Multiphoton detachment of H^- and the applicability of the Keldysh approximation

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We report results of calculations of photodetachment rates and photoelectron energy spectra for the negative hydrogen ion irradiated by a low-frequency field. We have carried out calculations in both one-electron and two-electron models (with electron-electron correlation included in the initial channel in the two-electron model). Our main focus is on exploring the differences between various approximations that arise from different treatments of the electron-field interaction, rather than from different treatments of the electron-electron interaction. Thus, for the most part, we describe the active electron by the one-electron model. Within this framework, the Keldysh theory yields results in remarkably good agreement with those obtained using Floquet theory. Furthermore Keldysh theory reproduces well the minima and "discontinuous" slopes of the rate at thresholds. The accuracy of the Keldysh approximation can be understood by examining the parameters that characterize the strength of the electron-field interaction. The Faisal-Reiss theory also gives good agreement with Floquet theory (unless the intensity is very high); the reason for this is more technical, having to do with the short range of the one-electron atomic potential. We have generalized the Keldysh and Faisal-Reiss theories to include electron-electron correlation by representing the ground-state ion with an accurate two-electron wave function. Correlation leads to a long-range interaction of the active electron with the atomic core, and thereby to more significant discrepancies between the Keldysh and Faisal-Reiss theories. We compare results obtained using the two-electron Keldysh theory with results, obtained by others, using two-electron perturbation theory and two-electron Floquet theory.

I. INTRODUCTION

In this paper we present results of various calculations of rates for multiphoton detachment of the negative hydrogen ion H^- by a low-frequency field. Our primary goal is to illustrate the differences between several approximations that arise through different treatments of the electron-field interaction, rather than through different treatments of the electron-electron interaction. Therefore, throughout most of this paper, we describe the active electron within a one-electron model. However, in Sec. IV, we also present results of a calculation based on using a many-parameter Hylleraas wave function to represent the unperturbed ground state of the ion; in this latter calculation, we take into account electron-electron correlation in the initial channel.

In our one-electron model of H^-, the atomic potential is taken to be the Yukawa potential

$$W(r)=-\beta\frac{e^{-r/a_0}}{r}\ , \tag{1}$$

where $\beta=1.1$ a.u. and where $a_0=1.0$ a.u. This model was used previously[1] to discuss multiphoton detachment of H^-. The potential $W(r)$ supports only one bound state, with a binding energy of $-0.027\,565\,4$ a.u., that is, about -0.750 eV, close to the binding energy -0.7551 eV for the real H^-. Furthermore, the solution of the Schrödinger equation for an electron moving in $W(r)$ yields a radial probability distribution that is close to the distribution obtained by integrating an accurate two-electron probability density for H^- over the coordinates of one of the electrons.[1] (Moreover, as we see in Sec. IV, the inclusion of electron-electron correlation does not lead to photodetachment rates that are very different from those obtained using the one-electron model.) Within the framework of the one-electron model, we compare estimates of multiphoton detachment rates obtained from perturbation theory, from a modified form of perturbation theory, from Keldysh theory,[2] from Faisal-Reiss theory,[3] and from Floquet theory.[4]

We take the results of Floquet theory as our benchmark. Roughly speaking, the Floquet ansatz is reliable when ionization (or photodetachment) takes place over a time interval long compared to cycle of the field, a condition that can be written as $\hbar/\Gamma_i \ll 2\pi/\omega$, where Γ_i is the

total ionization width of the initial bound state and ω is the frequency of the field. Over the intensity and frequency ranges considered here (intensities up to about 10^{12} W/cm^2, frequencies down to about 0.1 eV) this inequality is amply satisfied.

We find the agreement between the Keldysh and Floquet theories to be rather good, even excellent at very low frequencies. That the Keldysh approximation is so accurate may seem surprising, but can be understood by considering the parameters[5] that measure the strength of the electron-field interaction in the initial and final channels. In the length gauge the electron-field interaction is $-e\mathbf{F}(t)\cdot\mathbf{x}$ where $\mathbf{F}(t)\equiv\mathrm{Re}(\mathbf{F}_0e^{-i\omega t})$ is the (external) electric field, whose amplitude is $F_0\equiv(\mathbf{F}_0^*\cdot\mathbf{F}_0)^{1/2}$. The intensity of the field is $I\equiv cF_0^2/8\pi$, and the ponderomotive energy of an electron (mass μ, charge e) in this field is $P\equiv e^2F_0^2/4\mu\omega^2$. The dimensionless parameter which, in the length gauge, characterizes the strength of the interaction of the electron with the field in the initial state is roughly $\lambda_i^{(l)}\equiv|eF_0a_i/\delta E|$, where a_i is the characteristic orbital radius of the electron in the initial unperturbed state, and where δE is the smallest energy detuning from a one-photon resonance between the initial unperturbed level and other discrete levels to which a one-photon transition is not forbidden. For H^- there are no other discrete levels and so we take δE to be either the distance of the unperturbed ground-state level from the continuum threshold or $\hbar\omega$, whichever is the larger. The unperturbed ground-state energy $E_i^{(0)}$ is about -0.028 a.u., and the expectation value of r with respect to the unperturbed ground state is, to the nearest whole number, 3 a.u. Putting $a_i\approx3$ a.u. we have $\lambda_i^{(l)}<0.06$ for an intensity $I<10^{10}$ W/cm^2. Since $\lambda_i^{(l)}$ is small, it should be a reasonable approximation, in the length gauge, to neglect the field in the initial channel; in other words, we can replace the exact electron state vector $|\Psi(t)\rangle$ by the state vector

$$|\Phi_i(t)\rangle\equiv\exp(-iE_i^{(0)}t/\hbar)|\Phi_i\rangle$$

representing the unperturbed initial bound state.[6] On the other hand, the parameter $\lambda_f^{(l)}$ that measures the strength of the electron-field coupling in the final (continuum) state is not necessarily small. This parameter differs from $\lambda_i^{(l)}$ through the replacement of a_i by the excursion amplitude of a free electron, which is roughly $eF_0/\mu\omega^2$; in addition, we always have $\delta E=\hbar\omega$ (the separation of connecting levels). Hence $\lambda_f^{(l)}=e^2F_0^2/\mu\hbar\omega^3$, which can be rewritten as $4P/\hbar\omega$ where P is the ponderomotive energy. At the CO_2 laser frequency $\omega\approx4.3\times10^{-3}$ a.u., and at an intensity of 10^{10} W/cm^2, we have $\lambda_f^{(l)}\approx4$, which is not small, so the motion of the electron may be strongly influenced by the field in the final channel. However, the atomic potential should have only a weak influence on the motion of the electron in the final channel (it has a strong influence in the initial channel since it binds, albeit weakly, the electron). As discussed in further detail later, the neglect of the field in the initial channel, and the neglect of the atomic potential in the final channel, forms the basis of the Keldysh theory,[2] which we therefore expect to yield reasonably accurate results. Note, however, that although $\lambda_i^{(l)}$ vanishes as F_0 vanishes, this does not imply that the Keldysh theory becomes exact as F_0 decreases; as F_0 vanishes, so does the photodetachment rate, and although the Keldysh estimate of the rate also vanishes, the *relative* error does not. The remarkable accuracy of the Keldysh approximation for zero-range potentials (also supporting only one bound state) has been noted previously by others.[7,8]

In the velocity gauge, the electron-field interaction is $-(e/\mu c)\mathbf{A}(t)\cdot\mathbf{p}$, with $\mathbf{A}(t)\equiv\mathrm{Re}(\mathbf{A}_0e^{-i\omega t})$ the vector potential and $\mathbf{p}$ the canonical momentum. Note that $\mathbf{F}(t)=(-1/c)\partial\mathbf{A}(t)/\partial t$, and hence $\mathbf{F}_0=i(\omega/c)\mathbf{A}_0$. The parameter which characterizes the strength of the coupling in the initial state is $\lambda_i^{(v)}\equiv|ev_iF_0/\omega\delta E|$, where v_i is the characteristic orbital speed of the electron in the unperturbed ground state. Although $\lambda_i^{(l)}$ is small, $\lambda_i^{(v)}$ need not be small. For example, at the CO_2 laser frequency we have, taking $v_i\approx a_i|E_i^{(0)}|/\hbar\approx0.08$ a.u., the value $\lambda_i^{(v)}\approx0.4$ at $I=10^{10}$ W/cm^2. In any case, as long as $\hbar\omega\ll|E_i^{(0)}|$ we have $\lambda_i^{(l)}\ll\lambda_i^{(v)}$. Therefore, the approximation of $|\Psi(t)\rangle$ by $|\Phi_i(t)\rangle$ is less accurate in the velocity gauge; but this approximation forms the basis of the Faisal-Reiss theory[3]—the only difference from the Keldysh theory is the gauge in which the replacement of $|\Psi(t)\rangle$ by $|\Phi_i(t)\rangle$ is made. Accordingly, one may expect that, except in the weak-field limit $\lambda_i^{(v)}\ll1$, the Faisal-Reiss theory yields poorer results than does the Keldysh theory. In fact, however, the difference in the results obtained from the two theories, in the context of multiphoton detachment from a negative ion, is not very significant until the intensity is very high. The underlying reason that the discrepancy is so small is simple but technical, having to do with the short range of the atomic potential; this is explained more fully in Sec. II C. As in the Keldysh approximation, in the Faisal-Reiss approximation the atomic potential is neglected in the final channel, and the motion of the free electron in this channel is treated exactly. However, the parameter $\lambda_f^{(v)}$ which measures the coupling strength in the final channel, in the velocity gauge, is not the same as $\lambda_f^{(l)}$; we have $\lambda_f^{(v)}=|eF_0v_f/\hbar\omega^2|$, where v_f is the drift speed of the electron in the final state.

In lowest-order (Rayleigh-Schrödinger) perturbation theory the field is neglected in both the initial and final channels, but the atomic potential is fully included in both channels. Note that when $\lambda_i^{(l)}\ll1$ and $\lambda_i^{(l)}\ll\lambda_i^{(v)}$, the field-induced shift Δ_i of the initial unperturbed level relative to the continuum threshold is very nearly $-P$, where P, the ponderomotive energy, is $P\equiv e^2F_0^2/4\mu\omega^2$, as defined above. The ratio $P/\hbar\omega$ is a measure of the *increase* in the minimum number of photons which are required to detach the electron. Since the shift Δ_i is omitted in lowest-order perturbation theory, this theory overestimates the ionization rate when, roughly speaking, $P/\hbar\omega$ is an appreciable fraction of unity. Perturbation theory may be slightly improved by taking the energy shift Δ_i into account (still evaluating matrix elements in lowest order). However, this modified (Brillouin-Wigner) perturbation theory begins to break down when the ratio of the excursion speed of a free electron to the orbital speed of a bound electron exceeds roughly unity. This ra-

tio is essentially $1/\gamma$, where γ, the Keldysh parameter, is defined[2] as $(|E_i^{(0)}|/2P)^{1/2}$. For $\gamma<1$ the electron charge cloud is considerably distorted by the oscillating field, and the electron spends less time in the region of the nucleus, the region where it can most easily absorb (and emit) photons; consequently modified perturbation theory, which neglects this effect, also overestimates rates. Note that when $\gamma<1$, photodetachment occurs by the electron tunneling through the barrier formed by the atomic potential and the instantaneous applied electric field.[2] In fact, γ can be interpreted[2] as the ratio of the time it takes for the electron to pass through the barrier to the cycle time of the oscillating field.

The success of the Keldysh theory within the framework of our one-electron model suggests that it is worthwhile to extend this theory to include electron-electron correlation, at least in the initial channel. We have done this by representing the ground state of the ion by an accurate two-electron wave function. In the final channel, correlation is neglected; the outgoing photoelectron is described by a plane wave, while the remaining electron is represented by the ground-state wave function of atomic hydrogen. We have also generalized the Faisal-Reiss theory in the same way; correlation results in more significant discrepancies between the Keldysh and Faisal-Reiss theories, due in part to the fact that the interaction of the active electron with the atomic core is long range.

In Sec. II, we describe, in more detail, the various approximations used in treating the interaction of the electron with the radiation field, within the framework of the one-electron model. In Sec. III we present results obtained using this model. These results include total photodetachment rates as well as photoelectron energy spectra. In Sec. IV we outline the extension of the Keldysh and Faisal-Reiss theories to include correlation in the initial state, and we compare results obtained using these theories with results obtained by others using (two-electron) perturbation theory and Floquet theory.

II. DIFFERENT THEORIES

A. Floquet theory

The Floquet ansatz is the replacement of $|\Psi(t)\rangle$ by $e^{-iE_i t/\hbar}|\mathcal{F}(t)\rangle$ where the Floquet vector $|\mathcal{F}(t)\rangle$ is periodic in time with period $2\pi/\omega$. The quasienergy

$$E_i \equiv E_i^{(0)}+\Delta_i-i\Gamma_i/2 \tag{2}$$

is determined by solving the eigenvalue problem that results from substituting the Floquet ansatz into the time-dependent Schrödinger equation and imposing outgoing-wave boundary conditions. We make the dipole approximation and work throughout in the velocity gauge, wherein the electron-field interaction is

$$V(t)=-(e/\mu c)\,\mathbf{A}(t)\cdot\mathbf{p}\ , \tag{3}$$

with

$$\mathbf{A}(t)\equiv \mathrm{Re}(\,\mathbf{A}_0 e^{-i\omega t})\ , \tag{4a}$$

$$\mathbf{A}_0=-i(c/\omega)\mathbf{F}_0\ , \tag{4b}$$

$$\mathbf{F}_0=F_0[\cos(\xi/2)\hat{\mathbf{x}}+i\sin(\xi/2)\hat{\mathbf{y}}]\ , \tag{4c}$$

where $\hat{\mathbf{x}}$ and $\hat{\mathbf{y}}$ are unit vectors that define the polarization plane, with ξ the ellipticity parameter. It is convenient to introduce

$$\zeta(t)\equiv\frac{1}{\hbar}\int_0^t dt'\left[\frac{|\hbar\mathbf{k}_A(t')|^2}{2\mu}-P\right]\ . \tag{5}$$

Note that we have omitted, from $V(t)$, the $|\mathbf{A}(t)|^2$ term

$$(e^2/2\mu c^2)|\mathbf{A}(t)|^2\equiv|\hbar\mathbf{k}_A(t)|^2/2\mu\ ,$$

where

$$\hbar\mathbf{k}_A(t)\equiv e\,\mathbf{A}(t)/c \tag{6}$$

is the "quiver momentum;" the $|\mathbf{A}(t)|^2$ term has been absorbed into $|\Psi(t)\rangle$, through the phase factor $e^{-i(Pt/\hbar)-i\zeta(t)}$, whereby it contributes $-P$ to the energy shift Δ_i. If ω is well below the atomic-orbital frequency of the bound electron, we have $\Delta_i\approx -P$. The Hamiltonian of the electron is

$$H(t)=H_a+V(t)\ , \tag{7}$$

where H_a is the atomic Hamiltonian

$$H_a\equiv(\mathbf{p}^2/2\mu)+W\ , \tag{8}$$

with W the atomic binding potential.

Making the Fourier expansion

$$|\mathcal{F}(t)\rangle=\sum_n e^{-in\omega t}|\mathcal{F}_n\rangle\ , \tag{9}$$

and writing

$$V(t)=V_+e^{-i\omega t}+V_-e^{i\omega t}\ , \tag{10}$$

the harmonic components $|\mathcal{F}_n\rangle$ satisfy the homogeneous set of equations

$$(E_i+n\hbar\omega-H_a)|\mathcal{F}_n\rangle=V_+|\mathcal{F}_{n-1}\rangle+V_-|\mathcal{F}_{n+1}\rangle\ . \tag{11}$$

This coupled set of equations, together with outgoing wave boundary conditions on the harmonic components, form an eigenvalue problem for E_i. The imaginary part of this eigenvalue gives the total ionization rate $\Gamma_i/\hbar$. To the extent that the Floquet ansatz is valid, $\Gamma_i/\hbar$ must be equal to the sum of the partial N-photon ionization rates $\Gamma_{iN}/\hbar$, that is,

$$\Gamma_i=\sum_{N\,(\geq N_0)}\Gamma_{iN}\ , \tag{12}$$

where N_0, the minimum number of photons required to photodetach the electron, is the least value of N for which E_{iN}, defined by

$$E_{iN}\equiv E_i^{(0)}+\Delta_i+N\hbar\omega\ , \tag{13}$$

is positive. The drift momentum (that is, cycle-averaged momentum) of an electron that has absorbed N photons, and is moving along $\hat{\mathbf{n}}$, is $\hbar\mathbf{k}_N=\hbar k_N\hat{\mathbf{n}}$, where

$k_N=(2\mu E_{iN}/\hbar^2)^{1/2}$.

An electron moving freely through the field with drift momentum $\hbar\mathbf{k}_f$ is represented by

$$|\Phi_f(t)\rangle \equiv e^{-i(E_f t/\hbar)-i\Theta(t,\mathbf{k}_f)}|\mathbf{k}_f\rangle \ , \tag{14}$$

where $E_f=|\hbar\mathbf{k}_f|^2/(2\mu)$, where

$$\Theta(t,k)=-\frac{\hbar}{\mu}\int^t dt'\mathbf{k}\cdot\mathbf{k}_A(t) \ , \tag{15}$$

and where $|\mathbf{k}\rangle$ represents a plane wave, normalized here so that in $\mathbf{x}$ space (position space) $\langle\mathbf{x}|\mathbf{k}\rangle=e^{i\mathbf{k}\cdot\mathbf{x}}$. Note that

$$\langle\Phi_{f'}(t)|\Phi_f(t)\rangle=(2\pi)^3\delta^3(\mathbf{k}_{f'}-\mathbf{k}_f) \ .$$

The probability amplitude for finding the electron at $t\sim\infty$ in the state represented by $|\Phi_f(t)\rangle$, if at $t=0$ it was in the state represented by $|\Phi_i(t)\rangle$, is[5]

$$\mathcal{A}_{fi}=\langle\Phi_f(0)|\Phi_i\rangle-\frac{i}{\hbar}\int_0^T dt[\langle\Phi_f(t)|\mathcal{H}(t)^\dagger]|\Psi(t)\rangle \ , \tag{16}$$

where $T\sim\infty$ and where

$$\mathcal{H}(t)=H(t)-i\hbar\frac{d}{dt} \ . \tag{17}$$

Carrying out the operation with $\mathcal{H}(t)$ yields

$$\mathcal{A}_{fi}=\langle\mathbf{k}_f|\Phi_i\rangle-\frac{i}{\hbar}\int_0^T dt\, e^{(iE_f t/\hbar)+i\Theta(t,\mathbf{k}_f)}\langle\mathbf{k}_f|W|\Psi(t)\rangle \ , \tag{18}$$

and putting $|\Psi(t)\rangle\approx e^{-iE_i t/\hbar}|\mathcal{F}(t)\rangle$, neglecting the imaginary part of E_i, we arrive at

$$\mathcal{A}_{fi}=\langle\mathbf{k}_f|\Phi_i\rangle-\frac{i}{\hbar}\sum_N M_N\int_0^T dt\, e^{i(E_f-E_N)t/\hbar} \ , \tag{19}$$

where the matrix element M_N is[5,9]

$$M_N=\frac{\omega}{2\pi}\int_0^{2\pi/\omega}dt\, e^{iNt+i\Theta(t,\mathbf{k}_N)}\langle\mathbf{k}_N|W|\mathcal{F}(t)\rangle \ ; \tag{20}$$

we have put $\mathbf{k}_f=\mathbf{k}_N$ in M_N, anticipating energy conservation. The rate for the electron to emerge with drift momentum $\mathbf{k}_f$ and an energy spread dE is

$$\rho'(E_f)dE\frac{d}{dT}|\mathcal{A}_{fi}|^2 \ ,$$

where $\rho'(E_f)$ is the density-of-states factor. Dropping all finite (energy-nonconserving) terms in $(d/dT)|\mathcal{A}_{fi}|^2$, we find that the cycle-averaged rate for the electron to absorb $N\geq N_0$ photons and emerge into the solid angle $d\hat{\mathbf{n}}$ with drift energy E_{iN} and drift momentum $\hbar\mathbf{k}_N=\hbar k_N\hat{\mathbf{n}}$, is

$$\frac{1}{\hbar}\frac{d\Gamma_{iN}}{d\hat{\mathbf{n}}}=\frac{2\pi}{\hbar}\rho'(E_N)|M_N|^2 \ , \tag{21}$$

where the density of states factor is $\rho'(E_N)=(\mu k_N/8\pi^3\hbar^2)$. The Floquet vector is normalized so that[10]

$$\sum_n(-1)^n[\langle\bar{\mathcal{F}}_n|T^\dagger]|\mathcal{F}_n\rangle=1 \ , \tag{22}$$

where T is the time-reversal operator, and where the $|\bar{\mathcal{F}}_n\rangle$ are the harmonic components of the Floquet vector that corresponds to the reverse sense of rotation for the polarization of the field. Inserting the harmonic expansion, Eq. (9), into the right-hand side of Eq. (20), and writing $\Theta(t,\mathbf{k})=\rho\sin(\omega t-\chi)$, where

$$\rho e^{i\chi}\equiv-(e/\mu c\omega)\mathbf{k}\cdot\mathbf{A}_0 \ , \tag{23}$$

we can integrate over t to yield[5,9]

$$M_N=\sum_n e^{i(N-n)\chi}J_{N-n}(-\rho)\langle\mathbf{k}_N|W|\mathcal{F}_n\rangle \ , \tag{24}$$

where $J_n(z)$ is the regular Bessel function.

B. Keldysh theory

In the Keldysh theory $|\Psi(t)\rangle$ is replaced by $|\Phi_i(t)\rangle$ *in the length gauge.* Since we are working in the velocity gauge, we must replace $|\Psi(t)\rangle$ by

$$e^{-i(e/\hbar c)\Lambda(\mathbf{x},t)}|\Phi_i(t)\rangle \ ,$$

where $e^{-i(e/\hbar c)\Lambda(\mathbf{x},t)}$ is the unitary operator which transforms a state vector in the length gauge to one in the velocity gauge. Recalling that we remove the $|\mathbf{A}(t)|^2$ term, we have

$$\Lambda(\mathbf{x},t)=-\mathbf{A}(t)\cdot\mathbf{x}-(\hbar c/e)[\zeta(t)+Pt/\hbar] \tag{25}$$

and since

$$|\Psi(t)\rangle=e^{-iE_i t/\hbar}|\mathcal{F}(t)\rangle \ ,$$

where $E_i\approx E_i^{(0)}+\Delta_i$ with $\Delta_i\approx-P$, the Keldysh approximation amounts to replacing the Floquet vector $|\mathcal{F}(t)\rangle$ in Eq. (20) by $e^{i(e/\hbar c)\mathbf{A}(t)\cdot\mathbf{x}+i\zeta(t)}|\Phi_i\rangle$. This gives

$$M_N^K=\frac{\omega}{2\pi}\int_0^{2\pi/\omega}dt\, e^{iN\omega t+i\Theta(t,\mathbf{k}_N)+i\zeta(t)}\langle\mathbf{k}_N-\mathbf{k}_A(t)|W|\Phi_i\rangle\cdot \tag{26}$$

The usual form of the Keldysh approximation can be recovered by first noting that

$$\langle \mathbf{k}_N-\mathbf{k}_A(t)|W|\Phi_i\rangle=\langle \mathbf{k}_N-\mathbf{k}_A(t)|[H_a-(\mathbf{p}^2/2\mu)]|\Phi_i\rangle \tag{27a}$$

$$=\{E^{(0)}-(\hbar^2/2\mu)[\mathbf{k}_N-\mathbf{k}_A(t)]^2\}\langle \mathbf{k}_N-\mathbf{k}_A(t)|\Phi_i\rangle \tag{27b}$$

$$=-\hbar\left[\frac{d}{dt}[N\omega t+\Theta(t,\mathbf{k}_N)+\zeta(t)]\right]\langle \mathbf{k}_N-\mathbf{k}_A(t)|\Phi_i\rangle\ , \tag{27c}$$

where in arriving at the last step we used Eqs. (5), (6), and (15), together with

$$E_{iN}-E_i^{(0)}=N\hbar\omega-P\ .$$

It follows that

$$M_N^K=\frac{\omega}{2\pi}\int_0^{2\pi/\omega}dt\left[-i\hbar\frac{d}{dt}e^{iN\omega t+i\Theta(t,\mathbf{k}_N)+i\zeta(t)}\right]\langle \mathbf{k}_N-\mathbf{k}_A(t)|\Phi_i\rangle$$

$$=-\frac{\omega}{2\pi}\int_0^{2\pi/\omega}dt\,e^{iN\omega t+i\Theta(t,\mathbf{k}_N)+i\zeta(t)}\langle \mathbf{k}_N-\mathbf{k}_A(t)|e\mathbf{F}(t)\cdot\mathbf{x}|\Phi_i\rangle\ , \tag{28}$$

where in arriving at the second step we integrated by parts and used

$$-i\hbar\frac{d}{dt}\langle \mathbf{k}_N-\mathbf{k}_A(t)|\Phi_i\rangle=\left\langle \mathbf{k}_N-\mathbf{k}_A(t)\left|\mathbf{x}\cdot\frac{d\mathbf{k}_A(t)}{dt}\right|\Phi_i\right\rangle\ , \tag{29}$$

with $\hbar d\mathbf{k}_A(t)/dt=-e\mathbf{F}(t)$.

The integrand on the right-hand side of Eq. (28) oscillates as t varies, due to the phase factor, and the oscillation is rapid if either $N\gg1$ or $(P/\hbar\omega)\gg1$, as is the case for a field of low frequency. The phase is stationary at those values of t for which

$$E_i^{(0)}-(\hbar^2/2\mu)[\mathbf{k}_N-\mathbf{k}_A(t)]^2=0\ . \tag{30}$$

However, since $E_i^{(0)}$ is negative, there is no real solution of Eq. (30). Rather, there is a saddle point in the complex t plane, and by distorting the contour of integration around this saddle point, and evaluating the integral in Eq. (28) by the saddle-point method, Keldysh was able to derive his famous tunneling formula,[2] valid for frequencies sufficiently low that the Keldysh parameter $\gamma\equiv(|E_i^{(0)}|/2P)^{1/2}$ is small compared to unity. Note that the root-mean-square value of $\hbar\mathbf{k}_A(t)$ is $\sqrt{2\mu P}$. Hence, if $\gamma\ll1$, Eq. (30) is approximately satisfied at those real values of t for which $\mathbf{k}_A(t)\approx\mathbf{k}_N$; such points on the real t axis lie close to the saddle point, and therefore yield the greatest contribution to M_N^K. In other words, if $\gamma\ll1$, a photoelectron is most likely to emerge into the field with a drift momentum $\hbar\mathbf{k}_N$ close to the value of the quiver momentum $\hbar\mathbf{k}_A(t)$ at the moment t of escape.[11,12] (The photoelectron escapes with an instantaneous speed that is not significantly larger than the characteristic atomic orbital speed.) The size of the quiver momentum at the moment of escape depends strongly on the polarization of the field. In the case of linear polarization[11] the magnitude of the electric field $|F_0\cos(\omega t)|$ varies from zero to F_0, and the electron is ejected when the field has its maximum magnitude F_0 (unless F_0 is so large that significant ionization can occur[11] when $|\cos(\omega t)|\ll1$); but when the field has its maximum magnitude, the quiver momentum vanishes, and therefore the drift energy is at most of the order of magnitude of the binding energy. Therefore, for linear polarization, the photoelectron energy distribution is peaked near the threshold, corresponding to $N\approx N_0$ for the number N of photons absorbed. On the other hand, in the case of circular polarization,[12] $\hbar|\mathbf{k}_A(t)|=\sqrt{2\mu P}$ for all t, and therefore the drift energy is of order P, which can be very large at low frequencies. Thus, for circular polarization the energy distribution is peaked at $N\approx2N_0$ (at low frequencies). The energy distributions for both linear and circular polarization were calculated, on the basis of the Keldysh theory, by Nikishov and Ritus[13] and Perelomov, Popov, and Terent'ev;[14] for circularly polarized light the distribution is Gaussian and peaks at $N\approx N_c\equiv2N_0(1-2\gamma^2/3)$ with a full width at half maximum of about $2[N_c\ln(2)/(\sqrt{2}\gamma)]^{1/2}$.

Note that the momentum-space wave function $\langle \mathbf{k}|\Phi_i\rangle$ falls off rapidly as $k\equiv|\mathbf{k}|$ increases beyond $k_i\equiv(2\mu|E_i^{(0)}|/\hbar^2)^{1/2}$. To see this, we can express $\langle \mathbf{k}|\Phi_i\rangle$ as an integral in position space. This integral is small if the integrand oscillates. Now, the range of integration is essentially the range $1/k_i$ of the bound-state wave function $\langle \mathbf{x}|\Phi_i\rangle$, so that $e^{-i\mathbf{k}\cdot\mathbf{x}}$, and therefore the integrand, undergo at least one oscillation over the range of integration when $k/k_i>1$; hence $\langle \mathbf{k}|\Phi_i\rangle$ is small if $k/k_i>1$. Similarly, the matrix element $\langle \mathbf{k}|W|\Phi_i\rangle$ falls off rapidly when k exceeds the larger of k_i and $1/R_W$, where R_W is the characteristic range of the potential W. For the potential of Eq. (1) we have $R_W=1$ a.u., which is much shorter than the range of the bound state, but when we take into account angular correlation between the electrons (or polarization of the core) we have that R_W is greater than the range of the bound state (see Sec. IV). For a neutral atom we always have that R_W is much greater than the range of the bound state.

C. Faisal-Reiss theory

The Faisal-Reiss approximation amounts to replacing $|\Psi(t)\rangle$, in the velocity gauge, by

$$e^{+i[\zeta(t)+Pt/\hbar]}|\Phi_i(t)\rangle ,$$

where the phase factor $e^{+i[\zeta(t)+Pt/\hbar]}$ appears only because we remove the $|\mathbf{A}(t)|^2$ term. This is equivalent to replacing $|\mathcal{F}(t)\rangle$ in Eq. (20) by $e^{+i\zeta(t)}|\Phi_i\rangle$, and gives

$$M_N^{FR}=\frac{\omega}{2\pi}\int_0^{2\pi/\omega}dt\, e^{iN\omega t+i\Theta(t,\mathbf{k}_N)+i\zeta(t)}\langle \mathbf{k}_N|W|\Phi_i\rangle . \tag{31}$$

Thus the Keldysh and Faisal-Reiss N-photon matrix elements differ only through the replacement of the instantaneous mechanical momentum, in the matrix element on the right-hand side of Eq. (26), by the drift momentum. The usual form of the Faisal-Reiss approximation can be recovered upon noting that

$$\langle \mathbf{k}_N|W|\Phi_i\rangle=\langle \mathbf{k}_N|[H_a-(\mathbf{p}^2/2\mu)]|\Phi_i\rangle \tag{32}$$

$$=(E_i^{(0)}-E_{iN})\langle \mathbf{k}_N|\Phi_i\rangle , \tag{33}$$

whereby we obtain

$$M_N^{FR}=(P-N\hbar\omega)\langle \mathbf{k}_N|\Phi_i\rangle \times\frac{\omega}{2\pi}\int_0^{2\pi/\omega}dt\, e^{iNt+i\Theta(t,\mathbf{k}_N)+i\zeta(t)} . \tag{34}$$

The integral over t can be reduced to a generalized Bessel function.[3] We have $\zeta(t)=-(P/2\hbar\omega)\cos(\xi)\sin(2\omega t)$, and for circular polarization ($\xi=\pi/2$) we have $\zeta(t)=0$ so that the integral over t reduces to an ordinary Bessel function.

The Faisal-Reiss theory gives a result that is significantly different from the Keldysh theory only if $\langle \mathbf{k}_N-\mathbf{k}_A(t)|W|\Phi_i\rangle$ and $\langle \mathbf{k}_N|W|\Phi_i\rangle$ are significantly different. Now $\langle \mathbf{k}|W|\Phi_i\rangle$ falls of rapidly with increasing $k\equiv|\mathbf{k}|$ only if $kR_W>1$, provided that R_W (the range of the potential W) is shorter than the range of the bound state, which is true for the potential of Eq. (1). It follows that for this potential the Keldysh and Faisal-Reiss theories can give substantially different results only if the root-mean-square value of $\mathbf{k}_A(t)$ is larger than $1/R_W$, that is, if $P\gg(\hbar^2/\mu R_W^2)$; since $R_W\approx 1$ a.u., the intensity must be very high before there is a noticeable difference.[15]

D. Perturbation theory

In perturbation theory M_N is evaluated to lowest (nonvanishing) order in F_0, that is, to order $(F_0)^N$. Thus, in Eq. (24), we replace $J_{N-n}(-\rho)$ by the leading term in its power series expansion in ρ, we truncate the sum over n, restricting n to the range $0\le n\le N$, and for n in this range we evaluate the harmonic components $|\mathcal{F}_n\rangle$ to lowest (nonvanishing) order in the field F_0, that is, to order $(F_0)^n$. The "perturbative" harmonic components, which we denote by $|\mathcal{F}_n^{(0)}\rangle$, satisfy a set of equations obtained from Eq. (11) by omitting V_-, replacing E_i by the unperturbed value $E_i^{(0)}$, and putting $|\mathcal{F}_0^{(0)}\rangle=|\Phi_i\rangle$. Thus in place of Eq. (11) we have, for $n>0$,

$$(E_i^{(0)}+n\hbar\omega-H_a)|\mathcal{F}_n^{(0)}\rangle=V_+|\mathcal{F}_{n-1}^{(0)}\rangle,\quad |\mathcal{F}_0^{(0)}\rangle=|\Phi_i\rangle . \tag{35}$$

We can try to improve perturbation theory slightly by approximately including the shift Δ_i, taking here, for simplicity, $\Delta_i\approx -P$. Thus modified perturbation theory differs from normal perturbation theory only in that the harmonic components are evaluated by solving a set of equations that differ from the set given by Eq. (35) through the addition of $-P$ to $E_i^{(0)}$.

III. RESULTS (ONE-ELECTRON MODEL)

In Figs. 1–3 we show estimates of rates, obtained in the various approximations discussed above, for multiphoton detachment of our model H^- by linearly polarized light. These rates are *total* rates, integrated over all angles of the emergent photoelectron and summed over all channels. In Fig. 1 we show total rates versus intensity I at a fixed frequency of 0.49 eV; Fig. 2 is similar, but for a fixed frequency of 0.12 eV (the CO_2 laser frequency). We take the Floquet rates as the standard for comparison.

Focusing on Fig. 1, we note first that the minimum number of photons N_0 that must be absorbed to detach the electron at the frequency 0.49 eV is 2 at low intensities. However, at the two-photon threshold intensity (about 3.6×10^{11} W/cm^2) N_0 increases from 2 to 3. We

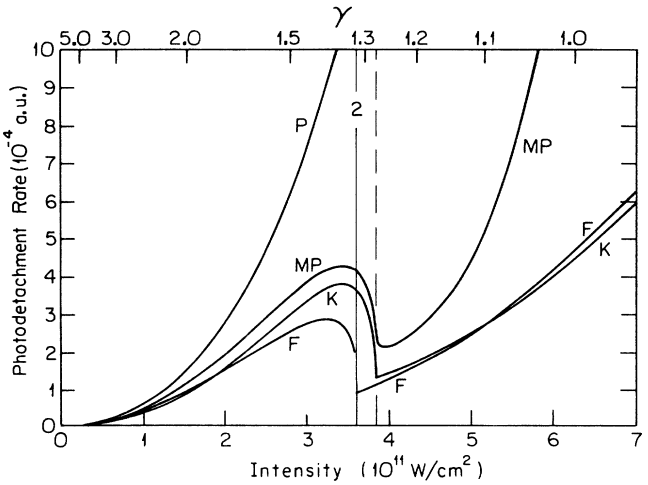

FIG. 1. Estimates of the total rate vs intensity for photodetachment of H^- within the framework of the one-electron model described in the text—the potential is given by Eq. (1). The radiation field has frequency 0.0180 a.u., that is, about 0.49 eV, and is linearly polarized. The notation is as follows: F, Floquet theory; K, Keldysh theory; P, perturbation theory; MP, modified perturbation theory. The solid vertical line marks the threshold intensity at which N_0, the minimum number of photons required to detach the electron, increases from 2 to 3. The dashed vertical line indicates the value of this threshold intensity when the ac Stark shift is neglected. The upper horizontal axis is the Keldysh parameter $\gamma\equiv|E_i^{(0)}|/2P$, where $E_i^{(0)}$ is the unperturbed binding energy and P is the ponderomotive energy.

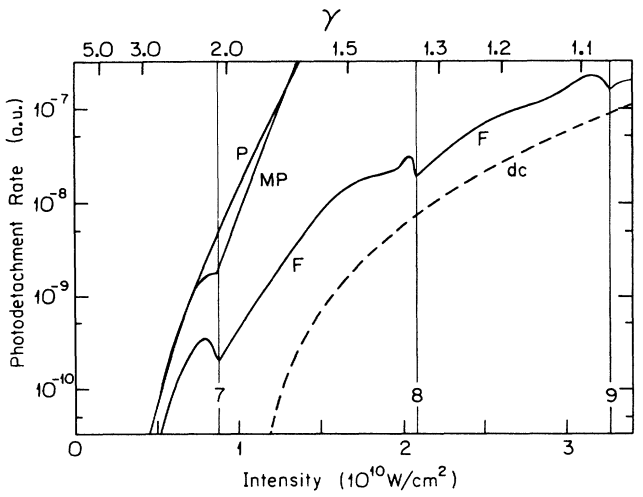

FIG. 2. Same as Fig. 1 but for the frequency 0.004 40 a.u., that is, about 0.12 eV. The rates calculated from Keldysh theory and Floquet theory are indistinguishable. The solid vertical lines mark thresholds, labeled by an integer n; at intensities between the $(n-1)$th and nth thresholds we have $N_0=n$. The curve labeled dc is the cycle-averaged rate for dc ionization.

see that ordinary (Rayleigh-Schrödinger) perturbation theory begins to seriously break down at an intensity of about 10^{11} W/cm^2, when $P/\hbar\omega$ is about 0.12. (The early breakdown of perturbation theory has been noted in numerous papers, for example, Refs. 1, 8, and 10.) Modified (Brillouin-Wigner) perturbation theory begins to seriously break down when the Keldysh parameter $\gamma\equiv(|E_i^{(0)}|/2P)^{1/2}$, which decreases with increasing I as $I^{-1/2}$, is *roughly* unity. In fact, we see from Fig. 1 that modified perturbation theory becomes inaccurate at an intensity slightly above the two-photon threshold intensity, when $\gamma\approx 1.1$. (The ratio of the excursion amplitude $\alpha\equiv|eF_0/\mu\omega^2|$ of a free electron to the binding radius a_i can be significantly larger than unity before modified perturbation theory is inadequate. In fact, $\gamma=1.1$ corresponds, with $a_i=3$ a.u., to $\alpha/a_i\approx 4$.) Both the Keldysh and Faisal-Reiss theories give results in very good agreement with Floquet theory, except near the two-photon threshold. We have not shown the Faisal-Reiss results—they would be barely distinguishable on the figure from those of Keldysh theory (the ratio of the Faisal-Reiss to Keldysh estimates is about 0.95 throughout the intensity range of Fig. 1). As I vanishes, the ratio of the Keldysh rate to the perturbative rate is about 0.77. All approximations but ordinary perturbation theory give rates that exhibit a minimum in the vicinity of the two-photon threshold. Note that the N-photon threshold intensity is the intensity for which $E_{iN}\equiv E_i^{(0)}+\Delta_i+N\hbar\omega$ vanishes. The exact two-photon threshold intensity is indicated by the solid vertical line on Fig. 1, and the minimum of the Floquet rate is exactly at this threshold intensity. That the Floquet rate has a minimum at each N-photon threshold intensity is, as noted earlier,[9] due to the fact that the phase space of the electron in the N-photon channel vanishes as this channel closes. The density of states factor $\rho'(E_N)\equiv\mu k_N/8\pi^3\hbar^2$ vanishes as an N-photon channel closes, and the N-photon partial rate vanishes at least as fast as k_N vanishes, faster if the minimum value of the orbital angular momentum quantum number, l_N say, in the N-photon channel is nonzero, since M_N vanishes as $(k_N)^{l_N}$—see Eqs. (20) and (21). The drop in the total rate near an N-photon threshold intensity simply reflects the vanishing of the contribution of the N-photon partial rate. The reason that Keldysh theory and modified perturbation theory give rates that have minima slightly displaced from the exact position of the three-photon threshold intensity is because, in these two theories, Δ_i is approximated by $-P$, and therefore the threshold appears displaced by the (small) ac Stark shift, Δ_i+P. Of course, in ordinary perturbation theory no channels close as I varies, since Δ_i is neglected. At the N-photon threshold intensity, the nonperturbative total rate, as a function of I, has a "discontinuous" slope; this is because k_N, and therefore the N-photon partial rate, have discontinuous slopes at the threshold intensity. Naturally, in a real experiment the discontinuity in the slope would be hard to detect since a real laser pulse has a profile of intensities as well as a frequency bandwidth. The reason that ordinary perturbation theory breaks down at a value of $P/\hbar\omega$ that is rather small compared to unity is that the rate varies rather rapidly as I increases toward the threshold of the lowest open channel, indicating the presence of a nearby singularity.

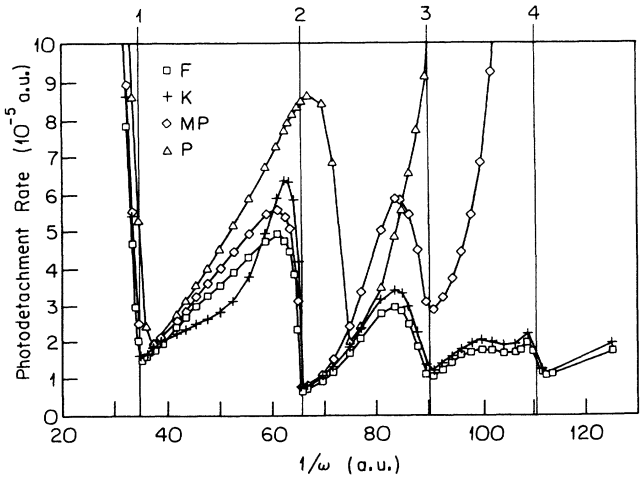

FIG. 3. Same as Figs. 1 and 2, but for a fixed intensity of 10^{11} W/cm^2 and a variable frequency ω.

Note, incidentally, that the total rate for ionization of neutral atoms varies smoothly as a multiphoton ionization threshold is scanned with intensity.[10] This is because, for neutral atoms, the square of the N-photon matrix element M_N diverges as $1/k_N$ as the N-photon threshold is approached from below, and hence the N-photon partial rate [see Eq. (21)] tends to a nonvanishing finite number; just above the N-photon threshold, the N-photon partial rate vanishes, but there are infinitely many resonances accumulating there and when the contributions of the open channels are averaged over these resonances, the total rate exhibits a relatively smooth behavior.

Returning to Fig. 1, observe that we have shown a

break in the Floquet rate right at the threshold. This is because of insufficient accuracy in our calculation. At a threshold, there are two Floquet eigenvalues that are both physically significant; they each lie on different sheets of the Riemann energy surface.[9] When a threshold is passed, these two eigenvalues switch roles—the one that was not physically significant before the threshold was passed becomes physically significant after the threshold is passed, and *vice versa*. When the photodetachment width is relatively large, as it is in Fig. 1, this switchover is difficult to follow numerically, and this is what we mean by a loss of accuracy.

Before turning to Fig. 2, we note that in Ref. 1 partial rates and angular distributions for multiphoton detachment at $\omega=0.49$ and 0.234 eV were presented, based on Floquet calculations using the one-electron model. We have redone these Floquet calculations of partial rates and angular distributions using a different method outlined in Ref. 10. We reproduced results close to those of Ref. 1 and we verified that the partial rates Γ_{in} sum to the total rate Γ_i when the normalization of Eq. (22) is used. (That the partial rates sum to the total rate was also shown in Ref. 1, but a different normalization was used, one that is not as convenient to implement numerically as the normalization chosen here.) Furthermore, we have verified that the Keldysh theory gives essentially the same results for the partial rates and angular distributions as reported in Ref. 1.

In Fig. 2, we present total rates for multiphoton detachment by light of frequency 0.12 eV. We also present the cycle-averaged rate for detachment by a dc field, where by "cycle-averaged" we mean we have taken the average over one cycle of the dc rate at the instantaneous field $F_0\cos(\omega t)$ (this average is independent of ω). In the low-intensity limit of multiphoton detachment, we have $N_0=7$. At $I\approx 0.9\times 10^{10}$ W/cm^2, N_0 increases to 8, at $I\approx 2.1\times 10^{10}$ W/cm^2, N_0 increases to 9, and at $I\approx 3.3\times 10^{10}$ W/cm^2, N_0 increases to 10. Both perturbation theory and modified perturbation theory break down somewhat below the seven-photon threshold intensity; at this threshold $\gamma\approx 2$ (and $\alpha/a_i\approx 9$). Both the Keldysh and Faisal-Reiss theories give rates that are indistinguishable (on the figure) from the Floquet rate. The ponderomotive energy P is inversely proportional to ω^2, and therefore, at the low frequency of 0.12 eV, P is relatively large. Thus the first few thresholds are reached at relatively low intensities. At these low intensities the ac-Stark shift, being only a very small fraction of P (less than one percent), is negligible. Therefore Δ_i is very close to P, and the minima of the rates obtained in Keldysh theory (and also Faisal-Reiss theory) occurs at almost exactly the correct threshold intensities, at least for the first few thresholds, where the intensity is low. We see that the cycle-averaged dc rate approaches the Floquet rate as I increases, that is, as γ decreases below unity; this is expected on the basis of the tunneling theory of Keldysh.[2] Incidentally, we did not have as much difficult calculating the rate at the thresholds seen in Fig. 2 as we did at the threshold in Fig. 1; this is because the rate is not as large near the thresholds in Fig. 2 (recall our discussion of the switchover of eigenvalues at a threshold in connection with Fig. 1).

In Fig. 3 we show the rates versus $1/\omega$ for I fixed at the value 10^{11} W/cm^2. At this intensity, the parameter $\lambda_i^{(I)}$ which characterizes the strength of the electron-field coupling in the initial channel, is small for all frequencies. At large values of ω, above the one-photon threshold frequency (where $N_0=1$), we have $\delta E=\hbar\omega$, and $\lambda_i^{(I)}=|eF_0a_i/\hbar\omega|$. As ω decreases, $\lambda_i^{(I)}$ increases, and when the one-photon threshold frequency is passed, $\lambda_i^{(I)}$ becomes the constant $|eF_0a_i/E_i^{(0)}|$. Once again, Keldysh theory is in good agreement with Floquet theory (as is Faisal-Reiss theory), except perhaps in the region between the one- and two-photon thresholds; in this region the *s-p-s* angular momentum channel is significant, and the *s*-wave phase shift of the outgoing photoelectron (which is neglected in the Keldysh-Faisal-Reiss theories) is nonnegligible. At large values of ω (above the one-photon threshold frequency), both forms of perturbation theory are rather accurate—naturally, the modified form is the more accurate. Ordinary perturbation theory begins to seriously break down between the one- and two-photon threshold frequencies, below about $\omega=\frac{1}{55}$ a.u., when $P/\hbar\omega\approx 0.12$, whereas modified perturbation theory does not begin to seriously break down until the frequency is between the two- and three-photon thresholds, below about $\omega=\frac{1}{75}$ a.u., when $\gamma\approx 1.8$ (and $\alpha/a_i\approx 10$). The minima in the rates at frequency thresholds are analogous to the minima at intensity thresholds, and, likewise, the slope in the rate at a frequency threshold is also discontinuous. Rates for photodetachment of H^- were recently measured[16] over a continuous frequency range, and minima were clearly observed. Presumably these experimentally observed minima, which have also been discussed theoretically by others,[17–19] occur at the frequency thresholds; this is difficult to confirm definitively since the intensity of the laser pulse used in the experiment fluctuated wildly in time so that the thresholds were ponderomotively shifted by uncertain amounts during the passage of the pulse. Note that in (ordinary) perturbation theory there is no ponderomotive shift of the frequency thresholds; in Fig. 2 we see that the unperturbed two-photon frequency threshold (where the perturbative rate has a minimum) occurs at a significantly lower frequency than does the shifted two-photon frequency threshold. Incidentally, the perturbative rate is very accurate at its two-photon minimum, though this may be fortuitous.

In Fig. 4 we show the photoelectron energy distributions for both linear and circular polarizations at a frequency of 0.12 eV and an intensity of 2×10^{11} W/cm^2. The Keldysh parameter γ is about 0.43, and so photodetachment takes place through tunneling. We show the N-photon partial rate, calculated using Keldysh theory, versus the number N of photons absorbed (lower scale) or versus the drift energy in the field, that is, the energy absorbed above the shifted threshold (upper scale). The energy distributions calculated using Faisal-Reiss theory are not shown but would not be much different (only slightly lower) than those of Fig. 4. Note that in the weak-field limit the minimum number of photons required for photodetachment is 7 ($|E_i^0|\approx 7\hbar\omega$), while at

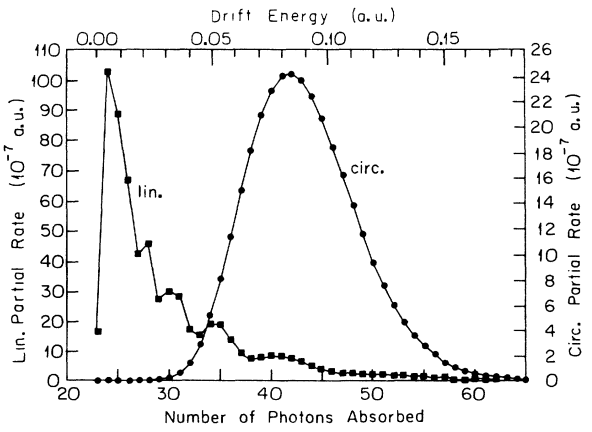

FIG. 4. Photoelectron energy spectra (calculated using the Keldysh theory within the framework of the one-electron model) at a frequency of 0.004 40 a.u., that is, about 0.12 eV, and at an intensity of 2×10^{11} W/cm^2. We show results for linear (lin.) or circular (circ.) polarization.

the intensity relevant to Fig. 4 we have $N_0=23$; the ponderomotive energy is $P\approx17\hbar\omega$, that is, about 2.0 eV, or, equivalently, about $2.7|E_i^{(0)}|$. We see that in the case of linear polarization the distribution peaks at $N=24$, that is, at $N\approx N_0$, so that the drift energy of the photoelectron in the field is small, comparable to $|E_i^{(0)}|$. (The reason that the partial rate is so low at $N=23$ is that 23 photons lift the electron only barely above threshold.) However, in the case of circular polarization the distribution peaks at $N=42$, that is, at $N=N_0+19\approx2N_0$, where $(N-N_0)\hbar\omega\approx P$, and hence the distribution peaks when the drift energy of the electron in the field is about the ponderomotive energy, in accord with the discussion of Sec. II B and Refs. 11–14. The full width at half maximum of the distribution is (in the case of circular polarization) about $13\hbar\omega$, which is the same as the width $2[\sqrt{2}N_0\ln(2)/\gamma]^{1/2}$ predicted in Ref. 13. The ponderomotive energy is about twice as large as $|E_i^{(0)}|$, and evidently, in the case of circular polarization, the photoelectron emerges into the field with a drift momentum that is about a factor of $\sqrt{2}$ larger than the characteristic momentum that this electron had in its initial bound state. In the experiments of Refs. 11 and 12, the photoelectron has a drift momentum many times larger than the characteristic bound-state momentum. Therefore, the question naturally arises as to where this momentum comes from, since a photon carries very little momentum. Note that (in the dipole approximation) no net momentum is removed from the radiation field, since the atomic core, in this case, the neutral atom, acquires a drift momentum that is equal and opposite to that acquired by the detached photoelectron. However, the interference of the electric field of the electron with the magnetic field of the radiation transmits significant momentum to the electron. As long as the electron is bound to the core, this momentum is balanced by the electrostatic binding force, but once the electron becomes free of the core it receives a net nonzero momentum. The heavy neutral core, on the other hand, moves freely off with a constant momentum-close to the instantaneous momentum it had at the moment of breakup. See the paper of Ref. 20 for further discussion of this point. We note that while the Keldysh theory accounts for this momentum transfer mechanism, the Faisal-Reiss theory does not; in the latter theory, the drift momentum comes from the bound-state momentum distribution, and this distribution falls off rapidly with increasing momentum, particularly if the orbital angular momentum quantum number of the bound state is large. Hence, although the Faisal-Reiss theory underestimates the ionization rate by only 10–20% for the parameters of Fig. 4, this theory seriously underestimates the rate when $P\gg|E_i^{(0)}|$, if the light is circularly polarized.

IV. TWO-ELECTRON MODEL

To include electron-electron correlation in the initial channel we use a Hylleraas-type wave function to represent the initial unperturbed bound state. Thus, with $\mathbf{x}_1$ and $\mathbf{x}_2$ the electron coordinates, the unperturbed ion is represented in position space by

$$\langle\mathbf{x}_1,\mathbf{x}_2|\Phi_i\rangle=\sum_{l,m,n}c_{lmn}s^l t^{2m}u^n e^{-qs/2}\ , \tag{36}$$

where, with $r_j=|\mathbf{x}_j|$, we have $s=r_1+r_2$, $t=r_1-r_2$, and $u=r_{12}\equiv|\mathbf{x}_1-\mathbf{x}_2|$. We have factored out the (antisymmetric) singlet spin component, so the spatial wave function $\langle\mathbf{x}_1,\mathbf{x}_2|\Phi_i\rangle$ is symmetric. As before, we define

$$|\Phi_i(t)\rangle\equiv e^{-E_i^{(0)}t/\hbar}|\Phi_i\rangle\ ,$$

but now $E_i^{(0)}$ is the unperturbed bound-state energy of the complete ion. Recalling that we are working in the velocity gauge, the Keldysh ansatz is

$$|\Psi(t)\rangle\approx e^{-i(e/\hbar c)\Lambda(\mathbf{x}_1,\mathbf{x}_2,t)}|\Phi_i(t)\rangle\ , \tag{37}$$

where we have introduced a gauge-transform function that is analogous to $\Lambda(\mathbf{x},t)$ of Eq. (25) but that depends on both electron coordinates:

$$\Lambda(\mathbf{x}_1,\mathbf{x}_2,t)=-\mathbf{A}(t)\cdot(\mathbf{x}_1+\mathbf{x}_2)-2(\hbar c/e)[\zeta(t)+Pt/\hbar]\ . \tag{38}$$

In the final channel the emergent photoelectron moves freely through the field with drift momentum $\hbar\mathbf{k}_f$ and drift energy $|\hbar\mathbf{k}_f|^2/2\mu$, while the other electron is bound in the $1s$ state of atomic hydrogen, with energy $E_{\mathrm{H}(1s)}^{(0)}=-0.5$ a.u. The state of the system, in the final channel, is represented in position space by

$$\langle\mathbf{x}_1,\mathbf{x}_2|\Phi_f(t)\rangle\equiv\frac{1}{\sqrt{2}}e^{-iE_f t/\hbar-i\Theta(t,\mathbf{k}_f)}\times(\langle\mathbf{x}_1,\mathbf{x}_2|e^{-i(e/\hbar c)\Lambda(\mathbf{x}_2,t)}|\mathbf{k}_f,\phi_{\mathrm{H}(1s)}\rangle+\{1\leftrightarrow2\})\ , \tag{39}$$

where $\Theta(t,\mathbf{k})$ was defined by Eq. (15), where $E_f=E_{\mathrm{H}(1s)}^{(0)}+|\hbar\mathbf{k}_f|^2/(2\mu)$ and where $\{1\leftrightarrow2\}$ implies an interchange of the two electrons in the preceding term, so that the wave function is properly symmetrized, and where

$$\langle\mathbf{x}_1,\mathbf{x}_2|\mathbf{k}_f,\phi_{\mathrm{H}(1s)}\rangle\equiv\langle\mathbf{x}_1|\mathbf{k}_f\rangle\times\langle\mathbf{x}_2|\phi_{\mathrm{H}(1s)}\rangle\ , \tag{40}$$

with $\langle\mathbf{x}|\phi_{\mathrm{H}(1s)}\rangle$ the wave function of ground-state atomic hydrogen, normalized so that

$$\langle\phi_{\mathrm{H}(1s)}|\phi_{\mathrm{H}(1s)}\rangle=1\ .$$

As before, we impose the normalization

$$\langle \Phi'_f(t)|\Phi_f(t)\rangle=(2\pi)^3\delta^3(\mathbf{k}_{f'}-\mathbf{k}_f)\ ;$$

but this is not exactly in accord with the choice of the normalization factor of $1/\sqrt{2}$ in Eq. (37) since the overlap

$$\langle \mathbf{k}_f|e^{-i(e/\hbar c)\Lambda(\mathbf{x},t)}|\phi_{\mathrm{H}(1s)}\rangle$$

is nonvanishing. However, since we are treating the two electrons as independent (we neglect correlation) in the final channel, it is consistent to neglect this overlap. One might consider adding to $|\mathbf{k}_f\rangle$ a term (proportional to $e^{-i(e/\hbar c)\Lambda(\mathbf{x}_1,t)}|\phi_{\mathrm{H}(1s)}\rangle$), so as to force the overlap to vanish, but we do not do this here. Combining Eqs. (16), (37), and (39) yields, in place of Eq. (26),

$$M_N^K=\sqrt{2}\frac{\omega}{2\pi}\int_0^{2\pi/\omega}dt\, e^{iNt+i\Theta(t,\mathbf{k}_N)+i\zeta(t)}\langle[\mathbf{k}_N-\mathbf{k}_A(t)],\phi_{\mathrm{H}(1s)}|\left[\frac{e^2}{r_{12}}-\frac{e^2}{r_1}-e\mathbf{F}(t)\cdot\mathbf{x}_2\right]|\Phi_i\rangle\ , \tag{41}$$

where the factor of $\sqrt{2}$ accounts for the presence of both electrons. We note that a similar generalization of the Keldysh theory to two electrons was carried out and applied[21] to the ionization of helium; in that application, the outgoing photoelectron was described by a Coulomb wave rather than a plane wave. Note also that the generalization of the Faisal-Reiss amplitude M_N^{ER} to two electrons, differs from Eq. (41) only through the replacement of $\mathbf{k}_N-\mathbf{k}_A(t)$ by $\mathbf{k}_N$ on the right-hand side of Eq. (41).

The spatial integration on the right-hand side of Eq. (41) can be simplified by using the identity

$$\mathcal{M}_1\equiv\langle \mathbf{k},\phi_{\mathrm{H}(1s)}|\left[\frac{e^2}{r_{12}}-\frac{e^2}{r_1}\right]|\Phi_i\rangle \tag{42a}$$

$$=\left[E_i^{(0)}-E_{\mathrm{H}(1s)}^{(0)}-\frac{\hbar^2}{2\mu}k^2\right]\langle \mathbf{k},\phi_{\mathrm{H}(1s)}|\Phi_i\rangle\ , \tag{42b}$$

with $k=|\mathbf{k}|$. Note that, as far as the matrix element $\mathcal{M}_1$ is concerned, the effective interaction of the photoelectron (electron 1) with the residual atom is determined by the integral over $\mathbf{x}_2$ of the interaction $e^2(r_{12}^{-1}-r_1^{-1})$ multiplied by the weight function $\langle \mathbf{x}_2|\phi_{\mathrm{H}(1s)}\rangle\langle \mathbf{x}_1,\mathbf{x}_2|\Phi_i\rangle$. If electron-electron angular correlation in the initial channel were neglected, this weight function would be spherically symmetric in $\mathbf{x}_2$, and all the multipole terms of $e^2(r_{12}^{-1}-r_1^{-1})$ would vanish upon integration over $\mathbf{x}_2$; in that case, the effective interaction of electron 1 would decay exponentially, the effective range being about 1 a.u., as for the one-electron potential $W(r)$. However, when angular correlation is included, multipole terms do contribute and the effective interaction falls off with increasing r_1 as a power of $1/r_1$; therefore, angular correlation, that is, the terms in the variable u in the Hylleraas wave function, Eq. (36), are expected to lead to a more significant (though still small) difference between the Faisal-Reiss and Keldysh theories than was found in studying the one-electron model.

The term in $e\mathbf{F}(t)\cdot\mathbf{x}_2$ on the right-hand side of Eq. (41) contributes only through the electron-electron angular correlation (the terms in u). Consequently, we expect the matrix element

$$\langle \mathbf{k},\phi_{\mathrm{H}(1s)}|\mathbf{F}(t)\cdot\mathbf{x}_2|\Phi_i\rangle\equiv\mathbf{k}\cdot\mathbf{F}(t)\mathcal{M}_2 \tag{43}$$

to be small compared to $\mathcal{M}_1$. Furthermore, $\mathcal{M}_2$ is pure imaginary while $\mathcal{M}_1$ is pure real, and the contributions of these two matrix elements to $\mathcal{M}_N^K$ are out of phase by $\pi/2$ [the integration over t in Eq. (41) does not alter the relative phase] so that there is no interference in the absolute magnitude $|M_N^K|$. Hence, we expect the contribution from $\mathcal{M}_2$ to $|\mathcal{M}_N^K|$ to be negligible. Both the matrix elements $\mathcal{M}_1$ and $\mathcal{M}_2$ can be evaluated in closed form.

In Table I we illustrate the convergence of the total binding energy $E_i^{(0)}$ with respect to the number of terms in the Hylleraas wave function of Eq. (36), and we also illustrate the convergence of the matrix elements $\mathcal{M}_1$ and $\mathcal{M}_2$ at three different values of k. The rate of convergence of the energy is fast, which is not unexpected since the energy estimate is a *variational* estimate, of *second* order in the error of the wave function. On the other hand, the errors in the estimates of the matrix elements are of first order in the error of the wave function, and accordingly the rate of convergence is slower. In fact, $\mathcal{M}_2$ has not converged, and is significantly effected by the inclusion of

TABLE I. Total binding energy, and overlap matrix elements $\mathcal{M}_1$ and $\mathcal{M}_2$, defined by Eqs. (42) and (43) of the text, for the ground state of H^-. The indices n, l, and m, are the powers of s, t^2, and u in the Hylleraas wave function, Eq. (36) of the text; the total number of terms N_p is indicated in the first column of the table. The nonlinear parameter q of the wave function was chosen to minimize the energy. The number k is the momentum, in a.u., of the outgoing photoelectron.

						$\mathcal{M}_1$			$i\mathcal{M}_2$		
N_p	l	m	n	q	Energy	$k=0.01$	$k=0.2$	$k=1$	$k=0.01$	$k=0.2$	$k=1$
2	0	1	0	1.3481	−0.512 293	40.7	28.5	1.65	0.000	0.00	0.00
8	1	1	1	1.6085	−0.526 816	37.5	26.9	1.82	0.038	0.66	0.42
12	1	2	1	1.5578	−0.527 541	43.7	28.1	1.79	0.040	0.69	0.43
18	2	2	1	1.6267	−0.527 686	45.1	28.1	1.78	0.035	0.61	0.43
24	2	3	1	1.6402	−0.527 722	47.1	28.0	1.78	0.034	0.60	0.43
36	2	3	2	1.6274	−0.527 745	47.8	27.9	1.78	0.031	0.54	0.37
48	3	3	2	1.7135	−0.527 749	47.8	27.9	1.78	0.025	0.44	0.33

terms in u^2. We could estimate these matrix elements using a variational procedure,[22] but we deemed this unnecessary since reasonable accuracy for $\mathcal{M}_1$ is obtained by including 24 terms in the Hylleraas wave function, and the relative contribution of the exact $\mathcal{M}_2$ to $|M_N^K|$ is almost certainly very small. Incidentally, $\mathcal{M}_2$ vanishes linearly with k as $k \rightarrow 0$.

We have carried out calculations of photodetachment rates using the "two-electron" Keldysh theory; in all calculations we used the 24-term Hylleraas wave function. In Fig. 5 we present results for two-photon detachment of H^- by *circularly* polarized light. We show the photodetachment rate versus the photoelectron kinetic energy ε (the frequency is varied) at the fixed intensity of 10^9 W/cm^2. This intensity is sufficiently low that the electron-field interaction can be accurately treated within perturbation theory. On the log-log scale of the figure, our (Keldysh) results for the rate are almost indistinguishable from the perturbation theory results of Fink and Zoller[23] and Liu and Starace.[24] The Fink-Zoller and Liu-Starace results were obtained by treating the atomic interaction of the two electrons within the adiabatic hyperspherical formulation. To indicate the discrepancies, we have plotted the ratio of the Liu-Starace estimate of the rate to that of ours. Note that the rate vanishes rapidly as the photoelectron energy ε does. An electron that absorbs two circularly polarized photons has (in the perturbative limit) an orbital angular momentum quantum number of 2, and therefore the rate vanishes with ε as $\varepsilon^{2,5}$ (recall our discussion of the minima seen in Fig. 1). This slope is confirmed by the results of Fig. 5. The photodetachment rate turns down as the photoelectron energy approaches $\hbar\omega$, close to the one-photon threshold.

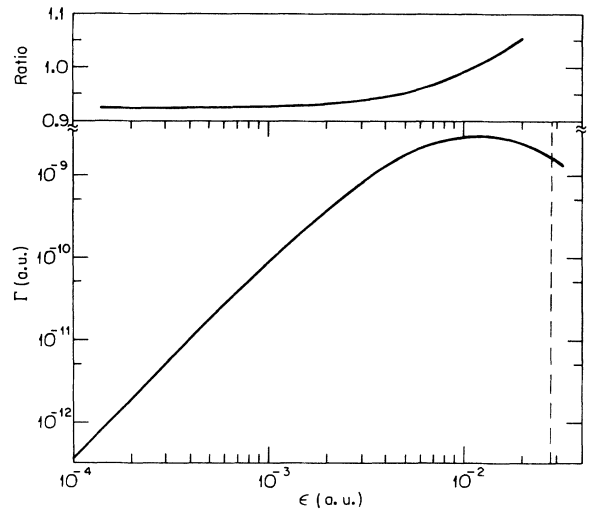

FIG. 5. Lower box: Photodetachment rate Γ (in a.u.) vs photoelectron energy ε (in a.u.) for two-photon detachment of H^- by circularly polarized light at the intensity 10^9 W/cm^2, calculated using the two-electron Keldysh theory. The vertical dashed line marks the value of ε (or, rather, ω) for which one-photon detachment becomes possible. Upper box: Ratio of the (two-electron) perturbation theory rates of Liu and Starace (Ref. 24) to the two-electron Keldysh rates; these results would be indistinguishable on the scale of the lower box.

In Fig. 6 we present results of two-electron Keldysh theory for two-photon detachment of H^- by *linearly* polarized light. As in Fig. 5, we show the photodetachment rate versus the photoelectron kinetic energy ε at the fixed low intensity of 10^9 W/cm^2. We also show the perturbation theory results of Liu and Starace[24] (based on the adiabatic hyperspherical formulation) as well as perturbation theory results that we obtained within the framework of the one-electron model; we have now multiplied the latter results by a factor of 2, so as to take into account that either of two electrons may be ejected. The two sets of perturbation theory results are qualitatively the same, and the quantitative agreement is rather good also, indicating the reliability of the one-electron model.[25] The agreement with the two-electron Keldysh results is not as good, but is still reasonable. As in Fig. 3, the discrepancy with Keldysh theory is probably due to the neglect, in the Keldysh theory, of the *s*-wave phase shift in the *s-p-s* channel; this phase shift is nonnegligible for ε well above zero. (The *d*-wave phase shift in the *s-p-d* channel is small, so a similar discrepancy does not occur in the case of circular polarization.) Note that the rate should vanish with vanishing ε as $\sqrt{\varepsilon}$; this slope is confirmed by the results of Fig. 6. Note also that the one-electron Keldysh results (not shown) have roughly the same shape as the two-electron Keldysh results, but are up to a factor of 1.7 higher; thus the agreement with the Liu-Starace results is not quite as good.

Finally, in Table II we present results of two-electron Keldysh theory for multiphoton detachment of H^- by linearly polarized light at two frequencies close to the CO_2 laser frequency, and at four different intensities. We also show the results of two-electron Faisal-Reiss theory, results of Mercouris and Nicolaides[26] obtained using two-electron Floquet theory, and results of various calculations (Floquet, Keldysh, and Faisal-Reiss) carried out within the framework of the one-electron model; once again, we have multiplied the "one-electron results" by a

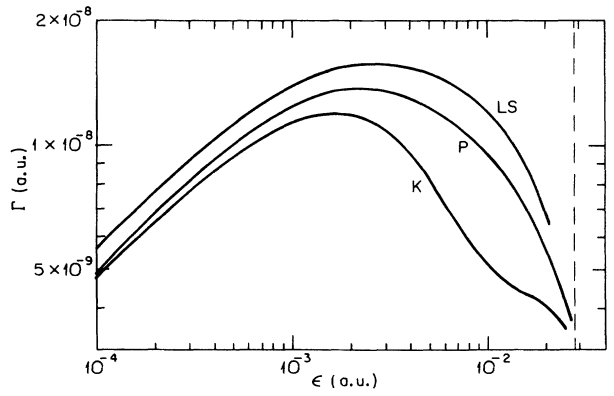

FIG. 6. Photodetachment rate Γ (in a.u.) vs photoelectron energy ε (in a.u.) for two-photon detachment of H^- by linearly polarized light at the intensity 10^9 W/cm^2, calculated using *K*, two-electron Keldysh theory; *P*, one-electron perturbation theory; LS, two-electron perturbation theory (Liu-Starace, Ref. 24).

TABLE II. Total rate Γ (in a.u.) for ionization of H^- by linearly polarized light of various intensities I (in 10^9 W/cm^2), and frequencies ω (in 10^{-3} a.u.). Column (a): two-electron Floquet results of Mercouris and Nicolaides, including electron-electron correlation (Ref. 26). Columns (b) and (c): two-electron Keldysh and Faisal-Reiss results, respectively, based on a 24-parameter Hylleraas wave function for the initial state. Columns (d)–(f): results obtained within the framework of the one-electron model. (d): Floquet, (e): Keldysh, (f): Faisal-Reiss. Results in columns (d)–(f) have been multiplied by 2 to take into account two electrons. Note that, due to numerical roundoff error, we cannot obtain the very small Floquet rates at $I=1.402\times10^9$ W/cm^2. Here $a[b]$ denotes $a\times10^b$.

I	ω	(a)	(b)	(c)	(d)	(e)	(f)
1.402	4.0	3.3[−11]	2.6[−16]	8.1[−16]		1.1[−15]	1.1[−15]
1.402	4.3	3.3[−11]	3.1[−15]	8.8[−15]		1.2[−14]	1.1[−14]
2.805	4.0	6.4[−11]	6.2[−14]	1.7[−13]	2.4[−13]	2.3[−13]	2.2[−13]
2.805	4.3	6.2[−11]	4.0[−13]	1.1[−12]	1.4[−12]	1.5[−12]	1.4[−12]
11.22	4.0	2.1[−10]	4.8[−10]	1.2[−9]	1.6[−9]	1.6[−9]	1.5[−9]
11.22	4.3	2.7[−10]	8.8[−10]	2.1[−9]	2.7[−9]	2.8[−9]	2.6[−9]
25.25	4.0	3.7[−9]	5.2[−8]	1.1[−7]	1.5[−7]	1.6[−7]	1.5[−7]
25.25	4.3	4.5[−9]	5.1[−8]	1.0[−7]	1.4[−7]	1.4[−7]	1.4[−7]

factor of 2. Our various one-electron results (columns *d*, *e*, and *f*) are in good agreement with each other, and, incidentally, agree closely with the results reported by Reiss in Ref. 3. (In Ref. 3, Reiss calculated rates for multiphoton detachment of H^- by a CO_2 laser, using a one-electron model with a very simple wave function to describe the H^- ground state.) Our "one-electron" results never differ by more than a factor of 3 or 4 from our two-electron Keldysh and Faisal-Reiss results. Our one- are much smaller than two-electron results are in part due to the differences in the values of the binding energy of the active electron used in the one- and two-electron calculations; the frequency is close to the seven-photon threshold, whose exact position depends on the value of the binding energy, and since the rate varies rapidly in the vicinity of this threshold the rate is sensitive to the binding energy. Note that, in both our one- and two-electron calculations, we have $N_0=8$, except for $\omega=0.43$ a.u. at the intensities 1.402 and 2.805×10^9 W/cm^2, where we have $N_0=7$. In contrast to the one-electron model, the two-electron Keldysh and Faisal-Reiss results are noticeably different, by a factor of 2 or 3; this may be due to the fact that correlation extends the range of the atomic interaction of the active electron, as noted earlier in this section. Our results are much smaller than those of Mercouris and Nicolaides [column (a)] at low intensities (1.402 and 2.805×10^9 W/cm^2). We cannot establish the source of such large discrepancies; however, we note that, due to roundoff error, we found it very difficult to calculate an accurate Floquet rate within our one-electron model when the rate is very small, as it is at low intensities—in fact, we were unable to calculate an accurate rate at the intensity 1.402×10^9 W/cm^2. The weak field Floquet results of Crance[25] are also in disagreement with our results.

V. CONCLUSION

We have examined various approximations for treating the electron-field interaction in the calculation of rates for multiphoton detachment of H^- by low-frequency light. Ordinary perturbation theory is inadequate once the ponderomotive energy is an appreciable fraction of $\hbar\omega$. Perturbation theory can be modified by taking into account the energy shift of the bound level relative to the continuum, and this leads to an improvement. However, modified perturbation theory is inadequate once the intensity is so high that the Keldysh parameter γ is of order unity or below. The Keldysh theory gives results in remarkably good agreement with accurate multielectron perturbation theory calculations, at low intensities, and with single-electron Floquet calculations, at all intensities. We do not expect the Keldysh theory, in its present form, to give accurate results near the resonance regions of H^-, and certainly this theory is not accurate in treating systems having many bound states, such as the hydrogen atom.[10] Note that the Keldysh approximation yields a vanishing dipole moment in the initial channel, so that, within this approximation, $\sum_n \langle \mathcal{F}_{n-N}|e\mathbf{x}|\mathcal{F}_n\rangle$ vanishes for all integer N; this cancellation is perhaps one reason why the numerical calculation of Nth order harmonic generation rates tends to be less stable at high intensities than the corresponding calculation of ionization rates.

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