Time-independent theory of multiphoton ionization of an atom by an intense field

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We formulate a computationally feasible method for calculating multiphoton ionization rates for atoms exposed to intense fields in the intensity regime where perturbation theory ceases to apply. The method is based on the time-independent picture of ionization, which starts with the Floquet ansatz. The question of the gauge of the radiation field is discussed in some detail. Various expressions for the ionization amplitude are derived from a variational principle, with the radiation field expressed in the velocity gauge. We consider in particular an approximation in which the wave vector developing from the initial state is replaced by a trial vector that is the Floquet expansion truncated just below the threshold for ionization, and in which the wave vector developing into the final state is replaced by a trial vector that is just the wave vector appearing in the Kroll-Watson low-frequency approximation in scattering theory. We have applied this approximation to hydrogen and we present some results for both nonresonant and resonant multiphoton ionization. We argue that the experimentally observed resonance structure in the above-threshold peaks of the ionization signal occurs through the electron jumping from one dressed-state energy-eigenvalue curve to another.

I. INTRODUCTION

With the advent of high-power, low-frequency lasers it has become possible to observe atomic electrons that absorb many photons, far more than the minimum number N_0 required for the atom to ionize.[1-4] A variety of theories[5-6] have been introduced to describe the crude features of the ionization process.[7] However, a more sophisticated approach is required to account for the detailed aspects of the *atomic* dynamics of multiphoton ionization. (We are not concerned here with the macroscopic, essentially classical, motion of the electron after it leaves the atom and travels out through the laser focus.) In the present paper we describe a fairly ambitious but computationally feasible approach which is based on the time-independent picture of multiphoton ionization. We treat the radiation field as classical and spatially homogeneous, and we assume the atomic system to have only one active electron. We aim at presenting a fairly comprehensive, but certainly not exhaustive, prescription within this framework.

In the time-independent picture, the ionization rate is constant in time and the probability for the electron to remain in the initial bound state follows the law of exponential decay. One cannot obtain detailed information about the time evolution of the electron probability density without numerically solving the time-dependent Schrödinger equation. Significant progress toward that end has been made by Kulander,[8] who calculated the time-dependent probability for electron loss from the ground state of a hydrogen atom exposed to an intense field. However, because of the limitation on computer time, the size of the grid chosen for the calculation was insufficient to obtain information about partial rates for ionization into specific continuum channels. Other, possibly more powerful, methods are being developed[9] for solving the time-dependent Schrödinger equation, but the computer time required for such calculations may still be prohibitive, except in circumstances where complete ionization takes place in a short time, less than, say, 100 cycles; a time-independent approach is less costly.

To pass to a time-independent picture of multiphoton ionization, we must assume that the radiation field is monochromatic (frequency ω) and that the electron has a well-defined energy. The passage is accomplished by the Floquet ansatz:[10] The exact wave vector $|\Psi_i(t)\rangle$, which represents the electron initially bound in state i, is approximated by a trial state vector

$$|F_i(t)\rangle = \exp(-i\varepsilon_i t/\hbar)\,|\psi_i(\tau)\rangle\ ,$$

where $|\psi_i(\tau)\rangle$ is a periodic function of $\tau=\omega t$ with period 2π, and where ε_i is a complex quasienergy with a negative imaginary part $-\Gamma_i/2$. Although $|F_i(t)\rangle$ satisfies the time-dependent Schrödinger equation so long as the Hamiltonian is harmonic, the harmonicity is disrupted during the time that the radiation field is turned on. Consequently, $|F_i(t)\rangle$ does not satisfy the boundary condition that the electron is initially localized in state i. In fact, $|F_i(t)\rangle$ is nonnormalizable; more importantly, $\langle \mathbf{x}\,|F_i(t)\rangle$ explodes exponentially as the radial electron coordinate $r=|\mathbf{x}|$ increases. This is a consequence of representing the electron as having a spectrum of discrete energies $\varepsilon_i+n\hbar\omega$. In reality, the electron has a spectrum of real energies $\mathrm{Re}(\varepsilon_i)+n\hbar\omega$ that are broadened with an induced width Γ_i. The electron should be represented by a normalized wave packet that initially describes the electron as localized in a bound state, and that at large times is peaked in energy space at the points $\mathrm{Re}(\varepsilon_i)+n\hbar\omega$, with each peak having a width at least equal to Γ_i. If Γ_i could simply be set to zero, $\langle \mathbf{x}\,|F_i(t)\rangle$ would not explode

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as r increases. However, we are not free to choose ε_i; it is an eigenvalue, and $|\psi_i(\tau)\rangle$ is the solution to a homogeneous eigenvalue equation. To circumvent this problem, we introduce a source term into the equation satisfied by $|\psi_i(\tau)\rangle$; this permits us to replace ε_i by $\mathrm{Re}(\varepsilon_i)$. The modified $|F_i(t)\rangle$ does not explode exponentially; rather, $\langle \mathbf{x}|F_i(t)\rangle$ now behaves as a physically meaningful superposition of outgoing waves as r increases, *provided that the radiation field is represented in the velocity gauge.* Note that $|F_i(t)\rangle$ contains no information about how the field was turned on, and so the normalization of $|F_i(t)\rangle$, which in fact depends on the gauge, must be fixed by the state of the electron just after the field has been turned on. We show how this is done. To derive the N-photon ionization amplitude we start from a variational principle. We derive (in the velocity gauge) two different expressions for this amplitude, based on approximating $|\Psi_i(t)\rangle$ by the modified trial $|F_i(t)\rangle$. We also derive further approximations that result from truncating the harmonic expansion of $|\psi_i(\tau)\rangle$. In particular, we reformulate an approximation that we introduced earlier,[11] the "threshold-truncated approximation" (TTA), and we present results of nonperturbative calculations based on this approximation.

The TTA starts with the replacement of $|\Psi_i(t)\rangle$ by $|F_i(t)\rangle$ and the subsequent truncation of the harmonic expansion of $|\psi_i(\tau)\rangle$, whose nth term represents the absorption of n photons. We truncate this series at the (N_0-1)th term, that is, just below the threshold for ionization. While this is a fairly severe approximation, the resulting trial wave function is normalizable (in the velocity gauge) and therefore easy to handle. Moreover, we attempt to compensate for the error in this trial wave function by choosing a reasonable trial approximation to the electron state vector $|\Psi_f(t)\rangle$, which develops into the final unbound scattering state where the electron has absorbed $N \geq N_0$ photons. The radiation field is taken to be a low-frequency field that oscillates slowly, and we suppose that the atomic scattering time is short compared to the cycle time of the field. Thus the electron scatters from the atomic potential as if the field is absent; the field acts only in the asymptotic region where the electron moves as a free particle with an instantaneous mechanical momentum that oscillates. In the length gauge the canonical momentum of a free particle in a field is just the instantaneous mechanical momentum, and therefore in this gauge we can, at each time t, approximate $|\Psi_f(t)\rangle$ by a scattering state eigenvector of the atomic Hamiltonian which at asymptotically large distances approaches the state vector of an otherwise free electron moving through the field. (The corresponding wave vector in the velocity gauge may be obtained from a gauge transformation.) This approximation is equivalent to the low-frequency approximation made by Kroll and Watson in their classic paper[12] on potential scattering in a laser field. In our picture of the N-photon ionization process the electron first climbs the Floquet ladder to the (N_0-1)th step, through the evolution of our approximate $|\Psi_i(t)\rangle$; it then absorbs a single photon and jumps into the continuum, after which it absorbs a further $N-N_0$ photons through the evolution of our approximate $|\Psi_f(t)\rangle$. Our approximate N-photon ionization amplitude is just the time average (over one cycle) of the matrix element for the electron to jump into a specific continuum channel from the (N_0-1)th step of the Floquet ladder [see Eq. (4.40)]. We previously calculated generalized cross sections, in the length gauge, for ionization of hydrogen in the weak-field limit and we compared[11] our results rather favorably with those obtained from perturbation theory. We present here results of a more detailed study of ionization in the weak-field limit, and we illuminate differences between the length and velocity gauges which arise because of approximations made in arriving at our weak-field ionization amplitude. We have also extended our calculations to ionization of hydrogen by moderately strong fields, beyond the weak-field limit, and we report some results below. We show results for both nonresonant and resonant ionization by strong fields, and we present a (limited) comparison with recent experimental data.[3] We have explored the influence of an intermediate resonance by expressing the electron wave vector $|\Psi_i(t)\rangle$ as a linear combination of two Floquet eigenvectors whose quasienergy eigenvalues vary with intensity and almost coincide (to within an integral multiple of $\hbar\omega$) at the resonance intensity. The time-dependent coefficients of the two eigenvectors are determined from the solution of two coupled differential equations. Recently Freeman *et al.*[4] observed, in an experiment where xenon was irradiated by a short pulse, secondary peak structure in the above-threshold peaks of the ionization signal. They attributed this structure to resonance enhancement; we argue further that the structure could not occur without the electron jumping from one dressed-state energy-eigenvalue curve to another.

In Sec. II we describe the radiation field and its interactions, in the length and velocity gauges, with the electron. In Sec. III we examine the Floquet ansatz and its physical significance. We derive a modified Floquet equation whose solution does not explode exponentially in coordinate space. In Sec. IV we introduce a variational principle for the ionization amplitude. The input to this variational principle is the solution to the modified Floquet equation, or an approximation to it. We show that provided one works in the velocity gauge a finite expression for the ionization amplitude is obtained; the corresponding expression in the length gauge is infinite. We discuss in some detail the TTA, which follows from using a trial wave function obtained by truncating at the ionization threshold the Floquet expansion in harmonic components. In Sec. V we present numerous results of calculations based on the TTA. In Appendix A we describe the state of the electron, in each of the two gauges, immediately after the field has been turned on, assuming no depopulation of the initial state has occurred during the rise time of the field. From this we may determine the appropriate normalization of the (modified) Floquet wave vector. In Appendix B we discuss the asymptotic motion of the electron, before the field is turned off. In Appendix C we briefly discuss perturbation theory. In Appendix D we derive the time-dependent coupled equations for the Floquet wave vectors at intensities where there is an avoided crossing of the eigenvalue curves.

II. RADIATION FIELD AND ITS INTERACTION WITH THE ELECTRON

We describe the radiation field as a classical, monochromatic field that is spatially homogeneous over atomic dimensions and has the electric field vector

$$\mathbf{F}(\tau)=\mathrm{Re}(F_0\hat{\boldsymbol{\epsilon}}e^{-i\tau}) \tag{2.1}$$

and vector potential

$$\mathbf{A}(\tau)=(c/\omega)\mathrm{Im}(F_0\hat{\boldsymbol{\epsilon}}e^{-i\tau}) . \tag{2.2}$$

Here $\tau=\omega t$, with ω the frequency and t the time, F_0 is the real electric field amplitude, and $\hat{\boldsymbol{\epsilon}}$ is the (possibly complex) unit polarization vector. The field amplitude does vary in space over the macroscopic dimensions of the focal region of the laser, and of course it varies in time during the intervals that the laser is turned on and off. We confine our attention in this study to a macroscopically small volume inside the laser focus, a volume which contains many atoms but over which the spatial variation of the field is negligible. We do not consider what happens after the active electron leaves this volume and scatters out of the laser focus. In particular, we do not take into account the effect[2] of spatial variations of the laser intensity on the energy spectrum and angular distribution of the electron. It is not difficult to correct for this.[2]

Initially, before the field is turned on, the electron is bound to an atomic potential W in the state i represented by

$$|\Phi_i^{(0)}(t)\rangle=e^{-iE_it/\hbar}|\Phi_i\rangle , \tag{2.3}$$

where $-E_i$ is the binding energy. At some finite time, say, $t=0$, the field is turned on. We assume that the field is turned on sufficiently slowly so that its resulting bandwidth $\Delta\omega$ is small compared to the frequency ω, so that it is realistic to treat the field as monochromatic.

In the length gauge the interaction of the electron with the field is

$$V^{(l)}(\tau)=-e\mathbf{F}(\tau)\cdot\mathbf{x} , \tag{2.4a}$$

while in the velocity gauge this interaction is

$$V^{(v)}(\tau)=-(e/\mu c)\mathbf{A}(\tau)\cdot\mathbf{p} , \tag{2.4b}$$

where e and μ are the charge and mass of the electron and $\mathbf{x}$ and $\mathbf{p}$ are its position and canonical momentum. (Here and elsewhere we sometimes insert a superscript v or l to distinguish the gauge.) The electron is governed by the time-dependent Schrödinger equation,

$$i\hbar\frac{d}{dt}\left|\Psi(t)\right\rangle=H(\tau)|\Psi(t)\rangle , \tag{2.5}$$

where, with $V(\tau)$ equal to $V^{(l)}(\tau)$ or $V^{(v)}(\tau)$, we have

$$H(\tau)=H_a+V(\tau) , \tag{2.6a}$$

where H_a is the atomic Hamiltonian,

$$H_a=(p^2/2\mu)+W . \tag{2.6b}$$

The instantaneous mechanical momentum of the electron is $\mu\,d\mathbf{x}/dt=(i/\hbar)[H(\tau),\mu\mathbf{x}]$, which in the length gauge is just the canonical momentum $\mathbf{p}$ but which in the velocity gauge is $\mathbf{p}-\hbar\mathbf{k}_A(\tau)$, where $\hbar\mathbf{k}_A(\tau)=e\,\mathbf{A}(\tau)/c$. In the velocity gauge $\mathbf{p}$ is the *mean* mechanical momentum, averaged over one cycle. Note that we have omitted the term $V_0(\tau)=e^2\mathbf{A}^2(\tau)/(2\mu c^2)$ from the interaction $V^{(v)}(\tau)$, while $V^{(l)}(\tau)$ includes $V_0(\tau)$. We may rewrite $V_0(\tau)$ as $\hbar^2\mathbf{k}_A^2(\tau)/2\mu$, which is the instantaneous kinetic energy of a free electron that is oscillating in the field, but is on average at rest. Since $V_0(\tau)$ is spatially independent over atomic dimensions it cannot influence the microscopic motion of the electron. It can only influence (through its macroscopic spatial variation) the motion of the electron as it travels through the laser focus *after* it has left the potential W to which it was bound. Were we to include $V_0(\tau)$ in $V^{(v)}(\tau)$ we could formally remove it through a gauge transformation,[13] which would amount to absorbing it in an irrelevant phase factor multiplying the electron wave function,

$$|\Psi(t)\rangle\rightarrow e^{-i(Pt/\hbar)-i\zeta(\tau)}|\Psi(t)\rangle . \tag{2.7}$$

Here P is the ponderomotive energy shift of a free electron, which is just the time average over one cycle of $V_0(\tau)$, that is,

$$P=e^2F_0^2/(4\mu\omega^2) , \tag{2.8}$$

and $\zeta(\tau)$ is a periodic function, with period 2π, defined by

$$\zeta(\tau)=\frac{1}{\hbar}\int_0^t dt'[V_0(\tau')-P] . \tag{2.9}$$

Substituting the right-hand side of Eq. (2.7) into both sides of Eq. (2.5) results in modifying $H(\tau)$ by subtracting the term $V_0(\tau)$. The interaction $V^{(l)}(\tau)$ shifts the (cycle-averaged) energy of a free electron upwards by P, and shifts the initial bound energy level by an amount $\Delta_i^{(l)}$ which may be negative. For frequencies well below the smallest resonant frequency we expect $|\Delta_i^{(l)}|\ll P$. The interaction $V^{(v)}(\tau)$ does not shift the energy of a free electron,[14,15] but it shifts the initial energy level by $\Delta_i^{(v)}=\Delta_i^{(l)}-P$. A wave vector $|\Psi^{(l)}(t)\rangle$ in the length gauge is related to the corresponding wave vector $|\Psi^{(v)}(t)\rangle$ in the velocity gauge by[16]

$$|\Psi^{(v)}(t)\rangle=e^{i(Pt/\hbar)+i\zeta(\tau)+i\mathbf{x}\cdot\mathbf{k}_A(\tau)}|\Psi^{(l)}(t)\rangle . \tag{2.10}$$

III. FLOQUET ANSATZ

In this section we discuss the Floquet ansatz, and in particular its physical significance. Starting from the equation satisfied by the Floquet wave vector, we develop a modified equation whose solution does not diverge exponentially in coordinate space.

A. "Ingoing" wave vector

The wave vector $|\Psi_i(t)\rangle$ which develops from the initial *bound* state i can be approximated by the Floquet form[10]

$$|F_i(t)\rangle=e^{-i\varepsilon_it/\hbar}|\psi_i(\tau)\rangle , \tag{3.1}$$

where $|\psi_i(\tau)\rangle$ is periodic with period 2π. The complex quasienergy ε_i may be expressed as $\varepsilon_i = E_i + \Delta_i - i\Gamma_i/2$, where Δ_i is the (gauge-dependent) shift in the initial energy E_i and where Γ_i is the positive (gauge-independent) width. The harmonicity of the Hamiltonian $H(t)$ is destroyed during the interval that the field is turned on; were it not for this, the Floquet ansatz would be exact. Stated another way, turning on the field introduces an essential boundary at the *finite* time $t=0$; once the field is on, the atom begins to disintegrate, and it totally breaks up after a characteristic time $\hbar/\Gamma_i$.

The Floquet wave vector $|\psi_i(\tau)\rangle$ contains no information about the way the field was turned on. Therefore the replacement of $|\Psi_i(t)\rangle$ by $|F_i(t)\rangle$ is justified only if the initial state is not significantly depopulated during the rise time t_0 of the field. A necessary condition is $2\pi/\omega \ll t_0 \ll \hbar/\Gamma_i$. If t_0 is long, and significant depletion occurs during the rise time, we may easily (in principle) account for the accumulated depletion provided that the state of the electron evolves adiabatically as the laser intensity I varies; we simply include the parametric dependence of ε_i and $|\psi_i(\tau)\rangle$ on I and we make the change

$$\exp(-i\varepsilon_i t/\hbar) \rightarrow \exp(-i\int^t \varepsilon_i dt'/\hbar)$$

in Eq. (3.1). However, in order to integrate ε_i over time we need some knowledge of how the field was turned on, that is, we need to know how I varies with time. If the laser frequency is close to a resonant frequency, we must assume that I varies very slowly with time and that the frequency bandwidth of the field is small compared to the detuning from resonance. Otherwise, the electron state will evolve nonadiabatically into a superposition of two or more Floquet states with time-dependent mixing coefficients that are determined from coupled differential equations; this situation will be considered in Sec. V when we present some results for resonant multiphoton ionization by a short pulse.

To simplify our discussion we assume throughout this section and Sec. IV that the initial atomic bound state is not significantly depopulated during the rise time of the field. As we just mentioned, in Sec. V we illustrate how our formulation can be generalized to treat resonant multiphoton ionization, a circumstance in which depletion does occur during the rise time. We envisage for the present that the field is turned on at $t=0$, reaches its peak intensity at $t=t_0$, and has constant intensity thereafter. In theory, we assume that the field is not turned off until $t=\infty$. In practice, all that is necessary is that the field be turned off some time after the electron has left the range of the atomic potential since the mean motion of a free electron is unaffected by the (spatially homogeneous) field and how it is turned off, as long as the field is turned off adiabatically. In Appendix A we discuss the state of the electron immediately after the field has been fully turned on (assuming that no depopulation of the initial atomic state has occurred). We show there that for t not much greater than t_0 the state of the electron is represented by

$$|\Phi_i^{(l)}(t)\rangle = \exp(-i\Delta_i^{(l)}t/\hbar)\,|\Phi_i^{(0)}(t)\rangle \tag{3.2a}$$

in the length gauge, and by

$$|\Phi_i^{(v)}(t)\rangle = \exp\{-i[(\Delta_i^{(v)}t/\hbar) - \zeta(\tau) - \mathbf{x}\cdot\mathbf{k}_A(\tau)]\}\,|\Phi_i^{(0)}(t)\rangle \tag{3.2b}$$

in the velocity gauge.

We may expand the Floquet wave vector in the harmonic series

$$|\psi_i(\tau)\rangle = \sum_n e^{-in\tau}\,|\psi_{in}\rangle\ , \tag{3.3}$$

which describes the electron as having a spectrum of discrete but complex energies $\varepsilon_i + n\hbar\omega$, averaged over one cycle. In reality the electron has a spectrum of real energies,

$$E_{in} = E_i + \Delta_i + n\hbar\omega\ ,$$

which are broadened by at least the natural width Γ_i. A harmonic component $|\psi_{in}\rangle$ represents an electron that has absorbed a total of n real and virtual photons, and has a discrete but unphysical energy $E_{in} - i\Gamma_i/2$ which approaches the physical energy E_{in} in the weak-field limit.[17] If $n \geq N_0$ (where N_0 is the smallest integer for which $E_{in} > 0$) the electron is unbound. Since an unbound electron cannot be localized in space if it is monochromatic in energy, the $|\psi_{in}\rangle$, $n \geq N_0$, are nonnormalizable even in the weak-field limit, and so $|\psi_i(\tau)\rangle$ is nonnormalizable. In contrast, $|\Psi_i(t)\rangle$ is a normalized wave packet which describes the electron as initially localized in the bound state i. Turning on the field gives the electron an energy bandwidth $\hbar\Delta\omega$, and after a characteristic time $\hbar/\Gamma_i$ the electron is free and is represented by a normalized superposition of free-particle wave packets, where each component wave packet is peaked in energy space at one of the E_{in}, with a width $\hbar\Delta\omega$. (The peaks are broadened further if ionization occurs over a range of intensities.) Recalling that $\hbar\Delta\omega \gg \Gamma_i$, a necessary condition for the electron energy spectrum to resemble a discrete one is that

$$\Gamma_i \ll |E_{in}|\ , \tag{3.4}$$

an inequality which is satisfied for all n if it is satisfied for $n=N_0$ and $n=N_0-1$. This inequality is just the condition[18] for the spatial spreading of the electron wave packet to be negligible during the characteristic time that the atom ionizes.

The Floquet approximation to $|\Psi_i(t)\rangle$ is a useful simplification if inequality (3.4) is satisfied. However, this simplification comes with a high price—the nonnormalizability of $|\psi_i(\tau)\rangle$, which introduces obvious mathematical difficulties. Note that the initial boundary condition, that immediately after the field is turned on the electron is in a state represented by $|\Phi_i^{(g)}(t)\rangle$, where g is the gauge, cannot be satisfied. Since $|\psi_i(\tau)\rangle$ describes a localized bound electron, the best we can do is to require that the cycle average of the probability for the electron to be in the initial state at time $t>0$ is $\exp(-\Gamma_i t/\hbar)$; that is,

$$\frac{1}{2\pi}\int_0^{2\pi} d\tau\,|\langle\Phi_i^{(g)}(t)\,|\,\psi_i^{(g)}(\tau)\rangle|^2 = 1\ . \tag{3.5}$$

This relation fixes the overall normalization factor for $|\psi_i(\tau)\rangle$, a factor which depends on the state of the electron *after* the field has been turned on.

For $t > t_0$ we have

$$\left[H(\tau)-\frac{i\hbar d}{dt}\right]e^{-i\varepsilon_i t/\hbar}|\psi_i(\tau)\rangle=0\ , \tag{3.6}$$

and if we substitute the expansion (3.3) into this equation we obtain

$$(\varepsilon_i+n\hbar\omega-H_a)|\psi_{in}\rangle=V_+|\psi_{i,n-1}\rangle+V_-|\psi_{i,n+1}\rangle\ , \tag{3.7}$$

where $V_\pm=V_\pm^{(g)}$ with

$$V_+^{(l)}=-(eF_0/2)(\hat{\boldsymbol{\varepsilon}}\cdot\mathbf{x})\ , \tag{3.8a}$$

$$V_+^{(v)}=-(eF_0/2i\omega\mu)(\hat{\boldsymbol{\varepsilon}}\cdot\mathbf{p})\ , \tag{3.8b}$$

and with $V_-^{(g)}=[V_+^{(g)}]^\dagger$. As noted already, a component $|\psi_{in}\rangle$ describes an electron which has an energy $E_{in}-\Gamma_i/2$ and has absorbed a total of n real and virtual photons. There are infinitely many ways that an electron can absorb net n photons; for example, it may first emit 6 photons, then absorb $n+6$ photons. Thus $|\psi_{in}\rangle$ is composed of infinitely many waves,

$$|\psi_{in}\rangle=\sum_m|\psi_{in,m}\rangle\ , \tag{3.9}$$

where here the subscript m signifies the number of *real* photons which the electron has absorbed; the number of *virtual* photons absorbed is $n-m$, which may be negative (corresponding to emission). Only the $m=n$ component, that is, $|\psi_{in,n}\rangle$, describes the electron on the energy shell. In the weak-field regime we have $0\le m\le n$. An electron which has absorbed m real photons will move out to infinity with a mean physical momentum $\hbar k_{im}=(2\mu E_{im})^{1/2}$, which in the Floquet approximation becomes complex, equal to

$$\hbar\kappa_{im}=[(2\mu)(E_{im}-i\Gamma_i/2)]^{1/2}\ . \tag{3.10}$$

We pause for a moment to define the branch of the square root. We follow standard convention and draw a series of overlapping branch cuts along the real axis in the energy plane. The nth cut begins at the threshold $-n\hbar\omega$ and extends to the right along the real axis to infinity. The physical energy sheet is defined so that if E lies on this sheet just above the positive real axis, the square root of E is positive. The point $E_i+\Delta_i-i\Gamma_i/2$ is defined to lie on the unphysical sheet reached from the physical sheet by crossing the real energy axis downwards between the N_0th and (N_0-1)th thresholds. Returning to our electron which has absorbed m real photons, we expect that since the mean momentum is the canonical momentum in the velocity gauge, we have

$$\langle\mathbf{x}|\psi_{in,m}^{(v)}\rangle\sim e^{i\kappa_{im}r}/r,\quad r\sim\infty \tag{3.11}$$

where $r=|\mathbf{x}|$. (We have ignored an angle-dependent factor, and also the logarithmic distortion which would arise if W were to have a Coulomb tail.) We have attached the gauge superscript v to $|\psi_{in,m}\rangle$ to emphasize that the asymptotic form (3.11) does not hold in the length gauge, for in the latter gauge the canonical momentum is the instantaneous momentum. Moreover, the interaction $V^{(l)}(\tau)$ diverges in coordinate space for $r\sim\infty$. [Of course, $V^{(v)}(\tau)$ diverges in momentum space for $p\sim\infty$, but not as fast as $p^2/2\mu$.] If $m<N_0$ we have $\mathrm{Im}(\kappa_{im})>0$ and $\langle\mathbf{x}|\psi_{in,m}^{(v)}\rangle$ decays as r increases. If $m\ge N_0$, we have $\mathrm{Re}(\kappa_{im})>0$ so that $\langle\mathbf{x}|\psi_{in,m}^{(v)}\rangle$ behaves as an outgoing wave; however, since $\mathrm{Im}(\kappa_{im})<0$ this wave explodes exponentially as r increases, which is clearly unphysical and stems from the quasienergy having the imaginary part $\Gamma_i/2$, a consequence of neglecting wave-packet localization. It follows that the components $\langle\mathbf{x}|\psi_{in}^{(v)}\rangle$ explode exponentially for all n, except in the zero-width limit $\Gamma_i\to0$. Since the length- and velocity-gauge solutions of Eq. (3.6) are related by the gauge transformation, Eq. (2.10), the $\langle\mathbf{x}|\psi_{in}^{(l)}\rangle$ must also explode exponentially. However, provided that inequality (3.4) is satisfied we ought to be able to neglect Γ_i. We cannot simply put $\Gamma_i=0$ in ε_i in Eq. (3.7), since the coupled equations (3.7) are homogeneous, and ε_i is an eigenvalue which is determined by the condition for a regular outgoing wave solution to exist. What we can do is to modify Eq. (3.7) by introducing the zero-field eigensolution as an inhomegeneous term. The eigenvalue becomes an input parameter to these inhomogeneous equations, and we take this parameter to be the shifted energy; we neglect the width.[19] We calculate the shift by first solving Eq. (3.7) for the eigenvalue ε_i; this can be done relatively easily[20,21] since the eigenvalue is unchanged by the rotation $r\to re^{i\theta}$, which transforms the functions $\langle\mathbf{x}|\psi_{in}\rangle$ into functions that decay as r increases if $\mathrm{Im}(e^{i\theta}\kappa_{iN_0})>0$.[22]

B. Modified Floquet equation

To modify Eq. (3.7) as suggested, it is convenient to introduce the projection operator

$$\hat{P}=|\Phi_i\times\Phi_i|\delta_{n0}\ ,$$

where the Kronecker δ operator δ_{n0} acts with reference to the subscript n of $|\psi_{in}\rangle$, and where $|\Phi_i\rangle$ represents the initial state of the electron in the zero-field limit. Premultiplying both sides of Eq. (3.7) by $\hat{P}$ gives, recalling that $H_a|\Phi_i\rangle=E_i|\Phi_i\rangle$,

$$(\varepsilon_i-E_i)\hat{P}|\psi_{in}\rangle=\hat{P}[V_+|\psi_{i,n-1}\rangle+V_-|\psi_{i,n-1}\rangle]\ . \tag{3.12}$$

The correct normalization of $|\psi_i(\tau)\rangle$ is determined from Eq. (3.5), but we can temporarily normalize $|\psi_i(\tau)\rangle$ so that $\langle\Phi_i|\psi_{i0}\rangle=1$; then $\hat{P}|\psi_{in}\rangle$ simplifies to $|\Phi_i\rangle\delta_{n0}$. We can later renormalize $|\psi_i(\tau)\rangle$ correctly. We now use Eq. (3.12) to rewrite the right-hand side of Eq. (3.7) as

$$\begin{aligned}&[\hat{P}+(1-\hat{P})][V_+|\psi_{i,n-1}\rangle+V_-|\psi_{i,n+1}\rangle]\\&\quad=(\varepsilon_i-E_i)|\Phi_i\rangle\delta_{n0}+(1-\hat{P})[V_+|\psi_{i,n-1}\rangle\\&\qquad\qquad+V_-|\psi_{i,n+1}\rangle]\ .\end{aligned}$$

At this point we put $\Gamma_i=-i\eta$, where η is positive but infinitesimal; this moves ε_i from the unphysical sheet to a nearby point on the physical sheet just above the positive

real axis. We have $\varepsilon_i - E_i = \Delta_i + \eta$ and Eq. (3.7) becomes the set of inhomogeneous equations

$$(E_{in} + i\eta - H_a)\,|\,\psi_{in}\rangle = \Delta_i\,|\,\Phi_i\rangle\delta_{n0} + (1-\delta_{n0}\,|\,\Phi_i\times\Phi_i\,|\,)[V_+\,|\,\psi_{i,n-1}\rangle + V_-\,|\,\psi_{i,n+1}\rangle]\ . \tag{3.13}$$

Thus the Floquet wave vector becomes

$$|\,F_i(t)\rangle = e^{-i(E_i+\Delta_i)t/\hbar}\,|\,\psi_i(\tau)\rangle\ , \tag{3.14}$$

where the harmonic components of $|\,\psi_i(\tau)\rangle$ are now determined from Eq. (3.13). It may be verified from Eq. (3.13) that, in contrast to Eq. (3.6), we have

$$\left[H(\tau) - i\hbar\frac{d}{dt}\right]\Big|F_i(t)\Big\rangle = -\lambda_i e^{-i(E_i+\Delta_i)t/\hbar}\,|\,\Phi_i\rangle\ , \tag{3.15}$$

where λ_i depends implicitly on $|\,\psi_{i,\pm1}\rangle$,

$$\lambda_i = \Delta_i - \langle\Phi_i\,|\,V_+\,|\,\psi_{i,-1}\rangle - \langle\Phi_i\,|\,V_-\,|\,\psi_{i,1}\rangle\ . \tag{3.16}$$

In the weak-field limit λ_i vanishes through first order in the intensity[23] and hence remains small even at moderate intensities. Introducing the nonlocal operator

$$\Pi(\tau) = |\,\Phi_i\times\Phi_i\,|\,(e^{-i\tau}V_+\delta_{n,-1} + e^{i\tau}V_-\delta_{n,1})\ , \tag{3.17}$$

we can rewrite the right-hand side of Eq. (3.15) as

$$-\Delta_i e^{-(E_i+\Delta_i)t/\hbar}\,|\,\Phi_i\rangle + \Pi(\tau)\,|\,F_i(t)\rangle\ ,$$

which explicitly displays the source term $\Delta_i\,|\,\Phi_i\rangle$. In the velocity gauge the solution of the coupled equations (3.13) is such that each component $\langle\mathbf{x}\,|\,\psi_{in}^{(v)}\rangle$ is regular at $r=0$ and behaves for $r\sim\infty$ as a superposition of physical (nonexploding) *outgoing* waves, of the form $\exp[ik_{im}r]/r$. This asymptotic boundary condition cannot be satisfied in the length gauge because the interaction $V^{(l)}(\tau)$ diverges for $r\sim\infty$. Note that while the length- and velocity-gauge solutions of Eq. (3.6) are connected by the gauge transformation of Eq. (2.10), the length- and velocity-gauge solutions of Eq. (3.15) are not, except in the weak-field limit.

C. Outgoing wave vector

We now turn to $|\,\Psi_f(t)\rangle$, which describes an unbound electron that scatters from the atomic potential, in the presence of a radiation field that is turned on $t=0$, and emerges as a free particle moving through the field with mean momentum $\hbar\mathbf{k}_f$. The wave vector $|\,\Psi_f(t)\rangle$ is nonnormalizable and can be expressed exactly in the Floquet form[12]

$$|\,\Psi_f(t)\rangle = e^{-i\varepsilon_f t/\hbar}\,|\,\psi_f(\tau)\rangle\ , \tag{3.18}$$

when $t>t_0$, where $|\,\psi_f(\tau)\rangle$ has period 2π. The energy ε_f is real and positive, and in the length gauge is shifted by the ponderomotive energy—we have $\varepsilon_f = E_f + P$, where $E_f = \hbar^2\mathbf{k}_f^2/2\mu$—while in the velocity gauge the energy is unshifted, $\varepsilon_f = E_f$. Whereas $|\,\Psi_i(t)\rangle$ satisfies a boundary condition at $t=0$, when the field is turned on, $|\,\Psi_f(t)\rangle$ satisfies a boundary condition at $t=\infty$, when the field is turned off. Turning off the field at $t=\infty$ does not disrupt the harmonicity of the Hamiltonian $H(\tau)$, and consequently the ansatz (3.18) is exact for $t>t_0$. Of course, turning on the field does disrupt the harmonicity of $H(\tau)$, and Eq. (3.18) ceases to be valid for $t<t_0$. The precise form of $|\,\Psi_f(t)\rangle$ for $t<t_0$ is of no interest to us, but it is affected by the way the field is turned on. What does interest us, however, is the fact that turning on the field profoundly alters the form of $|\,\Psi_f(t)\rangle$ since the electron, whose energy ε_f is close to one of the values E_{in}, can now emit n photons and be temporarily captured into the initial bound state i. Consequently, when $t>t_0$, $|\,\Psi_f(t)\rangle$ exhibits resonance poles[24] at energies $\varepsilon_f = E_{in} - i\Gamma_i/2$.

In Appendix B we discuss the state of the electron after the electron has escaped from the influence of the atomic potential but before the field is turned off. At such times the electron state is represented in the velocity gauge by $\exp[-i\theta(\tau)]\,|\,\mathbf{k}_f;t\rangle$ where, if $|\,\mathbf{k}\rangle$ is the eigenvector of the canonical momentum operator $\mathbf{p}$ with eigenvalue $\hbar\mathbf{k}$,

$$|\,\mathbf{k}_f;t\rangle = e^{-iE_f t/\hbar}\,|\,\mathbf{k}_f\rangle\ , \tag{3.19}$$

$$\theta(\tau) = -\frac{\hbar}{\mu}\int_0^t dt'\,\mathbf{k}_f\cdot\mathbf{k}_A(\tau')\ . \tag{3.20}$$

We also introduce in Appendix B the eigenvectors

$$|\,\Phi_f^{(0)}(t)\rangle = e^{-iE_f t/\hbar}\,|\,\Phi_{\mathbf{k}_f}^-\rangle\ , \tag{3.21}$$

$$|\,\Phi_f^{(v)}(t)\rangle = e^{-i\theta(\tau)}\,|\,\Phi_f^{(0)}(t)\rangle\ , \tag{3.22}$$

$$|\,KW_f^{(v)}(t)\rangle = e^{-iE_f t/\hbar}e^{-i\theta(\tau)+i\mathbf{x}\cdot\mathbf{k}_A(\tau)}\,|\,\Phi_{\mathbf{k}_f(\tau)}^-\rangle\ , \tag{3.23}$$

where $|\,\Phi_{\mathbf{k}}^-\rangle$ is the eigenvector of the atomic Hamiltonian H_a which satisfies out-asymptote boundary conditions and approaches $|\,\mathbf{k}\rangle$ at large distances, and where $\mathbf{k}_f(\tau) = \mathbf{k}_f - \mathbf{k}_A(\tau)$. Thus $|\,\Phi_f^{(0)}(t)\rangle$ represents the final state of the electron unperturbed by the field, while $|\,\Phi_f^{(v)}(t)\rangle$ differs from $|\,\Phi_f^{(0)}(t)\rangle$ only by a time-dependent phase factor. Each of the vectors of Eqs. (3.21)–(3.23) represents at asymptotically large distances the same physical state as does $|\,k_f;t\rangle$. The vectors $|\,\Phi_f^{(v)}(t)\rangle$ and $|\,KW_f^{(v)}(t)\rangle$ incorporate, to some extent, the effects of both the atomic potential and the interaction with the field. In the low-frequency limit $|\,KW_f^{(v)}(t)\rangle$ exactly describes the scattering of an electron from the atomic potential in the presence of the field, as first shown by Kroll and Watson [12,25]. The length-gauge forms of the vectors of Eqs. (3.21)–(3.23) are given by the transformation of Eq. (2.10).

IV. EXPRESSIONS FOR THE IONIZATION AMPLITUDE

We work in the velocity gauge throughout this section. We first introduce a variational principle, and using as input the exact solution to the modified Floquet equation,

Eq. (3.15), we derive two different expressions for the ionization rate. We subsequently derive the "truncated-threshold approximation" by using as input the truncated Floquet expansion in harmonic components. Our approximate expressions for the ionization amplitude are not, in general, gauge invariant. We note, however, that it may be possible to develop gauge-invariant expressions following the suggestion of Rosenberg.[26]

A. Variational principle

Let $|\Psi_{i,\mathrm{tr}}(t)\rangle$ be a trial approximation to $|\Psi_i(t)\rangle$ which satisfies the boundary condition

$$|\Psi_{i,\mathrm{tr}}(t)\rangle \rightarrow |\Phi_i^{(0)}(t)\rangle, \quad t\rightarrow 0 \tag{4.1}$$

where $|\Phi_i^{(0)}(t)\rangle$ represents the initial field-free state. Let $|\Psi_{f,\mathrm{tr}}(t)\rangle$ be a trial approximation to $|\Psi_f(t)\rangle$ which satisfies the boundary condition

$$|\Psi_{f,\mathrm{tr}}(t)\rangle \rightarrow |\Phi_f^{(v)}(t)\rangle, \quad t\rightarrow \infty \tag{4.2}$$

where $|\Phi_f^{(v)}(t)\rangle$ is orthogonal to $|\Phi_i^{(0)}(t)\rangle$ and is defined by Eq. (3.22), with its asymptotic form given by Eq. (B5). The amplitude $A_{fi}(T)$ for finding the electron at the large positive time T in the state represented by $|\Phi_f^{(v)}(t)\rangle$, if at $t=0$ it was in the state represented by $|\Phi_i^{(0)}(t)\rangle$, is[27]

$$\begin{aligned} A_{fi}(T) &= \langle \Phi_f^{(v)}(T) \mid U(T,0) \mid \Phi_i^{(0)}(0)\rangle \\ &= A_{fi,\mathrm{tr}}(T)+R_{fi,\mathrm{tr}}(T) , \end{aligned} \tag{4.3}$$

where $U(t,t')$ is the evolution operator and where, defining

$$\mathcal{H}(t)=H(\tau)-i\hbar\frac{d}{dt} , \tag{4.4}$$

and writing $|\mathcal{H}(t)\psi\rangle$ for $\mathcal{H}(t)\,|\,\psi\rangle$, we have

$$A_{fi,\mathrm{tr}}(T)=\langle \Phi_f^{(v)}(T) \mid \Psi_{i,\mathrm{tr}}(T)\rangle - \frac{i}{\hbar}\int_0^T dt\,\langle \Psi_{f,\mathrm{tr}}(t) \mid \mathcal{H}(t)\Psi_{i,\mathrm{tr}}(t)\rangle \tag{4.5a}$$

$$=\langle \Psi_{f,\mathrm{tr}}(0) \mid \Phi_i^{(0)}(0)\rangle - \frac{i}{\hbar}\int_0^T dt\,\langle \mathcal{H}(t)\Psi_{f,\mathrm{tr}}(t) \mid \Psi_{i,\mathrm{tr}}(t)\rangle , \tag{4.5b}$$

$$R_{fi,\mathrm{tr}}(T)=-\frac{1}{\hbar^2}\int_0^T dt\int_0^t dt'\langle \mathcal{H}(t)\Psi_{f,\mathrm{tr}}(t) \mid U(t,t') \mid \mathcal{H}(t')\Psi_{i,\mathrm{tr}}(t')\rangle . \tag{4.6}$$

It is apparent that the remainder $R_{fi,\mathrm{tr}}(T)$ is bilinear in the errors in $|\Psi_{i,\mathrm{tr}}(t)\rangle$ and $|\Psi_{f,\mathrm{tr}}(t)\rangle$, and so $A_{fi,\mathrm{tr}}(T)$ is a variational estimate of $A_{fi}(T)$ in the sense that it is stationary with respect to variations in the trial vectors about the exact vectors. Were we to choose $|\Psi_{f,\mathrm{tr}}(t)\rangle$ to be

$$\exp(-iE_f t/\hbar)\,|\,\psi_f^{(v)}(\tau)\rangle$$

we would have $\mathcal{H}(t)\,|\,\Psi_{f,\mathrm{tr}}(t)\rangle=0$, so that $R_{fi,\mathrm{tr}}(T)$ would vanish and $A_{fi,\mathrm{tr}}(T)$ would appear to be exact. This is not so. In writing down Eqs. (4.3)–(4.6) we replaced $U(t,T)\,|\,\Phi_f^{(v)}(T)\rangle$ by $|\Psi_{f,\mathrm{tr}}(t)\rangle$; this is justified in the limit $T\sim\infty$ but not, with the present choice of $|\Psi_{f,\mathrm{tr}}(t)\rangle$, for finite values of T because the limit is sensitive to the contribution from the resonance poles which arise from the capture-escape mechanism discussed in Sec. III C above. It might appear that $R_{fi,\mathrm{tr}}(T)$ would also vanish if we were to choose $|\Psi_{i,\mathrm{tr}}(t)\rangle$ to be

$$\exp(-i\varepsilon_i t/\hbar)\,|\,\psi_i^{(v)}(\tau)\rangle ,$$

with $|\psi_i^{(v)}(\tau)\rangle$ the *unmodified* vector satisfying Eq. (3.6), that is, $\mathcal{H}(t)\,|\,\Psi_{i,\mathrm{tr}}(t)\rangle=0$; but this cannot be true since $|\Psi_{i,\mathrm{tr}}(t)\rangle$ is not exact. The resolution of this paradox lies in the observation that expressions (4.3)–(4.6) were derived[27] using the Hermiticity of $H(\tau)$, a property that can be safely used only when the trial wave vectors are normalizable. Thus the subsequent passage to nonnormalizable trial wave vectors, through some kind of limit process, must be taken with caution. The values of $A_{fi,\mathrm{tr}}(T)$ and $R_{fi,\mathrm{tr}}(T)$ must be insensitive to this limit process. Clearly this would not be the case were we to allow $|\Psi_{i,\mathrm{tr}}(t)\rangle$ to approach the exponentially exploding unmodified Floquet wave vector since the spatial integrals would become infinite. On the other hand, as we shall see, the spatial integrals remain finite when we take $|\Psi_{i,\mathrm{tr}}(t)\rangle$ to be the nonnormalizable but nonexploding modified Floquet wave vector.

B. Ionization rate

The rate for ionization to a group of states f with density $\rho'(E_f)$ in the energy interval $(E_f,\ E_f+dE)$ is, for $T\sim\infty$,

$$\dot{P}_{fi}=\rho'(E_f)dE\frac{d}{dT}\,|\,A_{fi}(T)\,|^2 . \tag{4.7}$$

In the examples that we consider below we find that $A_{fi,\mathrm{tr}}(T)$ can be expressed in the form

$$A_{fi,\mathrm{tr}}(T)=-\frac{i}{\hbar}\int_0^T dt\,e^{i(E_f-E_i-\Delta_i)t/\hbar}\sum_n M_n e^{-in\tau} , \tag{4.8}$$

where M_n is an ionization matrix element. Using the result

$$\int_0^\infty dt\,e^{i\Omega t}=iP\left[\frac{1}{\Omega}\right]+\pi\delta(\Omega) , \tag{4.9}$$

we obtain, for $T\sim\infty$, the expression

$$A_{fi,\mathrm{tr}}(T)=-\pi i\sum_n \delta(E_f-E_{in})M_n , \tag{4.10}$$

where we have neglected the finite principal part since it does not contribute to the rate for ionization to a group of states within a narrow energy bandwidth. Only terms with $n \geq N_0$ contribute to the sum on the right-hand side of Eq. (4.10) since $E_f = \hbar^2 \mathbf{k}_f^2 / 2\mu$ is positive. Noting that

$$\frac{d}{dT} A_{fi,\mathrm{tr}}(T) = -\frac{i}{\hbar} \sum_n M_n e^{i(E_f - E_{in})T/\hbar} , \tag{4.11}$$

we obtain for the approximate N-photon ionization rate the result

$$\dot{P}_{fN,i} = (2\pi/\hbar)\rho'(E_{iN}) \mid M_N \mid^2 , \tag{4.12}$$

where $N \geq N_0$; we have neglected interference terms in the ionization signal arising from the absorption of different numbers of photons, since such interference terms vanish when the signal is averaged over a cycle.

C. Floquet approximation

We now develop expressions for $A_{fi,\mathrm{tr}}(T)$ based on choosing $\mid \Psi_{i,\mathrm{tr}}(t) \rangle$ to be the modified Floquet wave vector $\mid F_i(t) \rangle$; we determine the harmonic components of $\mid \psi_i^{(v)}(\tau) \rangle$ by solving Eq. (3.13). Recall that the solution of Eq. (3.13) yields an approximation to $\langle \mathbf{x} \mid \psi_i^{(v)}(\tau) \rangle$ which behaves as a superposition of nonexploding outgoing waves as the radial coordinate increases. We note, however, that our $\mid \Psi_{i,\mathrm{tr}}(t) \rangle$ does not satisfy the boundary condition (4.1) unless we take into account that the field is turned on at $t = 0$. If we do this, the field amplitude varies with time and destroys the harmonicity of $\mid \psi_i^{(v)}(\tau) \rangle$ during the interval $[0, t_0]$. However, the contribution from this interval to the integral over t on the right-hand side of either Eq. (4.5a) or (4.5b) is finite and comparable to the finite part [cf. $P(1/\Omega)$ in Eq. (4.9)] which we throw away. We may therefore change the lower limit on the integral over t from zero to t_0 with no real further loss in accuracy. We formulate two different expressions for $A_{fi,\mathrm{tr}}(T)$ based on the choices (i)

$$\mid \Psi_{f,\mathrm{tr}}(t) \rangle = e^{i\theta(\tau)} \mid \Phi_f^{(v)}(t) \rangle = \mid \Phi_f^{(0)}(t) \rangle ,$$

where $\theta(\tau)$ was defined by Eq. (3.20), and[28] (ii)

$$\mid \Psi_{f,\mathrm{tr}}(t) \rangle = \mid \Phi_f^{(v)}(t) \rangle ,$$

where in both examples $E_f = E_{fN}$. The trial vector (i) does satisfy the correct boundary condition (4.2) since the phase $\theta(\tau)$ is "harmless" and the state represented by trial vector (i) is at any time t identical to the state represented by $\mid \Phi_f^{(v)}(t) \rangle$.

We start with choice (i). From Eqs. (B4) and (4.5b), recalling that

$$\left[H_a - i\hbar \frac{d}{dt} \right] e^{-iE_f t/\hbar} \mid \Phi_{\mathbf{k}_f}^- \rangle = 0 , \tag{4.13}$$

and that $\langle \Phi_f^{(0)}(0) \mid \Phi_i^{(0)}(0) \rangle = 0$, we have

$$A_{fi,\mathrm{tr}}(T) = -\frac{i}{\hbar} \int_{t_0}^{T} dt \langle \Phi_{\mathbf{k}_f}^- \mid V^{(v)}(\tau) \mid \psi_i^{(v)}(\tau) \rangle \times e^{i(E_f - E_i - \Delta_i^{(v)})t/\hbar} . \tag{4.14}$$

The harmonic expansion of $\mid \psi_i^{(v)}(\tau) \rangle$ yields

$$\langle \Phi_{\mathbf{k}_f}^- \mid V^{(v)}(\tau) \mid \psi_i^{(v)}(\tau) \rangle = \sum_n M_n e^{-in\tau} , \tag{4.15}$$

where in the present example M_n has the form

$$M_n = \langle \Phi_{\mathbf{k}_f}^- \mid V_-^{(v)} \mid \psi_{i,n+1}^{(v)} \rangle + \langle \Phi_{\mathbf{k}_f}^- \mid V_+^{(v)} \mid \psi_{i,n-1}^{(v)} \rangle . \tag{4.16}$$

Each of the two terms on the right-hand side of Eq. (4.16) is infinite. The reason is the following: $\langle \mathbf{x} \mid \Phi_{\mathbf{k}_f}^- \rangle$ is a standing wave, containing both ingoing and outgoing waves of the form $\exp(\pm ik_{iN}r)/r$, while $\langle \mathbf{x} \mid \psi_{i,n\pm1}^{(v)} \rangle$ are superpositions of outgoing waves that include the component $\exp(ik_{iN}r)/r$. The latter component interferes destructively with the outgoing part of $\langle \mathbf{x} \mid \Phi_{\mathbf{k}_f}^- \rangle$ (which becomes ingoing upon complex conjugation) in each of the integrands of the two integrals $\langle \Phi_{\mathbf{k}_f}^- \mid V_{\mp}^{(v)} \mid \psi_{i,n\pm1} \rangle$. Therefore each integrand has a nonoscillatory part which is not cut off by $V_{\pm}^{(v)}$ and therefore does not vanish as r increases. Consequently, the two integrals are divergent. However, it turns out that the on-shell matrix element M_N, which is the only matrix element we need, is finite. In other words, when $n = N$ the divergent parts of the two terms on the right-hand side of Eq. (4.16) cancel. To see this, we use Eq. (3.13) to rewrite M_n in the form

$$M_n = \langle \Phi_{\mathbf{k}_f}^- \mid (E_{in} + i\eta - H_a) \mid \psi_{in}^{(v)} \rangle . \tag{4.17}$$

Now $E_{in} + i\eta - H_a$ annihilates the component of $\langle \mathbf{x} \mid \psi_{in}^{(v)} \rangle$ which behaves as $\exp(ik_{in}r)/r$ for large r, or, more precisely, it turns this component into a function which vanishes as $1/r^3$ as r increases. Consequently, if $n = N$ there is no destructive interference and the part of the integrand of $\langle \Phi_{\mathbf{k}_f}^- \mid (E_{iN} + i\eta - H_a) \mid \psi_{iN}^{(v)} \rangle$ which does not vanish as r increases is oscillatory; the integral can therefore be made to converge by, for example inserting a convergence factor $\exp(-\eta r)$ into the integrand. It follows that M_N is finite. Recently, a calculation[29] of the matrix element M_1 for one-photon ionization of hydrogen was performed in perturbation theory, at one order beyond lowest order. Incidentally, it follows from Eq. (4.17) that

$$M_N = \langle \Phi_{\mathbf{k}_f}^- \mid (H_a^\dagger - H_a) \mid \psi_{iN}^{(v)} \rangle ,$$

a form which clearly illustrates the significance of the non-Hermiticity of H_a.

We note here that one sometimes sees in the literature an expression for the N-photon ionization matrix element which differs from the right-hand side of Eq. (4.16) only in that the superscript v is replaced by l, that is, the length gauge is used. However, except in the weak-field limit, such an expression is infinite;[29] $\langle \mathbf{x} \mid \psi_{iN}^{(l)} \rangle$ does not behave for large r as a superposition of outgoing waves $\exp(ik_{im}r)/r$, and we cannot repeat in the length gauge the proof we just gave for the finiteness of M_N.

Before continuing, we note one further point. Suppose we were to choose $\mid \psi_{i,\mathrm{tr}}(t) \rangle = \mid \Phi_i^{(0)}(t) \rangle$ and

$$|\Psi_{f,\mathrm{tr}}(t)\rangle=\exp(-iE_f t/\hbar)\,|\psi_f^{(v)}(\tau)\rangle\ .$$

Using Eq. (4.5a) we would obtain

$$A_{fi,\mathrm{tr}}(T)=-\frac{i}{\hbar}\int_{t_0}^{T}dt\,\langle\psi_f^{(v)}(\tau)\,|\,V^{(v)}(\tau)\,|\,\Phi_i\rangle \times e^{i(E_f-E_i)t/\hbar}\ . \tag{4.18}$$

Inserting the harmonic expansion of $|\psi_f^{(v)}(\tau)\rangle$ and $V^{(v)}(\tau)$ into the integrand yields

$$A_{fi,\mathrm{tr}}(T)=-\frac{i}{\hbar}\int_{t_0}^{T}dt\,e^{i(E_f-E_i)t/\hbar}\sum_n M_n e^{in\tau}\ , \tag{4.19}$$

where now

$$M_n=\langle\psi_{f,n-1}^{(v)}\,|\,V_-\,|\,\Phi_i\rangle+\langle\psi_{f,n+1}^{(v)}\,|\,V_+\,|\,\Phi_i\rangle\ . \tag{4.20}$$

Note, however, that if we neglect the finite part of the integral over t in Eq. (4.19) we obtain $E_f=E_i+n'\hbar\omega=E_{in'}-\Delta_i^{(v)}$, where $n'=-n$. In other words, the energy of the emergent electron is unshifted. The source of the discrepancy is that the components $|\psi_{f,n}^{(v)}\rangle$ have resonance poles at energies E_f close to E_{fm}, $m\geq N_0$, as noted above. We would obtain the correctly shifted energy $E_f=E_{in'}$ by choosing $|\Psi_{i,\mathrm{tr}}(t)\rangle$ to be $|\Phi_i^{(v)}(t)\rangle$ rather than $|\Phi_i^{(0)}(t)\rangle$, but this would change the structure of M_n; infinitely many components $|\psi_{fm}^{(v)}\rangle$ would explicitly contribute to M_n. Moreover, the "finite" part of the integral over t is very large and cannot be neglected.

We now consider choice (ii) above. From Eqs. (B1), (B2), and (4.5b), and again using Eq. (4.13) and

$$\langle\Phi_f^{(v)}(0)\,|\,\Phi_i^{(0)}(0)\rangle=0\ ,$$

we have

$$A_{fi,\mathrm{tr}}(T)=-\frac{i}{\hbar}\int_{t_0}^{T}dt\,\langle\Phi_{\mathbf{k}_f}^{-}\,|\,[V^{(v)}(\tau)-\Delta_f^{(v)}(\tau)]\,|\,\psi_i^{(v)}(\tau)\rangle \times e^{i\theta(\tau)}e^{i(E_f-E_i-\Delta_i^{(v)})t/\hbar}\ . \tag{4.21}$$

Thus the effective interaction is now $V^{(v)}(\tau)$ minus the instantaneous energy shift. We have the harmonic expansion

$$\langle\Phi_{\mathbf{k}_f}^{-}\,|\,[V^{(v)}(\tau)-\Delta_f^{(v)}(\tau)]\,|\,\psi_i^{(v)}(\tau)\rangle e^{i\theta(\tau)}=\sum_n M_n e^{-in\tau}\ , \tag{4.22}$$

where now

$$M_n=\frac{\omega}{2\pi}\int_0^{2\pi/\omega}dt\,\langle\Phi_{\mathbf{k}_f}^{-}\,|\,[V^{(v)}(\tau)-\Delta_f^{(v)}(\tau)]\,|\,\psi_i^{(v)}(\tau)\rangle \times e^{i\theta(\tau)+in\tau}\ . \tag{4.23}$$

We showed above that, with choice (i), M_N is the matrix element for a transition induced by the interaction of the electron with the field, that is, $V_{\pm}^{(v)}$. We now show that, with choice (ii), M_N is the matrix element for a transition induced by the interaction of the electron with the atomic potential W. However, the reader uninterested in mathematical manipulation can skip directly to the result, Eq. (4.32), which can in fact be obtained more simply, as explained later. Rewriting

$$\Delta_f^{(v)}(\tau)e^{i\theta(\tau)}=-i\hbar\frac{d}{dt}e^{i\theta(\tau)} \tag{4.24}$$

and integrating by parts the term in the time in the time derivative in Eq. (4.23) (noting the surface term vanishes because the integrand is periodic), we obtain

$$M_n=\frac{\omega}{2\pi}\int_0^{2\pi/\omega}dt\,e^{i\theta(\tau)+in\tau} \times\left\langle\Phi_{\mathbf{k}_f}^{-}\,\middle|\,\left[V^{(v)}(\tau)+n\hbar\omega-i\hbar\frac{d}{dt}\right]\middle|\,\psi_i^{(v)}(\tau)\right\rangle\ . \tag{4.25}$$

From Eq. (3.15) we have

$$-i\hbar\frac{d}{dt}\left|\psi_i^{(v)}(\tau)\right\rangle=[E_i+\Delta_i^{(v)}-H(\tau)]\,|\,\psi_i^{(v)}(\tau)\rangle -\lambda_i^{(v)}\,|\,\Phi_i\rangle\ , \tag{4.26}$$

and combining this result with Eq. (4.25) yields

$$M_n=\frac{\omega}{2\pi}\int_0^{2\pi/\omega}dt\,e^{i\theta(\tau)+in\tau} \times\langle\Phi_{\mathbf{k}_f}^{-}\,|\,(E_{in}-H_a)\,|\,\psi_i^{(v)}(\tau)\rangle\ . \tag{4.27}$$

We now specialize to $n=N$. Since $H_a\,|\,\Phi_{\mathbf{k}_f}^{-}\rangle=E_{iN}\,|\,\Phi_{\mathbf{k}_f}^{-}\rangle$ we have

$$M_N=\frac{\omega}{2\pi}\int_0^{2\pi/\omega}dt\,e^{i\theta(\tau)+iN\tau} \times\langle\Phi_{\mathbf{k}_f}^{-}\,|\,(H_a^{\dagger}-H_a)\,|\,\psi_i^{(v)}(\tau)\rangle\ . \tag{4.28}$$

Note again that H_a in non-Hermitian in the matrix element of Eq. (4.28) because of the destructive interference of the outgoing part of $\langle\mathbf{x}\,|\,\Phi_{\mathbf{k}_f}^{-}\rangle$ with the component of $\langle\mathbf{x}\,|\,\psi_i^{(v)}(\tau)\rangle$ that behaves as an outgoing wave $\exp(ik_{iN}r)/r$. We can express $|\,\Phi_{\mathbf{k}_f}^{-}\rangle$, up to a normalization factor, as

$$|\,\Phi_{\mathbf{k}_f}^{-}\rangle=|\,\mathbf{k}_f\rangle+G_a^{-}(E_f)W\,|\,\mathbf{k}_f\rangle\ , \tag{4.29}$$

where $G_a^{-}(E)=(E-i\eta-H_a)^{-1}$, the ingoing Green's operator for the atomic Hamiltonian. [Note that Eq. (4.29), and therefore Eqs. (4.32) and (4.33), are invalid for a potential W with a Coulomb tail.] Since $\langle\mathbf{x}\,|\,G_a^{-}(E_f)W\,|\,\mathbf{k}_f\rangle$ behaves as an ingoing wave, the second term on the right-hand side of Eq. (4.29) does not contribute to the matrix element of Eq. (4.28) and we can replace $|\,\Phi_{\mathbf{k}_f}^{-}\rangle$ by $|\,\mathbf{k}_f\rangle$ in this matrix element. If we use Eq. (B3) to write

$$H_a e^{-i\theta(\tau)}\,|\,\mathbf{k}_f\rangle=[W-V^{(v)}(\tau)]e^{-i\theta(\tau)}\,|\,\mathbf{k}_f\rangle +\left[E_f+i\hbar\frac{d}{dt}\right]e^{-i\theta(\tau)}\left|\mathbf{k}_f\right\rangle\ , \tag{4.30}$$

we can replace $\exp[i\theta(\tau)]\langle \Phi^-_{\mathbf{k}_f} | H_a^\dagger$ in Eq. (4.28) by the adjoint of the right-hand side of Eq. (4.30). Upon integrating the term in the time derivative by parts, and noting that $E_f - N\hbar\omega = E_i + \Delta_i^{(v)}$, we obtain

$$M_N = \frac{\omega}{2\pi}\int_0^{2\pi/\omega} dt\, e^{i\theta(\tau)+iN\tau} \times \left\langle \mathbf{k}_f \left| \left[W + E_i + \Delta_i^{(v)} - H(\tau) + i\hbar\frac{d}{dt} \right] \right| \psi_i^{(v)}(\tau) \right\rangle . \quad (4.31)$$

Combining Eqs. (4.26) and (4.31), and neglecting the term $\lambda_i^{(v)}\langle \mathbf{k}_f | \Phi_i \rangle$, which is small if indeed $\lambda_i^{(v)}$ is small, we obtain the desired result,

$$M_N = \frac{1}{2\pi}\int_0^{2\pi} d\tau\, e^{i\theta(\tau)+iN\tau}\langle \mathbf{k}_f | W | \psi_i^{(v)}(\tau)\rangle . \quad (4.32)$$

Writing $\theta(\tau) = \rho\cos(\tau-\chi)$, where

$$\rho = \exp(-i\chi)(e/\mu\omega^2)F_0\hat{\boldsymbol{\varepsilon}}\cdot\mathbf{k}_f ,$$

with ρ and χ both real, the harmonic expansion of $|\psi_i^{(v)}(\tau)\rangle$ yields

$$M_N = \sum_n e^{i(N-n)\chi} J_{N-n}(-\rho)\langle \mathbf{k}_f | W | \psi_{in}^{(v)}\rangle . \quad (4.33)$$

This last expression was used[21] to calculate partial rates for multiphoton detachment of the negative hydrogen ion. It is interesting that two trial vectors which differ only by a "harmless" phase lead to expressions for the ionization amplitude that have a very different structure, cf. Eqs. (4.16) and (4.33). As may be expected, the trial vector (i) is not as accurate as (ii) when the field is strong; the trial vector (ii) incorporates, through the phase factor $\exp[-i\theta(\tau)]$, the absorption and emission of virtual photons. It will be shown elsewhere that, for $n = N$, the right-hand side of Eq. (4.16) is a factor $J_0(-\rho)$ larger than the right-hand side of Eq. (4.33), provided that inequality (3.4) is satisfied. We could directly obtain Eq. (4.32) by taking $|\psi_{f,\mathrm{tr}}(t)\rangle$ equal to $e^{-i\theta(\tau)}|\mathbf{k}_f;t\rangle$ in Eq. (4.5b), provided we neglect the term $\langle \mathbf{k}_f | \Phi_i\rangle$ coming from $\langle \psi_{f,\mathrm{tr}}(0) | \Phi_i^{(0)}(0)\rangle$. This term is finite, and of the same order as the finite part of the integral over t which we have already neglected.

To evaluate M_N, in either of the forms (4.16) or (4.33), we must solve Eq. (3.15) for $|\psi_i^{(v)}(\tau)\rangle$. The numerical solution of this equation is complicated by the presence of the operator $\Pi(\tau)$, which is nonlocal in coordinate space and precludes the direct numerical integration[30] of the coupled differential equations (3.13) for the $\langle \mathbf{x} | \psi_n^{(v)}\rangle$. Nevertheless, these equations can be solved in a number of ways, for example, by expansion of the $\langle \mathbf{x} | \psi_n^{(v)}\rangle$ in a discrete (not necessarily normalizable) set of basis functions that have outgoing wave character. The coefficients of the basis functions satisfy an inhomogeneous matrix equation. However, the expansion which results from projecting $|\psi_i^{(v)}(\tau)\rangle$ onto $|\Phi^-_{\mathbf{k}_f}\rangle$ (with either $V_\pm^{(v)}$ or W sandwiched in between) does not converge. The reason for this is that $\langle \mathbf{x} | \Phi^-_{\mathbf{k}_f}\rangle$ contains an outgoing wave, which becomes an ingoing wave upon complex conjugation. The projection amounts to expanding an ingoing wave in terms of outgoing waves, which is like working against the grain of the wood. Nevertheless, convergence can be accomplished by Padé summation or by other means.[31] Rather than pursue this further (it will be taken up again in future work) we now describe an approximation which is free of this difficulty.

D. Truncated Floquet expansion

The convergence problem just alluded to arises from the nonnormalizable outgoing wave components of (the modified) $|\psi_i^{(v)}(\tau)\rangle$. However, we can approximate $|\psi_i^{(v)}(\tau)\rangle$ by a normalizable vector by truncating the harmonic expansion of $|\psi_i^{(v)}(\tau)\rangle$ at the (N_0-1)th term,

$$|\psi_i^{(v)}(\tau)\rangle = \sum_{n<N_0} |\psi_{in}^{(v)}\rangle e^{-in\tau} , \quad (4.34)$$

where the $|\psi_{in}^{(v)}\rangle$ are now determined from the following coupled equations, obtained by truncating the homogeneous equations (3.7),

$$(E_{in} - H_a)|\psi_{in}^{(v)}\rangle = V_+^{(v)}|\psi_{i,n-1}^{(v)}\rangle + V_-^{(v)}|\psi_{i,n+1}^{(v)}\rangle , \quad n < N_0 - 1$$
$$(E_{i,N_0-1} - H_a)|\psi_{i,N_0-1}^{(v)}\rangle = V_+^{(v)}|\psi_{i,N_0-2}^{(v)}\rangle . \quad (4.35)$$

It may be verified that Eqs. (4.34) and (4.35) combine to give

$$\left[E_i + \Delta_i^{(v)} + i\hbar\frac{d}{dt} - H(\tau)\right]\left|\psi_i^{(v)}(\tau)\right\rangle = -e^{-iN_0\tau}V_+^{(v)}|\psi_{i,N_0-1}^{(v)}\rangle , \quad (4.36)$$

which we can rewrite as

$$\left[H(\tau) - i\hbar\frac{d}{dt}\right]\left|F_i^{(v)}(t)\right\rangle = e^{-iE_{iN_0}t/\hbar}V_+^{(v)}|\psi_{i,N_0-1}^{(v)}\rangle ,$$

to contrast with Eqs. (3.6) and (3.15). Since E_{in} is negative for $n < N_0$ we can demand that the $\langle \mathbf{x} | \psi_{i,n}^{(v)}\rangle$ are regular at the origin and decay exponentially as r increases. Thus the set of homogeneous coupled equations (4.35) form a real eigenvalue equation, with the shift $\Delta_i^{(v)}$ determined from the condition for the existence of regular decaying solutions.[32] The width automatically vanishes.

We therefore choose the trial vector

$$|\Psi_{i,\mathrm{tr}}(t)\rangle = \exp[-i(E_i + \Delta_i^{(v)})t/\hbar]\,|\psi_i^{(v)}(\tau)\rangle ,$$

with $|\psi_i^{(v)}(\tau)\rangle$ now determined from Eqs. (4.34) and (4.35). Since this $|\psi_i^{(v)}(\tau)\rangle$ is normalizable it does not present any computational difficulties. In fact, rather than impose the normalization condition (3.5), we can impose the simpler requirement that the time average (over one cycle) of $\langle\psi_i^{(v)}(\tau)|\psi_i^{(v)}(\tau)\rangle$ be unity. Furthermore, two (truncated) Floquet eigenvectors $|\psi_i^{(v)}(\tau)\rangle$ and $|\psi_j^{(v)}(\tau)\rangle$ with different (real) quasienergy eigenvalues $\varepsilon_i^{(v)}$ and $\varepsilon_j^{(v)}$ are orthogonal in the sense that if N_{0i} and N_{0j} are the values of N_0 for ionization from states i and j, respectively, we have

$$\int_0^{2\pi} d\tau\, e^{i(N_{0i}-N_{0j})\tau}\langle\psi_j^{(v)}(\tau)|\psi_i^{(v)}(\tau)\rangle = \sum_{n<0}\langle\psi_{j,n+N_{0j}}^{(v)}|\psi_{i,n+N_{0i}}^{(v)}\rangle = \delta_{ji} \ . \tag{4.37}$$

To establish the orthogonality[33] we premultiply both sides of Eq. (4.36) by $\exp[i(N_{0i}-N_{0j})\tau]$ and by the bra $\langle\psi_j^{(v)}(\tau)|$ and we integrate over τ from 0 to 2π. This yields

$$\int_0^{2\pi} d\tau\, e^{i(N_{0i}-N_{0j})\tau}\left\langle\psi_j^{(v)}(\tau)\left|\left[\varepsilon_i^{(v)}+i\hbar\frac{d}{dt}-H(\tau)\right]\right|\psi_i^{(v)}(\tau)\right\rangle=0 \ ; \tag{4.38a}$$

there is no term on the right-hand side of this equation because the term in $\exp(-iN_{0j}\tau)\langle\psi_j^{(v)}(\tau)|$ vanishes upon integrating over τ. Interchanging i and j and taking the adjoint of Eq. (4.38a) we obtain

$$\int_0^{2\pi} d\tau\, e^{i(N_{0i}-N_{0j})\tau}\left\langle\psi_i^{(v)}(\tau)\left|\left[\varepsilon_j^{(v)}+i\hbar\frac{d}{dt}-H(\tau)\right]\right|\psi_j^{(v)}(\tau)\right\rangle^{\dagger}=0 \ . \tag{4.38b}$$

Subtracting Eq. (4.38b) from Eq. (4.38a), the terms $H(\tau)$ disappear since here $H(\tau)$ is Hermitian (the truncated Floquet eigenvectors are normalizable) and the terms in the time derivative[34] combine upon integration over τ to give a surface term proportional to $(N_{0i}-N_{0j})\hbar\omega$; it follows that

$$[(N_{0i}-N_{0j})\hbar\omega+\varepsilon_j^{(v)}-\varepsilon_i^{(v)}]\int_0^{2\pi} d\tau\, e^{i(N_{0i}-N_{0j})\tau}\langle\psi_j^{(v)}(\tau)|\psi_i^{(v)}(\tau)\rangle=0 \ ,$$

from which Eq. (4.37) follows immediately.

However, truncating the harmonic expansion at the (N_0-1)th term is fairly drastic, and so we try to compensate for this by improving the trial approximation vector $|\Psi_{f,\mathrm{tr}}(t)\rangle$ to $|\Psi_f(t)\rangle$, which represents the development into the final unbound state f. Rather than take $|\Phi_f^{(v)}(t)\rangle$ for $|\Psi_{f,\mathrm{tr}}(t)\rangle$, we choose the Kroll-Watson scattering wave vector $|KW_f^{(v)}(t)\rangle$ [cf. Eq. (3.23)], which provides an accurate description of those harmonic components $|\psi_{fn}^{(v)}\rangle$ of $|\psi_f^{(v)}(\tau)\rangle$ for which $|n\hbar\omega| \ll E_f$. We observe that the field is turned on at $t=0$ so that $|\Psi_{f,\mathrm{tr}}(0)\rangle=|\Psi_{\mathbf{k}_f}^-\rangle$, but as before we neglect the contribution from the interval $[0,t_0]$ to the integral over t in Eq. (4.5b). We have

$$M_n=\frac{\omega}{2\pi}\int_0^{2\pi/\omega} dt\, e^{in\tau}\left[\left\langle\psi_i^{(v)}(\tau)\left|\left[H(\tau)-E_f-i\hbar\frac{d}{dt}\right]e^{i\mathbf{x}\cdot\mathbf{k}_A(\tau)-i\theta(\tau)}\right|\Phi_{\mathbf{k}_f(\tau)}^-\right\rangle\right]^{\dagger} .$$

Integrating by parts the term in the time derivative yields, for $n=N$,

$$M_N=\frac{\omega}{2\pi}\int_0^{2\pi/\omega} dt\, e^{iN\tau+i\theta(\tau)}\left\langle\Phi_{\mathbf{k}_f(\tau)}^-\left|e^{-i\mathbf{x}\cdot\mathbf{k}_A(\tau)}\left[H(\tau)^{\dagger}-(E_i+\Delta_i)-i\hbar\frac{d}{dt}\right]\right|\psi_i^{(v)}(\tau)\right\rangle \ . \tag{4.39}$$

Noting that $H(\tau)$ is Hermitian since $|\psi_i^{(v)}(\tau)\rangle$ is normalizable, and using Eq. (4.36), we arrive at

$$M_N=\frac{1}{2\pi}\int_0^{2\pi} d\tau\, e^{i(N-N_0)\tau+i\theta(\tau)} \times\langle\Phi_{\mathbf{k}_f(\tau)}^-|e^{-i\mathbf{x}\cdot\mathbf{k}_A(\tau)}V_+^{(v)}|\psi_{i,N_0-1}^{(v)}\rangle \ , \tag{4.40}$$

a result obtained previously[11] in the length gauge. The advantage of using the velocity gauge is that expression (4.40) for M_N remains finite even in the strong-field limit; in contrast, the corresponding length-gauge expression[11] for M_N is finite only in the weak-field limit.[32] We refer to the approximation of Eq. (4.40) as the "threshold-truncated approximation" (TTA). In discussing the weak-field limit we consider the TTA in both its velocity-gauge and length-gauge forms, but unless the gauge is specified otherwise it is understood to be the velocity gauge.

The integration over τ in Eq. (4.40) serves to project out all but one of the harmonic components $|\psi_{fn}^{(v)}\rangle$ of

$$\exp(iE_f t/\hbar)|KW_f^{(v)}(t)\rangle \ ;$$

the component which survives is the (N_0-N)th, corresponding to the emission of $N-N_0$ photons. (To simplify notation we do not distinguish between trial and exact harmonic components.) We see, noting Eq. (3.23), that the expression on the right-hand side of Eq. (4.40) is just the spatially integrated matrix element of the one-photon absorption operator $V_+^{(v)}$ sandwiched between the trial

harmonic components $|\psi_{i,N_0-1}\rangle$ and $|\psi_{f,N_0-N}\rangle$. Now $|N_0-N|\hbar\omega$ is almost equal to E_f, and is therefore not small compared to E_f, so that we cannot expect the Kroll-Watson approximation to give a highly accurate trial component $|\psi_{f,N_0-N}\rangle$ (except if $N=N_0$ and the field is weak, since both the trial and exact $|\psi_{f0}\rangle$ approach $|\Phi_{\mathbf{k}_f}^-\rangle$.) Nevertheless, the Kroll-Watson approximation allows us to at least partially build the effects of both the field and the atomic potential into $|\Psi_{f,\mathrm{tr}}(t)\rangle$. The merit of the TTA must of course be judged against the comparison of its predictions with exact results. At present, exact results are available only for the weak-field limit; as we shall see in Sec. V, at least in this limit the TTA is reasonable. We conclude this section with a few remarks on other approximations.

E. High-Rydberg initial state; $N=1$

If the initial state is a high-Rydberg state we can have (with the parameters β defined in Appendix A) $\beta_i^{(v)}\ll 1$ and $\beta_i^{(l)}\gtrsim 1$, the quasistrong regime. Consider one-photon ionization to a state with $\beta_f^{(v)}\ll 1$ and $\beta_f^{(l)}=P/\hbar\omega\gtrsim 1$. These values for the final state parameters can be realized if the electron is ejected with an energy not far above threshold (which happens for a sufficiently low frequency) and if the intensity is sufficiently high. Working in the velocity gauge, and neglecting $\Delta_i^{(v)}$, we are led to the reasonable choices

$$|\Psi_{i,\mathrm{tr}}(t)\rangle=e^{-iE_i t/\hbar}|\Phi_i\rangle\ , \tag{4.41a}$$

$$|\Psi_{f,\mathrm{tr}}(t)\rangle=e^{-iE_f t/\hbar}|\Phi_{\mathbf{k}_f}^-\rangle\ . \tag{4.41b}$$

We thereby obtain

$$M_1=\langle\Phi_{\mathbf{k}_f}^-|V_+^{(v)}|\Phi_i\rangle\ . \tag{4.42}$$

Replacing $V_+^{(v)}$ by $V_+^{(l)}$ in the above matrix element yields a different value for M_1 unless $\beta_f^{(l)}\ll 1$. To obtain the expression which in the length gauge corresponds to Eq. (4.42) we must gauge-transform the trial vectors of Eqs. (4.41).

F. Another limit; $N=1$

We now consider one-photon ionization from a state in which the electron is tightly bound and both $\beta_i^{(v)}\ll 1$ and $\beta_i^{(l)}\ll 1$. We choose the frequency so that the electron is ejected with an energy not far above threshold so that $\beta_f^{(v)}\ll 1$. However, we choose a sufficiently intense field that $\beta_f^{(l)}\gtrsim 1$. We obtain Eq. (4.42) once again.

G. Reiss approximation

Many other approximate expressions for the N-photon ionization amplitude can be derived by choosing different trial vectors. For example, without commenting at this stage on its justification, we choose trial vectors

$$|\psi_{i,\mathrm{tr}}(t)\rangle=e^{-iE_i t/\hbar}|\Phi_i\rangle\ , \tag{4.43a}$$

$$|\Psi_{f,\mathrm{tr}}(t)\rangle=e^{-i(E_f+P)t/\hbar-i\zeta(\tau)-i\theta(\tau)}|\mathbf{k}_f\rangle\ . \tag{4.43b}$$

Here $|\Psi_{i,\mathrm{tr}}(t)\rangle$ represents the initial bound state unperturbed by the field, and $|\Psi_{f,\mathrm{tr}}(t)\rangle$ represents a free electron moving through the field, with the $\mathbf{A}^2$ interaction $V_0(\tau)$ now included in the velocity gauge. These trial vectors are a considerable simplification to those used in the TTA. Inserting the trial vectors of Eqs. (4.43) into the right-hand side of Eq. (4.5a) leads to the approximate amplitude considered by several authors,[6] and popularized by Reiss[6],

$$M_N=\frac{1}{2\pi}\int_0^{2\pi}d\tau\, e^{i(N-N_0)\tau+i\theta(\tau)+i\zeta(\tau)}\times\langle\mathbf{k}_f|[V^{(v)}(\tau)+V_0(\tau)]|\Phi_i\rangle\ . \tag{4.44}$$

V. RESULTS

We now present results of an application of the TTA to the calculation of rates for multiphoton ionization of hydrogen initially in its ground state. We begin by considering the weak-field limit. In this limit we have $\Delta_i\to 0$ and

$$|\psi_{in}\rangle\to|\Phi_{in}\rangle\equiv G_a(E_i+i\eta+n\hbar\omega)V_\pm\cdots\times G_a(E_i+i\eta\pm\hbar\omega)V_\pm|\Phi_i\rangle\ , \tag{5.1}$$

where $G_a(E)$ is the atomic Green's operator, and where the choice of sign is plus if $n>0$ and minus if $n<0$. We can apply the TTA in both the velocity *and* length gauges in the weak-field limit, but we do not expect to obtain the same result in both gauges when $N>N_0$ since we truncate the harmonic expansion of $|\psi_i(\tau)\rangle$ at the (N_0-1)th term. In lowest-order perturbation theory,[35,36] which is exact in the weak-field limit, we have the matrix element

$$M_N=\langle\Phi_{\mathbf{k}_f}^-|V_+|\Phi_{i,N-1}\rangle\ , \tag{5.2}$$

where $|\Phi_{i,N-1}\rangle$ is given by Eq. (5.1). The value of this matrix element is the same in both gauges for all $N\geq N_0$ (see Appendix C). Note that in the weak-field limit M_{N_0} is the same in the TTA as it is in lowest-order perturbation theory. In Table I we present results of calculations of partial rates in the form of ratios of approximate results to exact results (with the latter obtained in lowest-order perturbation theory). We show results obtained in the TTA, in both gauges, and we also show results which follow from modifying the TTA in the velocity gauge by replacing $|\Phi_{\mathbf{k}_f(\tau)}^-\rangle$ in Eq. (4.40) either by

$$e^{i\mathbf{x}\cdot\mathbf{k}_A(\tau)}|\Phi_{\mathbf{k}_f(\tau)}^-\rangle\to|\Phi_{\mathbf{k}_f}^-\rangle\ , \tag{5.3}$$

which corresponds to using[28] the trial wave vector $|\Phi_f^{(v)}(t)\rangle$ of Eq. (2.17), or by

$$e^{i\mathbf{x}\cdot\mathbf{k}_A(\tau)}|\Phi_{\mathbf{k}_f(\tau)}^-\rangle\to|\mathbf{k}_f\rangle\ , \tag{5.4}$$

which amounts to neglecting the atomic potential in the final state. (Either replacement leads to a considerable simplification in the evaluation of M_N.) In the last column of Table I we show results obtained from the Reiss approximation, Eq. (4.44). All of the versions of

TABLE I. Ratios of approximate to exact estimates of partial rates for N-photon ionization of H by circularly polarized light of wavelength λ in the weak-field limit. The lowest value of N for each wavelength is the minimum number N_0 of photons required to ionize the atom. The entries in each column correspond to (a) threshold-truncated approximation of Eq. (4.40), in velocity gauge; (b) same approximation in length gauge; (c) same as (a), but with replacement of Eq. (5.3); (d) same as (a), but with replacement of Eq. (5.4); (e) Reiss approximation, Eq. (4.44). The exact results at $\lambda=265$ nm were taken from Gontier *et al.* (Ref. 35).

λ (nm)	N	(a)	(b)	(c)	(d)	(e)
50	1	1.0	1.0	1.0	1.7	1.7
50	2	0.44	0.65	0.41	0.94	0.94
50	3	0.31	0.66	0.32	0.72	0.72
80	1	1.0	1.0	1.0	0.34	0.34
80	2	0.33	0.41	0.24	0.48	0.48
80	3	0.20	0.36	0.17	0.38	0.38
120	2	1.0	1.0	1.0	6.6	0.0046
120	3	0.36	1.0	0.45	2.6	0.0062
120	4	0.22	1.2	0.32	1.5	0.0062
140	2	1.0	1.0	1.0	1.7	0.029
140	3	0.40	0.58	0.32	0.96	0.035
140	4	0.24	0.53	0.21	0.65	0.035
230	3	1.0	1.0	1.0	0.93	0.0048
230	4	0.46	0.54	0.30	0.77	0.01
265	3	1.0	1.0	1.0	0.0062	0.000 062
265	4	0.58	0.47	0.23	0.29	0.0058
265	5	0.31	0.31	0.14	0.28	0.0078
265	6	0.21	0.30	0.10	0.25	0.0080
265	7	0.16	0.29	0.09	0.21	0.0082
265	8	0.13	0.29	0.07	0.19	0.0078
300	4	1.0	1.0	1.0	2.0	0.0022
300	5	0.52	0.63	0.37	1.1	0.0047
350	4	1.0	1.0	1.0	0.0086	0.000 029
350	5	0.78	0.54	0.28	0.26	0.0022

TABLE II. Partial differential rates (in a.u.), divided by the Nth power of the intensity, for N-photon ionization of H in the weak-field limit by linearly polarized light of wavelength λ, with ejection in the direction of the polarization vector. The exact results, where available, were taken from Gontier *et al.* (Ref. 35). The entries in each column correspond to (a) exact results; (b) the TTA of Eq. (4.40), in the velocity gauge; (c) same approximation in length gauge; (d) Reiss approximation, Eq. (4.44). The numbers in square brackets represent powers of ten.

λ (nm)	N	(a)	(b)	(c)	(d)
265	3	7.2[1]	7.2[1]	7.2[1]	1.4[0]
265	4	7.9[3]	2.2[3]	2.2[3]	5.1[0]
265	5	1.0[5]	1.5[4]	1.8[4]	7.1[−1]
265	6	7.8[5]	1.1[5]	1.1[5]	9.7[1]
265	7	4.5[6]	6.9[5]	5.5[5]	8.9[2]
355	4		5.5[4]	5.5[4]	2.1[0]
355	5		1.4[6]	1.7[6]	1.8[3]
532	6		7.0[8]	7.0[8]	6.0[4]
532	7		8.2[10]	7.3[10]	5.5[6]
532	8		2.5[12]	3.2[12]	9.1[7]
532	9		1.0[14]	1.3[14]	1.4[10]
532	10		4.6[15]	5.2[15]	5.1[11]
532	11		2.2[17]	1.8[17]	1.2[13]

the TTA yield results that, with a few exceptions, are of the correct order of magnitude. While there are significant differences between the various TTA results there is no one version of the TTA which appears to be *significantly* more accurate than the others, although overall the use of the Kroll-Watson wave function in the unmodified TTA leads to superior results. We see that the Reiss approximation does not yield results that are of the correct order of magnitude when N_0 is greater than about 2, and this is a consequence of retaining just a single term (the $n=0$ term) in the harmonic expansion of $|\psi_i^{(v)}(\tau)\rangle$; the electron reaches the continuum threshold by climbing up the harmonic components of

$$\exp[-i\zeta(\tau)-i\theta(\tau)]\,|\,\mathbf{k}_f\rangle$$

that correspond to electron energies below the continuum threshold, but the strong coupling of the below-threshold components of $|\Psi_{f,\mathrm{tr}}(t)\rangle$ to the initial bound state i has been neglected. We note here that although the Reiss approximation is intended as a strong-field approximation, the feature that is responsible for its failure in the weak-field limit remains present in the strong-field limit. In Table II we show results for partial differential rates for N-photon ionization by linearly polarized light, with ejec-

tion in the direction of the polarization vector. We see that both the velocity- and length-gauge forms of the TTA yield results that again are of the correct order of magnitude. While the TTA is not nearly as simple to use as the Reiss approximation, estimates of rates into continuum channels labeled by positive $S \equiv N - N_0$ can be more readily obtained using the TTA than using lowest order perturbation theory. In fact, once $|\psi_{i,N_0-1}\rangle$ has been calculated, the effort involved in using the TTA to estimate rates into positive S channels is no more than that required to obtain the rate into the $S=0$ channel. In contrast, to estimate rates into positive S channels using lowest-order perturbation theory one must calculate $|\psi_{i,N}\rangle$, which is nonnormalizable and significantly complicates the calculation.

Continuing our discussion of the weak-field limit we

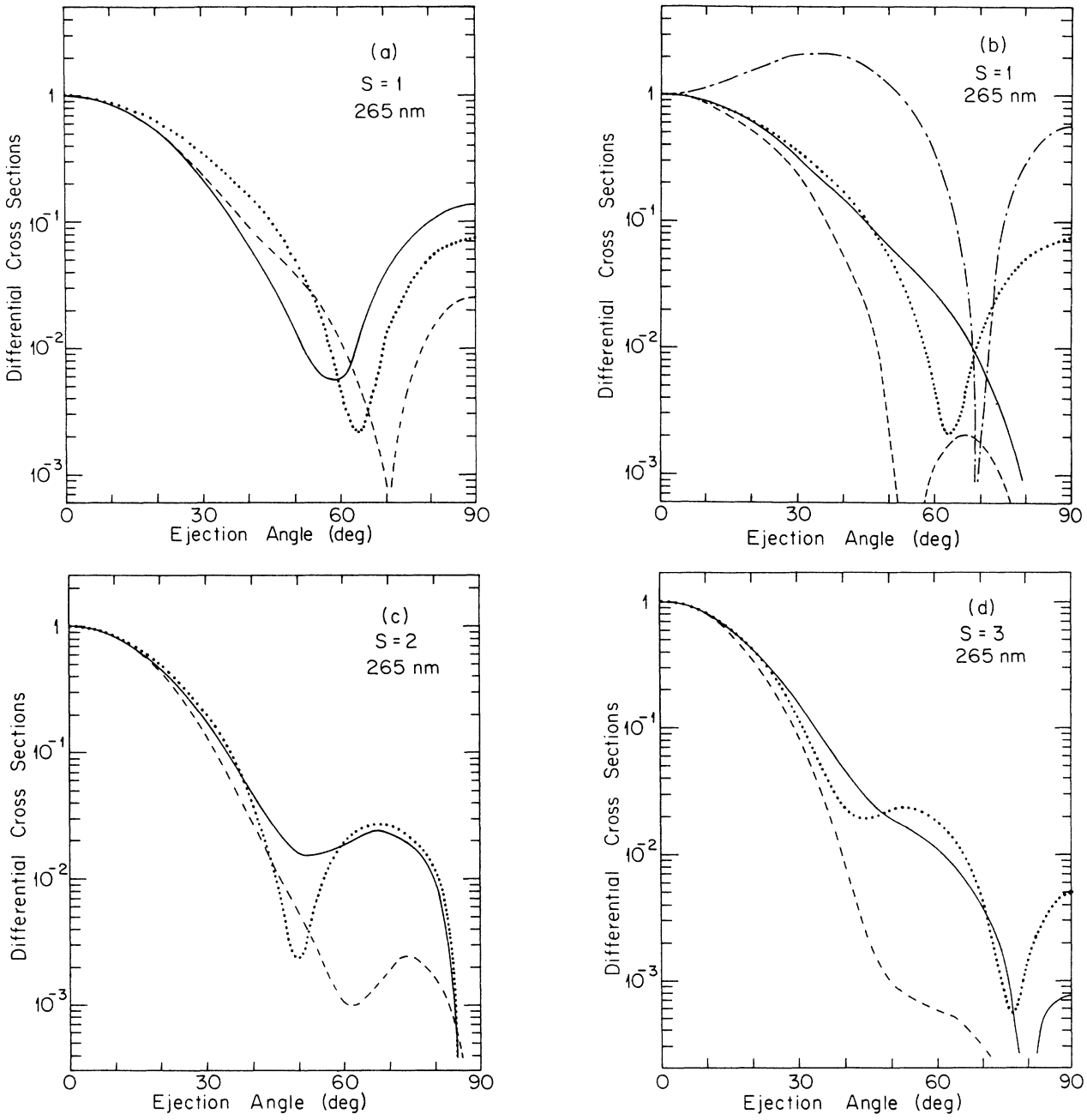

FIG. 1. Angular distributions for N-photon ionization of H by a weak linearly polarized field of wavelength 265 nm as a function of angle measured relative to the polarization vector. We have normalized the angular distributions to unity in the forward direction. In each figure the curve · · · · represents the exact results. We have (a) ——, $N=4$ ($S=1$), TTA, in velocity gauge, according to Eq. (4.40); — — —, same approximation in length gauge; (b) ——, $N=4$, ($S=1$), TTA, in velocity gauge, modified according to Eq. (5.3); — — —, TTA, in velocity gauge, modified according to Eq. (5.4); —·—·—·, Reiss approximation, Eq. (4.44); (c) $N=5$ ($S=2$), otherwise same as (a); (d) $N=6$ ($S=3$), otherwise same as (a).

turn to angular distributions. In Fig. 1 we show angular distributions for ionization of H by a weak linearly polarized field of wavelength 265 nm ($N_0=3$). We show exact results and those obtained in the velocity- and length-gauge forms of the TTA for the $S=1$, 2, and 3 channels. We also show, for the $S=1$ channel, results obtained using the two modified versions of the TTA discussed above, as well as the Reiss approximation. From symmetry considerations the differential cross section vanishes when the angle θ of the ejected electron, measured relative to the polarization vector, is 90°, provided that N is odd. (Note that $\langle \mathbf{x} \mid \Phi_i \rangle$ is even under $\hat{\boldsymbol{\varepsilon}}\cdot\mathbf{x} \rightarrow -\hat{\boldsymbol{\varepsilon}}\cdot\mathbf{x}$.) This prediction is confirmed by all of the approximations. However, the two modified forms of the TTA predict that the differential cross section at $\theta=90°$ vanishes for all $S \geq 1$, even when N is even. The unmodified TTA and the Reiss approximation both correctly predict a nonzero differential cross section at $\theta=90°$ when N is even, but for quite different reasons [the influence of the atomic potential W in the TTA, and the influence of the ponderomotive potential $V_0(\tau)$ in the Reiss approximation]. The velocity-gauge form of the TTA appears to give the best agreement with the exact angular distributions. This is, in fact, typical, and we have found that in general the TTA (velocity-gauge form) reproduces the overall envelopes of the exact angular distributions reasonably well, although it often fails to reproduce the detailed structure.

We now turn to strong fields. We have solved Eqs. (4.35) by expanding the $\langle \mathbf{x} \mid \psi_{in}^{(v)} \rangle$ in spherical harmonics, with the radial parts expanded in a discrete basis set composed of Sturmian functions. The coefficients of the basis functions satisfy a homogeneous matrix eigenvalue equation.[37] Only spherical harmonics with orbital angular momentum quantum number ≤ 7, and harmonic components $\mid \psi_{in}^{(v)} \rangle$ with photon index in the range $-3 \leq n (\leq N_0 - 1)$, were included. This was sufficient to ensure convergence over the range of intensities studied. In Fig. 2 we show the angular distributions obtained in the TTA for ionization of H into the $S=1$, 2, and 3 channels by a linearly polarized field of wavelength 532 nm ($N_0=6$) and intensity 5×10^{12} W/cm^2, and we compare our results with the experimental data of Feldmann *et al.*[3] (Actually, the intensity of the field is not really strong, and lowest-order perturbation theory still applies. The angular distributions in lowest-order perturbation theory have been recently calculated by Kracke *et al.*,[38] who obtained very good qualitative agreement with the experimental data of Feldmann *et al.*[3]) The overall envelopes of the experimentally observed angular distributions are reproduced reasonably well by the TTA but the structure in the $S=1$ channel is only barely hinted at and the structure in the $S=2$ and 3 channels is missing altogether. The modified versions of the TTA, modified according to Eqs. (5.3) and (5.4), do not give as good overall agreement, nor does the Reiss approximation, as indicated in Fig. 2(c).

To determine the intensity at which lowest-order perturbation theory breaks down we have calculated the index of nonlinearity $K(\theta)$, defined here as the derivative with respect to $\ln I$ (where I is of the intensity) of the logarithm of the differential cross section for ejection at angle

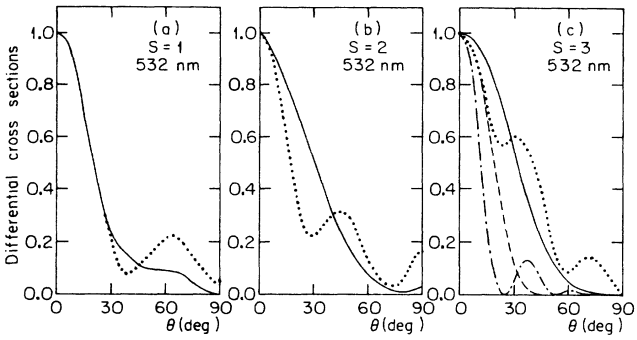

FIG. 2. Angular distributions, normalized to unity in the forward direction, for N-photon ionization of H by linearly polarized light of wavelength 532 nm and intensity 5×10^{12} W/cm^2. The angle θ is measured relative to the polarization vector. The curves are · · · ·, experimental data of Feldmann *et al.* (Ref. 3); ——, TTA; — — —, TTA modified according to Eq. (5.3); —·—·—·, Reiss approximation. We have (a) $N=7$ ($S=1$); (b) $N=8$ ($S=2$); (c) $N=9$ ($S=3$). Results obtained using the TTA modified according to Eq. (5.4) are almost indistinguishable from the curve — — — in (c).

θ into a specific continuum channel. In Fig. 3 we show $K(\theta=0)$ for ionization of H by linearly polarized light of wavelength 532 nm. In the weak-field limit the index of nonlinearity is just the number N of photons absorbed in the specific channel. The intensity at which $K(\theta)$ begins to deviate from a straight line is the intensity at which lowest-order perturbation theory becomes invalid. Note that, in Fig. 3, $K(\theta=0)$ turns downwards at high intensities; this is because the ionization potential increases as the intensity increases, and the threshold is approached at which the minimum number N_0 of photons required for ionization increases by 1.[15] The departure from the weak-field limit is not always signaled by a downward turn in $K(\theta)$. In Fig. 4 we show $K(\theta=0)$ for ionization by linearly polarized light of wavelength 355 nm and we see that $K(\theta=0)$ turns upwards at high intensities. This

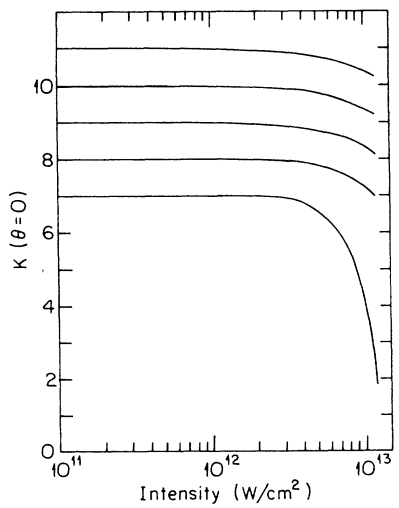

FIG. 3. Index of nonlinearity vs laser intensity for ejection into the forward direction by light of wavelength 532 nm.

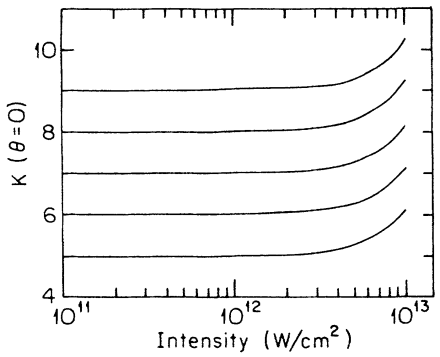

FIG. 4. Same as Fig. 3 but for the wavelength 355 nm.

is because the wavelength 355 nm is close to the wavelength for an intermediate three-photon resonant transition between the $1s$ and $2p$ states, and the resonance wavelength moves towards 355 nm as the intensity increases, thereby enhancing the cross section.

Unfortunately, the laser intensities used in the experiment of Feldmann *et al.*[3] were not sufficiently high to observe resonance enhancement of the ionization rate at 355 nm. Nevertheless, we now explore this intermediate resonance in more detail. For generality we begin by discussing the case where there is an intermediate m-photon resonant transition between the bound states i and j. The electron is excited into a superposition of the bound states i and j as the field is turned on, and its state can best be described in terms of the two dressed states represented by the Floquet eigenvectors $|\psi_i^{(v)}(\tau)\rangle$ and $|\psi_j^{(v)}(\tau)\rangle$, which are determined by solving Eqs. (4.35). We have $m=N_{0i}-N_{0j}$. The (real) quasienergy eigenvalues $\varepsilon_i^{(v)}$ and $\varepsilon_j^{(v)}$ differ by very nearly $m\hbar\omega$, and in the weak-field limit we have—recall Eq. (5.1)—

$$|\psi_{in}^{(v)}\rangle \rightarrow |\Phi_{in}^{(v)}\rangle \ , \tag{5.5a}$$

$$|\psi_{jn}^{(v)}\rangle \rightarrow |\Phi_{jn}^{(v)}\rangle \ . \tag{5.5b}$$

As the intensity varies, the shifts $\Delta_i^{(v)}$ and $\Delta_j^{(v)}$ also vary, and the resonance frequency may move closer to the laser frequency ω. In other words, if we plot $\varepsilon_i^{(v)}$ and $\varepsilon_j^{(v)}-m\hbar\omega$ versus intensity, these curves may approach one another. However, rather than cross, the curves in general repel each other and there is an avoided crossing. Following the usual nomenclature,[39] we refer to the intensity I_r at which the curves are at closest approach as the "crossing" point; at this point the frequency ω is resonant with the m-photon $i\rightarrow j$ transition. As the intensity passes through the crossing point the characters of the dressed-state eigenvectors interchange, that is, at intensities well above the crossing point we have (assuming that the field is still relatively weak)

$$|\psi_{in}^{(v)}\rangle \rightarrow |\Phi_{j,\,n-m}^{(v)}\rangle \ , \tag{5.6a}$$

$$|\psi_{jn}^{(v)}\rangle \rightarrow |\Phi_{i,\,n+m}^{(v)}\rangle \ . \tag{5.6b}$$

In the lower portion of Fig. 5 we have plotted $\varepsilon_{1s}^{(v)}$ and $\varepsilon_{2p}^{(v)}-3\hbar\omega$ versus intensity for the case where H is exposed to light of wavelength 355 nm; as noted above, there is a three-photon resonance with the $1s\rightarrow 2p$ transition. (The

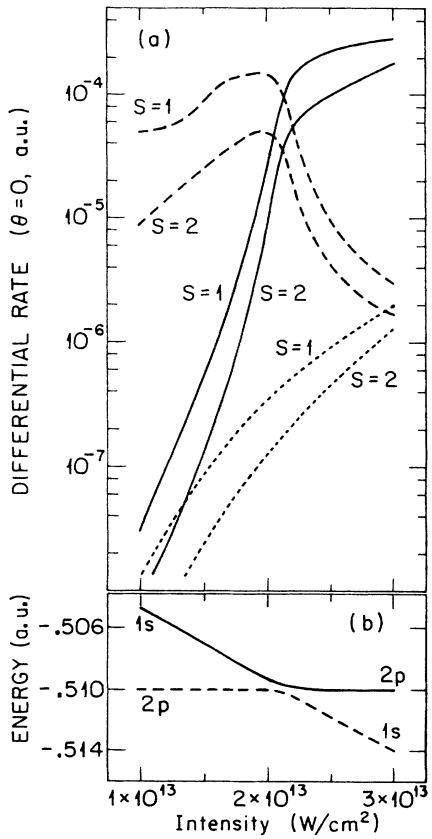

FIG. 5. Ionization of H by linearly polarized light of wavelength 355 nm. At this wavelength there is an intermediate three-photon resonance with the $1s\rightarrow 2p$ transition. (a) Partial differential rates for ejection in the direction of the polarization vector. The solid and dashed curves refer to ionization from the dressed states which in the weak-field limit approach the $1s$ and $2p$ bound states, respectively. The dotted curves are the ionization rates from the $1s$ state calculated in lowest-order perturbation theory. (b) The quasienergy eigenvalues for the two dressed states: ——, $\varepsilon_{1s}^{(v)}$; — — —, $\varepsilon_{2p}^{(v)}-3\hbar\omega$.

index n of $|\psi_{jn}^{(v)}\rangle$ was restricted to the same range, except shifted downwards by m, as the index n of $|\psi_{in}^{(v)}\rangle$.) Note that $\Delta_{2p}^{(v)}$ is small at intensities below the crossing point, and $\Delta_{1s}^{(v)}$ is small at intensities above the crossing point. This is because the interaction $V^{(v)}(\tau)$ induces only a small shift in an excited state (and an even smaller shift in a highly excited state—and no shift in the continuum). In the upper portion of Fig. 5 we show the rates (calculated in the TTA) for ionization into the $S=1$ and 2 channels from each of the two dressed states. At intensities well below I_r the rates for ionization from the dressed state which develops from the $2p$ bound state far exceed those for ionization from the dressed state which develops from the $1s$ bound state; this is because the former dressed state is predominantly the $2p$ bound state, from which ionization occurs rapidly. However, as the crossing point is approached, the rates for ionization from the dressed state which evolves from the $2p$ state reach their maxima, and decrease as the intensity is increased further because $|\psi_{2p}^{(v)}(\tau)\rangle$ acquires $1s$ character.[40] In contrast, rates for ionization from the dressed state which evolves

from the $1s$ state rise very rapidly as the intensity increases towards the crossing point because $|\psi_{1s}^{(v)}(\tau)\rangle$ picks up more and more of the $2p$ component; these rates continue to rise as the crossing point is passed, but less steeply because $|\psi_{1s}^{(v)}(\tau)\rangle$ has fully acquired $2p$ character. The ionization rates calculated in lowest-order perturbation theory, which are also shown in Fig. 5(a), approach the TTA rates at intensities well above I_r but are too small in the vicinity of the resonance.

The electron starts out on the quasienergy eigenvalue curve corresponding to the dressed state that approaches the initial bound state in the limit of vanishing intensity. If the intensity is varied slowly, and if the energy gap at the crossing point between the initial eigenvalue curve and another one is larger than the laser bandwidth, the electron will adiabatically follow the initial eigenvalue curve, and ionization will proceed from the corresponding dressed state. On the other hand, if the intensity varies sufficiently rapidly in the vicinity of the crossing point, the electron will jump across the gap, onto the other eigenvalue curve, even if the energy gap is large compared to the laser bandwidth. To be specific, if t_r is the transit time through the crossing point (defined as the time taken for the intensity to vary from a point on the left of the crossing point at which the energy gap is roughly twice the minimum energy gap to a similar point on the right) the electron will jump curves if the fluctuation energy $\hbar/t_r$ is comparable to or larger than the energy gap.[39] No matter how the intensity varies, if the laser bandwidth is comparable to or larger than the energy gap at the crossing point the electron must be described, at all but very low intensities, by a superposition of dressed states which represents the atom flopping back and forth between the bound states i and j as it ionizes. It is clear from Fig. 5 that were the electron to remain on its initial eigenvalue curve, the ionization rate would continue to rise as the intensity increases above I_r, and consequently the ionization yield, as a function of intensity, would also continue to rise, unless depletion effects were to enter. On the other hand, if the electron were to jump to the other eigenvalue curve as the crossing point is passed, the ionization rate, and therefore the yield, would drop as the intensity increases above I_r. In the recent experiment of Freeman *et al.*,[4] xenon atoms were ionized by a short powerful pulse. (If the pulse is short, the spatial inhomogeneity of the laser focus does not affect the motion of the electron on its way to the detector because the light disappears before the electron has a chance to travel a sufficient distance to experience its spatial inhomogeneity.) During the rise and fall of the pulse, as the intensity varied, Rydberg states shifted in and out of resonance. Freeman *et al.* resolved each peak in the ionization signal (corresponding to ionization to a specific continuum channel) into a series of subpeaks, with each subpeak corresponding to an energy shift at which there is an intermediate resonance with a transition to a Rydberg state. Since subpeaks were observed, rather than simply a monotonic increase in the yield, we conclude that the electron jumped eigenvalue curves as each crossing point was passed.

To study the dynamics near a curve crossing we have solved the time-dependent Schrödinger equation within a two-state model. We approximate the wave vector of the electron by the superposition

$$|R(t)\rangle = a_i(t)e^{-iS_i(t)}|\psi_i(\tau)\rangle + a_j(t)e^{-iS_j(t)}|\psi_j(\tau)\rangle \ , \tag{5.7}$$

where, with $l=i$ or j,

$$S_l(t) = \frac{1}{\hbar}\int_0^t dt' \varepsilon_l \ , \tag{5.8}$$

and where the eigenvalues ε_l and eigenvectors $|\psi_l(\tau)\rangle$ are explicitly dependent on the intensity I and implicitly dependent on the time t through $I=I(t)$. The boundary condition on the coefficients $a_l(t)$ is $a_l(0)=\delta_{li}$, and these coefficients satisfy the coupled equations (see Appendix D)

$$\frac{d}{dt}a_i(t) = \frac{dI(t)}{dt}e^{im\tau + iS_{ij}(t)}U_{ij}(I)a_j(t) \ , \tag{5.9a}$$

$$\frac{d}{dt}a_j(t) = \frac{dI(t)}{dt}e^{-im\tau + iS_{ji}(t)}U_{ji}(I)a_i(t) \ , \tag{5.9b}$$

where $S_{ij}(t)=S_i(t)-S_j(t)$ and where

$$U_{ij}(I) = -\sum_{n<0}\left\langle \psi_{i,\,n+N_{0i}} \left| \frac{d}{dI} \right| \psi_{j,\,n+N_{0j}} \right\rangle \ , \tag{5.10}$$

with $U_{ji}(I)=-U_{ij}(I)$. At intensities far from the crossing point the eigenvectors vary relatively slowly with I and so the coupling potential $U_{ij}(I)$ is relatively small. However, at intensities near the crossing point, where the eigenvectors undergo a rapid change of character, $U_{ij}(I)$ is large (see Appendix D).

The total ionization yield is the sum of the yields from the two dressed states. This sum is incoherent provided that the laser bandwidth is small compared to the energy gap at the crossing point,[41] since in principle one could tell by an energy measurement which dressed state the electron emerged from. If M_{iN} and $M_{j,\,N-m}$ denote the matrix elements for ionization from the two dressed states, the total N-photon ionization yield over a time interval dt, during which the intensity changes by dI, is, for the incoherent process,

$$dY_N(I,\theta) = (2\pi/\hbar)\rho'(E_{iN})(dI/dt)^{-1}dI \times [\,|a_i(t)M_{iN}|^2 + |a_j(t)M_{j,\,N-m}|^2\,] \ , \tag{5.11}$$

where, of course, M_{iN} and $M_{j,\,N-m}$ depend explicitly on I. If the laser bandwidth is comparable to or larger than the energy gap at the crossing point the atom will ionize as it undergoes Rabi oscillations and the process is coherent; we must replace the expression in square brackets on the right-hand side of Eq. (5.11) by

$$|a_i(t)M_{iN} + e^{iS_{ij}(t)+im\tau}a_j(t)M_{j,\,N-m}|^2 \ .$$

We have solved Eqs. (5.9) for the coefficients $a_l(t)$ in several cases. We allowed for depletion of the dressed states by including the half widths $\Gamma_l/2$ in the quasienergy eigenvalues, with Γ_l calculated at each value of I from

$$\Gamma_l=(2\pi/\hbar)\sum_{N\geq N_{0l}}\int d\Omega\,\rho'(E_{iN})\,|M_{l,N-n}|^2\,, \qquad (5.12)$$

where $n=m\delta_{lj}$ and where the integration is over the full solid angle into which the electron is ejected. In Fig. 6 we show the yield, integrated over angle, for three-photon ionization of H into the $S=0$ channel by linearly polarized light in the form of a pulse

$$I(t)=I_0\exp(-t^2/t_p^2)\,,$$

whose pulse duration is $t_p=0.2$ ps, whose peak intensity is $I_0=5\times10^{13}$ W/cm^2, and whose wavelength is either 204 or 206 nm. There is an intermediate two-photon resonance with the $1s\rightarrow3s$ transition at the wavelength 204 nm, and the laser bandwidth ($\sim1/t_p$) is (marginally) smaller than the energy gap at the crossing point of the eigenvalue curves. We calculated[42] the yield from the incoherent sum, Eq. (5.11), and show in Fig. 6 the envelope of a histogram of yield versus intensity for each wavelength. (Since the pulse is symmetric, the time t is a double-valued function of the intensity I, and two time intervals contribute to dI.) The peak in the yield at intensities between $(1-2)\times10^{13}$ W/cm^2 for the wavelength 204 nm is due to the intermediate $1s\rightarrow3s$ transition which becomes resonant at intensities in this range. Two photons are insufficient to excite the $3s$ state at the wavelength 206 nm, and the detuning from resonance increases as the intensity increases so that there is no resonance enhancement during the rise and fall of the pulse at this wavelength. After the pulse has died out, ionization is only 2% complete for 206 nm, while it is 72% complete for 204 nm. The probability that the electron jumps eigenvalue curves as the intensity increases past the crossing point (for 204 nm) was calculated to be 73% when depletion was not taken into account; this estimate is significantly smaller than the 93% predicted by the short-pulse approximation, Eqs. (D8). (This latter approximation improves for smaller t_p.) However, when depletion was taken into account, the population on the initial eigenvalue curve relative to that on the other curve was found to be significantly smaller at intensities $I>I_r$ then suggested by a jump probability of 73%. This is because ionization proceeds rapidly from the initial curve at intensities $I>I_r$. All this said, the probability for the electron to jump curves is sufficiently high that the yield decreases as I increases above I_r, and this decrease is accelerated by depletion. The electron may, of course, jump back to the initial eigenvalue curve as the intensity *decreases* past the crossing point; we observed no return jump in the case of the present pulse. Whether or not the return jump were to occur with high probability, the yield would exhibit a peak (rather than a shoulder) since the intensity is decreasing when the return jump occurs. The sharp rise in the yield near the peak intensity (for both wavelengths) occurs because the derivative dI/dt, whose inverse appears as a factor in the expression (5.11) for the yield, vanishes at the peak intensity.

In Fig. 7 we show the eigenvalue curves for H irradiated by linearly polarized light of wavelength 384.5 nm ($N_0=5$). This is a more complicated situation; there are several intermediate resonances, corresponding to four-

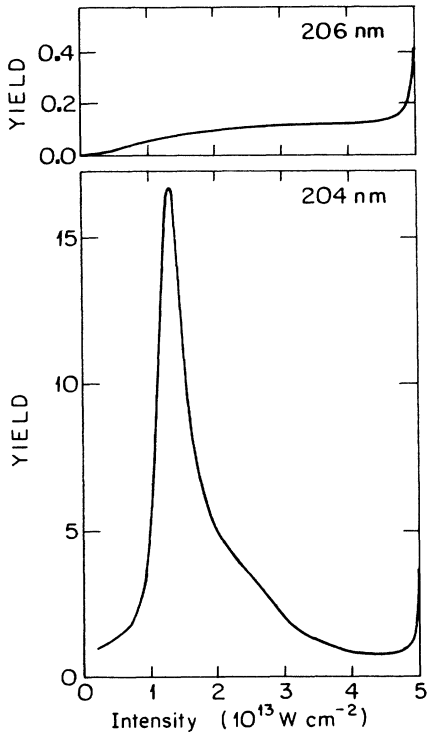

FIG. 6. Yield vs intensity for ionization of H into the $S=0$ channel by a 0.2-ps Gaussian pulse of wavelength 204 or 206 nm. There is an intermediate two-photon $1s\rightarrow3s$ resonance at 204 nm. The units of the yield are $(4\pi/3)\times10^{-3}$ a.u.

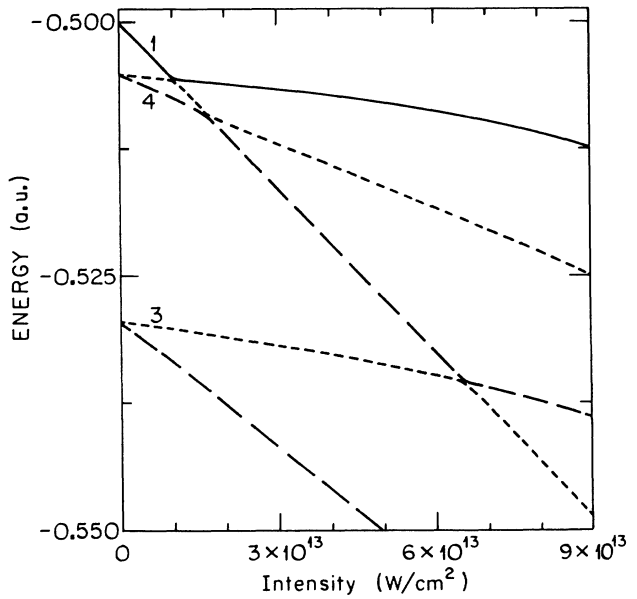

FIG. 7. Quasienergy eigenvalue curves vs intensity for H exposed to a linearly polarized field of wavelength 384.5 nm. The curves are $\varepsilon_1^{(v)}$, $\varepsilon_3^{(v)}-4\hbar\omega$, and $\varepsilon_4^{(v)}-4\hbar\omega$, where the subscript is the principal quantum number (in the zero-field limit), which is indicated on the figure. For each of the principal quantum numbers 3 and 4 there are two curves, which converge in the zero-field limit.

photon transitions from the ground state to bound states with principal quantum numbers 3 and 4. Parity considerations restrict the angular momentum of the electron in its resonant intermediate states to s and d, and because of the special degeneracy of the hydrogen atom these s and d states are mixed by the field. The mixing is due, in lowest order, to the emission *and* subsequent reabsorption of a photon, or vice versa, which allows for a change in the orbital angular momentum of zero or two units. (In contrast, Stark mixing in a static electric field occurs, in lowest order, through the emission *or* absorption of a single *zero*-frequency photon; the photon is not reabsorbed or reemitted, and a change of one unit in the orbital angular momentum is allowed.) The appropriate combinations of s and d states in the zero-field limit follow from an application of degenerate-level perturbation theory. There are two linearly independent combinations, and therefore two eigenvalue curves for each of the two principal quantum numbers 3 and 4. Note that all the crossings are avoided, in contrast to the case of a hydrogen atom in a static electric field. In the latter case, the energy eigenvalue curves may actually cross, a consequence of an underlying dynamical symmetry[43] reflected in the fact that the time-independent Schrödinger equation for an electron in a static field, which is a scalar equation, is separable in parabolic coordinates. Unlike the static field case, the time-independent Schrödinger equation for an electron in a monochromatic radiation field is a vector equation, and this vector equation is not separable in parabolic coordinates. Note that while the crossings are avoided, the gaps at the crossing points are sometimes extremely small; the gap at 1.07 W/cm^2 in Fig. 7 is only 5×10^{-6} a.u. (compared to a gap of 4×10^{-4} a.u. at the resonance discussed above at the wavelength 204 nm). In this circumstance the jump probability at the crossing is expected to be close to unity, and it is appropriate to describe the electron by a superposition of "diabatic" state vectors (see Appendix D). The probability for an electron to remain on a diabatic quasienergy curve is close to unity, and in the absence of any mixing of diabatic states there is no resonance enhancement of the ionization signal. Even if there is some mixing of the diabatic states, the resonance peak in the yield, as a function of intensity, will be narrow.

VI. CONCLUSION

Starting from the Floquet ansatz, we have formulated a time-independent theory for multiphoton ionization by moderately intense fields—intensities for which the field induced width Γ_i in the initial atomic energy level is far less than the laser frequency ω. The theory yields a *finite* amplitude for N-photon ionization provided that the *velocity* gauge is used. In the most complete form of the theory, a set of inhomogeneous equations must be solved for the harmonic components of the electron wave vector. However, in our application we have introduced a further approximation, the truncation of the harmonic expansion at the ionization threshold. This approximation (the TTA) leads to a substantial simplification—the wave vector is normalizable—but it also leads to a loss in accuracy. Thus the TTA can only be relied upon to give the overall envelope of the angular distributions, and not the detailed structure. Nevertheless, we believe the TTA to be a significant improvement over existing simpler theories. Indeed, the replacement of the TTA trial initial state vector by the unperturbed initial atomic bound state vector can lead to an error of several orders of magnitude in the generalized cross sections, even at nonresonant frequencies. Of the various trial final state vectors we examined, the Kroll-Watson form seems to give the most accurate results on the whole. Furthermore, we have shown that the TTA can be readily adapted to the treatment of resonant multiphoton ionization, when two or more Floquet eigenvectors are coupled together in a time-dependent way.

Presumably, we could improve significantly on the TTA by solving the inhomogeneous equations for the harmonic components, without truncation at the ionization threshold.[44] It might be sufficient to incorporate, in the trial initial state vector, absorption of just two or three photons above threshold, with higher absorption accounted for by the Kroll-Watson trial final state vector, which should be far more accurate at energies of $2\hbar\omega$ or $3\hbar\omega$ above threshold. This is currently under investigation.

The situation where a threshold is approached at which N_0 increases by unity is complicated because at an intensity just above this threshold there are an infinite number of resonances (if the atomic potential has a Coulomb tail). Furthermore, an eigenvalue on a particular unphysical energy sheet casts "shadow" eigenvalues on infinitely many other unphysical energy sheets, and, as a threshold is passed, the eigenvalue closest to the physical energy axis changes. These points will be discussed in detail elsewhere.

ACKNOWLEDGMENT

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APPENDIX A: ELECTRON STATE JUST AFTER FIELD TURN-ON

Here we examine the state of the electron immediately after the field has been turned on, assuming that the initial state i is not appreciably depopulated during the interval $0\le t\le t_0$ that the intensity rises. Now, the parameter β_i, which characterizes the strength of the coupling of the initial state to the radiation field, is a gauge-dependent quantity which may be defined as follows: Let $\Delta\Omega_i$ be the detuning of the laser frequency ω from a one-photon resonance, that is, $\hbar\Delta\Omega_i=|\Delta E_i-\hbar\omega|$, where ΔE_i is the energy separation between the initial level and the particular level for which, of all bound levels optically coupled by one photon to the initial level, $\Delta\Omega_i$ is smallest.[45] We define β_i as the ratio of the characteristic coupling energy to the detuning energy $\hbar\Delta\Omega_i$. In the length gauge the characteristic coupling energy is eF_0a_i, where a_i is the characteristic radius of the initial state, so that $\beta_i=\beta_i^{(l)}\equiv|eF_0a_i/\hbar\Delta\Omega_i|$. In the velocity gauge the

characteristic coupling energy is $eF_0 v_i/\omega$, where v_i is the characteristic mean speed of the electron in the initial state, so that $\beta_i = \beta_i^{(v)} \equiv |eF_0 v_i/\omega\hbar\Delta\Omega_i|$. If we work in the length gauge, and if $\beta_i^{(l)} \ll 1$, the initial state i will not be significantly perturbed until the field has been on for a long time, longer than t_0. All that happens while the field is turned on is that the electron energy is shifted adiabatically by the small amount $\Delta_i^{(l)}$. Thus immediately after the field has been fully turned on we may represent the electron by the state vector $|\Phi_i^{(l)}(t)\rangle$ given by

$$|\Phi_i^{(l)}(t)\rangle = \exp(-i\Delta_i^{(l)}t/\hbar)\,|\Phi_i^{(0)}(t)\rangle\ . \tag{A1a}$$

Now, this being the case, the state vector $|\Phi_i^{(v)}(t)\rangle$, which represents the electron in the velocity gauge immediately after the field has been turned on, is, using Eq. (2.10), just

$$|\Phi_i^{(v)}(t)\rangle = \exp\{-i[(\Delta_i^{(v)}t/\hbar) - \zeta(\tau) - \mathbf{x}\cdot\mathbf{k}_A(\tau)]\}\,|\Phi_i^{(0)}(t)\rangle\ . \tag{A1b}$$

The spatial distributions $|\langle\mathbf{x}|\Phi_i^{(l)}(t)\rangle|^2$ and $|\langle\mathbf{x}|\Phi_i^{(v)}(t)|^2$ are clearly equal to $|\langle\mathbf{x}|\Phi_i\rangle|^2$, the spatial distribution before the field was turned on. Here, of course, $|\mathbf{x}\rangle$ denotes an eigenvector of the operator $\mathbf{x}$ with eigenvalue $\mathbf{x}$. Let $|\hbar\mathbf{k}\rangle$ denote the eigenvector of the canonical momentum operator $\mathbf{p}$ with eigenvalue $\hbar\mathbf{k}$. Recalling that the velocity operator is $\mathbf{p}/\mu$ in the length gauge and $[\mathbf{p}-\hbar\mathbf{k}_A(\tau)]/\mu$ in the velocity gauge, we observe that the velocity distributions

$$|\langle\mathbf{k}|\Phi_i^{(l)}(t)\rangle|^2$$

and

$$|\langle\mathbf{k}+\mathbf{k}_A(\tau)|\Phi_i^{(v)}(t)\rangle|^2$$

are equal to $|\langle\mathbf{k}|\Phi_i\rangle|^2$, the velocity distribution before the field was turned on. We note, however, that $|\Phi_i^{(v)}(t)\rangle$ differs significantly from $|\Phi_i^{(l)}(t)\rangle$ through the spatially dependent phase $\nu(\mathbf{x},\tau) = \mathbf{x}\cdot\mathbf{k}_A(\tau)$. The characteristic value of this phase is $(ea_iF_0/\hbar\omega)$, that is, $(\Delta\Omega_i/\omega)\beta_i^{(l)}$. Hence if the detuning is large compared to the photon frequency, as will be the case for frequencies well below the smallest resonant frequency, $\nu(\mathbf{x},\tau)$ can be significant even if $\beta_i^{(l)} \ll 1$. At frequencies far from resonance, the detuning is comparable to the transition energy ΔE_i. Now roughly speaking, if ΔE_i is comparable to the energy splitting between the initial level and its nearest neighbor, the orbital period of the electron in the initial state is $\hbar/\Delta E_i$, and therefore $v_i = a_i\Delta E_i/\hbar$. Using this result and putting $\Delta\Omega_i = \Delta E_i$ we see that, far from resonance, the characteristic value of $\nu(\mathbf{x},\tau)$ is just $\beta_i^{(v)}$. To summarize, if $\beta_i^{(l)} \ll 1$ the electron state immediately after the field has been turned on is represented (i) in the length gauge by the eigenvector $|\Phi_i\rangle$ of the unperturbed atomic Hamiltonian H_a, but with the eigenvalue E_i shifted slightly by $\Delta_i^{(l)}$, and (ii) in the velocity gauge by a vector which, if $\beta_i^{(v)} \gtrsim 1$, differs significantly from $|\Phi_i\rangle$ by the multiplicative phase factor $e^{i\nu(\mathbf{x},\tau)}$.

We call the regime in which $\beta_i^{(l)} \ll 1$ and $\beta_i^{(v)} \gtrsim 1$, or vice versa, the quasi-strong-field regime, to be distinguished from the very strong-field regime in which both $\beta_i^{(l)}$ and $\beta_i^{(v)}$ are greater than or comparable to unity. We have just seen that if $\beta_i^{(l)} \ll 1$ the spatial and velocity distributions of the electron are not immediately altered by turning on the field; in other words, the electron orbital motion is not appreciably perturbed during the rise time of the field. The phase factor $\nu(\mathbf{x},\tau)$, which incorporates *virtual* absorption and emission of photons, accounts for the fact that while the velocity operator has shifted by the amount $\hbar\mathbf{k}_A(\tau)/\mu$ the electron velocity is the same after as before the field was turned on. In the case where $\beta_i^{(l)}$ is smaller than unity, but not much smaller, the atom will be significantly polarized, i.e., the spatial and velocity distributions of the electron will be significantly altered by turning on the field; but if the field has a low nonresonant frequency so that the number m of photons required to excite (or deexcite) the atom is large, the excitation rate will be low, proportional to $[\beta_i^{(l)}]^m$, and consequently the population of the (polarized) initial state will not be immediately altered by turning on the field. Suppose, however, that the laser frequency is nearly resonant with the transition frequency between the initial level and another bound level, which we denote as j. In this case both $\beta_i^{(l)}$ and $\beta_i^{(v)}$ are large, and in fact the electron will undergo excitation into the state j the moment the field is turned on. The electron is now best described in terms of two *dressed* states,[46] which are uncoupled to each other. These dressed states, denoted as $i\pm j$, are each a linear superposition of the states i and j and we must now introduce (in each gauge) two coupling strength parameters $\beta_{i\pm j}$, which measure the coupling strength of the dressed states to the radiation field. Provided that $\beta_{i\pm j}$ are both small in one gauge, the population will remain in the dressed states while the field is turned on. Note that if the laser is turned on slowly and if its bandwidth $\Delta\omega$ is small compared to the detuning $\Delta\Omega_i$, the initial electron state will evolve adiabatically, as the field is turned on, into just one of the two dressed states, determined by the sign of the detuning.[46] However, if the turn-on is not adiabatic the electron will arrive in a superposition of dressed states, which describes the population flopping back and forth between the bound states i and j, at approximately the Rabi frequency.

Consider now an atom that is initially in a high-Rydberg state, with principal quantum number n. We have $a_i = n^2a_0$ and $v_i = v_0/n$, where $a_0 = \hbar^2/\mu e^2$ and $v_0 = e^2/\hbar$. For sufficiently large n we have $\beta_i^{(v)} \ll 1$ and $\beta_i^{(l)} \gtrsim 1$. Consequently, we obtain

$$|\Phi_i^{(v)}(t)\rangle = \exp(-i\Delta_i^{(v)}t/\hbar)\,|\Phi_i^{(0)}(t)\rangle\ , \tag{A2a}$$

$$|\Phi_i^{(l)}(t)\rangle = \exp\{-i[(\Delta_i^{(l)}t/\hbar) + \zeta(\tau) + \mathbf{x}\cdot\mathbf{k}_A(\tau)]\}\,|\Phi_i^{(0)}(t)\rangle\ . \tag{A2b}$$

The velocity distribution of the electron is now significantly altered by turning on the field; the electron vibrates with a velocity $\hbar\mathbf{k}_A(\tau)/\mu$, the same as a free electron in the oscillating field. In fact, of course, the electron is almost free, being only tenuously held at a very large distance a_i from the atomic nucleus. In the limit $n \to \infty$ we have $\beta_i^{(v)} \to 0$, and hence $\Delta_i^{(v)} \to 0$ (and

$\Delta_i^{(l)}=\Delta_i^{(v)}+P\rightarrow P$). In this limit the electron becomes a free electron, displaced infinitely far from the nucleus, and it cannot absorb a real photon.

Let us finally consider an electron which has positive energy, and which is incident from infinity with mean speed v_i. This unbound electron can emit and absorb photons as it passes near to the atomic nucleus. (A bound electron in a high-Rydberg state has only a very small probability of being near to the atomic nucleus.) The coupling-strength parameter in the velocity gauge is, putting[47] $\Delta\Omega_i=\omega$, just $\beta_i^{(v)}=(eF_0v_i/\hbar\omega^2)$. Now $\beta_i^{(l)}$ is $(\omega/v_i)\beta_i^{(v)}$ multiplied by the characteristic value of $|\mathbf{x}|$. But what do we take for the characteristic value of $|\mathbf{x}|$? In the weak-field limit we take the wavelength $(1/\Delta k)$, where Δk is the change in the wave number of the electron when it absorbs a photon. (This is the characteristic distance over which absorption takes place, as may be checked using perturbation theory.) If $\hbar\omega\lesssim\mu v_i^2$ we have $\Delta k\sim\omega/v_i$, while if $\hbar\omega\gtrsim\mu v_i^2$ we have $\Delta k\sim(2\mu\omega/\hbar)^{1/2}$. [The characteristic distance can also be determined as follows: The time required for a photon to be absorbed is roughly $2\pi/\omega$, and during this time the electron covers a distance of order v_i/ω provided that $\hbar\omega\ll\mu v_i^2/2$ so that the mean speed of the electron is not significantly altered by the absorption of a photon. If $\hbar\omega\gtrsim\mu v_i^2/2$ the uncertainty $\Delta\mathbf{p}$ in the mean momentum $\mathbf{p}$ is comparable to $\mathbf{p}$ and we have $(\Delta\mathbf{p})^2/2\mu\sim\hbar\omega$ so that the effective distance over which absorption occurs is $\hbar/|\Delta\mathbf{p}|\sim(\hbar/2\mu\omega)^{1/2}$.] It follows that in the weak-field limit

$$\beta_i^{(l)}=\begin{cases}\beta_i^{(v)} & \text{if } \hbar\omega\lesssim\mu v_i^2\\ (\hbar\omega/\mu v^2)\beta_i^{(v)} & \text{if } \hbar\omega\gtrsim\mu v_i^2\ .\end{cases} \tag{A3}$$

However, if the field is not weak, we must take into account the oscillation of the electron in the field. A free electron oscillates with an amplitude $(eF_0/\mu\omega^2)$, and this is larger than $1/\Delta k$ if $\delta>1$, where

$$\delta=\begin{cases}(\hbar\omega/\mu v_i^2)\beta_i^{(v)} & \text{if } \hbar\omega\lesssim\mu v_i^2\\ (\hbar\omega/\mu v_i^2)^{1/2}\beta_i^{(v)} & \text{if } \hbar\omega\gtrsim\mu v_i^2\ .\end{cases} \tag{A4}$$

If $\delta>1$ we must take the amplitude $(eF_0/\mu\omega^2)$ to be the characteristic value of $|\mathbf{x}|$, and in this case $\beta_i^{(l)}=P/\hbar\omega$.[48] Note that if $\delta>1$ the ratio $\beta_i^{(l)}/\beta_i^{(v)}$ is equal to δ if $\hbar\omega\lesssim\mu v_i^2$ or to $(\hbar\omega/\mu v_i^2)^{1/2}\delta$ if $\hbar\omega\gtrsim\mu v_i^2$; in either case we have $\beta_i^{(l)}>\beta_i^{(v)}$. It is apparent from Eq. (A4) that in the high-frequency limit $\hbar\omega\gg\mu v_i^2$ we can have $\delta>1$, and therefore $\beta_i^{(l)}>1$, and yet $\beta_i^{(v)}\ll1$; this is the quasi-strong-field regime. Note that, whether the field is weak or strong, the shift $\Delta_i^{(v)}$ in the energy of an unbound electron is zero since the electron spends most of its time as a free electron, far from the atomic potential.[14] Hence we have $\Delta_i^{(l)}=P$, a result noted already. To summarize, if $\beta_i^{(v)}\ll1$ the electron state immediately after the field has been turned on is represented (i) in the velocity gauge by the eigenvector $|\Phi_i\rangle$ of the unperturbed atomic Hamiltonian H_a, but with the eigenvalue E_i shifted by an amount $\Delta_i^{(v)}$ which is small and vanishes if E_i is positive, and (ii) in the length gauge by a vector which, if $\beta_i^{(l)}\gtrsim1$, differs significantly from $|\Phi_i\rangle$ by the multiplicative phase factor $e^{-i\nu(\mathbf{x},\tau)}$.

In general, we have $\beta_i^{(v)}>\beta_i^{(l)}$ if the electron is tightly bound, and $\beta_i^{(l)}>\beta_i^{(v)}$ if the electron is loosely bound, or unbound. Evidently, the gauge in which the electron can be represented by an eigenvector of H_a, immediately after the field is turned on, is dictated by the initial state—the length gauge if the electron is tightly bound, the velocity gauge if the electron is loosely bound, or unbound. Similar remarks apply to the final state, in discussing the states of the electron just before and after the field is turned off. We can introduce coupling strength parameters $\beta_f^{(l)}$ and $\beta_f^{(v)}$ for the final state, in analogy to those for the initial state. We are interested here of course in ionization, where the electron is finally unbound. We know that if $\beta_f^{(v)}\ll1$ we can represent the final unbound state of the electron in the velocity gauge by an eigenvector of H_a.

APPENDIX B: ASYMPTOTIC MOTION

Ultimately the electron is free and infinitely far from the atomic potential W, and its mean momentum is then constant in time. The state vector of a free electron moving with momentum $\hbar\mathbf{k}_f$ in the *absence* of a field is just $|\mathbf{k}_f;t\rangle=\exp(-iE_ft/\hbar)|\mathbf{k}_f\rangle$, where $E_f=\hbar^2\mathbf{k}_f^2/2\mu$ and $\mathbf{p}|\mathbf{k}_f\rangle=\hbar\mathbf{k}_f|\mathbf{k}_f\rangle$. Since $\mathbf{p}$ commutes with $V^{(v)}(\tau)$, the state vector of a free electron moving through a field with mean momentum $\hbar\mathbf{k}_f$ differs, in the velocity gauge, from $|\mathbf{k}_f;t\rangle$ only by a time-dependent factor, $\exp[-i\theta(\tau)]$, say. This is true even in the strong-field limit $\beta_f^{(v)}\gtrsim1$. In fact, we can always transform to the inertial frame where the mean momentum of the free electron vanishes so that $\beta_f^{(v)}$ vanishes. The phase $\theta(\tau)$ is

$$\theta(\tau)=\frac{1}{\hbar}\int_0^t dt'\Delta_f^{(v)}(\tau)\ , \tag{B1}$$

where $\Delta_f^{(v)}(\tau)$ is the instantaneous energy shift of a free electron, which vanishes when averaged over one cycle, and is given by

$$\begin{aligned}\Delta_f^{(v)}(\tau)&=(\hbar^2/2\mu)[\mathbf{k}_f-\mathbf{k}_A(\tau)]^2-V_0(\tau)-E_f\\&=-(\hbar^2/\mu)\mathbf{k}_f\cdot\mathbf{k}_A(\tau)\ .\end{aligned} \tag{B2}$$

It is easy to verify that, if $H_0=\mathbf{p}^2/2\mu$,

$$\left[H_0+V^{(v)}(\tau)-i\hbar\frac{d}{dt}\right]e^{-i\theta(\tau)}|\mathbf{k}_f;t\rangle=0\ , \tag{B3}$$

a well-known result. (Often $\exp[-i\theta(\tau)]|\mathbf{k}_f;t\rangle$ is referred to as the "Volkov" vector.) In the length gauge, since $\mathbf{p}$ does not commute with $V^{(l)}(\tau)$, the state vector of a free electron moving with mean momentum $\hbar\mathbf{k}_f$ is not an eigenvector of $\mathbf{p}$ with constant eigenvalue $\hbar\mathbf{k}_f$; it *is* an eigenvector of $\mathbf{p}$, but with an eigenvalue equal to the instantaneous mechanical momentum $\hbar\mathbf{k}_f(\tau)=\hbar[\mathbf{k}_f-\mathbf{k}_A(\tau)]$, which is not constant.

We now introduce an eigenvector of the atomic Hamiltonian H_a, namely,

$$|\Phi_f^{(v)}(t)\rangle=e^{-i\theta(\tau)}|\Phi_f^{(0)}(t)\rangle=e^{-i\theta(\tau)-E_ft/\hbar}|\Phi_{\mathbf{k}_f}^-\rangle\ , \tag{B4}$$

where $|\Phi_{\mathbf{k}_f}^-\rangle$ is an eigenvector of H_a, with eigenvalue

E_f, which satisfies out-asymptote boundary conditions and approaches $|\mathbf{k}_f\rangle$ at large distances. At asymptotically large times the electron is at an asymptotically large distance from the atomic nucleus and

$$|\Phi_f^{(v)}(t)\rangle \rightarrow e^{-i\theta(\tau)}|\mathbf{k}_f;t\rangle \ . \tag{B5}$$

Note that the asymptotic limit, Eq. (B5), is accomplished by adiabatically switching off the atomic potential W at large times. This is done by multiplying W by the switching factor $\exp(-\eta t/\hbar)$, where η is positive but infinitesimal. Effectively, this introduces an energy spread η and corresponds to constructing a normalized wave packet which represents a localized electron moving far away from the atomic potential at large times. In the limit that η vanishes this wave packet becomes the non-normalizable $|\Phi_f^{(v)}(t)\rangle$.

In the velocity gauge, $|\Phi_f^{(v)}(t)\rangle$ correctly describes at asymptotically large distances an electron moving through the field, even if $\beta_i^{(v)} \gtrsim 1$. The gauge transform of $|\Phi_f^{(v)}(t)\rangle$, namely, $|\Phi_f^{(l)}(t)\rangle$, is given by Eq. (2.10), and it is *not* an eigenvector of H_a when $\beta_i^{(v)} \gtrsim 1$ because the phase $\nu(\mathbf{x},\tau)$ cannot be neglected. This has important consequences[13,29] for the formulation of a time-independent theory in the length gauge, since the field cannot be turned off in a time-independent formalism. However, we can construct an eigenvector of H_a which does have the correct asymptotic form in the length gauge. It is

$$|KW_f^{(l)}(t)\rangle = e^{-i\xi(t)}|\Phi^-_{\mathbf{k}_f(\tau)}\rangle \ , \tag{B6a}$$

where

$$\xi(t) = \frac{\hbar}{2\mu}\int_0^t dt' \mathbf{k}_f^2(\tau) \ ; \tag{B6b}$$

recall that $\mathbf{k}_f(\tau) = \mathbf{k}_f - \mathbf{k}_A(\tau)$. Thus $|KW_f^{(l)}(t)\rangle$ is an eigenvector of H_a with eigenvalue $\hbar^2\mathbf{k}_f^2(\tau)/2\mu$, and it asymptotically approaches the asymptotic form of $|\Phi_f^{(l)}(t)\rangle$. Observe that $|KW_f^{(l)}(t)\rangle$ is the scattering wave vector used by Kroll and Watson[12,25] in their low-frequency approximation for electron scattering from a potential in the presence of a radiation field. The frequency is considered low if the time $2\pi/\omega$ required for the electron to absorb a photon is large compared to the quantum collision time $\hbar/E_f$. In this limit the electron absorbs a photon while scattering from the tail of the potential, when it is almost free. Consequently, at each time t the electron state is represented by the wave vector for the electron to scatter from the potential with asymptotic momentum equal to the instantaneous mechanical momentum $\hbar\mathbf{k}_f(\tau)$. The gauge transform of $|KW_f^{(l)}(t)\rangle$, namely, $|KW_f^{(v)}(t)\rangle$, is given by Eq. (2.10),

$$|KW_f^{(v)}(t)\rangle = e^{-iE_f t/\hbar}e^{-i\theta(\tau)+i\mathbf{x}\cdot\mathbf{k}_A(\tau)}|\Phi^-_{\mathbf{k}_f(\tau)}\rangle \ ; \tag{B7}$$

it is not an eigenvector of H_a, the reason originating in the fact that in the velocity gauge the canonical momentum is not the instantaneous mechanical momentum.

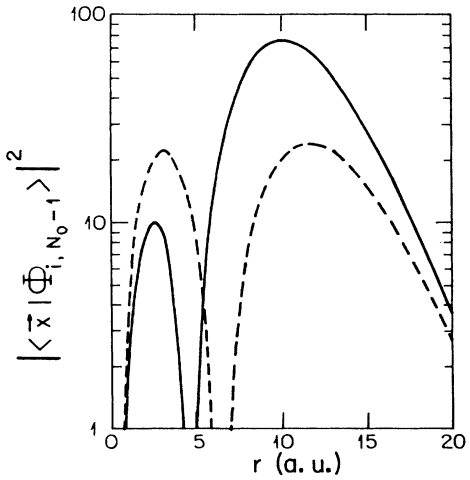

FIG. 8. Probability density $|\langle \mathbf{x}|\Phi_{i,N_0-1}\rangle|^2$ (in arbitrary units) vs the radial coordinate $r = |\mathbf{x}|$, with $\mathbf{x}$ pointing along the polarization vector, for a weak, linearly polarized field of wavelength 532 nm ($N_0 = 6$). We have ——, velocity gauge; — — —, length gauge.

APPENDIX C: GAUGE INVARIANCE IN THE WEAK-FIELD LIMIT

The lowest- (nonvanishing) order perturbation theory expression for M_N can be derived in each gauge directly from the original unmodified Floquet formalism by taking the weak-field limit, whereby gauge invariance is preserved. We therefore expect that

$$\langle \Phi^-_{\mathbf{k}_f}|V_+^{(v)}|\Phi^{(v)}_{i,N-1}\rangle = \langle \Phi^-_{\mathbf{k}_f}|V_+^{(l)}|\Phi^{(l)}_{i,N-1}\rangle \ , \tag{C1}$$

where $E_f = E_i + N\hbar\omega$ and where $|\Phi_{in}^{(g)}\rangle$ is defined by Eq. (5.1). However, the explicit proof of (C1) requires some manipulation. Indeed, it is perhaps remarkable that (C1) holds since $\langle \mathbf{x}|\Phi_{in}^{(v)}\rangle$ is quite different from $\langle \mathbf{x}|\Phi_{in}^{(l)}\rangle$. For example, in Fig. 8 we show the variation with radial coordinate of $\langle \mathbf{x}|\Phi^{(g)}_{i,N_0-1}\rangle$ in each gauge for linearly polarized light of wavelength 532 nm ($N_0 = 6$). A general proof of Eq. (C1), to all orders of perturbation theory, has been given by Haller and Landovitz and by Aharonov and Au.[49]

APPENDIX D: DYNAMICS NEAR A CURVE CROSSING

We derive here[39] the coupled equations (5.9) for the coefficients of the dressed-state eigenvectors, assuming that there is an intermediate m-photon resonance (where $m = N_{0i} - N_{0j}$). We substitute $|R(t)\rangle$ from Eq. (5.7) into the Schrödinger equation

$$i\hbar\frac{d}{dt}\Big|\Psi(t)\Big\rangle = H(t)|\Psi(t)\rangle \ , \tag{D1}$$

where the total time derivative is

$$\frac{d}{dt}=\frac{\partial}{\partial t}\bigg|_I+\frac{dI(t)}{dt}\frac{\partial}{\partial I}\bigg|_t . \tag{D2}$$

Noting that

$$i\hbar\frac{d}{dt}e^{-iS_l(t)}=\varepsilon_l e^{-iS_l(t)} , \tag{D3}$$

and using Eq. (4.36), observing that the time derivative in Eq. (4.36) is the partial derivative (with I held fixed), we obtain

$$\sum_{l=i,j} e^{-iS_l(t)}\left[\left[\frac{da_l(t)}{dt}+a_l(t)\frac{dI(t)}{dt}\frac{\partial}{\partial I}\right]|\psi_l(\tau)\rangle-e^{-iN_{0l}\tau}V_+^{(v)}\,|\,\psi_{l,N_{0l}-1}\rangle\right]=0 . \tag{D4}$$

Now at the crossing point the (total) time derivative of the phase $m\tau+S_{ij}(t)$ is the frequency gap, which is very small. Therefore in the region of the crossing point $S_{ij}(t)$ is nearly equal to $-m\tau$ up to a constant. We premultiply the left-hand side of Eq. (D4) by $e^{iS_i(t)}\langle\psi_i(\tau)|$, and we integrate over τ treating $m\tau+S_{ij}(t)$, $a_l(t)$, and the time derivatives of $a_l(t)$ and $I(t)$, as constant over the cycle time $2\pi/\omega$. Using the orthonormality relation, Eq. (4.37), we obtain Eq. (5.9a) after observing that the terms in

$$\exp[iS_{il}(t)-iN_{0l}\tau]\langle\psi_i(\tau)|$$

vanish upon integration over τ and that, with

$$U_{ij}(I)=\frac{1}{2\pi}\int_0^{2\pi}d\tau\,e^{i(N_{0j}-N_{0i})\tau}\times\left\langle\psi_i(\tau)\left|\frac{\partial}{\partial I}\right|\psi_j(\tau)\right\rangle , \tag{D5}$$

we have

$$U_{ij}(I)+U_{ji}(I)=\frac{1}{2\pi}\frac{\partial}{\partial I}\int_0^{2\pi}d\tau\,e^{i(N_{0j}-N_{0i})\tau}\times\langle\psi_i(\tau)\,|\,\psi_j(\tau)\rangle=0 , \tag{D6}$$

where the last step follows from Eq. (4.37). We see from (D6) that $U_{ii}(I)=0$. We can obtain Eq. (5.9b) similarly by first premultiplying the left-hand side of Eq. (D4) by $e^{iS_j(t)}\langle\psi_j(\tau)|$.

If the intensity varies very rapidly, the coefficients $a_l(t)$ will change appreciably in absolute magnitude only during a very short time interval when the intensity passes through the crossing point. In this case we can treat the phase $m\tau+S_{ij}(t)$ as a constant, equal to α, say, and Eqs. (5.9) reduce to the equations

$$\frac{d}{dI}a_i(t)=U_{ij}(I)e^{i\alpha}a_j(t) , \tag{D7a}$$

$$\frac{d}{dI}e^{i\alpha}a_j(t)=-U_{ij}(I)a_i(t) , \tag{D7b}$$

which have the solution

$$a_i(t)=\cos\left[\int^I dI'U_{ij}(I')\right] , \tag{D8a}$$

$$e^{i\alpha}a_j(t)=-\sin\left[\int^I dI'U_{ij}(I')\right] . \tag{D8b}$$

If the light propagates as a short powerful pulse, the intensity necessarily varies rapidly in time, and then (D8) provides a rather accurate solution to Eqs. (5.9). The main contributions to the integrals over I' in Eqs. (D8) come from the region of the crossing point, where $U_{ij}(I')$ is large.

We can expand the "adiabatic" state vectors $|\psi_l(\tau)\rangle$, $l=i$ and j, in terms of "diabatic" state vectors $|d_l(\tau)\rangle$, where $|d_l(\tau)\rangle$ is defined as the solution to Eq. (4.36) when the atomic Hamiltonian H_a is replaced by $Q_lH_aQ_l$, where $Q_l=1-|\Phi_{l'}\rangle\langle\Phi_{l'}|$, with $l'\neq l$, $l'=i$ or j. The projection operator Q_l removes the intermediate resonance from $|\psi_l(\tau)\rangle$ and the diabatic quasienergy eigenvalue curves actually cross at the resonance intensity I_r. We have

$$|\psi_l(\tau)\rangle\approx\begin{cases}|d_l(\tau)\rangle, & I<I_r\\ |d_{l'}(\tau)\rangle, & I>I_r\end{cases} \tag{D9}$$

and, noting that the diabatic state vectors approximately satisfy an orthonormality relation similar to Eq. (4.37), we can write

$$|\psi_i(\tau)\rangle\approx\cos\chi(I)\,|\,d_i(\tau)\rangle+\sin\chi(I)\,|\,d_j(\tau)\rangle , \tag{D10a}$$

$$|\psi_j(\tau)\rangle\approx-\sin\chi(I)\,|\,d_i(\tau)\rangle+\cos\chi(I)\,|\,d_j(\tau)\rangle , \tag{D10b}$$

where the mixing angle $\chi(I)$ is zero for $I\ll I_r$ and $\pi/2$ for $I\gg I_r$. Although the diabatic state vectors vary with intensity, their variation is relatively slow, and to a first approximation can be neglected; we thereby obtain

$$U_{ij}(I)\approx-\frac{d\chi(I)}{dI} , \tag{D11}$$

and the short-pulse approximation (D8) yields $|a_i(\infty)|=0$ and $|a_j(\infty)|=1$, that is, a jump probability of unity.

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[12]N. M. Kroll and K. M. Watson, Phys. Rev. A **8**, 804 (1973). See also the review by L. Rosenberg, Adv. At. Mol. Phys. **18**, 1 (1982). The Kroll-Watson wave function was used in a theory of two-color multiphoton ionization by M. Dörr and R. Shakeshaft, Phys. Rev. A **36**, 421 (1987).

[13]R. Shakeshaft, Z. Phys. D **8**, 47 (1988).

[14]One may always transform to an inertial frame in which a free electron is (on the average) at rest. In that frame we have, in the velocity gauge, $\mathbf{p}=0$ and therefore $V^{(v)}(\tau)=0$.

[15]H. G. Muller, A. Tip, and M. J. van der Wiel, J. Phys. B **16**, L679 (1983).

[16]See, e.g., C. Cohen-Tannoudji, B. Diu, and F. Laloe, *Quantum Mechanics* (Hermann-Wiley, Paris, 1977), Vol. 1, p. 315.

[17]Note that associated with each eigenvector $|\psi_i(\tau)\rangle$ are a denumerably infinite number of physically indistinguishable eigenvectors which differ from $|\psi_i(\tau)\rangle$ in that the index n of the harmonic components $|\psi_{in}\rangle$ is shifted by an integer l; the quasienergy eigenvalue ε_i transforms to $\varepsilon_i+l\hbar\omega$ under the change $n\rightarrow n-l$, and in the weak-field limit it is the harmonic component $|\psi_{il}\rangle$ which approaches $|\phi_i\rangle$. See S.-I. Chu, Ref. 10.

[18]R. Shakeshaft, J. Opt. Soc. Am. B **4**, 705 (1987).

[19]Though phrased somewhat differently, this idea is contained in Chap. 8 of M. L. Goldberger and K. M. Watson, *Collision Theory* (Krieger, New York, 1975).

[20]S.-I. Chu and J. Cooper, Phys. Rev. A **32**, 2769 (1985).

[21]R. Shakeshaft and X. Tang. Phys. Rev. A **36**, 3193 (1987).

[22]For reviews of complex coordinate techniques, see B. R. Junker, Adv. At. Mol. Phys. **18**, 207 (1982); W. P. Reinhardt, Ann. Rev. Phys. Chem. **33**, 223 (1982).

[23]X. Tang and R. Shakeshaft, Z. Phys. D **5**, 27 (1987).

[24]I. J. Berson, J. Phys. B **8**, 3078 (1975); R. Shakeshaft, Phys. Rev. A **28**, 667 (1983); A. Giusti-Suzor and P. Zoller, *ibid.* **36**, 5178 (1987); L. Dimou and F. H. M. Faisal, Phys. Rev. Lett. **59**, 872 (1987).

[25]For an alternative derivation of the form of $|KW_f^{(v)}(t)\rangle$ see R. Shakeshaft, Phys. Rev. A **28**, 667 (1983).

[26]L. Rosenberg, Phys. Rev. A **34**, 978 (1986).

[27]See, e.g., L. Spruch, in *Lectures in Theoretical Physics,* edited by S. Geltman, K. T. Mahanthappa, and W. E. Britten (Gordon and Breach, New York, 1969), Vol. XIC, p. 77.

[28]The use of the trial wave vector $|\phi_f^{(v)}(t)\rangle$ was first suggested in the context of multiphoton ionization by M. Jain and N. Tzoar, Phys. Rev. A **18**, 538 (1978) and more recently by S. Basile, F. Trombetta, G. Ferrante, R. Burlon, and C. Leone, *ibid.* **37**, 1050 (1988). It has been used in the context of scattering in a laser background by P. Cavaliere, G. Ferrante, and C. Leone, J. Phys. B **13**, 4495 (1980); L. Rosenberg, Phys. Rev. A **34**, 4567 (1986).

[29]M. Edwards and R. Shakeshaft, Z. Phys. D **8**, 51 (1988).

[30]M. Aymar and M. Crance, J. Phys. B **14**, 3585 (1981).

[31]S. Klarsfeld and A. Maquet, Phys. Lett. **73A**, 100 (1979); J. Phys. B **12**, L553 (1979); B. R. Johnson and W. P. Reinhardt, Phys. Rev. A **29**, 2933 (1984); T. N. Rescigno and C. W. McCurdy, *ibid.* **31**, 624 (1985); R. Shakeshaft and X. Tang, *ibid.* **35**, 3945 (1987).

[32]We note that in the length gauge the truncated set of coupled equations corresponding to Eq. (4.35) does not admit regular decaying solutions, except in the weak-field limit, because $V_{\pm}^{(l)}$ diverges for $r-\infty$ and so tunneling is always possible, and the *effective* width in the energy is nonzero.

[33]The orthogonality of Floquet eigenvectors was discussed by H. Sambe, Phys. Rev. A **7**, 2203 (1973), though without regard to the fact that an eigenvector is normalizable only when its harmonic expansion is truncated. The appearance of the weight factor $\exp[i(N_{0i}-N_{0j})\tau]$ in our orthogonality state-

ment is a reminder of this fact.

[34]Regarded as an operator in the space of functions that are periodic in τ with period 2π the time derivative is Hermitian. See Sambe, Ref. 33.

[35]Y. Gontier, N. K. Rahman, and M. Trahin, Phys. Rev. A **34**, 1112 (1986).

[36]See also, for example, E. Karule, J. Phys. B **11**, 441 (1978); S. Klarsfeld and A. Maquet, Phys. Lett. **78A**, 40 (1980); M. Aymar and M. Crance, J. Phys. B **13**, L287 (1980); Y. Gontier, M. Poirier, and M. Trahin, *ibid.* **13**, 1381 (1980); R. Shakeshaft, Phys. Rev. A **34**, 5119 (1986); M. Edwards, X. Tang, and R. Shakeshaft, *ibid.* **35**, 3758 (1987).

[37]The matrix equation is similar to that described by A. Maquet, S.-I. Chu, and W. P. Reinhardt, Phys. Rev. A **27**, 2946 (1983).

[38]G. Kracke, H. Marxer, J. T. Broad, and J. S. Briggs, Z. Phys. D **8**, 103 (1988).

[39]This analysis is similar to the usual discussion of a Landau-Zener crossing of molecular energy curves in a slow atomic collision. See N. F. Mott and H. S. W. Massey, *The Theory of Atomic Collisions,* 3rd ed. (Oxford, New York, 1965), p. 353; B. H. Bransden, *Atomic Collision Theory,* 2nd ed. (Benjamin-Cummings, Reading, 1983), pp. 392–400.

[40]Of course, these rates might eventually increase again as the intensity increases to a sufficiently high value, though even then the yields might not increase if substantial depletion occurs.

[41]Recall that, by assumption, the ionization widths are smaller than the laser bandwidth, and are therefore smaller than the energy gap if the laser bandwidth is.

[42]For both wavelengths we have $N_{0l}=3$. Only spherical harmonics with orbital angular momentum quantum number ≤ 1, and harmonic components $|\psi_{ln}^{(v)}\rangle$ with $-1\leq n\leq 2$ and $|\psi_{jn}^{(v)}\rangle$ with $-3\leq n\leq 0$, were included.

[43]K. Helfrich, Theor. Chim. Acta **24**, 271 (1972).

[44]As an alternative to solving the inhomogeneous equations for the harmonic components, we have recently demonstrated that a finite ionization amplitude can be constructed from the solution to the homogeneous equations (3.7) by analytic continuation through a computationally practical technique. R. M. Potvliege and R. Shakeshaft, Phys. Rev. A **38**, 1098 (1988).

[45]If there is no bound level which is optically coupled (by one photon) to the initial level we take ΔE_i to be the distance of the initial energy level from the continuum threshold. If there is a near m-photon resonance, with $m>1$, we should raise the coupling energy to the power m in the definition of β_i, and replace $\hbar\Delta\Omega_i$ by the product of m factors $\Delta E_i-n\hbar\omega$ with $n=1,2,\ldots,m$.

[46]See, e.g., M. H. Mittleman, *Theory of Laser-Atom Interactions* (Plenum, New York, 1982).

[47]One could discretize the continuum by putting the system in a box whose linear dimensions are large but finite, equal to L, say. However, the coupling between any two of these discrete levels vanishes as $L\rightarrow\infty$ because of the normalization factor $L^{-3/2}$. Consequently, transitions occur only between *groups* of levels, separated in energy by $\hbar\omega$ (or an integral multiple thereof). Degeneracy effects are smoothed out, so that the coupling in the continuum varies smoothly with ω, and the detuning is simply ω.

[48]We ignore factors of 2, π, etc., in defining the parameters. Incidentally, recall that the speed v_i which enters $\beta_i^{(v)}$ is the characteristic *mean* speed; were we to take instead the amplitude of the instantaneous speed, that is, $eF_0/\mu\omega$, we would obtain $\beta_i^{(v)}=P/\hbar\omega=\beta_i^{(l)}$.

[49]K. Haller and L. F. Landovitz, Phys. Rev. D **2**, 1498 (1970); Y. Aharonov and C. K. Au, Phys. Rev. A **20**, 1553 (1979).