46 ns	4.42[13]	4.02[13]	981 fs	362 fs
								2	4	8	13.1 ns	1.33[14]	9.90[13]	3.95 ps	1.33 ps
Wavelength: 532 nm								0	5	6	2.52 ns	2.76[13]	2.62[13]	2.93 ps	964 fs
								0	5	7	2.31 ns	2.76[13]	2.69[13]	2.62 ps	856 fs
0	4	8	5.13 ns	8.81[13]	9.79[13]	1.60 ps	524 fs	0	5	8	2.67 ns	2.76[13]	2.75[13]	2.98 ps	971 fs
1	4	8	30.2 ns	1.76[14]	1.96[14]	4.31 ps	1.54 ps	1	5	7	24.7 ns	5.53[13]	5.20[13]	14.0 ps	4.76 ps
0	5	6	58.5 ns	1.10[14]	1.21[14]	14.6 ps	4.84 ps	1	5	8	18.1 ns	5.53[13]	5.16[13]	10.3 ps	3.51 ps
0	5	7	49.9 ns	1.10[14]	1.23[14]	12.3 ps	4.05 ps	2	5	8	378 ns	1.66[14]	1.52[14]	95.0 ps	24.9 ps
0	5	8	55.3 ns	1.10[14]	1.25[14]	13.5 ps	4.43 ps	0	6	8	33.6 ns	3.32[13]	3.40[13]	29.9 ps	9.88 ps
1	5	7	847 ns	2.20[14]	2.52[14]	102 ps	33.6 ps	1	6	8	527 ns	6.63[13]	6.35[13]	252 ps	82.9 ps
1	5	8	585 ns	2.20[14]	2.50[14]	70.4 ps	23.4 ps								
0	6	8	1.09 μs	1.32[14]	1.55[14]	211 ps	70.1 ps	Wavelength: 1064 nm ($\omega = 0.0428$ a.u.)								
1	6	8	27.0 μs	2.64[14]	3.08[14]	2.77 ns	876 ps								
								0	4	5	43.4 ps	1.10[13]	8.94[12]	146 fs	48.6 fs
Wavelength: 620 nm ($\omega = 0.0856$ a.u.)								0	4	8	109 ps	1.10[13]	1.01[13]	322 fs	108 fs
								1	4	8	342 ps	2.20[13]	1.80[13]	502 fs	190 fs
0	3	4	63.9 ps	4.18[13]	3.90[13]	48.0 fs	16.4 fs	2	4	8	2.47 ns	6.60[13]	4.01[13]	1.71 ps	617 fs
0	4	5	1.04 ns	5.57[13]	5.51[13]	571 fs	188 fs	0	5	6	553 ps	1.38[13]	1.20[13]	1.40 ps	459 fs
0	4	8	2.11 ns	5.57[13]	5.95[13]	1.09 ps	355 fs	0	5	7	528 ps	1.38[13]	1.25[13]	1.30 ps	423 fs
1	4	8	10.8 ns	1.12[14]	1.16[14]	2.58 ps	927 fs	0	5	8	626 ps	1.38[13]	1.28[13]	1.51 ps	490 fs
2	4	8	131 ns	3.34[14]	3.49[14]	12.9 ps	3.75 ps	→1	5	7	4.48 ns	2.75[13]	2.34[13]	5.59 ps	1.92 ps
0	5	6	20.2 ns	6.97[13]	7.30[13]	8.40 ps	2.77 ps	1	5	8	3.41 ns	2.75[13]	2.33[13]	4.24 ps	1.46 ps
0	5	7	17.6 ns	6.97[13]	7.46[13]	7.19 ps	2.36 ps	0	6	7	8.31 ns	1.65[13]	1.54[13]	16.3 ps	5.39 ps
0	5	8	19.7 ns	6.97[13]	7.57[13]	7.98 ps	2.61 ps	0	6	8	6.31 ns	1.65[13]	1.58[13]	12.2 ps	4.01 ps
→1	5	7	256 ns	1.39[14]	1.50[14]	51.5 ps	17.1 ps	1	6	8	78.6 ns	3.30[13]	2.87[13]	82.4 ps	27.4 ps
1	5	8	180 ns	1.39[14]	1.48[14]	36.3 ps	12.2 ps								
0	6	7	475 ns	8.36[13]	9.26[13]	155 ps	51.3 ps	Wavelength: 20 μm ($\omega = 0.00228$ a.u.)								
0	6	8	334 ns	8.36[13]	9.38[13]	108 ps	35.6 ps								
1	6	8	7.13 μs	1.67[14]	1.83[14]	1.22 ns	390 ps	0	19	20	37.1 ns	7.89[9]	5.02[9]	217 ns	74.0 ns
								0	19	22	5.59 ns	7.89[9]	5.17[9]	31.9 ns	10.8 ns
								1	19	21	124 ns	1.58[10]	6.41[9]	573 ns	193 ns

ment) $m+2$, and principal quantum numbers $n=l+1$, $l+2$, or $l+3$. We stress that it may not be reliable for states or wavelengths significantly outside these ranges. In particular, stabilization of states whose initial l value is greater than $m+1$ is often spoiled by interaction with other states as the intensity varies. Also, the scaling law breaks down when $\hbar\omega$ is too close to E_i. Nevertheless, Eq. (1) may remain a good guide for estimating the magnitude of $I_{\min}$ for higher-lying circular states, as is illustrated by the results for the states with $m=19$ that are given in the table [14].

The good quantitative agreement between I_{sc} and $I_{\min}$ might be somewhat coincidental. However, the increase of $I_{\min}$ with ω^3, m, and $l - m$ can be understood on physical grounds. The ω^3 dependence of $I_{\min}$ is in agreement with the results of previous theoretical investigations of the stabilization of states with $m = 0$. For example, it is known that the Floquet ionization rates of the low-lying s-states of H start to decrease when the laser intensity I increases past a value where the dimensionless parameter $\Xi \equiv (\hbar\omega/2P)^{1/2}$ is approximately 1 [3]; the ω^3 scaling law follows from the frequency dependence of the ponderomotive energy $P = e^2I/(2\mu\omega^2\epsilon_0 c)$. Equation (1) implies that the peak ionization rates of the excited states considered in this work occurs at an intensity where $\Xi \approx [m(l + 1 - m)!/2]^{-1/2}$, that is, at a larger intensity than that where $\Xi = 1$. It is not surprising that the frequency dependence of $I_{\min}$ is in ω^3, since in the high-frequency regime the characteristic dimensionless quantity that can be constructed from P (the intensity parameter of the problem) is the "Reiss parameter" $P/\hbar\omega \propto I/\omega^3$. According to the high-frequency theory, photoelectron emission at an angle θ_{ej} from the polarization direction is suppressed if $\Xi/2 \ll \cos\theta_{ej}$ [1, 9, 15]. The same conclusion was also reached by Pont and Shakeshaft [6, 16], who argued that in the high intensity high frequency limit adiabatic stabilization is primarily a kinematical effect originating from the reduction of the angular phase space into which the photoelectron can be ejected by a single elastic collision with the nucleus. The constraint $\cos\theta_{ej} \lesssim \Xi/2$ on the ejection angle correctly implies that $I_{\min}$ increases with m, since the intensity must be larger when $m \gg 0$ than when $m = 0$ for θ_{ej} to be more restricted by this constraint than it is, even in first-order perturbation theory, by the $\sin^{2m}\theta_{ej}$ factor in the differential cross section [16]. That $I_{\min}$ increases with $l - m$ is also explained by this argument. Indeed, the symmetry properties of the spherical harmonics allow the photoelectron to be ejected with $\theta_{ej} \approx \pi/2$ when $l = m + 1$, but not when $l = m$; in consequence, the restriction on θ_{ej} becomes severe (and photoionization is quenched) at a larger value of Ξ, i.e., at a lower intensity, when $l = m$. The difference between the two cases diminishes at large values of m, the angular distribution being then strongly peaked about $\pi/2$ even for $l = m$. The photoelectron cannot be ejected with $\theta_{ej} \approx \pi/2$ when $l = m + 2$, either; however, such states are increasingly mixed with (much broader) states with $l = m$, as the intensity increases, so preventing $I_{\min}$ from being as small as it would be without mixing.
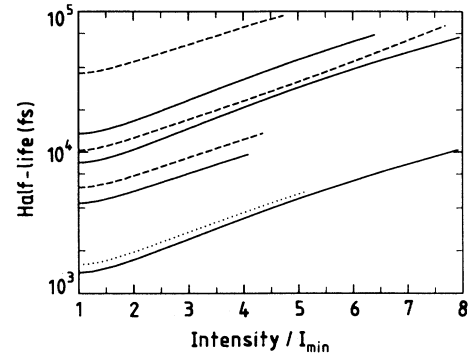

FIG. 2. Half-life ($\hbar \ln 2/\Gamma$) of a hydrogen atom vs the intensity of the incident laser field. On the horizontal scale, 1 corresponds to the intensity where the lifetime reaches its minimum value. The initial states (n,l,m) and incident wavelengths are, from top to bottom, (8,6,5) 620 nm; (8,5,5) 532 nm; (8,6,5) 842 nm; (6,5,5) 620 nm; (7,6,5) 1064 nm; (8,5,4) 532 nm; (8,4,4) 532 nm; and (6,5,5) 1064 nm. The different styles of curve correspond to different values of l.

We have no satisfactory explanation for another remarkable relation between the results presented in Table I, namely that the minimum lifetime $\tau_{\min}$ is in general about three times longer than the lifetime obtained in first-order perturbation theory at the intensity $I_{\min}$. Together with Eq. (1), this "rule" can be used to obtain a rough estimate of the lifetime of the state in the nonperturbative regime, given its rate of ionization in a weak field.

We cannot confirm the power-law increase with intensity of the lifetime for $I \gg I_{\min}$, which is predicted by the high-frequency theory, since Sturmian-Floquet calculations for intensities much higher than $I_{\min}$ are too demanding numerically. Figure 2 suggests that the rate of increase of the lifetime for $I \gtrsim I_{\min}$ depends essentially on the angular momentum of the initial state rather than on ω or m. However, more work is necessary to establish this trend. It should be noted that the increase of the lifetime with the laser intensity is not rapid. Simulations taking into account the spatial extension of a realistic laser pulse indicate that an experimental detection of adiabatic stabilization would require the target excited atoms to be produced within a very small volume of the laser beam, because of this slow increase.

Finally, we mention that recent calculations using model potentials show that the lifetimes of the circular states of alkali metals are very close to those for hydrogen, and that the lifetime of hydrogen in a circularly polarized field also passes by a minimum value; however, the minimum occurs at much higher intensity (and much larger ionization rate) than in the case of linear polarization. This will be described elsewhere.

[1] See, e.g., the recent review by K. Burnett, V. C. Reed, and P. L. Knight, J. Phys. B **26**, 561 (1993).

[2] M. Dörr, R. M. Potvliege, and R. Shakeshaft, Phys. Rev. Lett. **64**, 2003 (1990); R. Shakeshaft, R. M. Potvliege, M. Dörr, and W. E. Cooke, Phys. Rev. A **42**, 1656 (1990); M. Pont and M. Gavrila, Phys. Rev. Lett. **65**, 2362 (1990); L. Dimou and F. H. M. Faisal, Phys. Rev. A **46**, 4442 (1992).

[3] M. Dörr, R. M. Potvliege, D. Proulx, and R. Shakeshaft, Phys. Rev. A **43**, 3729 (1991).

[4] R. Potvliege and R. Shakeshaft, in *Electronic and Atomic Collisions (Proceedings of the XVIIth International Conference on the Physics of Electronic and Atomic Collisions, Brisbane, 1991)*, edited by W. R. MacGillivray, I. E. McCarthy, and M. C. Standage (Hilger, Bristol, 1992), p. 497.

[5] R. J. Vos and M. Gavrila, Phys. Rev. Lett. **68**, 170 (1992).

[6] M. Pont and R. Shakeshaft, Phys. Rev. A **44**, R4110 (1991).

[7] K. C. Kulander, K. J. Schafer, and J. L. Krause, Phys. Rev. Lett. **66**, 2601 (1991).

[8] A more detailed description of the method can be found in R. Potvliege and R. Shakeshaft, *Atoms in Intense Laser Fields* [Adv. At. Mol. Opt. Phys. Suppl. **1**, 373 (1992)].

[9] M. Gavrila, in *Atoms in Intense Laser Fields* [Adv. At. Mol. Opt. Phys. Suppl. **1**, 435 (1992)].

[10] The Sturmian-Floquet ac-Stark shift of this state agrees well with Vos's and Gavrila's result [5].

[11] As seen from Fig. 1, the lifetime at low intensity, in the perturbative regime, is underestimated by the high-frequency calculations. This is a direct consequence of approximating the wave function of the outgoing photoelectron by a plane wave. The ionization amplitude is not gauge-invariant when this "Born approximation" is made. For example, in this approximation the lifetime of the state of Fig. 1, calculated in first-order perturbation theory at 1×10^{10} W/cm^2 for 1064 nm wavelength, is 191 fs if the length form of the dipole operator is used, 7.55 ps if the velocity form is used, or 283 ps if the acceleration form is used (as in Ref. [5]). The exact (gauge-invariant) first-order result is 6.47 ns.

[12] M. Pont (private communication).

[13] A. Buchleitner and D. Delande (unpublished).

[14] Equation (1) implies that the excursion amplitude of the electron at the intensity I_{min} scales like $[m(l + 1 - m)!/\omega]^{1/2}$. It is about 73 a.u. for the Rydberg state $(n = 20, l = 19, m = 19)$ at 20 μm wavelength.

[15] M. Pont, Phys. Rev. A **44**, 2152 (1992).

[16] R. Shakeshaft, Comments At. Mol. Phys. **28**, 179 (1992).