Photoionization spectra by intense linear, circular, and elliptic polarized lasers

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We study the characteristics of ionization rates of an atom by an intense laser field using an extension of Keldysh theory. High-order semianalytical expressions are obtained for linear, circular, and elliptically polarized fields. We compare the features of the new rates with Keldysh analytical results as functions of frequency and electric field strength. The directionality of photoelectron emissions is compared for elliptical, circular, and linearly polarized cases. Laser polarization has significant effects on the ionization spectra and directionality of photoelectron emission.

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I. INTRODUCTION

Over the past decade, there has been much interest in highly nonlinear nonperturbative interactions of atoms and molecules with intense few-cycle laser pulses with intensity [1] approaching the atomic unit of intensity $I_0 = 5 \times 10^{16}$ W cm^{-2} or equivalently the electric field strength $E_0 = 5 \times 10^9$ V cm^{-1}. The study of electron dynamics [2] in this nonlinear nonperturbative regime leading to new processes such as above-threshold ionization (ATI), multiphoton ionization (MPI) [3], and high-order-harmonic generation (HHG) [4] has brought remarkable advancement in the field of attosecond physics [5,6]. The pioneering work of Keldysh [7] provides the basic theoretical description of the tunnel ionization by intense linearly polarized light where the photon energy $\hbar\omega$ is lower than the ionization potential [8] I_p. For photon energy higher than I_p the model describes multiphoton ionization where the transition from bound states to free ionized states is accompanied by the absorption of several quanta of order N_m. The Keldysh theory [9–11] determines whether the photoionization is in the tunneling or multiphoton region through the adiabaticity parameter $\gamma = \sqrt{I_p/2U_p}$, where $U_p = e^2E^2/4m\omega^2$ is the classic ponderomotive energy. The three-step model by Corkum [3,12] provides a classical description of the electron in the strong electromagnetic field, E, by initially tunneling to the continuum with initial zero velocity [3,4] or nonzero velocity [12] and then recolliding with the parent ion, hence resulting in the maximum return energy or equivalently the photon energy, $N_m\hbar\omega_0 = I_p + 3.17U_p$. Recolliding electrons can be used for molecular [13] or nuclear imaging [14]. Their high energy and corresponding wavelengths, $\lambda = 2\pi\hbar/p$, become comparable to molecular distances, thus providing new tools for imaging and requiring further the development of electron control through intense light-matter interactions [15,16].

Recent experiments [17] have led to the generation of high-energy attosecond light sources from intense laser-matter interaction. The generation of attosecond pulses is based on a highly nonlinear response, HHG, where the electron is driven to the Volkov continuum state, returns and recollides with the parent ion [18], emitting a plateau of harmonics where a maximum energy cutoff is located at $I_p + 3.17U_p$. The nature of the cutoff and whether other values beside $I_p + 3.17U_p$ are possible is a subject of current interest as explored by Milošević and A. F. Starace [19] who showed that linearly polarized intense pulses with a static field perpendicular to the linear polarization laser field can induce a plateau extending towards high energy x-ray photons. Yuan and Bandrauk [20] showed numerically that molecular high-order-harmonic generation (MHOHG) can have the maximum elliptically polarized harmonic energies of $I_p + 13.5U_p$ [20] for certain internuclear distances and also the relative pulse carrier envelope phase (CEP). Their model shows that MHOHG is not only contributed by the recollision of the electron with the parent ion of H_2^+ but also by recollision with the neighboring ion. Constant *et al*. [21] have recently proposed this as a new route for attosecond pulse generation in a dissociative molecular medium. Milošević *et al*.. [22] and Borca *et al*. [23] have shown, respectively, that circularly and elliptically polarized harmonics can be also generated if a linearly polarized laser field is orientated at an appropriate angle relative to a static electric field.

In this paper, we examine analytically and to higher order the role of polarization, strength, and frequency of the laser field on atomic polarized photoionization rates. We present general semianalytical expression of photoionization rates for the hydrogen atom by intense circular, elliptical polarization fields [24,25]. We generalize Keldysh's result for linear polarized light from small photoelectron momentum to an arbitrary value of momentum. We compare the general results of linearly polarized light and the approximate expression of the Keldysh theory. We analyze the variations of the spectrum of the photoionization rates with laser field strength and frequency. The main results are detailed spectra and directionality of photoelectron emission for intense circular and elliptical polarizations, which are qualitatively different from the results with linear polarized laser fields.

II. KELDYSH THEORY FOR LINEAR POLARIZATION

For a linearly polarized electric field of strength E, the ionization rate for small momentum was first derived by Keldysh [7]. The theory is valid for small momenta such that terms higher than $(\frac{p}{\sqrt{2mI_p}})^2$ are negligible. This restriction also implies a limitation on the laser field strength E (since $\frac{d\mathbf{p}}{dt} \simeq e\mathbf{E}$) and hence the Keldysh parameter $\gamma = \sqrt{I_p/2U_p}$.

A. Ionization rate for small momentum

The general ionization rate is obtained by integration over the three-dimensional momenta

$$w = \frac{2\pi}{\hbar}\int_{-\infty}^{\infty}\sum_{n=-\infty}^{\infty}|L_n(\mathbf{p})|^2\delta[\hbar\Omega(p)-n\hbar\omega]\frac{d^3p}{(2\pi\hbar)^3}, \quad (1)$$

with the ionization amplitude (integrated over one period T of the laser oscillations)

$$L_n(\mathbf{p}) = \frac{1}{2\pi}\int_{-T/2}^{T/2} V_0(\mathbf{\Pi}(t))e^{iS(\mathbf{p},t)}e^{-i[\Omega(p)-n\omega]t}\omega dt. \quad (2)$$

The $V_0(\mathbf{\Pi}(t)) = \int \psi_s(\mathbf{r})e\mathbf{E}\cdot\mathbf{r}\exp[-\frac{i}{\hbar}\mathbf{\Pi}(t)\cdot\mathbf{r}]d^3r$ is the transition matrix element between the starting state of the atom with ψ_s and the photoelectron, and $S(\mathbf{p},t) = \int_0^t \frac{1}{\hbar}[I_p + \frac{1}{2m}\mathbf{\Pi}(\tau)^2]d\tau$ is the action phase during photoionization. Here, $\mathbf{\Pi}(t) = \mathbf{p} - e\mathbf{A}(t)$ and $\hbar\Omega(p) = I_p + U_p + \frac{p^2}{2m}$, with $d^3p = p^2dpd\Omega_a$ and $d\Omega_a = \sin\Theta d\Theta d\Phi$.

The celebrated Keldysh formula [9] for a strong field ionization rate is then obtained from Eqs. (1) and (2) as

$$w = 8\omega\sqrt{\frac{2I_p}{\hbar\omega}}\xi^{3/2}\exp\left[2n_0\left(\frac{\gamma\sqrt{1+\gamma^2}}{2\gamma^2+1} - \sinh^{-1}\gamma\right)\right] \times \sum_{n=\langle n_0+1\rangle}^{\infty}\exp[2\Delta n(\xi - \sinh^{-1}\gamma)]\mathcal{D}(\sqrt{2\xi\Delta n}), \quad (3)$$

where the summation over n starts with the photon integer number n_0+1, $\Delta n = n - n_0$, $\xi = \frac{\gamma}{\sqrt{1+\gamma^2}}$, $U_p = \frac{e^2E^2}{4m\omega^2}$ is the ponderomotive energy, and $n_0(E,\omega) = \frac{I_p+U_p}{\hbar\omega}$. The Dawson integral in Eq. (3) is defined as

$$\mathcal{D}(y) = \int_0^y \exp(z^2 - y^2)dz, \quad (4)$$

$$y^2 = 2\xi\Delta n. \quad (5)$$

Equation (3) is derived by a residue theorem following Ref. [9] instead of using the saddle point method, which includes all (two) poles. This gives results that are twice as large as the single pole Keldysh result [9]. Furthermore the integrand in Eq. (4) must be multiplied by $[1+(-1)^n\cos(zg_n)]$, where $g_n = 4\sqrt{2B(1+\gamma^2)(n-n_0)}$. However, the rapid oscillation term $\cos(zg_n)$ has a negligible effect on the results.

B. General ionization rate for arbitrary momentum

We have extended the Keldysh theory to arbitrary momenta, giving the more general result that is semianalytical,

$$w = \frac{m2\pi}{(2\pi\hbar^2)^2}\int_0^{\pi}\sum_{n==n_0}^{\infty}|L(\mathbf{p}_n)|^2 p_n\sin\Theta d\Theta, \quad (6)$$

where

$$|L(\mathbf{p}_n)|^2 = \left(\frac{4\hbar\omega I_p}{eE}\right)^2\frac{\pi a}{\eta}\left|\frac{e^{iS(\mathbf{p}_n,u_+)}}{\cos\omega t_+} + \frac{e^{iS(\mathbf{p}_n,u_-)}}{\cos\omega t_-}\right|^2, \quad (7)$$

with double saddle points

$$u_\pm = -\gamma\chi a_z \pm \gamma\sqrt{(\chi a_z)^2 - (1+\chi^2)}, \quad (8)$$

$$\omega t_+ = \sin^{-1}u_+, \quad (9)$$

$$\omega t_- = \pi - \sin^{-1}u_-, \quad (10)$$

where $a_z = \cos\Theta$, $\eta = 1+\chi^2\sin^2\Theta$, and the corresponding phase for each saddle point is

$$S(u_+) = \frac{\Omega}{\omega}\sin^{-1}u_+ - B(4\gamma\chi a_z + u_+)v_+, \quad (11)$$

$$S(u_-) = \frac{\Omega}{\omega}(\pi - \sin^{-1}u_-) + B(4\gamma\chi a_z + u_-)v_-, \quad (12)$$

where $\hbar\Omega = I_p + U_p + \frac{p^2}{2m}$, $B = \frac{U_p}{\hbar\omega}$, and $v_\pm = \sqrt{1-u_\pm^2}$.

The momentum p_n depends on the frequency ω and n through the parameter

$$\chi = \frac{p_n}{\sqrt{2mI_p}} = \sqrt{\frac{\hbar\omega}{I_p}(n-n_0)}, \quad (13)$$

where $n_0 = \frac{I_p+U_p}{\hbar\omega}$ is the threshold. Equation (13) follows the energy conservation from $\frac{p_n^2}{2m} = n\hbar\omega - (I_p+U_p)$ which clearly describes the photoelectric effect, with a threshold that serves as the effective work function, i.e., the sum of ionization energy I_p and the ponderomotive energy U_p. Note that the photoelectron kinetic energy (and p_n^2) is proportional to the photon frequency. The strong field regime is characterized by the Keldysh parameter $\gamma = \sqrt{\frac{I_p}{2U_p}}$. In the limit U_p becomes significant compared to I_p; i.e., the threshold becomes dependent on the laser intensity, $I = \frac{\varepsilon_0E^2}{2}$ and $\gamma = \sqrt{\frac{I_p}{2U_p}} \lesssim 1$.

Figure 1 shows that the new analytical results, Eq. (6), agree quite well with the Keldysh results at lower frequencies ω up to a quite high electric field E, according to the condition $\chi = \frac{p_n}{\sqrt{2mI_p}} \ll 1$, where $p_n^2 = 2m\hbar\omega(n-n_0)$. However, there are significant deviations for large frequency ω and large field strength E. This shows that the present high-order theory is necessary for the high frequency and high field regimes.

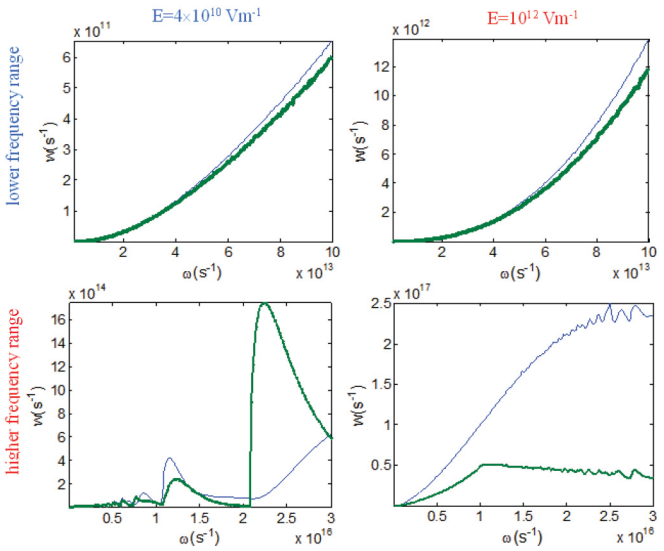

FIG. 1. (Color online) Comparison of ionization rates $w(\mathrm{s}^{-1})$ between double pole Eq. (7) (thick line) and Keldysh Eq. (3) (thin line) for linear polarization, low and high frequencies $\omega(\mathrm{s}^{-1})$. and electric fields $E(\mathrm{V\,m}^{-1})$.

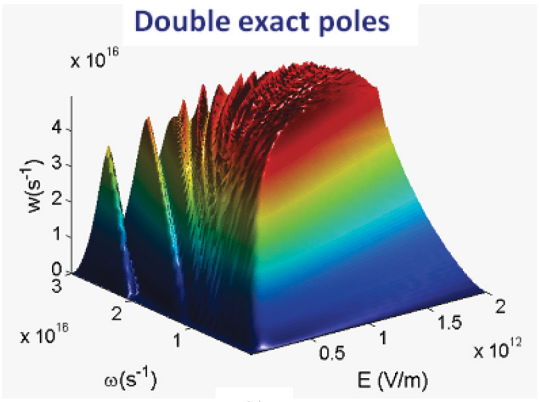

FIG. 2. (Color online) Linear polarization ionization rates w (s^{-1}) vs frequency ω (s^{-1}) and electric field E (V m^{-1}) for (a) double exact poles from Eq. (7), (b) single exact pole from Eq. (7), and (c) Keldysh Eq. (3).

Figure 2(a) reports the ionization rate for a range of frequencies ω and electric fields E. The general trend is that the rate increases with both the frequency and the electric field. At frequencies greater than about 10^{16} s^{-1} or wavelengths less than about $\lambda = 0.2$ μm, the rate shows clear oscillations [not found in the Keldysh theory, Eq. (3)] especially at small fields, E. The rates show saturation with increasing electric field and frequency. Since there are oscillations when only one pole is considered [Fig. 2(b)], therefore the oscillations in the general result Eq. (6) and Fig. 4 are NOT due to the beating or interference of the two terms $\frac{e^{iS(\mathbf{p}_n,u_+)}}{\cos\omega t_+}$ and $\frac{e^{iS(\mathbf{p}_n,u_-)}}{\cos\omega t_-}$ in Eq. (7) associated with the two *exact poles* $u_\pm$ (that include the momentum to all orders). However, the result with the two poles, Eq. (6), gives slightly different oscillations and with the rate 2 (instead of 4) times that of a single pole, a result of the incoherent sum of the two pole terms with different phases. The Keldysh result [Fig. 2(c)] shows ripples instead of oscillations, since the terms associated with the second pole have negligible effect. The Keldysh Eq. (3) predicts a rate about 50 times larger than the higher order theory. This discrepancy is due to the breakdown of the small momentum approximation used in the Keldysh theory requiring higher corrections $\frac{p}{\sqrt{2mI_p}}$.

The angular dependence of the photoionization rate obtained from Eq. (6) is shown in Fig. 3:

$$\frac{dw}{d\Omega_a} = \frac{m}{(2\pi\hbar^2)^2}\sum_{n=n_0}^{\infty}|L(\mathbf{p}_n)|^2 p_n. \tag{14}$$

The differential rate is obtained by differentiating the rate w with respect to the solid angle Ω_a.The differential rate $\frac{dw}{d\Omega_a}$ is maximum at $\Theta = 0$, parallel to the field E, which is along the z axis, and then becomes 50 times smaller at $\Theta = \pi/4$ and about 100 times smaller in the perpendicular x-y plane ($\Theta = \pi/2$). These new results describe the high directionality of the photoelectron emission by intense linear polarized light.

III. ELLIPTICAL POLARIZATION

For elliptical polarized light $\mathbf{E} = E(\alpha\cos\omega t, \beta\sin\omega t, 0) = \frac{1}{2}E[(\hat{x}\alpha + i\hat{y}\beta)e^{-iwt} + \text{c.c.}]$, where α and β determine the ellipticity $\epsilon = \alpha/\beta$, the integration over Φ, the azimuthal angle, should be included:

$$w = \frac{m}{(2\pi\hbar^2)^2}\int_0^{2\pi}\int_0^{\pi}\sum_{n=n_0'}^{\infty}|L(\mathbf{p}_n)|^2 p_n \sin\Theta d\Theta d\Phi, \tag{15}$$

where

$$L(\mathbf{p}_n) = \frac{1}{2\pi}\oint\frac{V_0(\mathbf{\Pi}_n(u))}{\sqrt{1-u^2}}e^{iS(\mathbf{p}_n,u)}du \tag{16}$$

$$= \frac{1}{2\pi}\int_{-\pi}^{\pi}V_0(\mathbf{\Pi}(s))\exp iS(\mathbf{p}_n,s)ds, \tag{17}$$

$$S(\mathbf{p},s) = ns - \frac{U_p}{\hbar\omega}\frac{(\alpha^2-\beta^2)\sin 2s}{2} - \frac{eE}{\hbar m\omega^2}[\alpha p_x\cos s + \beta p_y \sin s], \tag{18}$$

with $u = \sin s$ and $s = \omega t$. The angular momentum components $\mathbf{p} = p(\sin\Theta\cos\Phi, \sin\Theta\sin\Phi, \cos\Theta)$ define the subsequent electron momentum as it follows the field predominantly confined in the x-y plane, with $p(a_x,a_y,a_z)$ that can be found from $\frac{d\mathbf{p}}{dt} = e(\mathbf{E} + \frac{1}{m}\mathbf{p}\times\mathbf{B})$. Neglecting the magnetic Lorentz force at nonrelativistic speed defines $\frac{d\mathbf{p}}{dt} \approx e\mathbf{E}$, giving $a_z = 0$, and finite transverse components $\frac{a_y}{a_x} \approx \frac{\beta}{\alpha}\frac{u}{v}$, with $v = \sqrt{1-u^2}$. This corresponds to setting $\Theta = \pi/2$ in the unit vector of $\mathbf{p}$, i.e., $a_x = \frac{1}{\sqrt{1+(\frac{\beta u}{\alpha v})^2}}, a_y = \frac{1}{\sqrt{1+(\frac{\alpha v}{\beta u})^2}}$. We note that a further contribution to the electron dynamics may come from gradients of the ponderomotive energy U_p as discussed

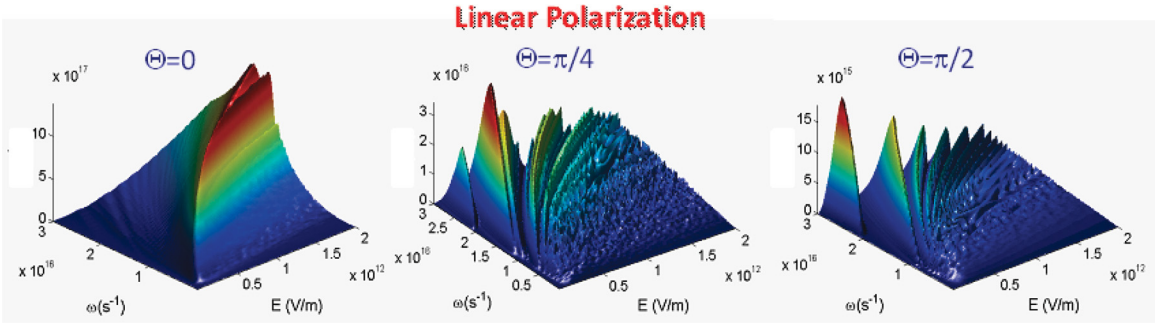

FIG. 3. (Color online) Differential ionization rate $dw/d\Omega_a$ for linearly polarized laser field, at different detection angle Θ.

in filamentation processes [26]. As shown in this work, at nonrelativistic energies and slow varying pulse envelopes, such corrections are small perturbations on electron velocities and are neglected in the present analysis. This is consistent with the dipole approximation adopted here.

The transition matrix element between an initial bound state ψ_s and a continuum Volkov state $\psi_p(\mathbf{r},t) = \exp\{\frac{i}{\hbar}[\mathbf{\Pi}(t)\cdot\mathbf{r} - \int_0^t \frac{\mathbf{\Pi}(\tau)^2}{2m}d\tau]\}$, with $\mathbf{\Pi}(t) = \mathbf{p} - e\mathbf{A}(t)$, is

$$V_0(t) = e\int \psi_s(\mathbf{r})Er\sin\theta C(t,\phi)e^{-i\Xi}r^2dr\sin\theta d\theta d\phi, \quad (19)$$

where

$$\Xi(\theta,\phi) = \frac{1}{\hbar}[Qr\sin\theta + Pr\cos\theta], \quad (20)$$

$$C(t,\phi) = \alpha\cos\omega t\cos\phi + \beta\sin\omega t\sin\phi, \quad (21)$$

$$P = p_z, \quad (22)$$

$$Q(t,\phi) = (p_x + eA_x\sin\omega t)\cos\phi + (p_y - eA_y\cos\omega t)\sin\phi. \quad (23)$$

In $Q(t,\phi)$, the radiation momenta $eA_x = \alpha\frac{eE}{\omega}$ and $eA_y = \beta\frac{eE}{\omega}$ are due to the x- and y-component fields of the elliptically polarized laser, respectively.

For a hydrogenic atom, $\psi_s(\mathbf{r}) = R(r)Y(\theta,\phi) = R(r)\Theta(\theta)\frac{1}{\sqrt{2\pi}}e^{im\phi}$. Assuming an initial $1s$ state, $\psi_s(\mathbf{r}) = \sqrt{\frac{1}{\pi a^3}}e^{-r/a}[R(r) = \sqrt{\frac{4}{a^3}}e^{-r/a}]$, we may perform the r integration and obtain a semianalytical expression:

$$V_0(\mathbf{\Pi}(t)) = \sqrt{\frac{1}{\pi a^3}}eE6a^4\int C(t,\phi)G(\phi)d\phi, \quad (24)$$

with

$$G(\phi) = \int \frac{\sin^2\theta d\theta}{\left[1 + i\frac{a}{\hbar}(Q\sin\theta + P\cos\theta)\right]^4} \quad (25)$$

$$= \frac{1}{6A^3}\left[\frac{3}{\sqrt{A}}(P'^2 - 1 - 4Q'^2)\left(i\pi + 2\tanh^{-1}\frac{Q'}{\sqrt{A}}\right) + 26Q' - \frac{4Q'^3}{P'^2 - 1}\right], \quad (26)$$

where

$$A = P'^2 + Q'^2 - 1, \quad (27)$$

$$P' = \frac{ia}{\hbar}P,\ Q' = \frac{ia}{\hbar}Q. \quad (28)$$

The above expressions have been computed numerically and are illustrated in Figs. 4 and 5 for various fields. The saddle point method $S' = 0$ gives, on the other hand,

$$\frac{\Omega}{\omega B} + (\alpha^2 - \beta^2)(2u_s^2 - 1) + 4\gamma\chi\{\alpha a_x u_s - \beta a_y\sqrt{1 - u_s^2}\} = 0. \quad (29)$$

This gives four saddle points (roots) u_s that do not have a simple analytical form. The transcendental Eq. (29) reduces to Eq. (11) for linear polarization by setting $\alpha = 1$ and $\beta = 0$, with $\hbar\Omega = I_p + U_p(\alpha^2 + \beta^2) + p^2/2m$. Each root has the corresponding derivative

$$S''(u_s) = \frac{1}{\hbar\omega\sqrt{1 - u_s^2}}\left[4U_p u_s(\alpha^2 - \beta^2) + \sqrt{\frac{4U_p}{m}}\left(\alpha p_x + \beta p_y\frac{u_s}{\sqrt{1 - u_s^2}}\right)\right]. \quad (30)$$

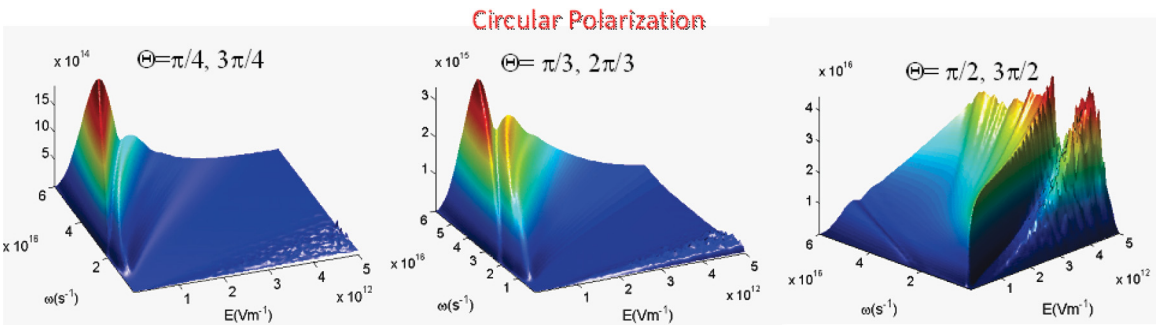

FIG. 4. (Color online) Differential ionization rate $dw/d\Omega_a$ for circularly polarized laser field; $\alpha = \beta = 1/\sqrt{2}$ at different detection angle Θ. The results are independent of azimuthal angle Φ due to symmetry of the circularly polarized laser.

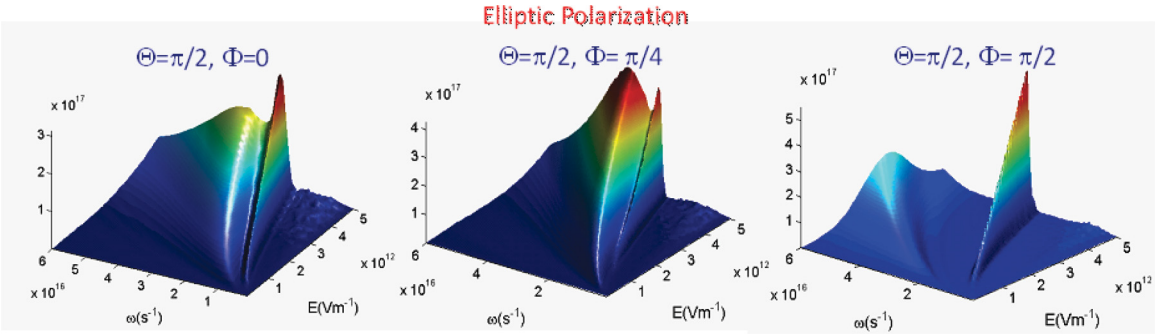

FIG. 5. (Color online) Differential ionization rate $dw/d\Omega_a$ for elliptically polarized laser field; $\alpha = 1/\sqrt{5}$ and $\beta = 2/\sqrt{5}$ at different detection directions Φ and $\Theta = \pi/2$.

For circular polarized fields, the differential photoionization rate $dw/d\Omega$ (Fig. 4) at $\Theta = \pi/2$ is about 30 times smaller than the rate for linear polarization at $\Theta = 0$. However, there are no rapid oscillations despite the fourth-order root in the saddle point formula, Eq. (29). The calculated spectra do not vary with Φ due to the circular symmetry of atoms as opposed to molecular spectra [12], where ionization rates depend on laser-molecule orientation.

Ellipticity $\epsilon = \alpha/\beta$ has significant effects on the differential ionization rate spectra and their intensity dependence (Fig. 5). There is a clear minimum threshold frequency of the laser required for photoionization for each value of the electric field that is almost independent of the azimuthal angle Φ. This feature due to the asymmetry, the term $\alpha^2 - \beta^2$ in the phase S, does not appear in the circular polarized case, since then $\alpha = \beta$ in Eq. (29), thus reducing it to a two pole problem. The electron is ejected mainly in the x-y plane (when $\Theta = \pi/2$), as expected. The rate along the y direction ($\Phi = \pi/4$) is slightly greater than that along the x direction since $\beta > \alpha$. The results are similar to a recent analysis [27] for initial nonzero angular momentum states.

IV. CONCLUSION

We have obtained general semianalytical expressions for atomic photoionization rates driven by intense linear, circular, and linearly polarized laser fields. The new result for linear polarization is different from that of the Keldysh theory in the high field and high frequency regime. Due to the occurrence of multiple poles in the transition amplitudes, we find oscillating features as the result of interference in the high frequency and high intensity regime, not only in the rate w but also in the differential rate $dw/d\Omega$, especially at larger angles Θ. Such features do not appear in the circular and elliptic polarization field cases. The differential rates for circular polarized fields are about ten times smaller than those for the spectra for elliptical polarization which show different asymmetry as a function of the azimuthal angle ϕ in the perpendicular x-y plane. This asymmetry is absent in atoms for circular polarization (Fig 4) but appears in diatomic molecules for circular polarization, due to different ionization rates parallel and perpendicular to molecular symmetry axes [28]. Thus the molecular circular polarization response mimics atomic elliptical polarization ionization.

The theory developed above can be extended to calculate higher-order terms in the perturbative formalism. For example, the second-order term in the atomic transition amplitudes gives the effects of laser-induced recollision and HHG driven by intense circular and elliptically polarized laser fields. Orientation-dependent ionization rates in molecules [20,28] offer a new challenge for developing nonperturbative analytic expressions for these highly nonlinear processes as a function of laser-molecule orientation for new applications in molecular imaging on ultrashort time scales.

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