Tunneling Ionization of Atomic Hydrogen by an Intense Low-Frequency Field

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We present results of Floquet calculations of rates for ionization of H by high-intensity light with wavelengths up to 1064 nm. If the frequency, ω, is well below the atomic-orbital frequency, ω_{at}, the rates approximately obey a law similar to that given by the Keldysh tunneling theory, and they approach the dc rate as the intensity increases. If $\omega > \omega_{at}$, there is no tunneling regime and the rates eventually decrease toward zero as the intensity increases.

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In this Letter we report results of Floquet calculations of rates for ionization of atomic hydrogen by intense radiation. For frequencies, ω, well below the atomic-orbital frequency, ω_{at}, the rates approach the dc rates as the intensity increases, and at intensities well below the "critical" intensity for static-field ionization, the rates follow a tunneling-ionization law that has an exponential behavior similar to that predicted by Keldysh.[1] On the other hand, if $\omega > \omega_{at}$, there is no tunneling regime, and the rates eventually decrease toward zero as the intensity increases; this atomic stability is in accord with earlier predictions.[2]

We define the electric field as

$$\mathbf{F}(t) = F[\cos(\omega t)\hat{\mathbf{z}} + \tan(\zeta/2)\sin(\omega t)\hat{\mathbf{x}}], \qquad (1)$$

with the x-z plane the polarization plane and with $\zeta = 0$ ($\pi/2$) for linear (circular) polarization. The intensity is $I \equiv (cF^2/8\pi)\operatorname{sech}^2(\zeta/2)$. The maximum magnitude of the instantaneous field $\mathbf{F}(t)$ is F; for circularly polarized light $|\mathbf{F}(t)| = F$ for all t. A free electron (charge e, mass μ) oscillates in the field, and even if it has no drift velocity it has a cycle-averaged energy $P \equiv (e^2F^2/4\mu\omega^2)\operatorname{sech}^2(\zeta/2)$, the ponderomotive energy. Thus, to liberate an electron that is initially bound with unperturbed energy $E^{(0)}$, the electron must absorb an energy that is at least $P - E^{(0)} - \Delta_{ac}$, where Δ_{ac} is the usual ac Stark shift.

Ionization occurs very rapidly, within a fraction of a cycle, when, simultaneously, $I > I_{cr}$ and $\omega \ll \omega_{cr}$, where I_{cr} and ω_{cr} are critical values of the intensity and frequency. It is well known from field-ionization studies[3] that I_{cr} is reached when the peak of the potential barrier for an electron in both an atomic field and a static electric field $F\hat{\mathbf{z}}$ is lowered to the dc-Stark-shifted energy $E_{dc} \equiv E^{(0)} + \Delta_{dc}$. Then the electron simply flows over the top of the barrier. The characteristic time it takes for the electron to reach the barrier peak is the atomic-orbital period, $2\pi/\omega_{at}$. This time must be short compared to the cycle time $2\pi/\omega$ for the field to be considered static, and therefore $\omega_{cr} \approx \omega_{at}$. In determining I_{cr} for ground-state atomic hydrogen, we must take into account the exceptional symmetry of a hydrogen atom in the field $F\hat{\mathbf{z}}$; this symmetry is expressed by the separability[4] of the Schrödinger equation in parabolic coordinates $\xi = r + z > 0$ and $\eta = r - z > 0$. For H(1s) the complete wave function is $\psi_1(\xi)\psi_2(\eta)/(\xi\eta)^{1/2}$, where[4]

$$-\frac{\hbar^2}{2\mu}\frac{d^2\psi_1(\xi)}{d\xi^2} + V_1(\xi)\psi_1(\xi) = \frac{E_{dc}}{4}\psi_1(\xi), \qquad (2a)$$

$$V_1(\xi) = -\frac{Z_1 e^2}{2\xi} - \frac{\hbar^2}{8\mu\xi^2} + \frac{eF\xi}{8}, \qquad (2b)$$

and $\psi_2(\eta)$ satisfies a similar equation, with ξ replaced by η and $V_1(\xi)$ replaced by $V_2(\eta)$; the potentials $V_1(\xi)$ and $V_2(\eta)$ are of the same form except for the sign of the term in eF. The separation constants Z_1 and Z_2 sum to Z. Note that $V_1(\xi)$ has a barrier, whose maximum is located on the positive ξ axis at the point given by $dV_1(\xi)/d\xi = 0$. The critical field, F_{cr}, is obtained by setting $V_1(\xi) = E_{dc}/4$ at the barrier maximum; at this field the electron can flow along the ξ axis over the top of the barrier.[4] Expressing Z_1 in a perturbation expansion[4] in powers of F, and calculating the dc Stark shift,[5] we obtain a transcendental equation for F_{cr} which we solve numerically[6] to give, for H(1s), $F_{cr} \approx 0.15$ a.u. or, equivalently, $I_{cr}\cos^2(\zeta/2) \approx 8 \times 10^{14}$ W/cm^2. For $I < I_{cr}$ the electron can tunnel out through the barrier $V_1(\xi)$, provided that the tunneling time is short compared to the cycle time. As shown by Keldysh,[1] the ratio of the tunneling time to the cycle time is $\gamma \equiv (E^{(0)}/2P)^{1/2}$, and so tunneling can occur if $\gamma < 1$. There is no tunneling regime if ω is of order, or greater than, ω_{at} since, as noted above, $\omega_{cr} \approx \omega_{at}$; in fact, at the critical field γ is, in general,[6] of order ω/ω_{at}, and therefore if $I < I_{cr}$ and $\omega > \omega_{at}$, we have $\gamma > 1$.

Let $|\Psi(t)\rangle \equiv e^{-iEt/\hbar}|\mathcal{F}(t)\rangle$ denote the state vector of the atom. The electron-field interaction is $V(t) = -(e/\mu c)\mathbf{A}(t)\cdot\mathbf{p}$, where $\mathbf{p}$ is the canonical momentum of the electron, and $\mathbf{A}(t) = (c/\omega)\Im m(\mathbf{F}e^{-i\omega t})$; we have neglected the spatial dependence of $\mathbf{A}(t)$ (the dipole approximation). The essence of the multiphoton picture is the Floquet *Ansatz*:[7] $|\mathcal{F}(t)\rangle$ is periodic in t, with period

$2\pi/\omega$. With this *Ansatz,* E becomes a quasienergy that can be written as $E = E^{(0)} + \Delta - i\Gamma/2$, where Δ and Γ are the field-induced shift and width of the level whose unperturbed energy is $E^{(0)}$; substituting for E, we see that the electron probability density, $|\langle \mathbf{x} | \Psi(t) \rangle|^2$, in any finite region of space decays in time as $e^{-\Gamma t/\hbar}$, and therefore $\Gamma/\hbar$ is the total ionization rate. Of course, the true energy of the electron is real—but it is not sharply defined since $|\Psi(t)\rangle$ is a nonstationary normalized wave packet with an energy distribution that has a width at least as large as Γ. The Floquet quasienergy is sharply defined, but it is complex to account for the energy width. Note that we have removed the $|\mathbf{A}(t)|^2$ term from $V(t)$ through a simple gauge transformation which amounts to multiplying $|\Psi(t)\rangle$ by the phase factor $\exp[-i(e^2/2\mu c^2\hbar)\int^t dt' |\mathbf{A}(t')|^2]$. Consequently, the continuum threshold does not shift—an electron at the threshold has zero drift momentum, so $(e/\mu c)\mathbf{A}(t)\cdot\mathbf{p}$ cannot shift the threshold—while Δ includes the ponderomotive energy P; thus $\Delta = \Delta_{\rm ac} - P$. For $\omega \ll \omega_{\rm at}$, we have $\Delta_{\rm ac} \approx \frac{1}{2}\,\mathrm{sech}^2(\zeta/2)\Delta_{\rm dc}$ when the two shifts are evaluated at the same value of F; the factor of $\frac{1}{2}\,\mathrm{sech}^2(\zeta/2)$ arises from the cycle averaging of $|\mathbf{F}(t)|^2$. We denote by N_0 the minimum number of photons that must be absorbed by the atom to ionize, that is, the smallest integer n for which $E^{(0)} + \Delta + n\hbar\omega > 0$, with $N_0^{(0)}$ denoting the value of N_0 in the weak-field limit (where $\Delta \rightarrow 0$). For $\omega \ll \omega_{\rm at}$, N_0 increases as I increases. Writing $V(t) = V_+ e^{-i\omega t} + V_- e^{i\omega t}$ and $|\mathcal{F}(t)\rangle = \sum_n e^{-in\omega t} |\mathcal{F}_n\rangle$, the time-dependent Schrödinger equation yields the following homogeneous set of coupled equations for the harmonic components $|\mathcal{F}_n\rangle$:

$$(E + n\hbar\omega - H_a)|\mathcal{F}_n\rangle = V_+ |\mathcal{F}_{n-1}\rangle + V_- |\mathcal{F}_{n+1}\rangle. \quad (3)$$

We solve for the eigenvalue E along the lines described previously.[8] Note that $|\mathcal{F}_n\rangle$ represents an electron that has absorbed n (real or virtual) photons.

We now present results of calculations of rates for ionization of H(1*s*) by circularly polarized light at intensities below $I_{\rm cr}$ and frequencies below $\omega_{\rm at} = 0.5$ a.u. We define $F_0 \equiv \frac{2}{3}(\sqrt{\mu}/e\hbar)|2E^{(0)}|^{3/2} = \frac{2}{3}$ a.u. (note that $F_{\rm cr} \approx 0.2F_0$), and in Fig. 1 we show the logarithm of $F\Gamma$ vs F_0/F at several different wavelengths. We also show the rate for ionization of H(1*s*) by a dc field of strength F (equal to the strength of the instantaneous field).[5] We see that as F increases the ac rates approach the dc rate from above; this is the main result of our paper. Note that the rates can be approximated by the form

$$\Gamma/\hbar \approx (CF_0/F)e^{-DF_0/F}, \quad (4)$$

where C and D depend only weakly on the frequency ω.

We interpret the approach of the ac rates towards the dc rate, as F increases, as the onset of tunneling. This is in qualitative accord with the tunneling theory of Keldysh,[1] which yields an ionization rate that differs only in the preexponential factor in Eq. (4). The discrepancy in the preexponential factor is expected since in the Keldysh theory the Coulomb interaction in the final state is neglected. Actually, we cannot easily determine the form of the preexponential factor, not only because the exponential behavior dominates, but also because (4) is not an asymptotic form, as we explain in a moment. The tunneling formula[9] for dc ionization of H(1*s*) has the form (4), and we have chosen to plot our data to reveal the similarity to this formula. In the dc-tunneling formula $C = 6$ a.u. and $D = 1$ (Keldysh also predicts $D = 1$); we estimate, from the 1064-nm data (see caption to Fig. 1), $C \approx 1.7$ a.u. and $D \approx 0.85$. The discrepancies in C and D should not be taken too seriously since at high fields the dc-tunneling formula overestimates the dc rates by slightly more than a factor of 2 at $F = 0.07$ a.u., for example. The dc-tunneling formula[9] becomes exact in the asymptotic limit $F/F_{\rm cr} \rightarrow 0$, not $F/F_{\rm cr} \rightarrow \infty$. However, the onset of tunneling in an oscillating field occurs at an intensity for which the Keldysh parameter γ is about 1. Thus, for the ac rates, Eq. (4) is not an asymptotic formula; it is accurate only over a finite range of F for which F is sufficiently large that $\gamma \ll 1$ and sufficiently small that $F/F_{\rm cr} \ll 1$ (so our estimation of C and D is somewhat ambiguous). We note, incidentally, that the ac Stark shifts, for circularly polarized light, converge toward the dc Stark shift as I increases.[6]

Equation (4) is for circular polarization. Since $\Gamma/\hbar$ is the rate averaged over one cycle, we expect the preexponential factor to depend significantly on the polarization; in the case of circular polarization $|\mathbf{F}(t)| = F$, all t,

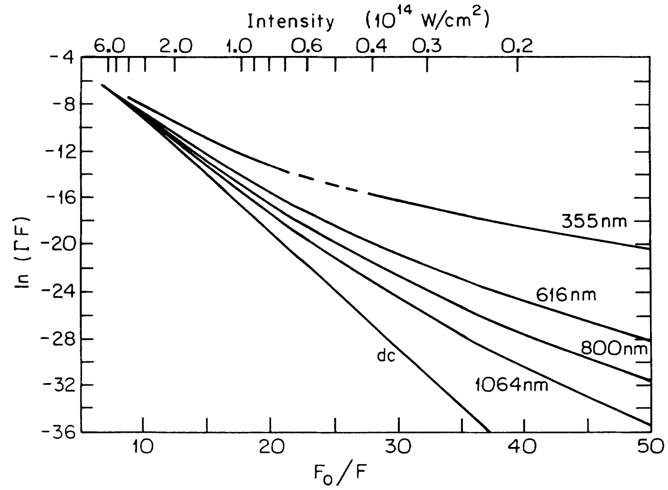

FIG. 1. Width Γ, multiplied by field strength F, in a.u., vs F_0/F, for ionization of H(1*s*) by circularly polarized light of various wavelengths (labeled in nm) or by a dc field (labeled dc). $F_0 = \frac{2}{3}$ a.u. The parameters C and D of Eq. (4) were taken to be the y-axis intercept and the slope of the 1064-nm curve extrapolated to $F_0/F = 0$. The broken line (for 355 nm) is a smooth interpolation through a region of intermediate resonances accumulating at a multiphoton ionization threshold.

and the electron can tunnel out at every moment of the cycle, while in the case of linear polarization the electron tunnels out primarily near the extremal points where $|\mathbf{F}(t)|$ is close to F. Following Delone and Krainov,[10] to generalize (4) to linear polarization, we replace F by $F|\cos(\omega t)|$ in Eq. (4), and average the resulting expression over one cycle; only the preexponential factor is changed—this factor becomes $C'(F_0/F)^{1/2}$, where $C' = (2/\pi D)^{1/2}C$. This suggests that for linear polarization we plot the logarithm of $F^{1/2}\Gamma$ vs F_0/F, and that we compare with a dc rate that is cycle averaged over the field $F\cos(\omega t)$; we have confirmed that the ac rates do indeed approach the cycle-averaged dc rate as I increases, if $\omega \ll \omega_{at}$.[6]

Since the tunneling rates are exponentially small, the static-field limit may be experimentally observable, if $\omega \ll \omega_{at}$, not in the tunneling regime but only in the regime above F_{cr}, when ionization occurs in a time of order $2\pi/\omega_{at}$, a time much smaller than the cycle time $2\pi/\omega$. This is consistent with recent experiments,[11-13] where ionization by very-low-frequency fields is observed only at intensities above the I_{cr} appropriate to the atom. Note that the Floquet method breaks down when $\Gamma > \hbar\omega$, for then it is inappropriate to describe the ionization process by a cycle-averaged rate.[14] Nevertheless, $\hbar/\Gamma$ still gives a good indication of the time required for ionization.

In Fig. 2 we plot the index of nonlinearity, $K = d(\ln\Gamma)/d(\ln I)$ vs I, for circularly polarized light. As I vanishes, K approaches $N_0^{(0)}$, in accord with perturbation theory. As I increases, K usually decreases (more rapidly at longer wavelengths), despite the fact that N_0 increases. An exception occurs for short wavelengths (e.g., 355 nm), when no excess photons are absorbed (until I is relatively high) so that the closing of a channel at a multiphoton ionization threshold has a significant effect. The reduction in K as I increases is due to the polarization of the atom by the field; as I increases, the electron spends less time in the region of the nucleus, the region where it can absorb photons, and therefore Γ does not rise as fast as perturbation theory predicts.[8]

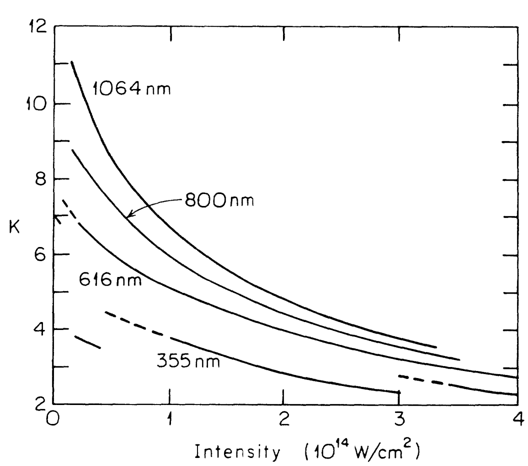

FIG. 2. Index of nonlinearity K (defined in text) vs intensity, for ionization of H(1s) by circularly polarized light. The broken lines commence at multiphoton ionization thresholds, and are smooth extrapolations through the resonance regions; note that there is a discontinuous jump in K corresponding to the closing of the lowest open channel.

To obtain an accurate rate, $\Gamma/\hbar$, we must allow the photon index n, in $|\mathcal{F}_n\rangle$, to become sufficiently large, but, in the case of circular polarization, it can be less than N_{ch}, the characteristic number of photons that the electron ultimately absorbs. N_{ch} can be estimated[11,12] in the tunneling regime by calculating the drift velocity delivered to a free electron that is released into a classical oscillating field with zero instantaneous velocity. For circular polarization[12] the drift energy is of order P, and so, taking into account that N_0 photons must be absorbed to produce a free electron with zero drift velocity, $N_{ch} > 2N_0$ when $P \gg |E^{(0)}|$. However, many of these photons are absorbed *after* the electron has escaped, and do not strongly affect the ionization rate; we find it is sufficient to take n up to $1.5N_0$. Of course, a statement as to when photons are absorbed is not gauge invariant; no experiment can decide whether the photons were absorbed before or after the electron escaped. The notion of tunneling is appropriate to the length gauge, where the electron-field interaction is $-e\mathbf{x}\cdot\mathbf{F}(t)$; in this gauge one imagines that *most* of the photons are absorbed after the electron tunnels out. This suggests that to calculate an accurate ac rate for ionization by circularly polarized light in the length gauge, one need not, in the tunneling regime, let n become much greater than the maximum angular momentum quantum number that must be included in the calculation of an accurate dc rate. Naturally, to calculate the photoelectron energy distribution, we must allow n to significantly exceed N_{ch}.

Finally, we briefly comment on the high-intensity limit for frequencies $\omega > \omega_{at}$. In Fig. 3 we show the rate for ionization of H(1s) by linearly polarized light of wavelength 70 nm ($\omega = 0.65$ a.u.). The rate at first rises as I increases, but peaks at $I = 1.1\times10^{16}$ W/cm^2, and then falls off. The peak occurs at an intensity for which the

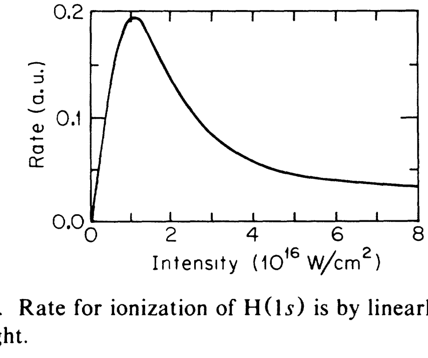

FIG. 3. Rate for ionization of H(1s) is by linearly polarized 70-nm light.

ratio of the excursion amplitude $|eF/\mu\omega^2|$ of a free electron to the binding radius a_0 ($=1$ a.u.) is slightly greater than unity (actually 1.33). At all frequencies the coupling strength of the electron to the field increases as I does, and this tends to enhance the ionization rate; but, as noted above, the field also polarizes the atom, and this effect tends to reduce the rate. For $\omega < \omega_{at}$ the inhibiting effect of polarization is seen in a reduction of the index of nonlinearity, but ionization still occurs through tunneling. For $\omega > \omega_{at}$, on the other hand, tunneling is not possible, and polarization defeats ionization. Of course, in practice saturation would occur before the peak in Fig. 3 could be reached. However, processes such as spontaneous decay, harmonic generation, and Raman scattering may increase atomic stability; this will be addressed in a future article.

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[1]L. V. Keldysh, Zh. Eksp. Teor. Fiz. **47**, 1945 (1965) [Sov. Phys. JETP **20**, 1307 (1965)].

[2]J. J. Gersten and M. H. Mittleman, J. Phys. B **9**, 2561 (1976); M. V. Fedorov and A. M. Movsesian, J. Phys. B **21**, L155 (1988); M. Gavrila and M. Pont (private communication).

[3]See, e.g., J. E. Bayfield and P. M. Koch, Phys. Rev. Lett. **33**, 258 (1974); T. W. Ducas, M. G. Littman, R. R. Freeman, and D. Kleppner, Phys. Rev. Lett. **35**, 366 (1975); R. F. Stebbings, C. J. Latimer, W. P. West, F. B. Dunning, and T. B. Cook, Phys. Rev. A **12**, 1453 (1975); W. E. Cooke and T. F. Gallagher, Phys. Rev. A **17**, 1226 (1978); M. G. Littman, M. M. Kash, and D. Kleppner, Phys. Rev. Lett. **41**, 103 (1978).

[4]H. A. Bethe and E. E. Salpeter, *Quantum Mechanics of One and Two Electron Atoms* (Academic, New York, 1957).

[5]We have extended the dc-shift and dc-width calculations of A. Maquet, S.-I. Chu, and W. P. Reinhardt [Phys. Rev. A **27**, 2946 (1983)] to higher field strengths.

[6]A more detailed description of our calculations, and further results, will be given elsewhere.

[7]See, e.g., Maquet, Chu, and Reinhardt, Ref. 5. See also, R. M. Potvliege and R. Shakeshaft, Phys. Rev. A **40**, 3061 (1989); **41**, 1609 (1990); M. Dörr, R. M. Potvliege, and R. Shakeshaft, Phys. Rev. A **41**, 558 (1990).

[8]Potvliege and Shakeshaft, Ref. 7; Dörr, Potvliege, and Shakeshaft, Ref. 7.

[9]L. D. Landau and E. M. Lifshitz, *Quantum Mechanics* (Pergamon, New York, 1965), 2nd ed., p. 275.

[10]Tunneling ionization, and the approach of ac rates to dc rates, has been discussed extensively for a model atom with a zero-range binding potential, though in this model there is no I_{cr} for which the electron can flow over the top of the barrier. See, e.g., N. B. Delone and V. P. Krainov, *Atoms in Strong Light Fields* (Springer, Berlin, 1985); W. Becker, J. K. McIver, and M. Confer, Phys. Rev. A **40**, 6904 (1989).

[11]T. F. Gallagher, Phys. Rev. Lett. **61**, 2304 (1988); T. F. Gallagher and T. J. Scholz, Phys. Rev. A **40**, 2762 (1989).

[12]P. B. Corkum, N. H. Burnett, and F. Brunel, Phys. Rev. Lett. **62**, 1259 (1989).

[13]S. Augst, D. Strickland, D. D. Meyerhofer, S. L. Chin, and J. H. Eberly, Phys. Rev. Lett. **63**, 2212 (1989).

[14]Note that the Floquet eigenvalue E is not unique; associated with any "dominant" eigenvalue, corresponding to the eigenvector that represents a physically realizable state, are infinitely many "shadow" eigenvalues on unphysical sheets of the Riemann energy surface. When $\Gamma > \hbar\omega$ it is not possible to distinguish shadow and dominant eigenvalues. See R. M. Potvliege and R. Shakeshaft, Phys. Rev. A **38**, 6190 (1988); M. Dörr and R. M. Potvliege, Phys. Rev. A **41**, 1472 (1990).