Multiphoton processes in an intense laser field. IV. The static-field limit

Robin Shakeshaft
Physics Department, University of Southern California, Los Angeles, California 90089-0484

R. M. Potvliege
Physics Department, University of Durham, South Road, Durham, DH1 3LE, England

Martin Dörr* and W. E. Cooke
Physics Department, University of Southern California, Los Angeles, California 90089-0484
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We explore the similarity between dc-field ionization and low-frequency multiphoton ionization of atoms. If the frequency ω of the light is below the characteristic atomic-orbital frequency ω_{at}, ionization of the atom occurs by tunneling provided that the intensity I is sufficiently high that the ratio of the tunneling time to the cycle time—this is essentially the "Keldysh parameter" γ—is less than unity. However, if I exceeds a critical intensity, I_{cr}, the electron flows over the top of the potential barrier rather than tunneling through it. I_{cr} depends on the magnetic quantum number, m, of the initial bound state, and is proportional to, but significantly less than, the characteristic atomic intensity. We give a simple approximate expression for I_{cr} in terms of m, valid in the absence of an exceptional symmetry (such as exists for hydrogen). We find that I_{cr} increases as m does; consequently, electrons with $m = 0$ are stripped first as the intensity rises, and the residual ion will be left in a state of alignment, in agreement with calculations of ionization rates for Xe [K. C. Kulander, Phys. Rev. A **38**, 778 (1988)]. We present results of Floquet calculations of rates for ionization of H(1s) by circularly or linearly polarized light in the wavelength range 355 to 1064 nm, at intensities somewhat below I_{cr}. At these wavelengths, the rates approach more or less the same value as I increases, in accord with the Keldysh tunneling theory. We show that, provided $\omega < \omega_{at}$, the ac shift and the ac width, respectively, tend to the dc shift and the dc width (cycle averaged over the instantaneous field) once I is sufficiently large that $\gamma < 1$. On the other hand, we show that for $\omega > \omega_{at}$ there is no tunneling regime; rather, in the absence of strong intermediate resonances, the ionization rate reaches a peak when $\gamma \approx 1$, and decreases toward zero as γ does. Presumably the Floquet picture becomes inadequate when the ionization width Γ approaches the photon energy $\hbar\omega$, for then ionization takes place in less than a cycle. We speculate as to how the Floquet picture breaks down and, finally, we show that the statement $\Gamma \approx \hbar\omega_{at}$ yields the correct Z scaling of I_{cr} for ionization in a Coulomb field.

I. INTRODUCTION

Atomic ionization by strong low-frequency radiation may closely resemble static-field ionization. This is apparent from measurements of photoelectron energy spectra, which can be readily interpreted by assuming that the photoelectrons emerge at a particular phase of the oscillating field.[1,2] A further indication is that the threshold for *measurable* ionization by low-frequency radiation often occurs at the "critical" field for static-field ionization.[3] In this paper we explore the onset of static-field ionization when an atom is irradiated by a strong low-frequency field.

The classical potential of an electron moving in both an atomic field and an applied static electric field $F\hat{\mathbf{z}}$ (a caret denotes a unit vector) has a barrier with a saddle point.[4] An initially bound electron can tunnel out through this barrier, although if F is small the tunneling ionization rate is exponentially small. However, the potential energy at the saddle point decreases as the strength F of the applied field increases, and when F reaches a critical value F_{cr} the saddle-point potential energy becomes equal to the atomic binding energy of the electron. Above the critical field the electron simply flows over the top of the barrier, and ionization occurs rapidly;[5,6] in fact, as we see later, complete ionization occurs within a time of the order of the atomic-orbital period of the initially bound electron.

We are interested in ionization by an oscillating electric field of frequency ω. We treat this field as spatially homogeneous (the dipole approximation is valid at low frequencies). Taking the polarization plane to be the x-z plane, we introduce the complex field amplitude $\mathbf{F}$, defined as

$$\mathbf{F} = F[\hat{\mathbf{z}} + i\tan(\zeta/2)\hat{\mathbf{x}}], \tag{1a}$$

where F is real and positive, and where ζ is the ellipticity parameter, with $\zeta = 0$ $(\pi/2)$ for linear (circular) polarization. The instantaneous electric field is

$$\mathrm{Re}(\mathbf{F}e^{-i\omega t}) = F[\cos(\omega t)\hat{\mathbf{z}} + \tan(\zeta/2)\sin(\omega t)\hat{\mathbf{x}}], \tag{1b}$$

and its maximum magnitude is F; in the case of circular polarization the instantaneous field has magnitude F at all times. The root-mean-square strength of the field is $F_{\mathrm{rms}} = F\sec(\zeta/2)/\sqrt{2}$, and the intensity is $I \equiv (cF^2/8\pi)\sec^2(\zeta/2)$. The cycle-averaged energy of a free electron that oscillates in this field but that is on average at rest (its drift velocity is zero) is the ponderomotive energy $P \equiv e^2F_{\mathrm{rms}}^2/(2\mu\omega^2)$, that is, $(2\pi e^2 I/\mu c\omega^2)$, where e and μ are the electron charge and mass.

If F is below F_{cr}, ionization occurs via the electron tunneling through the barrier formed by the atomic potential and by the maximum instantaneous field—provided that the time taken for the electron to pass through the barrier (the tunneling time) is short compared to the cycle time, so that the oscillating field is effectively a static field of strength F while tunneling occurs. Note that the tunneling time is not the ionization time. Indeed, if F is well below F_{cr} the probability for tunneling during one cycle is exponentially small, and although the photoelectron tunnels out at a particular phase of the cycle, ionization proceeds over many cycles—the ionization time is very much longer than the tunneling time. As pointed out by Keldysh,[7] the ratio of the tunneling time to the cycle time is essentially the parameter $\gamma \equiv \sqrt{|E^{(0)}|/2P}$, where $E^{(0)}$ is the initial unperturbed binding energy of the electron; therefore, tunneling commences roughly when γ drops below unity (which for rare gases usually occurs long before the saturation intensity is reached). To understand this interpretation of γ, first observe that the characteristic width of the barrier is $|E^{(0)}/eF|$ and that the characteristic speed at which the electron tunnels through the barrier is the atomic-orbital speed, which is approximately $|2E^{(0)}/\mu|^{1/2}$; the tunneling time is the barrier width divided by the tunneling speed, and dividing this time by the cycle time gives, to within a constant of order unity, γ. Since γ decreases as F increases, ionization by fields of relatively high frequency may occur via tunneling at sufficiently high field strengths. However, if ω is larger than the atomic-orbital frequency ω_{at}, there is no tunneling regime since γ does not decrease below unity until F exceeds F_{cr}. Rather, we find that if $\omega > \omega_{at}$ the ionization rate reaches a maximum as I increases, at the point where $\gamma \approx 1$, and as I increases further, the rate falls toward zero, provided that there are no strong intermediate resonances; this atomic "stability" has been predicted previously.[8] At very low frequencies tunneling may commence at very low field strengths, but apparently the photoelectron signal is hardly measurable until the critical field is reached, at least in the case of rare gases.[3] When the threshold for measurable ionization by a very low-frequency field occurs well below the critical field the ionization process is a *multiphoton* one. For example, ionization from high Rydberg states by microwave fields can occur well below the critical field through resonant multiphoton Landau-Zener transitions.[9] In *multiphoton* ionization, photoelectrons do not emerge at a particular phase of the field.

In Sec. II of this paper we obtain the value of the critical intensity, I_{cr}, at which very rapid ionization occurs, assuming that ω is below a critical frequency ω_{cr}. We see that I_{cr} is proportional to, but significantly less than, the characteristic atomic intensity, while ω_{cr} is equal to the characteristic atomic-orbital frequency, ω_{at}. We extend previous derivations of I_{cr} by taking into account the orbital angular momentum of the initially bound electron, and we express I_{cr} as a simple function of the magnetic quantum number m of the initial bound state. However, this expression is only valid in the absence of an exceptional symmetry, and so does not apply to atomic hydrogen[9] (or a hydrogenlike ion). Excluding hydrogen, I_{cr} increases as m increases. In Sec. III we briefly discuss the Floquet method for calculating the width Γ induced in a bound level by an oscillating field. The total ionization rate is $\Gamma/\hbar$. At intensities above I_{cr}, ionization occurs in a time of order $2\pi/\omega_{\mathrm{at}}$, and we have $\Gamma \approx (\hbar\omega_{\mathrm{at}}/2\pi)$. When Γ becomes comparable to, or exceeds, $\hbar\omega$, the Floquet method becomes inadequate to describe the details of the ionization process since ionization takes place in less than a cycle, while $\Gamma/\hbar$ is a cycle-averaged rate. Nevertheless, $\hbar/\Gamma$ still gives an indication of the time required for ionization. We discuss how the Floquet method breaks down when $\Gamma \approx \hbar\omega$. We also show that the statement $\Gamma \approx (\hbar\omega_{\mathrm{at}}/2\pi)$ gives the correct Z scaling of I_{cr} in the case of a hydrogenlike ion of atomic number Z. In Sec. IV we discuss ionization of atomic hydrogen. To determine I_{cr} accurately in the case of a hydrogen atom (or hydrogenlike ion) we must take into account the special symmetry expressed by the separability of the Schrödinger equation for an electron moving in both a pure Coulomb potential and a static field. This symmetry results in a considerably higher value of I_{cr} for H($1s$), about $8 \times 10^{14}\sec^2(\zeta/2)$ W/cm^2, than would be obtained on the basis of the classical potential. In fact, this I_{cr} exceeds the saturation intensity for H($1s$) for all but very short pulses. The critical intensities for the rare gases in their ground states are quite a bit lower; for example, $I_{\mathrm{cr}} \approx 9 \times 10^{13}\sec^2(\zeta/2)$ W/cm^2 for Xe. We note, incidentally, that calculations of multiphoton ionization rates for Xe have been carried out by Kulander,[10] and these calculations indicate the onset of tunneling at intensities below 10^{14} W/cm^2. Despite the fact that hydrogen is somewhat anomalous, it is the simplest atom for which calculations can be performed, and in Sec. IV we present results of Floquet calculations of rates for ionization of H($1s$) by linearly or circularly polarized light with frequencies well below ω_{at}. We also present estimates of energy shifts. We see that as I increases, the rates become insensitive to frequency (if $\omega < \omega_{\mathrm{at}}$), and for I large (but below I_{cr}) the rates

approximately obey a law that is similar to the tunneling law predicted by Keldysh.[7] More significantly, as I increases, both the ac rates and ac shifts approach the dc rates and dc shifts when the latter are evaluated at the instantaneous ac-field and averaged over one cycle. We conclude Sec. IV by briefly considering, for the purpose of contrast, the case $\omega > \omega_{at}$. We present results that suggest the ionization signal maximizes when $\gamma \approx 1$ if $\omega > \omega_{at}$.

II. CRITICAL FREQUENCY AND FIELD

Ionization of an atom by an oscillating electric field $\mathrm{Re}(\mathbf{F}e^{-i\omega t})$ takes only a fraction of a cycle when, simultaneously,

$$I > I_{cr}, \tag{2a}$$

$$\omega \ll \omega_{cr}, \tag{2b}$$

where I_{cr} and ω_{cr} are critical values of the intensity and frequency. It is well known from field ionization studies[4-6,11] that the critical intensity is reached when the classical potential barrier peak is lowered to the dc-Stark-shifted energy of the atom in its initial state. Then the electron simply flows over the top of the barrier. However, it must do so in a time t_{cr} that is short compared to the cycle time $2\pi/\omega$. The characteristic distance that the electron must travel to reach the barrier peak is the atomic binding radius, a, say. If v_{at} denotes the characteristic atomic orbital speed we have (assuming the electron is not significantly accelerated by the electric field, a point we return to below) $t_{cr} \approx 2\pi a/v_{at}$, the atomic-orbital period, and hence ω_{cr} is equal to $\omega_{at} \equiv v_{at}/a$, the atomic-orbital frequency. Actually, t_{cr} may be a few orbital periods because the barrier peak is a saddle point, and to flow over the barrier the electron must travel through a valley. However, we will see below that the valley is not narrow on the scale of the atomic radius a, and therefore we take ω_{cr} to be the atomic-orbital frequency, ω_{at}, simply keeping in mind that the electron must channel through the valley.

We can easily determine I_{cr}, assuming that the barrier peak occurs at a distance where the atomic potential is the pure Coulomb potential $-Ze^2/r$. Suppose that the maximum instantaneous field points along the z axis, so that the effective static field is $F\hat{z}$. If the electron initially has magnetic quantum number m, with the z axis the quantization axis, and if we write $F \equiv -e/r_1^2$, the classical potential is, in cylindrical coordinates (ρ, z),

$$V_{cl}(\rho, z) = \frac{\hbar^2 m^2}{2\mu\rho^2} - \frac{Ze^2}{r} - \frac{e^2 z}{r_1^2}, \tag{3}$$

where $r = \sqrt{\rho^2 + z^2}$. Differentiating with respect to ρ and z, and introducing dimensionless variables $\rho' = \rho/(\sqrt{Z}r_1)$, $z' = z/(\sqrt{Z}r_1)$, $r' = r/(\sqrt{Z}r_1)$, and $(m')^2 = \hbar^2 m^2/(\sqrt{Z^3}\mu r_1 e^2)$, we find that $V_{cl}(\rho, z)$ has a stationary point when $(\rho')^4 = (m')^2(r')^3$ and $z' = (r')^3$. At a stationary point we have

$$|m'| = \sqrt{r'}[1 - (r')^4], \tag{4}$$

and

$$V_{cl}(\rho, z) = -(\sqrt{Z}e^2/2r_1 r')[1 + 3(r')^4]. \tag{5}$$

We set $V_{cl}(\rho, z) = -E$, where here $E = E^{(0)} + \Delta_{dc}$ with $E^{(0)}$ the unperturbed energy of the bound electron and with Δ_{dc} the dc Stark shift. Solving for r_1 in terms of r', we obtain the critical field strength[11]

$$F_{cr} \equiv e/r_1^2 = A(m)(E^2/4Ze^3), \tag{6}$$

where the "enhancement factor"

$$A(m) \equiv \left(\frac{4r'}{1 + 3(r')^4}\right)^2 \tag{7}$$

depends on $|m|$ through r', as implied by Eq.(4).

Note that at a stationary point the classical potential has a minimum with respect to ρ:

$$\frac{1}{2V_{cl}}\frac{\partial^2 V_{cl}(\rho, z)}{\partial\rho^2} = -\left(\frac{1}{r}\right)^2. \tag{8a}$$

At the critical field r and r' are related by

$$r = \frac{2Ze^2 r'}{E\sqrt{A(m)}}. \tag{8b}$$

Since $|m'|$ is real and positive we require that $0 \le r' \le 1$. However, r' is not a single-valued function of $|m'|$; there are two values of r' in the range $0 \le r' \le 1$ which give the same value of $|m'|$. In fact, $|m'|$ vanishes at both $r' = 0$ and $r' = 1$, and has a maximum value of $(\frac{8}{9})3^{-1/4}$ at $r' = 1/\sqrt{3}$. For a fixed value of $|m'| < (\frac{8}{9})3^{-1/4}$, both values of r' give stationary points of $V_{cl}(\rho, z)$, but only values of r' in the range $r' > 3^{-1/4}$ give a saddle point which is a *maximum* along the z axis (and a minimum with respect to ρ). We discard the solution in the range $r' < 3^{-1/4}$ since it gives a well, that is, a minimum in both the ρ and z directions. Contour plots of the classical potential are shown in Fig. 1 for $m = 0$ and 1. Note that the saddle-point region is fairly flat.

If $m = 0$ we have $r' = 1$ and therefore $A(0) = 1$, which yields the same critical field strength that is well known in field-ionization studies.[4-6] However, if $m \neq 0$ we have $r' < 1$; writing $r' = 1 - \epsilon$ we have, if ϵ is small, $A(m) \approx 1 + 4\epsilon + 7\epsilon^2 > 1$. Thus I_{cr} is magnified by $A^2(m)$ when $m \neq 0$. This is a consequence of two facts:[12] (i) The atomic potential is spherically symmetric, and so the atomic energy is shared between motion along the z axis and motion in the plane perpendicular to the z axis, and (ii) when $m \neq 0$ the angular momentum barrier pushes the electron off the z axis, so that a stronger field is required to pull the electron along the z axis. The maximum allowed value of $A(m)$, reached at $r' = 3^{-1/4}$, is $4/\sqrt{3}$. We see that the critical intensity is magnified by about a factor of 5 for sufficiently large m. It follows that electrons with magnetic quantum number $m = 0$ are ejected first as the intensity rises. Therefore,

in the case of linear polarization, if an outer subshell is not fully stripped the residual ion will be left in a state of alignment.[10] This is consistent with the detailed numerical calculations of Kulander,[10] who found that for ionization of Xe by linearly polarized 1064-nm light the *rate* for removal of a $5p$ electron is about 20 times larger for $m = 0$ than for $m = 1$ at a fixed high intensity. Note that the angular momentum quantization axis, which lies along the direction of polarization, is fixed in space in the case of linear polarization. In the case of circular polarization, this axis rotates in space, and if $\hbar\omega$ is larger than the Stark splittings of the $m = 0$ and 1 levels, the rotation is too rapid for the atom to adiabatically follow the axis, so that an electron that is in a state with a definite value of m at one point of the cycle will be in a superposition of states with different values of m, in particular $m = 0$, at another point of the cycle; the alignment of the ion will then be reduced.

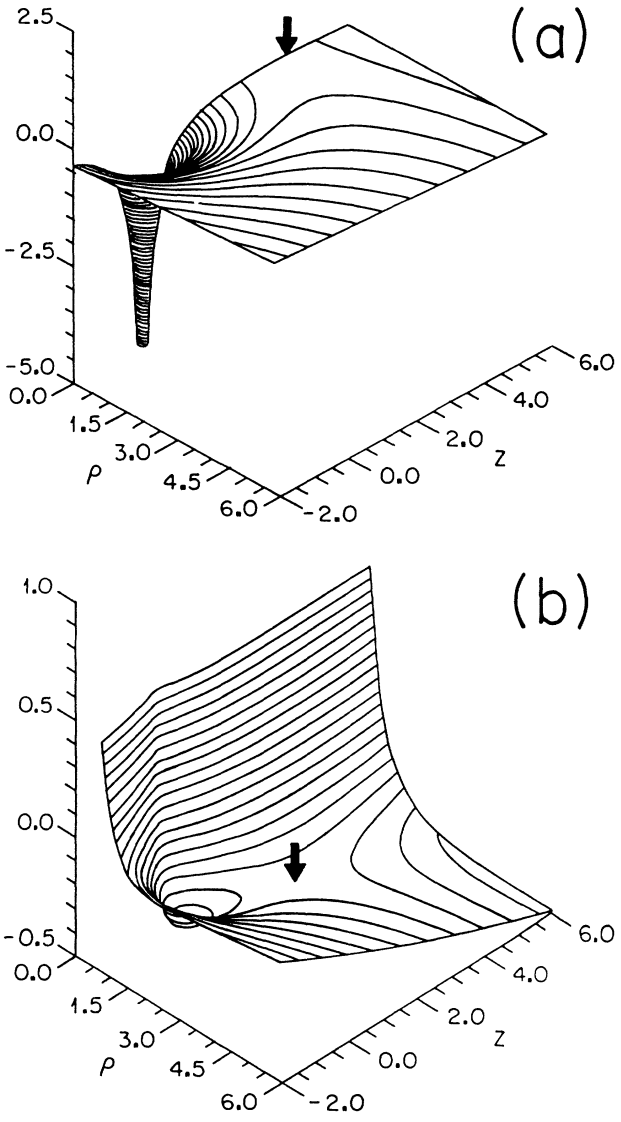

FIG. 1. The classical potential—see Eq. (2) of the text—for $Z = 1$, $e^2/r_1^2 = 0.07$ a.u., and (a) $m = 0$, and (b) $m = 1$. The potential is symmetric with respect to rotations about the z axis. When $m = 0$ there is an infinitely deep well at $r = 0$ and a saddle point (indicated by an arrow) on the z axis at $z = r_1$. When $m = 1$ there is a shallow well not far from the origin, and there is a saddle point (again indicated by an arrow) displaced off the z axis at a distance somewhat less than r_1 from the origin.

It is useful to introduce the number

$$n^* \equiv \sqrt{Z^2 e^2/2a_0|E|}, \tag{9}$$

where $a_0 = \hbar^2/\mu e^2$. We have

$$m/n^* = [1 - (r')^4]\sqrt{[1 + 3(r')^4]}. \tag{10}$$

After a little algebra, we find, for small m/n^*,

$$A(m) \approx 1 + \frac{|m|}{2n^*} + \frac{19}{64}\left(\frac{|m|}{n^*}\right)^2 \tag{11a}$$

$$\approx \frac{1 - \frac{3}{32}(|m|/n^*)}{1 - \frac{19}{32}(|m|/n^*)}, \tag{11b}$$

where the Padé extrapolation (11b) is more accurate than (11a). In Fig. 2 we plot $A(m)$ versus $|m|/n^*$; this is a universal curve (the same for all Z). Provided that the dc Stark shift is less than or comparable to $E^{(0)}$, which we believe to be the case,[13] n^* is of the order of the effective principal quantum number of the unperturbed atomic level. Hence, on physical grounds (namely, $|m| \leq l$, where l is the orbital angular momentum quantum number, which is less than or of the order of n^*) we expect $|m|/n^*$ to be less than or of the order of unity; in fact, the maximum value of $|m|/n^*$ is $2\sqrt{2}/3 \approx 0.94$, reached at $r' = 3^{-1/4}$. The comparison, in Fig. 2, of the exact value of $A(m)$ with the Padé approximation (11b) shows

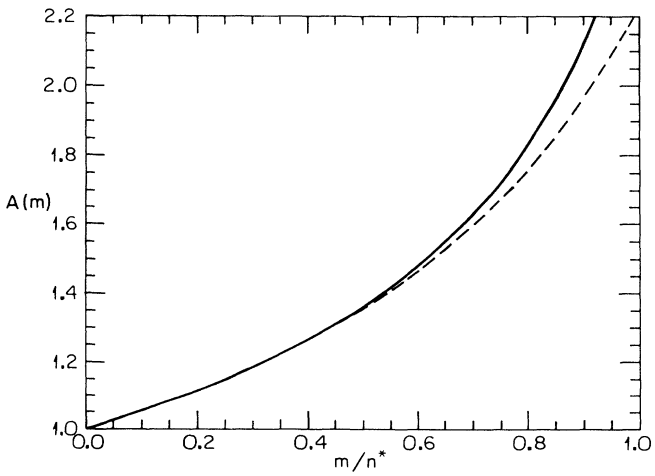

FIG. 2. Enhancement factor $A(m)$ of the critical field, vs m/n^*, for ionization of an atom in a state with magnetic quantum number m. (If the dc Stark shift is negligible, n^* is the effective principal quantum number of the state.) The dashed curve is the Padé approximation, Eq.(11b), of the text.

that this simple approximation is reliable over most of the range of interest. For the outer $(5p)$ subshell[13] of Xe, $n^* \approx 1$; putting $m = 1$ yields $A^2(m) \approx 5$. Note, incidentally, that the critical intensity $I_{cr} \equiv (cF_{cr}^2/8\pi)\sec^2(\zeta/2)$ for removing an $m = 0$ electron from Xe, in its ground state, is about $9 \times 10^{13}\sec^2(\zeta/2)$ W/cm^2.

Since n^* is comparable to the effective principal quantum number, we have $a \approx (n^*)^2 a_0/Z$ and $v_{at} \approx Zv_0/n^*$, where $v_0 = e^2/\hbar$. It follows that the critical frequency is

$$\omega_{cr} = \omega_{at} = \frac{Z^2}{(n^*)^3}\frac{e^2}{a_0\hbar}. \tag{12a}$$

From Eq. (8b) we see that the saddle point occurs at $r \approx 4a$, and it follows from Eq. (8a) that the sides of the valley do not rise steeply as ρ increases over distances that are small compared to a. Therefore we do not expect the electron to have great difficulty channeling through the valley along the z axis. Note that $F_{cr} = A(m)Z^3e/16(n^*)^4a_0^2 \approx [A(m)/16](Ze^2/a^2)$, which is proportional to but substantially less than the characteristic atomic field Ze^2/a^2, and therefore we were justified in neglecting the electric field in computing our order-of-magnitude estimate of t_{cr}. Note further that the "Keldysh" parameter[7] $\gamma \equiv \sqrt{E^{(0)}/2P}$ is of order $[16/A(m)](\omega/\omega_{at})$ at the critical field. It follows that if $\omega > \omega_{at}$ we have $\gamma > 1$ for $I < I_{cr}$, and therefore *there is no tunneling regime*; this is consistent with $\omega_{cr} = \omega_{at}$. It is perhaps worth emphasizing that the conditions for the onset of rapid ionization are no more than that (i) *the frequency be significantly less than the characteristic atomic-orbital frequency* and that (ii) *the field be greater than a critical field that is proportional to (but significantly less than) the characteristic atomic field.* The critical intensity is[11]

$$I_{cr} = \sec^2(\zeta/2)\frac{cA^2(m)Z^6e^2}{2^{11}\pi(n^*)^8a_0^4}, \tag{12b}$$

and this is proportional to (but significantly less than) the atomic intensity.

III. FLOQUET PICTURE

A. Floquet ansatz

Let $|\Psi(t)\rangle$ denote the state vector of an initially bound atomic electron that interacts with the oscillating electric field $\mathrm{Re}(\mathbf{F}e^{-i\omega t})$. The time-dependent Schrödinger equation is

$$i\hbar\frac{d}{dt}|\Psi(t)\rangle = [H_a + V(t)]|\Psi(t)\rangle, \tag{13}$$

where H_a is the atomic Hamiltonian and where $V(t)$ is the electron-field interaction. The essence of the multiphoton picture is the Floquet ansatz: $|\Psi(t)\rangle \equiv |\mathcal{F}(t)\rangle e^{-iEt/\hbar}$, where $|\mathcal{F}(t)\rangle$ is periodic in t, with period $2\pi/\omega$. Here E is 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 initial bound level whose unperturbed energy is $E^{(0)}$. The total ionization rate is $\Gamma/\hbar$. The minimum number of photons, N_0, which the atom must absorb to ionize is the smallest integer N for which $E^{(0)} + \Delta + N\hbar\omega$ is positive.

In the velocity gauge we have

$$V(t) = -(e/\mu c)\mathbf{A}(t)\cdot\mathbf{p}, \tag{14}$$

where $\mathbf{A}(t) \equiv (c/\omega)\mathrm{Im}(\mathbf{F}e^{-i\omega t})$, the vector potential, and where $\mathbf{p}$ is the canonical momentum operator. We have removed the $|\mathbf{A}(t)|^2$ term from $V(t)$ by a simple gauge transformation; this term would shift the continuous energy spectrum upwards by the ponderomotive energy $P \equiv (2\pi e^2 I/\mu c\omega^2)$, so its removal results in a downward contribution of P to the shift of the bound levels. Thus $\Delta = \Delta_{ac} - P$, where Δ_{ac} is the ac Stark shift. For $\omega \ll \omega_{at}$, we have $\Delta_{ac} \approx [\sec^2(\zeta/2)/2]\Delta_{dc}$, when the two shifts are evaluated at the same value of F; the factor of $\sec^2(\zeta/2)/2$ arises from the cycle-averaging of the square modulus of the instantaneous electric field. As I increases, so does $|\Delta|$ (if $\omega < \omega_{at}$), and N_0 may increase. Both Δ and the ionization width Γ are determined by solving an eigenvalue problem.[14] Expanding $|\mathcal{F}(t)\rangle$ in the harmonic series

$$|\mathcal{F}(t)\rangle = \sum_n e^{-in\omega t}|\mathcal{F}_n\rangle, \tag{15}$$

writing

$$V(t) = V_+e^{-i\omega t} + V_-e^{i\omega t}, \tag{16}$$

and substituting the Floquet ansatz into Eq. (14) we obtain the following set of coupled time-independent equations for the harmonic components $|\mathcal{F}_n\rangle$:

$$(E + n\hbar\omega - H_a)|\mathcal{F}_n\rangle = V_+|\mathcal{F}_{n-1}\rangle + V_-|\mathcal{F}_{n+1}\rangle. \tag{17}$$

These homogeneous equations, together with outgoing wave boundary conditions, form an eigenvalue problem. Note that the harmonic component $|\mathcal{F}_n\rangle$ represents an electron that has absorbed n real or virtual photons.

However, since the Riemann energy surface has infinitely many sheets, with branch points at the multiphoton ionization thresholds, the eigenvalue E has infinitely many branches; associated with any "dominant" eigenvalue, corresponding to the Floquet eigenvector that represents a physically realizable state, are infinitely many "shadow" eigenvalues on unphysical sheets of the Riemann energy surface.[15] At a fixed value of the intensity I the shadow eigenvalues usually correspond to Floquet eigenvectors that are of no physical significance, but as I increases, $|\Delta|$ increases, and at sufficiently high intensities the dominant eigenvalue passes a multiphoton ionization threshold; at this point the dominant eigenvalue changes roles with a nearby shadow eigenvalue, that is, the eigenvector that originally corresponded to the shadow eigenvalue acquires the physical character of the eigenvector that originally corresponded to the dominant eigenvalue, and the latter eigenvector acquires the un-

physical character originally possessed by the former.[15] Close to the threshold the exact state vector is a superposition of these two Floquet eigenvectors, but the separation of the dominant and nearby shadow eigenvalues at the threshold is of order Γ, and provided that the intensity sweeps past the threshold in a transit time $t_{\rm tr}$ that is short compared to $\hbar/\Gamma$, the switchover between shadow and dominant eigenvalues occurs diabatically, with no noticeable effect on the state of the atom. Now, an assumption underlying the Floquet ansatz is that the frequency bandwidth be very small compared to ω. It follows that the intensity should vary slowly over a cycle, and hence that the transit time $t_{\rm tr}$ should be much larger than the cycle time. However, under this restriction, if Γ is of the order of $\hbar\omega$ we have $t_{\rm tr} \gg 2\pi/\omega \approx \hbar/\Gamma$ and the atom ionizes during the transit time, in which case the passage past a threshold is not diabatic. In fact, if $\Gamma \gg \hbar\omega$ the separation $\hbar\omega$ of consecutive thresholds is much smaller than the separation of shadow and dominant eigenvalues, so it is not possible to distinguish between shadow and dominant eigenvalues. In this circumstance the atomic state vector is a superposition of many Floquet eigenvectors, with time-dependent coefficients that are not periodic; a single Floquet eigenvector cannot represent the dynamical ionization process. This indicates, not surprisingly, that the Floquet method is inadequate for treating ionization at intensities near or above the critical intensity; for as I approaches $I_{\rm cr}$ the ionization rate becomes of order $2\pi/\omega_{\rm at}$ and therefore the width Γ (whether this be the width of the dominant or shadow eigenvalue) exceeds $\hbar\omega$ (assuming, of course, that $\omega \ll \omega_{\rm at}$). The rate $\Gamma/\hbar$ is a cycle-averaged rate, which is unsuitable for describing ionization that takes place in less than a cycle; nevertheless, $\hbar/\Gamma$ does give a good indication of the time required for ionization.

B. Z scaling

We can define the critical intensity $I_{\rm cr}$ as the intensity for which $\hbar/\Gamma \approx 2\pi/\omega_{\rm at}$ (provided that $\omega \ll \omega_{\rm at}$). We now show that, in the case of a hydrogenlike ion of atomic number Z, this alternative definition of $I_{\rm cr}$ yields the same Z scaling of $I_{\rm cr}$ as given by Eq. (12b). For a hydrogenlike ion, $\Gamma \equiv \Gamma(Z, \omega/\omega_0, |\mathbf{F}|^2/F_0^2)$ depends on the three dimensionless variables, Z, $\omega/(e^2/\hbar a_0) \equiv \omega/\omega_0$, and $|\mathbf{F}|^2 a_0^3/(e^2/a_0) \equiv |\mathbf{F}|^2/F_0^2$, where $a_0 = (\hbar^2/\mu e^2)$. We define the critical field $F_{\rm cr}$ as the solution of

$$\Gamma(Z, \omega/\omega_0, F_{\rm cr}^2/F_0^2) = \hbar\omega_{\rm at}/2\pi; \tag{18}$$

the solution should be insensitive to ω for $\omega \ll \omega_{\rm at}$. The Schrödinger equation is (in the length gauge)

$$\left\langle \mathbf{x} \left| \left(\frac{\hbar^2}{2\mu}\nabla_{\mathbf{x}}^2 - \frac{Ze^2}{r} - e\mathbf{x}\cdot \mathrm{Re}(\mathbf{F}e^{-i\omega t}) - i\hbar\frac{d}{dt}\right) \right| \Psi(t) \right\rangle = 0, \tag{19}$$

subject to the boundary condition that at time $t = 0$ the electron is in the initial bound state. In terms of the scaled variables $\mathbf{x}' = Z\mathbf{x}$, $\mathbf{F}' = \mathbf{F}/Z^3$, $t' = Z^2 t$, and $\omega' = \omega/Z^2$, the Schrödinger equation and the boundary condition are independent of Z. Substituting the Floquet ansatz for $|\Psi(t)\rangle$ into Eq.(19), and transforming to the scaled variables, we see that to the extent that the Floquet ansatz is valid we have the scaling law

$$\Gamma(Z, Z^2\omega/\omega_0, Z^6|\mathbf{F}|^2/F_0^2)/Z^2 = \Gamma(1, \omega/\omega_0, |\mathbf{F}|^2/F_0^2). \tag{20}$$

[The shift $\Delta \equiv \Delta(Z, \omega/\omega_0, |\mathbf{F}|^2/F_0^2)$ satisfies a similar scaling law.] Now, since the solution of Eq.(18) is insensitive to ω, we are free to multiply ω by any factor β, provided that $\beta\omega \ll \omega_{\rm at}$. We choose $\beta = Z^2$, so that Eq.(18) becomes, dividing both sides by Z^2,

$$\Gamma(Z, Z^2\omega/\omega_0, F_{\rm cr}^2/F_0^2)/Z^2 = \hbar\omega_{\rm at}/2\pi Z^2; \tag{21}$$

the right-hand side of this equation is independent of Z, and comparing with the scaling law (20) we see that $F_{\rm cr}^2$, and therefore $I_{\rm cr}$, scale with Z as Z^6, in accord with Eq.(12b). (Even though the classical potential does not yield an accurate value of $I_{\rm cr}$ in the case of a hydrogenlike ion,[11] we expect it to yield the correct Z scaling.) Incidentally, in the weak-field limit the ionization width can be expressed as $(\mathcal{F}/\mathcal{F}_0)^{N_0}\Gamma'(Z, \omega/\omega_0)$, where $\mathcal{F}/\mathcal{F}_0 \equiv (|\mathbf{F}|/F_0)^2/(\omega/\omega_0)$ is a dimensionless photon flux (and where N_0 is the number of photons required to ionize the atom); it follows from the scaling law that $\Gamma'(Z, Z^2\omega/\omega_0)$ scales with Z as $1/Z^{4N_0-2}$.

IV. IONIZATION OF ATOMIC HYDROGEN

A hydrogen atom in a static electric field has an exceptional symmetry, expressed by the fact that the Schrödinger equation is separable in the parabolic coordinates $\xi = r + z$ and $\eta = r - z$.[4] The static-field problem reduces to the solution of the one-dimensional Schrödinger equations

$$-\frac{\hbar^2}{2\mu}\frac{d^2\psi_1(\xi)}{d\xi^2} + V_1(\xi)\psi_1(\xi) = \frac{E}{4}\psi_1(\xi), \tag{22a}$$

$$-\frac{\hbar^2}{2\mu}\frac{d^2\psi_2(\eta)}{d\eta^2} + V_2(\eta)\psi_2(\eta) = \frac{E}{4}\psi_2(\eta), \tag{22b}$$

where ξ and η are non-negative, and where $V_1(\xi)$ and

$V_2(\eta)$ are effective one-dimensional potentials that replace the classical potential. We have[4]

$$V_1(\xi) = -\frac{Z_1 e^2}{2\xi} + \frac{\hbar^2(m^2-1)}{8\mu\xi^2} + \frac{eF\xi}{8}, \tag{23a}$$

$$V_2(\eta) = -\frac{Z_2 e^2}{2\eta} + \frac{\hbar^2(m^2-1)}{8\mu\eta^2} - \frac{eF\eta}{8}, \tag{23b}$$

where Z_1 and Z_2 are separation constants that sum to Z. The potentials $V_1(\xi)$ and $V_2(\eta)$ are of the same form except for the sign of the term in eF. Recalling that e is negative (and F is positive) $V_1(\xi)$ 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 is obtained by setting $V_1(\xi) = E/4$ at the barrier maximum, for at this field the electron can flow along the ξ axis over the top of the barrier. Restricting ourselves to the ground state ($m = 0$), and using atomic units for the moment, the barrier maximum occurs at the point where

$$F\xi = 4Z_1/\xi + 2/\xi^2. \tag{24a}$$

Putting $V_1(\xi) = E/4$, and eliminating F using Eq. (24a), gives

$$\xi = -(2/E)[Z_1 + \sqrt{Z_1^2 - (3E/8)}] \tag{24b}$$

Now, through second order in an expansion in the perturbation ($eF\xi/8$), we have[4]

$$Z_1 \approx \frac{(-2E)^{1/2}}{2} - \frac{F}{2(-2E)} - \frac{9F^2}{8(-2E)^{5/2}}. \tag{24c}$$

Therefore the separation constant Z_1 can be determined to fairly high accuracy, even for rather large field strengths ($F < 0.2$ a.u.), provided we know E, that is, provided we know the dc Stark shift; the latter is relatively easy to calculate.[16] Inserting ξ from Eq. (24b) into Eq. (24a), and using Eq.(24c) for Z_1, yields a transcendental equation for F_{cr}, which we can solve numerically. We obtain (for the ground state of hydrogen) $F_{cr} \approx 0.15$ a.u., which is more than twice as large as the value (0.0625 a.u.) that would be obtained from Eq. (6), the value based on the classical potential. The reason for this anomalously large value of F_{cr} is that the motions along the ξ and η axes proceed independently, while for atoms other than hydrogen the tunneling along the z axis is coupled to the motion in the plane perpendicular to the z axis. The critical intensity for H(1s) is given by $I_{cr}\cos^2(\zeta/2) \approx 8 \times 10^{14}$ W/cm^2. At the critical field, the dc Stark shift for H(1s) is about 1.4 eV; the dc width is about 0.06 a.u., that is, about $0.8(\hbar\omega_{at}/2\pi)$, and so complete ionization of H(1s) by a dc field, at the critical field strength, occurs in a time very close to the atomic orbital period (closer than we have the right to expect).

Following the lines of previous work,[17] we have calculated the energy shift Δ and the ionization width Γ for H(1s) irradiated by circularly or linearly polarized light of various wavelengths, over an intensity range somewhat below the critical intensity. The results we report here are for wavelengths between 355 and 1064 nm, and for intensities up to 7×10^{14} W/cm^2. We have also calculated the dc shift, Δ_{dc}, and the dc width, Γ_{dc}, over a range of field strengths.[16]

Both the ac and dc shifts are approximately quadratic in the field strength F at values of F well beyond where perturbation theory is applicable.[18] In Fig. 3 we exhibit the departure from quadratic behavior by showing Δ_{dc}/F^2 and, for *circular* polarization, Δ_{ac}/F^2, versus $I \equiv (cF^2/4\pi)$. (Recall that $\Delta_{ac} = \Delta + P$ and that P is linear in F^2. Note also that, for the low frequencies of interest here, P is much larger than Δ_{ac}; for example, at 616 nm Δ_{ac} is only about 3% of P.) If $\omega \ll \omega_{at}$ (and if the light is circularly polarized) the difference between Δ_{dc} and Δ_{ac}, at the same value of I, is small, and this difference approaches zero at sufficiently high intensities. This is apparent from Fig. 3 for the wavelength 616 nm; for the wavelength 1064 nm we have not been able to calculate Δ_{ac} to sufficient accuracy at large intensities to show that Δ_{ac} approaches Δ_{dc}. An analysis of the perturbative expansion of Δ_{ac} may be instructive. In Table I we give the coefficients of the first few terms of the expansion of Δ_{ac} in powers of $F\sec(\zeta/2)$ for various wavelengths and for three different polarizations.[18] The coefficient of the first term (quadratic in F) is independent of polarization, while the coefficients of the higher terms decrease, in absolute magnitude, as the ellipticity ζ increases from 0 to $\pi/2$. The coefficients are negative for all terms below the order F^{2N_0}; at and above the order F^{2N_0} the coefficients are complex. Thus, at a given small intensity the absolute magnitude of the ac Stark shift is slightly larger for linear polarization than it is for circular polarization, the difference being entirely due to terms in F^4 or higher order. It is also clear from Table I that the expansion coefficients for Δ_{ac} converge to the expansion coefficients for Δ_{dc} as the wavelength increases; this was already demonstrated by Pan, Taylor, and Clark[18]

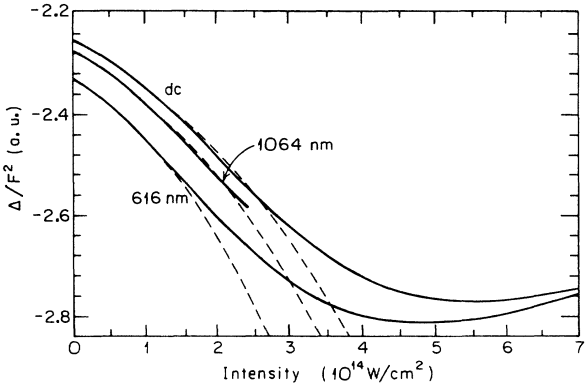

FIG. 3. Stark shifts, divided by the square of the field strength F, vs intensity $I = (cF^2/4\pi)$, for a dc field and for a circularly polarized field of wavelength 616 or 1064 nm. The dashed lines are the shifts obtained by summing the first four terms (the terms in F^2, F^4, F^6, and F^8) of the perturbation series.

TABLE I. Coefficients (in a.u.) of the expansion of the ac Stark shift of H(1s) in powers, N, of $F\sec(\zeta/2) = \sqrt{2}F_{\rm rms}$ (where $F_{\rm rms}$ is the root-mean-square field strength) for fields of (a) 616 nm, (b) 800 nm, (c) 1064 nm, (d) 10355 nm, and (e) "infinite" wavelengths (dc field). The entries (f) give the shortest wavelength (in μm) for which the coefficients are within 1 % of the dc-field limit. Results are given for linear, elliptic, and circular polarization, and for second, fourth, sixth, and eighth orders of perturbation theory. Numbers in square brackets denote powers of 10.

Field	$N=2$	$N=4$	$N=6$	$N=8$
		Linear polarization ($\zeta=0$)		
(a)	−1.165	−2.618[1]	−3.061[3]	−1.064[6]
(b)	−1.149	−2.382[1]	−2.264[3]	−5.032[5]
(c)	−1.139	−2.249[1]	−1.903[3]	−3.413[5]
(d)	−1.127	−2.093[1]	−1.546[3]	−2.199[5]
(e)	−1.127	−2.091[1]	−1.543[3]	−2.190[5]
(f)	1.1	2.9	4.8	6.9
		Elliptic polarization ($\zeta=\pi/3$)		
(a)	−1.165	−1.917[1]	−1.512[3]	−3.240[5]
(b)	−1.149	−1.762[1]	−1.176[3]	−1.780[5]
(c)	−1.139	−1.674[1]	−1.015[3]	−1.291[5]
(d)	−1.127	−1.569[1]	−8.501[2]	−8.909[4]
(e)	−1.127	−1.568[1]	−8.486[2]	−8.875[4]
(f)	1.1	2.7	4.5	6.4
		Circular polarization ($\zeta=\pi/2$)		
(a)	−1.165	−1.683[1]	−9.949[2]	−1.294[5]
(b)	−1.149	−1.556[1]	−8.128[2]	−8.557[4]
(c)	−1.139	−1.482[1]	−7.193[2]	−6.723[4]
(d)	−1.127	−1.395[1]	−6.181[2]	−5.020[4]
(e)	−1.127	−1.394[1]	−6.172[2]	−5.005[4]
(f)	1.1	2.7	4.1	5.7

for linear polarization. In Fig. 3 we compare the exact values of $\Delta_{\rm ac}/F^2$ with those obtained from the perturbation expansion through the term in F^8; evidently the perturbation expansion (truncated at the F^8 term) gives an accurate estimate of $\Delta_{\rm ac}$ up to nearly the intensity at which the curvature of $\Delta_{\rm ac}/F^2$ changes from convex to concave. At somewhat higher intensities $\Delta_{\rm ac}/F^2$ begins to rise as F increases, and this is clearly incompatible with the perturbation expansion, whose coefficients are negative. Note that the expansion through a fixed order (below F^{2N_0} remains accurate to higher F for lower ω, and yet the radius of convergence of the infinite power series approaches zero as ω does.

In Fig. 4 we show the total rate $\Gamma/\hbar$ for ionization of H(1s) by light of wavelength 616 nm, for both circular and linear polarization, versus intensity $I = (cF^2/8\pi)\sec^2(\zeta/2)$. The upper horizontal scale is $E^{(0)} + \Delta$, where $E^{(0)} = -0.499\,73$ a.u. and Δ is approximately the straight line $-1.03P$. The ratio $\omega/\omega_{\rm cr}$ is about 0.2. Note that we show the rate for ionization from the *diabatic* state, that is, the Floquet state whose atomic character is predominantly 1s. The peaks in the rate for ionization by linearly polarized light are due to resonance enhancement that occurs as Rydberg levels shift in and out of resonance with the field. Those Rydberg levels that are brought into resonance by circularly polarized light have very high angular momentum quantum numbers, and resonance enhancement is weakened by the centrifugal barrier that pushes the electron far away from the region of the nucleus — the region where the electron can absorb photons. Hence there are no prominent resonance peaks in the rate for ionization by circularly polarized light, and the resonance peaks that do exist cannot be resolved within our basis set. At very low values of I, in the perturbative regime, the minimum number of photons N_0 that the atom must absorb to ionize is 7. In this regime Γ increases rapidly with increasing I, as I^7; in other words, the index of nonlinearity K, defined as the derivative of $\ln(\Gamma)$ with respect to $\ln(I)$, is 7. However, as I increases further, K decreases, despite the fact that N_0 increases; this is true at all wavelengths, but the decrease of K is more rapid at longer wavelengths — see Fig.2 of Ref. 19 for a plot of the indices of nonlinearity at various wavelengths. One reason[17] that the slope of $\ln(\Gamma)$ decreases as I increases is that the electron charge cloud is forced by the field to oscillate, and the amplitude of oscillation increases as I does, so that the electron spends less time in the region of the nucleus. This intensity regime is the tunneling regime, as we see below. It may be useful to record at this point that the ratio of the excursion amplitude $\alpha \equiv |eF/\mu\omega^2|$ of a free electron oscillating in the field, to the atomic-orbital radius of the initially bound electron, is of order $(\omega_{\rm at}/\omega)/\gamma$.

In Fig. 5 we show the logarithm of $F\Gamma$ versus F_0/F for ionization of H(1s) by circularly polarized light of several different wavelengths. Here $F_0 \equiv \frac{2}{3}(\sqrt{\mu}/e\hbar)|2E^{(0)}|^{3/2}$,

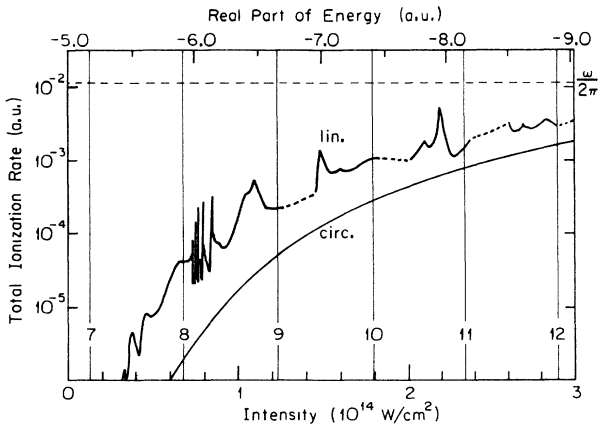

FIG. 4. Ionization width Γ, in a.u., vs intensity (lower horizontal scale) or vs the real part of the quasienergy (upper horizontal scale) for ground-state atomic hydrogen irradiated by circularly (circ.) or linearly (lin.) polarized light of wavelength 616 nm. (The short dashed lines span regions where there are too many resonances for us to calculate, to sufficient accuracy, the width for ionization by linearly polarized light.) The thin vertical lines mark the multiphoton ionization thresholds. At intensities between the $(n-1)$th and the nth thresholds, the minimum number of photons that must be absorbed for ionization to occur is n. As a multiphoton ionization threshold is passed, the "dominant" eigenvalue changes places with a nearby "shadow" eigenvalue, but this interchange is not visible on the figure. The dashed horizontal line marks the value that the width should reach for the ionization time $\hbar/\Gamma$ to equal one cycle, $2\pi/\omega$, of the field. See Ref. 17 for a full description of the calculation.

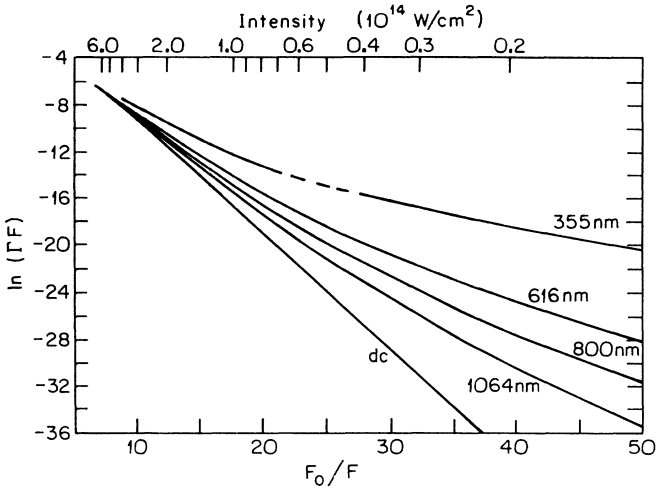

FIG. 5. Logarithm of the product of the width Γ and field strength F, both in a.u., vs F_0/F, for ionization of H($1s$) by either a dc field or a circularly polarized field of various wavelengths (labeled in nm). $F_0 = \frac{2}{3}$ a.u. We evaluated the parameters C and D of Eq. (25) from the y-axis intercept and the slope of the 1064-nm curve extrapolated to $F_0/F = 0$. The dashed line (for 355 nm) is a smooth interpolation through a region of intermediate resonances accumulating at a multiphoton ionization threshold.

that is $\frac{2}{3}$ a.u. (for a nucleus of infinite mass); F_0 is more than a factor of 4 larger than F_{cr}. We also show the rate for ionization of H($1s$) by a dc field of strength F (equal to the strength of the instantaneous ac field). As F increases (F_0/F decreases) the ac rates approach the dc rate from above. The rates can be approximated by the form

$$\Gamma/\hbar \approx (CF_0/F)e^{-(DF_0/F)}, \tag{25}$$

where C and D depend only weakly on the frequency ω; we estimate, from the 1064-nm data, $C \approx 1.7$ a.u. and $D \approx 0.85$.

We interpret the approach of the ac rates towards the dc rate, with increasing F, as the onset of tunneling. This is in qualitative accord with the tunneling theory of Keldysh,[7] a theory that has been developed further by many others, in particular Nikishov and Ritus[20] and Perelomov, Popov, and Terent'ev.[21] We find, from a visual fit to Eq. (25), that tunneling commences at an intensity for which $2P \approx |E^{(0)}|$, when the Keldysh parameter[7] γ is about 1. (This occurs at the following intensities: 6.5×10^{13} W/cm^2 at 1064 nm, 1.1×10^{14} W/cm^2 at 800 nm, and 1.9×10^{14} W/cm^2 at 616 nm.) We have chosen to plot our data to reveal the similarity to the form (25). This form is suggested by the dc-tunneling formula,[22] which is similar but with D equal to 1, close to our value of D, and with C equal to 6 a.u., more than a factor of 3 larger than our value for C. However, at high fields the dc-tunneling formula[22] overestimates the dc widths, by slightly more than a factor of 2 at $F = 0.07$ a.u. for example. The dc tunneling formula represents the dc-rate more accurately as F/F_{cr} *decreases*, and is exact in the asymptotic limit $F/F_{cr} \to 0$, not $F/F_{cr} \to \infty$. For an ac-field, tunneling does not occur until γ is less than roughly unity, and thus Eq. (25) is not an asymptotic form; rather, Eq. (25) is accurate only over a finite range of F for which F is sufficiently large that $\gamma \ll 1$ but sufficiently small that $F/F_{cr} \ll 1$. Therefore the parameters C and D cannot be unambiguously determined from a numerical study, and we should not regard the discrepancy with the dc values too seriously. (We chose to determine C and D as described in the caption to Fig. 5.) We note that tunneling ionization of a model atom, with a zero-range binding potential, has been discussed extensively in the literature,[23] and the convergence of the ac rates toward the dc rate has been demonstrated in several papers. For this zero-range model atom there is no critical intensity for which the electron can flow over the top of the barrier.

If tunneling ionization by a low-frequency field were no different from tunneling ionization by a static field, the parameters C and D in Eq. (25) would be independent of the polarization of the (oscillating) field. We do not expect the value of D to be strongly dependent on the polarization (since DF_0 is presumably a characteristic of the atom), though of course a small change in D is significant since it occurs in an exponential. However, 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 the electric field has magnitude F at all times, and the electron can tunnel out at every moment of the cycle, while in the case of linear polarization the electron is most likely to tunnel out during the times where the magnitude of the electric field is close to its extremal value F. Equation (25) can be generalized[21] to linear polarization by replacing F by $F|\cos(\omega t)|$ and averaging the resulting expression over one cycle using the result

$$\frac{2}{\pi}\int_0^{\pi/2} d\tau\,[F_0/F\cos(\tau)]e^{-D[F_0/F\cos(\tau)]} = (2F_0/\pi F)K_0(DF_0/F). \quad (26)$$

Noting that the modified Bessel function $K_0(x)$ has the asymptotic form $(\pi/2x)^{1/2}e^{-x}$ for large x, we see that only the preexponential factor is changed in Eq. (25) — this factor becomes $C'(F_0/F)^{1/2}$, where $C' = (2/\pi D)^{1/2}C$. Using our values of C and D gives $C' \approx 1.5$ a.u. Confirmation of this law, for linear polarization, is difficult due to the presence of numerous intermediate resonances. However, in Fig. 6 we show the logarithm of $F^{1/2}\Gamma$ versus F_0/F for ionization of H($1s$) by linearly polarized light at wavelengths 616 and 1064 nm. The 616-nm results are consistent with the law

$$\Gamma/\hbar \approx C'\sqrt{(F_0/F)}e^{-(DF_0/F)}, \quad (27)$$

provided we choose $C' \approx 0.4$ a.u. and $D \approx 0.75$ (though D is uncertain to at least 0.03, which results in a much larger uncertainty in C'). We also show, in Fig. 6, the result obtained by averaging the dc width over one cycle after this width is evaluated at the instantaneous field $F\cos(\omega t)$ at each moment of the cycle. (This cycle-averaged dc width is independent of the frequency ω.) We see that the ac width approaches the cycle-averaged dc width at sufficiently high field strengths. We can cycle-average the dc shift in the same way, and we find that at sufficiently high field strengths the ac shift approaches the cycle-averaged dc shift. In fact, we can state quite generally that for *arbitrary* polarization and for $\omega < \omega_{at}$ the ac shifts and ac widths approach the cycle-averaged dc shifts and dc widths as the field strength increases or as the frequency decreases.

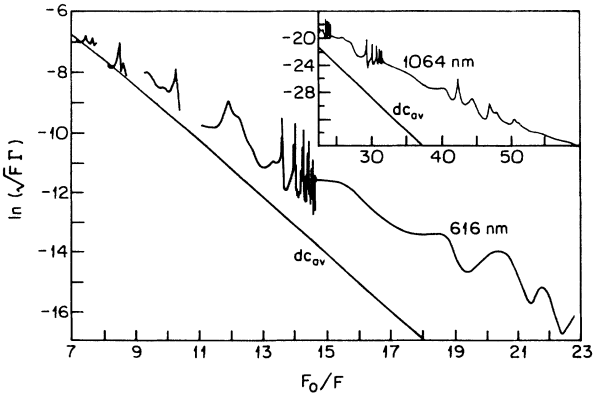

FIG. 6. Logarithm of the product of Γ and $\sqrt{F}$, in a.u., vs F_0/F, for ionization of H($1s$) by either a dc field or a linearly polarized field of wavelength 616 or 1064 nm. The dc width has been cycle-averaged over the instantaneous field. We evaluated the parameters C' and D of Eq. (27) from the y-axis intercept and the slope of the 616-nm curve extrapolated to $F_0/F = 0$. See Ref. 17 for a full discussion of the ac rates at intensities below the tunneling regime.

We saw from Fig. 4 that the rate for ionization by circularly polarized light is lower than that for linearly polarized light at the same value of the intensity I; this is not true, at least in the tunneling regime, when we compare rates at the same value F of the maximum instantaneous field [values of I differing by $\sec^2(\zeta/2)$]. In the tunneling regime the rate for ionization by circularly polarized light is slightly higher than that for ionization by linearly polarized light at the same value F of the maximum instantaneous field; this is because for circular polarization the electron can tunnel out at all moments of the cycle with equal probability. However, in the perturbative regime, circularly polarized light is less efficient, at long wavelengths (e.g., 1064 nm), than linearly polarized light at the same value of F; this is because fewer channels and intermediate resonances are accessible.[24]

We have not been able to carry out calculations of the ac width close to and beyond the critical intensity. To do so we would need a prohibitively large (or significantly more flexible) basis set; numerical instabilities have prevented us from going much beyond 3×10^{14}W/cm^2 at long wavelengths.[25] As we already mentioned, the Floquet method becomes inadequate when Γ is of the order of or exceeds $\hbar\omega$, but, nevertheless, $\hbar/\Gamma$ still gives an indication of the time required for ionization. We have been able to calculate the dc width for H($1s$) at intensities beyond I_{cr}, and we find that it continues to increase monotonically, but without any dramatic increase, as I passes I_{cr}.

Of course, since ionization is complete in a time of order of the atomic-orbital period at the critical intensity, the saturation intensity for a realistic laser pulse cannot be much greater than I_{cr}. We have carried out calculations of the ground-state population of hydrogen atoms as a function of time for atoms at the focus of a pulse that has a Gaussian temporal profile. We can ask what should the maximum intensity of a pulse of duration 100 fsec [full width at half maximum (FWHM)] be if at least 10% of the atoms (at the laser focus) are to experience this maximum intensity (before undergoing ionization)? The answer is, for circularly polarized light, 2.1×10^{14} W/cm^2 at 1064 nm or 1.8×10^{14} W/cm^2 at 616 nm. We can rephrase the question and ask what should the maximum pulse duration (FWHM) be if at least 10% of the atoms are to experience a specified maximum intensity? Taking the polarization to be circular again, and choosing a maximum intensity for which $\gamma = 1$ (roughly the intensity for the onset of tunneling), the answer is 1.5 nsec at 1064 nm or 80 fsec at 616 nm. If we specify the maximum intensity as 3×10^{14} W/cm^2 — an intensity

for which the 616-nm ac rate is only a factor of 1.9 larger than the cycle-averaged dc-rate — the answer is only 8 fsec at 616 nm. Note that at an intensity of about 5×10^{14} W/cm^2 the ionization rate is about $\omega/2\pi$ for wavelengths in the range 616–1064 nm, in which case the atoms ionize within roughly one cycle. We conclude that for pulses of duration of a few hundred fsec or longer, the saturation intensity for ionization of H($1s$) is no more than roughly 2×10^{14} W/cm^2, even when the spatial profile of the pulse is taken into account. This saturation intensity is below the critical intensity. On the other hand, since the critical intensities for rare gases are much lower than for hydrogen, the saturation intensities for rare gases can be comparable to I_{cr}.

We note again[19] that to obtain an accurate width Γ, we do not always need to include as many harmonic components $|\mathcal{F}_n\rangle$ as one would think based on the characteristic number N_{ch} of photons that the electron ultimately absorbs. As Gallagher[1] and Corkum, Burnett, and Brunel[2] have pointed out, N_{ch} can be estimated in the tunneling regime by calculating the drift velocity that a free electron has if it is released at the instant t_0 into the field of Eq. (1b). If $\mathbf{v}(t)$ is the instantaneous velocity of the free electron at time $t \geq t_0$, we have $\mu\, d\mathbf{v}(t)/dt = e\,\mathrm{Re}(\mathbf{F}e^{-i\omega t})$, and assuming the electron is released with zero instantaneous velocity, we have $\mathbf{v}(t) = \mathbf{u}(t) - \mathbf{u}(t_0)$, where

$$\mathbf{u}(t) = (eF/\mu\omega)[\sin(\omega t)\hat{\mathbf{z}} - \tan(\zeta/2)\cos(\omega t)\hat{\mathbf{x}}]. \tag{28}$$

Hence the drift (cycle-averaged) velocity of the free electron is $-\mathbf{u}(t_0)$, and the drift energy is $\frac{1}{2}\mu|\mathbf{u}(t_0)|^2$. For circular polarization ($\zeta = \pi/2$) we have $|\mathbf{u}(t_0)| = |eF/\mu\omega|$, independently of t_0, and so the drift energy is approximately P.[26] Taking into account that N_0 photons must be absorbed to produce a free electron with zero drift velocity, we see that in the case of circular polarization $N_{ch} \approx 2N_0$ when $P \gg |E^{(0)}|$, that is, when $\gamma \ll 1$. However, many of these photons are absorbed *after* the electron has tunneled out through the barrier, and do not strongly affect the ionization rate $\Gamma/\hbar$; we find[25] that it is sufficient to take a maximum value of about $1.5N_0$ for the photon index n in $|\mathcal{F}_n\rangle$ (this is true for circular or linear polarization). Of course, a statement as to when photons are absorbed is not gauge invariant;[27] no experiment can decide whether the photons were absorbed before or after the electron tunneled out. The notion of tunneling is appropriate to the length gauge, where the electron-field interaction is $-e\mathbf{x}\cdot\mathrm{Re}(\mathbf{F}e^{-i\omega t})$; in this gauge one imagines that *most* of the photons are absorbed after the electron has tunneled out. This suggests that to calculate, in the length gauge, an accurate ac rate for ionization by circularly polarized light (in the tunneling regime) one should not have to let the photon index n greatly exceed the maximum orbital angular momentum quantum number that must be included to calculate an accurate dc width. Naturally, to calculate the photoelectron energy distribution, we need to include very many harmonic components, significantly more than N_{ch}. Incidentally, the photoelectron energy distribution, for ionization at a fixed intensity such that $\gamma \ll 1$, has been calculated on the basis of the Keldysh theory by Nikishov and Ritus[20] and by Perelomov, Popov, and Terent'ev.[21] They showed that for linear polarization the distribution peaks near threshold, so that $N_{ch} \approx N_0$, while for circular polarization the distribution is a Gaussian with a peak at an energy[26,28] close to P, and with a full width at half maximum of $2[N_{ch}\ln(2)/(\sqrt{2}\gamma)]^{1/2}$ where $N_{ch} \approx 2N_0(1-2\gamma^2/3)$.

Finally, we briefly consider the behavior of the ionization rate with respect to intensity when the frequency exceeds the atomic-orbital frequency ω_{at}. In Fig. 7 we show total rates for ionization from the $1s$, $2s$, $4p$, and $4f$ levels versus γ, for a fixed frequency ($>\omega_{at}$) and linear polarization. The frequency is 0.65 a.u. (70 nm) for ionization from the $1s$ level, 0.17 a.u. (266 nm) from the $2s$ level, and 0.043 a.u. (1064 nm) from the $4p$ and $4f$ levels. Note that, except for the $1s$, $2s$, $2p$, and $3p$ levels, the *un*perturbed states within a given Rydberg manifold (specified by the principal quantum number) are, due to the degeneracy in hydrogen, superpositions of atomic states with the same parity but different orbital angular momentum quantum number.[29] Thus the odd-parity unperturbed states belonging to the Rydberg manifold with principal quantum number 4 are superpositions of $4p$ and $4f$ states. The label $4p$ implies that the $4p$ state is the dominant atomic state in that superposition; the label $4f$ has a similar meaning. (Of course, the perturbed states are superpositions of atomic states of both even and odd parity.) Looking at Fig. 7, we see that as the intensity increases (γ decreases) the rates each exhibit a peak at about $\gamma = 1$. With the exception of the rate from the $4f$

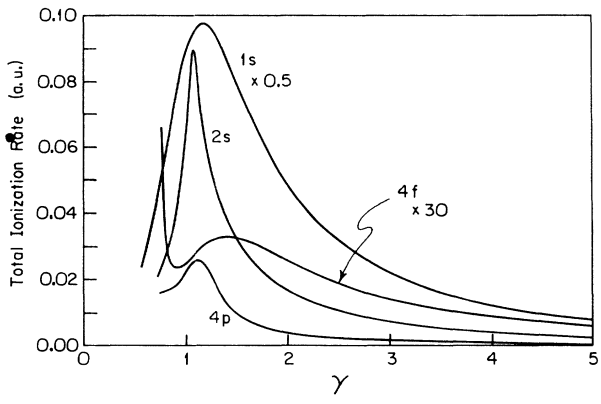

FIG. 7. Total rate $\Gamma/\hbar$ for ionization of H by "high"-frequency linearly polarized light, from four different levels, vs the Keldysh parameter γ. The rates for ionization from the $1s$ and $4f$ levels have been multiplied by the factors indicated. The frequency of the light is 0.65 a.u. for ionization from the $1s$ level, 0.17 a.u. from the $2s$ level, and 0.043 a.u. from the $4p$ and $4f$ levels. Note that the labels $4p$ and $4f$ are merely indicative, since the angular momentum quantum number is not a good quantum number for labeling unperturbed states, except for the $1s$, $2s$, $2p$, and $3p$ states (see text).

level (a point we return to shortly) these rates drop monotonically as the intensity increases beyond the value at which the peak occurs.[30] This monotonic decrease can be understood as follows: when the intensity increases, the strength of the coupling of the electron to the field also increases. However, with increasing intensity, the electron spends less time near the nucleus, where it can absorb photons. As noted in the discussion of Fig. 4 above, and depicted more clearly elsewhere,[19] if $\omega < \omega_{at}$, the index of nonlinearity decreases with increasing intensity; nevertheless, the ionization rate continues to increase since ionization occurs through tunneling. On the other hand, if $\omega > \omega_{at}$ tunneling is no longer possible (recall the last paragraph of Sec. II), and, rather than interpret γ as the ratio of the tunneling time to the cycle time, it is more appropriate when $\omega > \omega_{at}$ to interpret $1/\gamma$ as the ratio of the characteristic "quiver" speed of a free electron oscillating in the field to the characteristic atomic-orbital speed of the bound electron; thus $\gamma \approx 1$ implies the electron is almost free and therefore cannot easily absorb photons. The anomalous behavior of the rate from the $4f$ level — this rate exhibits a peak at $\gamma \approx 1$ but then exhibits a minimum and subsequently increases sharply as I increases further — is due to a three-photon intermediate resonance with a state that has $2s$ character. This resonance corresponds to an avoided crossing of the real parts of the $4f$ quasienergy and another quasienergy of $2s$ character[31] and is responsible for the rise in the $4f$ rate for larger I. A fuller account of the behavior of ionization at frequencies $\omega > \omega_{at}$ will be given elsewhere, but we end here by noting that a "stable" atom still oscillates, and therefore provides a continuous source of high-frequency radiation.

V. CONCLUSION

There is a critical intensity, I_{cr}, above which atoms undergo rapid ionization, in a time period of the order of the atomic-orbital period ($2\pi/\omega_{at}$), provided that the frequency ω is less than ω_{at}. We have derived a simple expression for I_{cr}, applicable to atoms other than hydrogen. The critical intensity for atomic hydrogen is significantly higher than for the rare gases; this is due to the exceptional symmetry of a hydrogen atom in a static field. We have performed Floquet calculations for ionization of H($1s$), and have shown that if $\omega < \omega_{at}$ tunneling ionization begins at roughly the intensity for which the Keldysh parameter γ is unity. The ac shift and ac width approach the cycle-averaged dc shift and dc width, respectively, as either the wavelength or intensity increases. The saturation intensity for H($1s$) turns out to be significantly below I_{cr}, except for unusually short pulses, but the saturation intensity for rare gases is comparable to I_{cr}. For $\omega > \omega_{at}$ there is no tunneling regime, and, in the absence of a strong intermediate resonance, the rate peaks at about $\gamma = 1$.

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*Present address: Physique Théorique, Free University of Brussels, Brussels, Belgium.

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[25] For circularly polarized light of wavelength 1064 nm our largest basis consisted of 70 Floquet blocks, with photon index ranging from −9 to 60, 4 angular momentum components per block, and 35 Sturmian functions per angular momentum component. For linear polarization at 616 nm our largest basis consisted of 46 Floquet blocks, with photon index ranging from −10 to 35, 10 angular momentum components per block, and 40 Sturmian functions per angular momentum component.

[26] The momentum $(2\mu P)^{1/2}$ can be far higher than the characteristic orbital momentum that the electron has in the bound state from which ionization occurs. However, since the residual ion has an equal but opposite drift momentum, no net momentum is removed from the field (in the dipole approximation). The momentum transmitted to the electron originates in the scattering from the atomic core as the initially bound electron orbits this core in the presence of the radiation field. See W. E. Cooke and R. Shakeshaft (unpublished) for further discussion. Note, however, that the scattering is a *slow* process on the atomic scale, taking place over many atomic orbital periods (but within a fraction of a cycle of the radiation field). The *sudden* (on the atomic scale) scattering mechanism suggested earlier by one of the present authors [R. Shakeshaft, Z. Phys. D **14**, 271 (1989)] is not applicable.

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[28] The suppression of electrons with drift energy below P, in the case of circular polarization, can also be understood in terms of the angular momentum barrier. An electron that has absorbed N photons has an orbital angular momentum quantum number at least as large as N, but if it has insufficient energy it will be unable to pass through the angular momentum barrier. See Ref. 20 above, and also P. H. Bucksbaum, M. Bashkansky, R. R. Freeman, T. J. McIlrath, and L. F. DiMauro, Phys. Rev. Lett **56**, 2590 (1986).

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