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(Received 3 December 2001; revised manuscript received 12 June 2002; published 18 September 2002)

In this paper, we generalize and improve the derivation of photoionization rate formula for one-electron atoms proposed by Keldysh [Zh. Eksp. Teor. Fiz. **47**, 1945 (1964) [Sov. Phys. JETP **20**, 1307 (1965)]]. More exact expressions of the photoionization rate in the tunneling regime have been obtained. In addition, we extend the derivation to photoionization rates of randomly oriented diatomic molecules.

DOI: 10.1103/PhysRevA.66.033401

PACS number(s): 33.80.Rv, 32.80.Rm

I. INTRODUCTION

Recently, owing to the rapid advancement of laser technology, the interaction of intense laser fields with atoms and molecules has been the subject of many theoretical and experimental investigations. Using the current high-intensity laser technology, we can approach the regime where the laser field interaction with electron far exceeds the binding energy of the valence electrons. In this case, many interesting phenomena of excitation and photoionization can be observed. For instance, overviews of the photoionization processes in the atomic systems are well documented in Refs. [1–3]. In molecules, more intriguing phenomena take place due to the additional degrees of freedom by many-body particle interactions. In atomic and molecular systems, above threshold ionization and dissociation [4], bond softening [5], vibrational population trapping [6], and charge resonance enhanced ionization [7] have been observed and are now well established. The other interesting phenomenon induced by high-power lasers is the Coulomb explosion of molecules and clusters, which has attracted considerable attention in recent years [8–17]. To understand these phenomena, it is important to investigate the photoionization processes in more detail.

A very important theory of one-electron atom photoionization by strong lasers has been given by Keldysh [18]. For the first time, using the first-order perturbation theory, he systematically derived the photoionization formula for the direct transition between the electric ground state and the Volkov continuum state which includes oscillatory motion of the free ionizing electron in the time-dependent linearly polarized electric field. The most important finding is that the adiabaticity parameter γ (Keldysh parameter) which was introduced during his derivation determines whether the photoionization process lies in the *tunneling* or *multiphoton* region. The Keldysh parameter is the ratio of the characteristic time that the electron takes to pass through the barrier formed by the electric field and static atomic potential to the cycle time of the oscillating electric field. In the tunneling limit $\gamma \rightarrow 0$, Keldysh formula reduces to the well-known ionization rate formula in a static electric field [19]. In addition, his formulas represent the main features of the photoionization process appropriately. For example, they describe the exponential dependence of the photoionization rate on the

amplitude of the incident electric field and the threshold peculiarities at the frequencies corresponding to thresholds for absorption of n photon quanta. This phenomenon is known as the above threshold ionization process which the famous Einstein relation for the atomic photoeffect did not include.

Soon after this work, Perelomov, Popov, and Terent'ev [20] developed the photoionization rate formulas in the linearly and circularly polarized electric fields on the basis of the Green's function method (PPT theory). Their theory is also based on the adiabatic approximation $\hbar\omega \ll I_0$ (ω , laser frequency; I_0 , ionization potential), as that of Keldysh. They obtained the formulas for the one-dimensional model in the δ -function-type potential and the actual three-dimensional atoms in the short-range potentials. Their three-dimensional photoionization rate formulas are applicable for hydrogen atoms with arbitrary initial ground states of orbital angular momentum l and projection quantum number m in the direction of the electric field [Eq. (54) of Ref. [20]].

Ammosov, Delone, and Krainov [21] derived the expressions for the tunnel ionization probabilities of arbitrary complex atoms and atomic ions (ADK theory). Their theory is essentially an extension of the PPT theory. They took into account that the states of the complex atoms are characterized by effective principal and orbital quantum numbers. The theories developed by Perelomov, Popov, and Terent'ev and Ammosov, Delone, and Krainov are based on one-dimensional models which extend the work of Keldysh.

In all of these theories (Keldysh, PPT, and ADK theories), the exponents of the formulas are the same and the photoionization rates show very similar behaviors; they are different from one another only with regard to the preexponential factors.

Currently, apart from the above-mentioned theories, there appeared two important analytical nonperturbative approaches for calculating the atomic photoionization probability. Faisal [22] considered an S -matrix theory in which the initial bound state is dressed by the laser field and the final ionization state is taken to be noninteracting. It is now recognized that Keldysh [18] and Faisal [22] theories are equivalent. Later, Reiss [23] established a rigorous basis for an extended version of the Keldysh theory in which systematic higher-order corrections can be applied to the Keldysh term. Depending on whether the length or the velocity gauge is used for describing the interaction between the atom and

the incident laser field, this scheme is known as the Keldysh or the Faisal-Reiss ansatz, respectively. These are well-known as the so-called Keldysh-Faisal-Reiss theory. They are categorized as “adiabatic” theories.

It is important to note that the Keldysh theory can be only applied to the *tunneling* regime, while in the *multiphoton* region, it breaks down. This is due to the fact that the intermediate resonance states which are recognized to be very important in the strong lasers were not taken into consideration in the theory and that the adiabatic assumption $\hbar\omega \ll I_0$ in Eq. (1.1) is necessary to use saddle-point method in the derivation. Using the higher-order perturbation theory, the applicability of Keldysh approximation to *multiphoton* ionization is discussed by Trombetta, Basile, and Ferrante [24] and Mittleman and Abranyos [25]. Their conclusion is that the Keldysh approximation is hardly adequate for describing multiphoton ionization of real atoms because of the many simplifications involved, however, at the same time it is of some use in obtaining preliminary information on some aspects of the process. For the tunneling ionization, details are well documented in Ref. [26].

Experimentally, there was an observation of pure 22-photon multiphoton ionization process, in contradiction with theoretical calculations which predict that a tunneling effect should appear in the laser-intensity range adopted ($1.06 \mu\text{m}$, 10^{15}W/cm^2) [27]. This is the first multiphoton ionization experiment in which the range $\gamma < 1$ was observed. On the contrary, there was an experimental observation of tunneling ionization in the multiphoton regime [28]. These two cases show that it is necessary to investigate carefully the competition between the tunneling and multiphoton ionizations.

Recently, we have witnessed some applications and improvements of Keldysh or other tunneling theories. For example, Pazdzersky and Yurovsky, and Pozdzersky, Usachenko, and Chernov, applied the Keldysh theory to study the tunneling photoionization in a bichromatic laser field [29].

Another remarkable improvement of Keldysh theory is to ameliorate the involved wave function of the final continuum spectrum, which was not taken into account by Keldysh. That is, in a neutral atom (e.g., hydrogen atom), Coulomb potential between the remaining charged nuclei and photo-

ionizing electron must be included properly. To simultaneously include the influence of both the laser field and Coulomb potential on the freely ionizing electron, a Coulomb-Volkov-type function is proposed for the continuum spectrum and is now being investigated intensively [30–37].

In addition, there are endeavors to construct the *molecular* photoionization rate formula by extending the *atomic* version in recent years. For example, Cornaggia and Hering [38] have derived the molecular single-ionization rate formula by extending the tunneling theory developed by Perelomov, Popov, and Terent’ev for atoms [20]. In Ref. [38], the molecular electronic wave function of the ionizing electron is taken into consideration.

Another interesting research for the tunneling ionization of the molecular system is to take into account the actual shape of the potential barrier properly. Otherwise, particularly in the complicated polyatomic molecules, significant errors can be made, because the barrier width and the barrier height influence the tunneling process prominently. The Keldysh adiabaticity model was extended by replacing the model zero-range potential with *ab initio* electrostatic potential energy surfaces and it was found that the conventional Keldysh parameter for the simple zero-range potential overestimates than that for the actual nonzero-range potential [39].

In spite of many arguments about the Keldysh theory as mentioned above, it remains to be a very important theory. This is due to the fact that it is one of the theories enabling us to derive easily analytical expressions by which insightful physical interpretation can be deduced. Therefore, we believe that it is worth to reexamine the Keldysh theory in the present work.

The purpose of this paper is to improve the original Keldysh theory and extend it to diatomic molecular photoionization processes in the *tunneling* regime. In accordance with Keldysh, we use Volkov function [40] as a final continuum state. We avoid using the saddle-point method to the integration of $L(\vec{p})$ of Eq. (15) in Ref. [18]; instead, we use the residue theorem for its evaluation. The dependence of the preexponential factor on the electron momentum \vec{p} ignored in Ref. [18], which is given by the following expression:

$$2\sqrt{\pi a_0^3} \frac{I_0}{eFa_0} \frac{\hbar\omega}{(1-u_s^2)^{1/2}} \exp\left[\frac{i}{\hbar\omega} \int_0^{u_s} \left\{ I_0 + \frac{1}{2m} \left(\vec{p} + \frac{e\vec{F}}{\omega} \nu \right)^2 \right\} \frac{d\nu}{(1-\nu^2)^{1/2}} \right], \quad (1.1)$$

will be incorporated in this work. It will be shown that such modifications allow us to obtain more exact expressions for the photoionization rate and to treat analytically the photoionization phenomenon for the molecular system.

It is important to note that we will not focus on *multiphoton* ionization in the present work. Our aim is to derive more exact expressions of photoionization rate, which can be applied efficiently in the *tunneling* regime.

The present paper is organized as follows. In Sec. II, fol-

lowing Ref. [18], we derive photoionization rate formulas for hydrogenlike one-electron atoms. Our derivation is different from that of Ref. [18] in the respect mentioned above. Our formulas work quite well in the tunneling ionization regime, which was assumed in the derivation of Ref. [18] and confirmed experimentally as well [41]. In Sec. III, based on the development made in Sec. II, we extend the derivation to the simple molecular systems: randomly oriented diatomic molecules. As an example, the photoionization formula for N_2

molecules is reported. In Sec. IV, we compare Keldysh's or other photoionization rates with our results numerically and discuss the validity of our formulas. For the atomic case, we focus on the $1s$ state of hydrogen atom as the initial state. For the molecular case, our attention is directed to the photoionization pathway to remove $\sigma_g 2p$ electron of N_2 molecule and to produce N_2^+ in its ground electronic state $X^2\Sigma_g^+$ [42–44]. Section V is devoted to the concluding remarks.

II. THEORY: ATOMIC CASE

A starting point of the Keldysh theory is to evaluate the rate of photoionization w_0 for direct transition from the atomic ground bound state to the continuum spectrum for one-electron atoms,

$$w_0 = \int \frac{d^3p}{(2\pi\hbar)^3} \frac{d}{dT} |c_{\vec{p}}(T)|^2 \Big|_{T \rightarrow \infty}, \quad (2.1)$$

where \vec{p} denotes the momentum of the freely ionizing electron and

$$c_{\vec{p}}(T) = \frac{i}{\hbar} \int_0^T dt \cos(\omega t) e^{-(i/\hbar)E_g t} \langle \psi_{\vec{p}}(\vec{r}, t) | \vec{d} \cdot \vec{F} | \psi_g(\vec{r}) \rangle \quad (2.2)$$

in the dipole approximation. Here, \vec{F} is the amplitude maximum of the incident linearly polarized electric field. The wave function $\psi_g(\vec{r})$ represents the initial electron ground state and $\psi_{\vec{p}}(\vec{r}, t)$, the final continuum state. We shall choose the $1s$ state of hydrogenlike atom for $\psi_g(\vec{r})$,

$$\psi_g(\vec{r}) = \sqrt{\frac{1}{\pi a^3}} \exp\left(-\frac{r}{a}\right), \quad a = \frac{a_0}{Z}, \quad (2.3)$$

and the Volkov function for $\psi_{\vec{p}}(\vec{r}, t)$,

$$\psi_{\vec{p}}(\vec{r}, t) = \exp\left\{ \frac{i}{\hbar} \left[[\vec{p} - e\vec{A}(t)] \cdot \vec{r} - \frac{1}{2m} \int_0^t dt' [\vec{p} - e\vec{A}(t')]^2 \right] \right\}, \quad (2.4)$$

where $\vec{A}(t) = -(\vec{F}/\omega)\sin(\omega t)$ and Z represents the effective nuclear charge.

Substituting Eq. (2.2) into Eq. (2.1) yields

$$w_0 = \frac{2}{\hbar^2} \lim_{T \rightarrow \infty} \text{Re} \int \frac{d^3p}{(2\pi\hbar)^3} \int_0^T dt \cos(\omega T) \cos(\omega t) V_0^* \left(\vec{p} + \frac{e\vec{F}}{\omega} \sin(\omega T) \right) V_0 \left(\vec{p} + \frac{e\vec{F}}{\omega} \sin(\omega t) \right) \times \exp \left[\frac{i}{\hbar} \int_T^t d\tau \left\{ I_0 + \frac{1}{2m} \left(\vec{p} + \frac{e\vec{F}}{\omega} \sin(\omega\tau) \right)^2 \right\} \right], \quad (2.5)$$

where $I_0 = -E_g = Z^2 e^2 / 2a_0$ (ionization potential) and

$$V_0(\vec{p}) = 8i(\pi a^3)^{1/2} e\hbar \vec{F} \cdot \vec{\nabla}_{\vec{p}} (1 + p^2 a^2 / \hbar^2)^{-2} \quad (2.6)$$

for $1s$ level of hydrogenlike atoms.

Carrying out the integration with respect to t and taking the infinity of T in Eq. (2.5) yield

$$w_0 = \frac{2\pi}{\hbar} \int \frac{d^3p}{(2\pi\hbar)^3} |L(\vec{p})|^2 \sum_{n=-\infty}^{\infty} \delta \left(\tilde{I}_0 + \frac{p^2}{2m} - n\hbar\omega \right), \quad (2.7)$$

where

$$L(\vec{p}) = -\frac{16ieI_0^3 \sqrt{\pi a^7}}{\pi\hbar} \oint du \frac{\vec{F} \cdot \left(\vec{p} + \frac{e\vec{F}}{\omega} u \right)}{\left\{ I_0 + \frac{1}{2m} \left(\vec{p} + \frac{e\vec{F}}{\omega} u \right)^2 \right\}^3} \times \exp \left[\frac{i}{\hbar\omega} \int_0^u \frac{d\nu}{\sqrt{1-\nu^2}} \left\{ I_0 + \frac{1}{2m} \left(\vec{p} + \frac{e\vec{F}}{\omega} \nu \right)^2 \right\} \right] \quad (2.8)$$

and $\tilde{I}_0 = I_0 + (e^2 F^2 / 4m\omega^2)$.

The singularity points u_s of the integrand of $L(\vec{p})$ can be determined by the following condition:

$$I_0 + \frac{1}{2m} \left(\vec{p} + \frac{e\vec{F}}{\omega} u_s \right)^2 = 0. \quad (2.9)$$

In order to evaluate $L(\vec{p})$, we let

$$j(u) = \int_0^u \frac{dv}{\sqrt{1-v^2}} \left\{ I_0 + \frac{1}{2m} \left(\vec{p} + \frac{e\vec{F}}{\omega} v \right)^2 \right\} \quad (2.10)$$

and expand the exponent and the denominator of the integrand of Eq. (2.8), which leads to

$$L(\vec{p}) = -\frac{16ieI_0^3\sqrt{\pi a^7}}{\pi\hbar} \oint du \frac{\vec{F} \cdot \left(\vec{p} + \frac{e\vec{F}}{\omega} u \right)}{j''(u_s)^3 (u-u_s)^3 (1-u^2)^{3/2}} \times \exp \left[\frac{i}{\hbar\omega} \left\{ j(u_s) + \frac{1}{2} j''(u_s)(u-u_s)^2 + \dots \right\} \right]. \quad (2.11)$$

Carrying out the contour integral in Eq. (2.11) using the residue theorem yields

$$L(\vec{p}) = \frac{16ieI_0^3\sqrt{\pi a^7} \vec{F} \cdot \left(\vec{p} + \frac{e\vec{F}}{\omega} u_s \right)}{\hbar^2 \omega j''(u_s)^2 (1-u_s^2)^{3/2}} \exp \left[\frac{i}{\hbar\omega} j(u_s) \right], \quad (2.12)$$

where

$$j''(u_s) = \frac{e}{m\omega\sqrt{1-u_s^2}} \vec{F} \cdot \left(\vec{p} + \frac{e\vec{F}}{\omega} u_s \right) \quad (2.13)$$

and

$$j(u_s) = \left(\tilde{I}_0 + \frac{p^2}{2m} \right) \sin^{-1} u_s - \frac{e^2 F^2}{4m\omega^2} u_s \sqrt{1-u_s^2} - \frac{e\vec{F} \cdot \vec{p}}{m\omega} (\sqrt{1-u_s^2} - 1). \quad (2.14)$$

Notice that the denominator of the integrand of Eq. (2.11), $(1-u^2)^{3/2}$, is approximated by $(1+\gamma^2)^{3/2}$ due to the assumption of small Keldysh parameter γ and low kinetic momentum p (cf. Appendix A).

A remarkable advantage of our method is that the singular point of the integrand and the zero point of $j'(u)$ do not necessarily have to coincide in Eq. (2.11) [18,45]. As a result, extension can be easily made when the Volkov function is replaced by Coulomb-Volkov function in which case the saddle-point is different from the zero point of $j'(u)$ [46].

The quantity $L(\vec{p})$ can be written as

$$L(\vec{p}) = \frac{4\hbar\omega I_0\sqrt{\pi a}}{eF \cos \theta_{pF} \sqrt{1-u_s^2}} \exp \left[\frac{i}{\hbar\omega} j(u_s) \right], \quad (2.15)$$

where $\cos \theta_{pF}$ is defined by

$$\vec{F} \cdot \left(\vec{p} + \frac{e\vec{F}}{\omega} u_s \right) = F \left| \vec{p} + \frac{e\vec{F}}{\omega} u_s \right| \cos \theta_{pF}. \quad (2.16)$$

Notice that at this point, our expression for $L(\vec{p})$ [Eq. (2.15)] is larger than that of Keldysh [Eq. (1.1)] by a factor of two, which arises from the contour integration made for evaluating Eq. (2.11), different from the saddle-point method utilized in Ref. [18].

In Ref. [18], dependence of $\cos \theta_{pF}$ and $\sqrt{1-u_s^2}$ on the electron momentum \vec{p} in Eq. (2.15) has been ignored, that is, $\cos \theta_{pF}=1$ and $\sqrt{1-u_s^2}=\sqrt{1+\gamma^2}$ were assumed. In the present paper, we shall take into account the \vec{p} dependence of these preexponential factors and examine what role they will play as well. For that purpose, we reduce $L(\vec{p})$ defined by Eq. (2.15) into the form being easily integrated with respect to \vec{p} in Eq. (2.7). The derivation is shown in Appendix A.

Substituting Eq. (A10) into Eq. (2.7) and integration with respect to \vec{p} leads to

$$w_0 = 4 \sqrt{\frac{2I_0\omega}{\hbar}} \left(\frac{\gamma}{\sqrt{1+\gamma^2}} \right)^{3/2} N(\gamma, \omega, I_0, \tilde{I}_0, B, C) \times \exp \left[-\frac{2\tilde{I}_0}{\hbar\omega} \left(\sinh^{-1} \gamma - \frac{\gamma\sqrt{1+\gamma^2}}{1+2\gamma^2} \right) \right]. \quad (2.17)$$

Equation (2.17) is an atomic photoionization formula including the \vec{p} dependence of the preexponential factors in Eq. (2.15).

The preexponential factor of Eq. (2.17) is slightly complicated due to the \vec{p} dependence of Eq. (2.15). It is essential to verify the effect of the preexponential factors on the photoionization rate. The preexponential factor $N(\gamma, \omega, I_0, \tilde{I}_0)$ in Eq. (2.17) and the photoionization rate formulas derived on the basis of different treatments for the preexponential factors are given in Appendix B.

III. DIATOMIC MOLECULAR CASE

In this section, based on the approach introduced in the preceding section, we show that the *molecular* photoionization rates can be derived analytically as well.

In general, as is clear from the derivation shown in the following, one can arbitrarily choose any molecular systems and their initial electronic bound states. In the present paper, we focus on the N_2 molecules as an example. Particularly, we consider the case of the molecular photoionization from the $\sigma 2p_z$ orbital of the N_2 molecules. In this case, the initial molecular state ψ_g is given by

$$\psi_g = N_{2p} [(2p_z)_1 + (2p_z)_2], \quad (3.1)$$

where $(2p_z)_1$ and $(2p_z)_2$ represent the atomic orbitals, and N_{2p} denotes the normalization constant. Here for simplicity of estimation, we use the simple molecular-orbital theory, that is, linear combination of atomic orbitals–molecular orbital method.

For the case of randomly oriented molecules, the photoionization rate w_0 is given by

$$w_0 = \frac{1}{3} \frac{2}{\hbar^2} \lim_{T \rightarrow \infty} \operatorname{Re} \int \frac{d^3 p}{(2\pi\hbar)^3} \int_0^T dt \cos(\omega T) \cos(\omega t) \vec{V}_0^* \left(\vec{p} + \frac{e\vec{F}}{\omega} \sin(\omega T) \right) \cdot \vec{V}_0 \left(\vec{p} + \frac{e\vec{F}}{\omega} \sin(\omega t) \right) \times \exp \left[\frac{i}{\hbar} \int_T^t d\tau \left\{ I_0 + \frac{1}{2m} \left(\vec{p} + \frac{e\vec{F}}{\omega} \sin(\omega\tau) \right)^2 \right\} \right], \quad (3.2)$$

where for the N_2 molecules

$$\vec{V}_0(\vec{p}) = \sqrt{2} e F \langle \exp(i\vec{p} \cdot \vec{r}/\hbar) | \vec{r} | N_{2p} [(2p_z)_1 + (2p_z)_2] \rangle, \quad (3.3)$$

using Eq. (3.1). It is worth noting that in the atomic case, $V_0(\vec{p})$ was a scalar as indicated by Eq. (2.6), while in the molecular case, its counterpart, $\vec{V}_0(\vec{p})$, is a vector owing to the directional nature of the molecular bond. The factor $1/3$ in Eq. (3.2) is introduced for the orientational average of the photoionization rate.

In the one-center approximation [47], Eq. (3.3) reduces to

$$\vec{V}_0(\vec{p}) = 2eF \langle \exp(i\vec{p} \cdot \vec{r}/\hbar) | \vec{r} | \psi_{2p_z} \rangle, \quad (3.4)$$

where

$$\psi_{2p_z} = \sqrt{\frac{1}{\pi a_2^5}} z \exp\left(-\frac{r}{a_2}\right). \quad (3.5)$$

Using the molecular-fixed coordinates, Eq. (3.4) leads to

$$\vec{V}_0(\vec{p}) = (2ie\hbar F) \vec{\nabla}_p \int d\vec{r} \exp\left(-\frac{i}{\hbar} \vec{p} \cdot \vec{r}\right) \psi_{2p_z}, \quad (3.6)$$

which after some algebra renders

$$\vec{L}(\vec{p}) = -\frac{2^5 \pi^{1/2} a_2^{5/2} I_0^3 e F}{\hbar \omega} \exp\left[\frac{i}{\hbar \omega} j(u_s)\right] \left[\frac{\hat{k}}{j''(u_s)^2 (1-u_s^2)^{3/2}} - \frac{3eF}{m \omega j''(u_s)^3 (1-u_s^2)^2} \left\{ \left(\vec{p} + \frac{e\vec{F}}{\omega} u_s \right) + \left(p_z + \frac{eF}{\omega} u_s \right) \hat{k} \right\} - \frac{\left(p_z + \frac{eF}{\omega} u_s \right) \left(\vec{p} + \frac{e\vec{F}}{\omega} u_s \right)}{m j''(u_s)^4 (1-u_s^2)^2} j'''(u_s) \right], \quad (3.10)$$

where

$$j''(u_s) = \frac{i\sqrt{2mI_0}}{\sqrt{1-u_s^2}} \frac{eF}{m\omega} \cos \theta_{pF}, \quad (3.11)$$

$$j'''(u_s) = \frac{e^2 F^2}{m\omega^2} \frac{1}{\sqrt{1-u_s^2}} + \frac{2i\sqrt{2mI_0} u_s}{(1-u_s^2)^{3/2}} \frac{eF}{m\omega} \cos \theta_{pF}. \quad (3.12)$$

$$\vec{V}_0(\vec{p}) = 64\pi e F a_2^5 N_{2p} \left[\frac{\hat{k}}{(1+k^2 a_2^2)^3} - \frac{6a_2^2 p_z \vec{p}}{\hbar^2 (1+k^2 a_2^2)^4} \right]. \quad (3.7)$$

Here, \hat{k} denotes the unit vector along the z direction, i.e., the molecular axis.

Repeating the derivation shown in the preceding section, we obtain

$$w_0 = \frac{2\pi}{3\hbar} \int \frac{d^3 p}{(2\pi\hbar)^3} |\vec{L}(\vec{p})|^2 \sum_{n=-\infty}^{\infty} \delta\left(\tilde{I}_0 + \frac{p^2}{2m} - n\hbar\omega\right), \quad (3.8)$$

where

$$\vec{L}(\vec{p}) = \frac{1}{2\pi} \oint du \vec{V}_0 \left(\vec{p} + \frac{e\vec{F}}{\omega} u \right) \times \exp \left[\frac{i}{\hbar \omega} \int_0^u \frac{d\nu}{\sqrt{1-\nu^2}} \left\{ I_0 + \frac{1}{2m} \left(\vec{p} + \frac{e\vec{F}}{\omega} \nu \right)^2 \right\} \right]. \quad (3.9)$$

Notice that the relation of $\hbar^2/a_2^2 = 2mI_0$ still holds for the molecular system as in the case for hydrogenlike atoms. After the contour integration as was done in Sec. II, we obtain

Using the same procedure adopted in Sec. II, the photoionization rate for the system of randomly oriented molecules is finally given by

$$w_0 = \frac{2^6}{3} \sqrt{\frac{2I_0 \omega}{\hbar}} \frac{\gamma^{3/2} (2\gamma^2 + 3)^2}{(1+\gamma^2)^{5/2}} N(\gamma, \omega, I_0, \tilde{I}_0, 1, 0) \times \exp \left[-\frac{2\tilde{I}_0}{\hbar \omega} \left(\sinh^{-1} \gamma - \frac{\gamma \sqrt{1+\gamma^2}}{1+2\gamma^2} \right) \right]. \quad (3.13)$$

In Eq. (3.13), the preexponential factor $N(\gamma, \omega, I_0, \tilde{I}_0, 1, 0)$ is given by Eq. (B1). Note that in Eq. (3.13) the \tilde{p} dependence of the preexponential factors is ignored.

It should be noted that the exponential factor for the randomly oriented molecular case [Eq. (3.13)] is exactly the same as that for the atomic case [Eq. (2.17)]. This indicates that ionization mechanisms of atomic and randomly oriented molecular systems are essentially the same because the ionization rate is essentially determined by the exponential factors. The difference between atomic and molecular systems arises only from the preexponential factors. In addition, it is important to note that in the case of the molecules, the Keldysh parameter γ can be defined in the same way as in the atomic system, Eq. (2.17).

It should be noted that in the tunneling limit ($\gamma \rightarrow 0$), comparing Eq. (3.13) for the molecular system and Eq. (2.17) with $B=1$ and $C=0$, we find that the photoionization rate is 48 times larger for the molecular system than for the atomic system. This is due to the different orbitals for the initial state and to the directional nature of the molecular bond.

The photoionization rates from σ_{2s} and π_{2p} states for the N_2 molecules can be evaluated in a similar manner and will not be presented in the present paper. In addition, application of the present method to other diatomic molecules is straightforward.

IV. RESULTS AND DISCUSSIONS

In this section, we demonstrate numerical results by using our analytical formulas [Eq. (2.17) for the atomic system and Eq. (3.13) for the molecular system] and compare them with those calculated by the Keldysh theory or numerical results of other authors.

In Fig. 1, we show the atomic ionization rate w_0 calculated by the Keldysh theory [Eq. (16) of Ref. [18]] and our theory [Eq. (2.17)] for a hydrogen atom with $Z=1$. For the estimation by the Keldysh theory, we have checked that sufficient convergence has been attained in the summation over n [Eq. (B1)]. Notice that our results for w_0 agree very well with that of Keldysh, although the latter underestimates than the former by a few factors. If we include whole preexponential factors, w_0 becomes smallest [solid line in Fig. 1(b)]. According to Ref. [41] the widely used Reiss and Keldysh methods underestimate the experimentally observed photoionization rates to a large extent. Therefore, Fig. 1 indicates that our formulas ameliorate the Keldysh's original ones.

In the range of tunneling limit ($\gamma \ll 1$), the photoionization rates w_0 obtained by the different approaches can be approximated as

$$w_0 = \frac{\sqrt{6\pi}}{4} \frac{I_0}{\hbar} \left(\frac{eF\hbar}{m^{1/2}I_0^{3/2}} \right)^{1/2} \exp\left\{ -\frac{4\sqrt{2m}I_0^{3/2}}{3e\hbar F} \right\} \times \left(1 - \frac{\gamma^2}{10} \right) \quad (\text{Keldysh}),$$

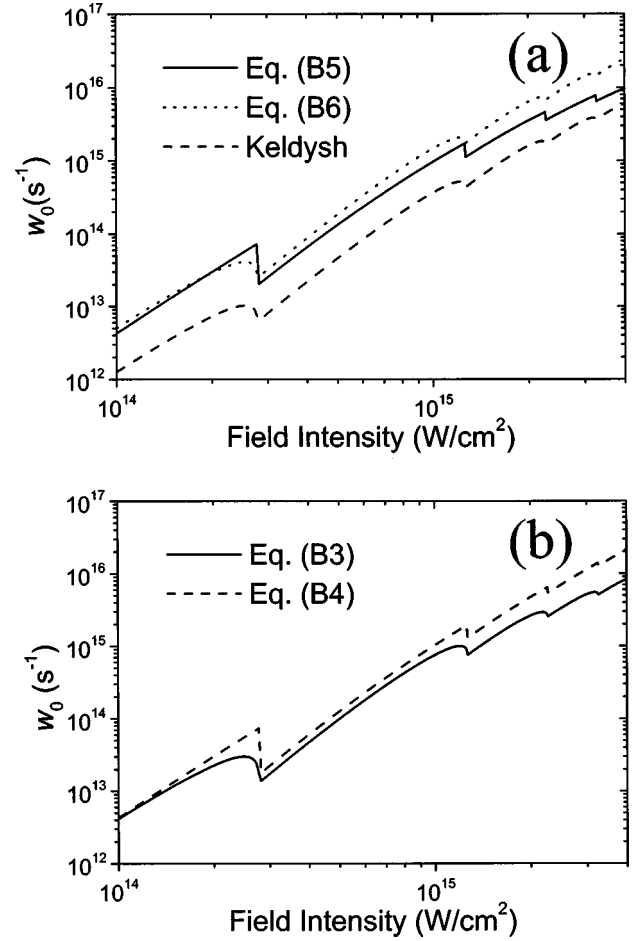


FIG. 1. Log-log plot of the calculated atomic photoionization rate w_0 . The calculation is performed using the Keldysh theory [Eq. (16) in Ref. [18]] and our theory [Eq. (2.17)]. The wavelength of the incident light is 248.0 nm [$\hbar\omega=5.0$ eV]. In order to ensure the convergence, the summation upper limit of Eq. (B1) was 500 for the higher intensity region.

$$w_0 = \frac{\sqrt{4\sqrt{2}\pi}}{3} \frac{I_0}{\hbar} \left(\frac{eF\hbar}{m^{1/2}I_0^{3/2}} \right)^{1/2} \exp\left\{ -\frac{4\sqrt{2m}I_0^{3/2}}{3e\hbar F} \right\} \times \left(1 - \frac{\gamma^2}{10} \right) \quad [\text{for the case Eq. (B5)}],$$

and

$$w_0 = \sqrt{3\sqrt{2}\pi} \frac{I_0}{\hbar} \left(\frac{eF\hbar}{m^{1/2}I_0^{3/2}} \right)^{1/2} \exp\left\{ -\frac{4\sqrt{2m}I_0^{3/2}}{3e\hbar F} \right\} \times \left(1 - \frac{\gamma^2}{10} \right) \quad [\text{for the case Eq. (B6)}.] \quad (4.1)$$

In Eq. (4.1), $\gamma \approx \hbar\omega/I_0$ is assumed, except for the Keldysh's tunneling formula. We notice that only the preexponential factors are different from each other and we can predict the magnitude of the difference of the preexponential constants. For instance, near the tunneling limit at the field intensity 4×10^{15} W/cm², the photoionization rate w_0 calculated by

Keldysh's formula [broken line in Fig. 1(a)] is the smallest and that estimated by Eq. (B5) [solid line in Fig. 1(a)] is smaller than that calculated by Eq. (B6) [dotted line in Fig. 1(a)]. This is consistent with Eq. (4.1).

From Fig. 1, we also notice that $d \log_{10}(w_0)/d \log_{10}(I)$ (I , laser intensity) is almost constant in the tunneling region, which is evident from Eq. (4.1). This indicates that the ionization potential I_0 or the effective nuclear charge Z can be estimated by measuring the slope $d \log_{10}(w_0)/d \log_{10}(I)$ using Eq. (4.1).

Now, let us compare our results and that calculated numerically based on the time-dependent Schrödinger equation by LaGattuta [48]. Our results including preexponential factors are quite similar to that obtained by LaGattuta, especially in the high-intensity range (around 1×10^{15} W/cm² or higher). In the low-intensity regime, large discrepancy is found. This tendency has already been pointed out by LaGattuta. This can be attributed to the fact that Keldysh and our theories cannot be applied to the multiphoton regime.

Figure 2 presents dependence of the atomic ionization rate w_0 on the effective charge Z . In the low field intensity range, Keldysh's and our results agree very well. However, in the high field intensity range, large discrepancy is found. Figure 2 shows that the ionization rate varies significantly with ionization energy. The lower the ionization energy is, the larger the ionization rate is. In other words, using Fig. 2, we can estimate the ionization rate as a function of laser intensity and ionization potential.

Next, we consider the molecular photoionization rates. Apart from the atomic photoionization, detailed studies of molecular photoionization will provide additional insights into the dynamics of photoionization processes in general [49,50]. Therefore, it is important to explore the molecular photoionization in more detail.

It is known that the Keldysh theory can also predict diatomic molecular photoionization rate to a certain extent. However, in order to estimate accurately diatomic or polyatomic molecular photoionization rates, it will be necessary to describe molecular states by including the directional nature of the molecular bond which is the most distinct characteristic from atoms. Along this line, DeWitt and Levis [51] conducted a time-of-flight mass spectrum experiment for benzene, naphthalene, and anthracene, and calculated their photoionization and dissociation probabilities in intense laser fields. From the theoretical or experimental interest, there are several investigations concerning photoionization rates or cross sections of N₂ molecule in recent years [52–55]. In connection with these works, we shall compare numerical results estimated by Eq. (3.13) and show its validity, which include the effect of the molecular bond to the photoionization rate.

The molecular ionization rates w_0 of N₂ molecule calculated by our theory [Eq. (3.13)] for various ionization potentials I_0 are demonstrated in Fig. 3. From He I photoelectron spectrum of N₂, it is known that the minimum energy necessary to remove $\sigma_g 2p$ electron to produce N₂⁺ in its ground electronic state $X^2\Sigma_g^+$ is 15.58 eV [56]; $I_0 = 15.58$ eV.

Let us compare our result with those calculated by Guo *et al.* by use of Ammosov-Delone-Krainov (ADK) model

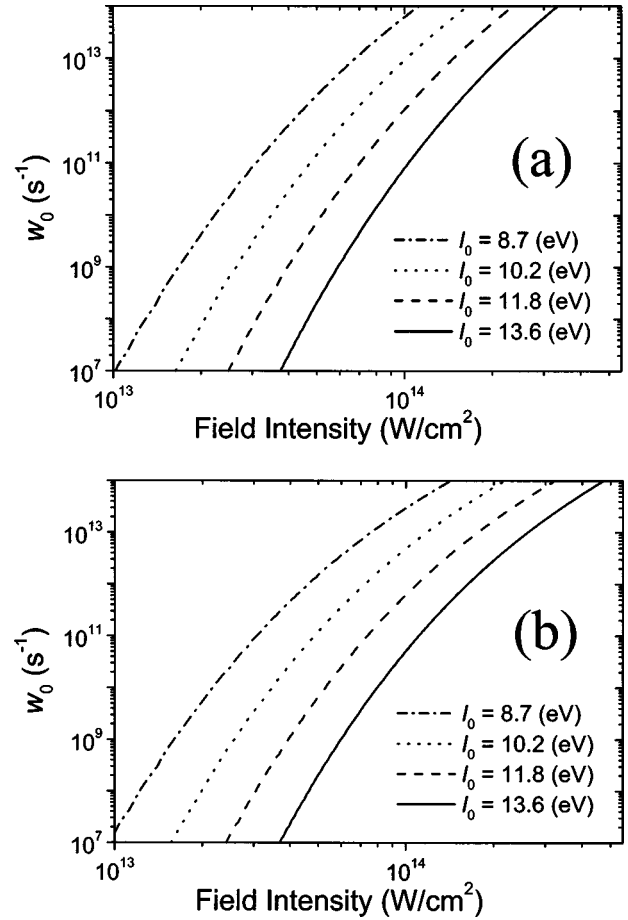


FIG. 2. Log-log plot of the dependence of the atomic ionization rate w_0 on the effective nuclear charge Z . The wavelength of the incident laser is 3000.0 nm (laser frequency $\omega = 0.413$ eV). The ionization potentials I_0 are 8.7, 10.2, 11.8, and 13.6 eV for $Z = 0.80, 0.87, 0.93,$ and 1.0 , respectively. Panel (a) was calculated by the Keldysh theory [Eq. (16) in Ref. [18]] and (b), by Eq. (2.17) with Eq. (B3). In order to ensure the convergence, in the highest intensity region, the upper limits of summation with regard to n in Eq. (B1) and the counterpart of Keldysh [Eq. (16) in Ref. [18]] were $n = 5 \times 10^4, 1 \times 10^5, 6 \times 10^6,$ and 7×10^6 for $I_0 = 8.7, 10.2, 11.8,$ and 13.6 eV, respectively.

[21] (Fig. 3 of Ref. [53]) and those estimated by DeWitt and Levis by use of orientationally averaged tunneling theory appropriate for diatomic molecules (Fig. 4 of Ref. [55]). In the whole range of the figure, our prediction agrees very well with that of ADK [53] and orientationally averaged tunneling theory [55]. Lower than the laser intensity 10^{13} W/cm², a minor discrepancy is found between our result and that of orientationally averaged tunneling theory [55]. As mentioned in Fig. 2, we can estimate the ionization rate as a function of laser intensity and ionization potential, which is demonstrated in Fig. 3 with various ionization potentials I_0 .

From Figs. 2 and 3, we can see that the photoionization rate increases very rapidly with decreasing ionization potential in the whole range of the laser intensity. This is due to the fact that under the condition of the same laser intensity, the potential barrier height and width are smaller for the system with smaller ionization potential. This suggests that

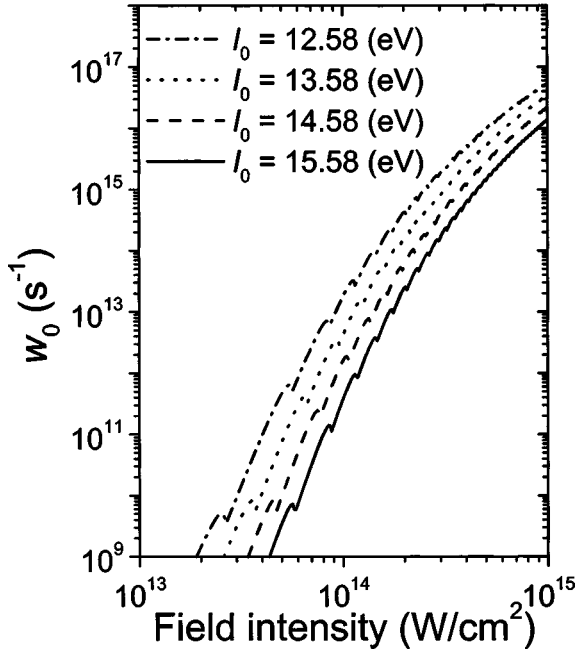


FIG. 3. Log-log plot of the calculated molecular photoionization rate w_0 . The calculation is performed using our theory [Eq. (3.13)] for $I_0 = 12.58, 13.58, 14.58,$ and 15.58 eV or $Z = 0.80, 0.85, 0.92,$ and 1.00 , respectively. The wavelength of the incident laser is 800.0 nm (laser frequency $\omega = 1.55$ eV). The solid line in the figure is compared with those calculated by Guo *et al.* [53] and by DeWitt *et al.* [55].

there will be a case where the indirect tunneling ionization process through an excited intermediate state (resonant tunneling ionization) may be more effective than (or at least comparable to) the direct tunneling ionization. In Fig. 4, we investigate the laser intensity and frequency regions where this is the case.

For estimation, we shall consider the indirect tunneling ionization from the ground g state to the ionization continuum via the intermediate state e . In this case, the indirect tunneling ionization rate w_{si} can be estimated from

$$w_{si} = \frac{W_{g \rightarrow e}(\omega)}{W_{g \rightarrow e}(\omega) + \gamma_e + w_e} w_e, \quad (4.2)$$

where w_e , $W_{g \rightarrow e}(\omega)$, and γ_e denote the tunneling ionization rate for the e state, optical absorption rate for $g \rightarrow e$, and the relaxation decay rate of the e state, respectively. Notice that

$$W_{g \rightarrow e}(\omega) = \frac{1}{2\hbar^2} \frac{\gamma_{eg} |\vec{d}_{eg} \cdot \vec{F}|^2}{\left(\omega_{eg} + \frac{|\vec{d}_{ee} - \vec{d}_{gg}| \cdot \vec{F}|^2}{4\hbar^2 \omega_{eg}} - \omega \right)^2 + \gamma_{eg}^2}, \quad (4.3)$$

where γ_{eg} represents the dephasing constant, and \vec{d}_{ee} and \vec{d}_{gg} denote the permanent dipole moments.

Figure 4 demonstrates laser intensity dependence of $W_{g \rightarrow e}(\omega)$, w_e , w_{si} , and direct tunneling ionization rate w_0 for the fixed photon frequency ω resonant with the energy gap ω_{eg} [1.0 eV for Fig. 4(a)]. Here for simplicity, we as-

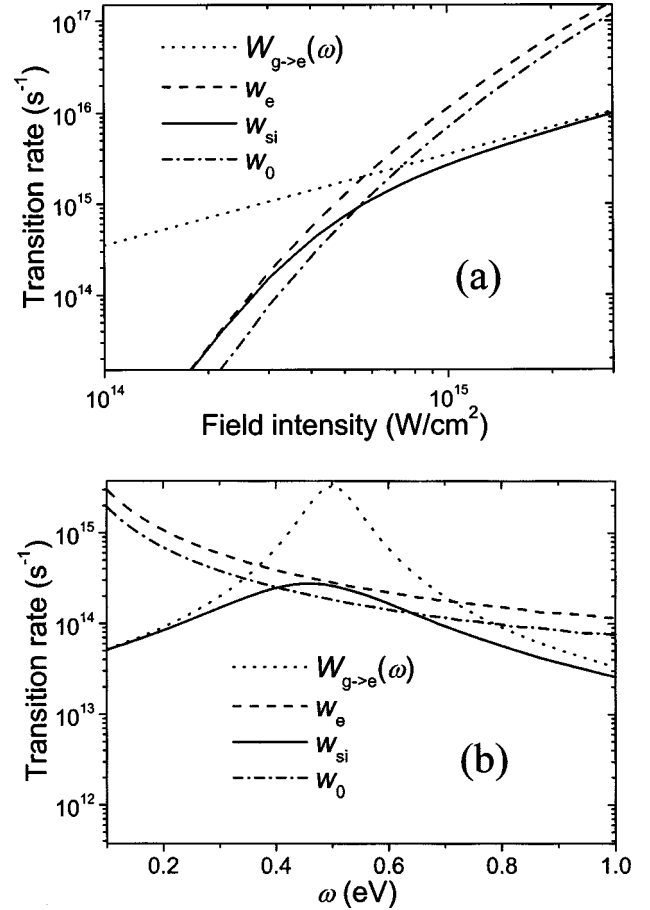


FIG. 4. Optical absorption rate $W_{g \rightarrow e}(\omega)$, ionization rate of the intermediate e state w_e , indirect photoionization rate w_{si} , and direct photoionization rate w_0 . The calculation is performed using Eqs. (4.2) and (4.3). The energy gaps ω_{eg} in Eq. (4.3) are 1.0 eV and 0.5 eV for (a) and (b), respectively. The laser intensity is 3×10^{14} W/cm² for (b). The ionization potential from the ground state to the ionization threshold is 13.6 eV. The indirect ionization rate w_e in Eq. (4.2) is estimated by using Eq. (2.17) with Eq. (B6). The magnitudes of the transition dipole moment \vec{d}_{eg} are set to be 1.0 a.u. and 0.2 a.u. for (a) and (b), respectively. The direction of the transition dipole moment is assumed to be along the laser polarization. The dephasing constants of the e state, γ_{eg} , are 4 times and 0.1 times of ω_{eg} for (a) and (b), respectively. The relaxation decay rate of the e state corresponds to 100 ps. (a) shows laser intensity dependence of the transition rates for the fixed photon frequency ω resonant with the energy gap ω_{eg} (1.0 eV). (b) shows laser frequency dependence of the transition rates for the fixed laser intensity (3×10^{14} W/cm²).

sume that the permanent dipole moments can be neglected: $\vec{d}_{ee} = \vec{d}_{gg} = \vec{0}$. In addition, we have used Eq. (B6) in order to calculate w_e and w_0 : we assume the tunneling ionization process of hydrogen atoms.

Now, let us concentrate on Fig. 4(a). From Eq. (4.2), it is expected that for the first absorption resonance, w_{si} would be negligibly small until a certain intensity of laser power is reached so that w_e and $W_{g \rightarrow e}(\omega)$ are comparable (at the field intensity 5×10^{14} W/cm²). In the case of Fig. 4(a), $W_{g \rightarrow e}(\omega)$

is large due to the resonance transition between g and e states, while w_e is much smaller due to the small tunneling probability in the low laser intensity range ($< 5 \times 10^{14}$ W/cm²) and slightly larger than w_0 due to the larger tunneling probability. Therefore, w_{si} and w_e are comparable in the low-intensity regime. Beyond this limit (around 5×10^{14} W/cm²), w_{si} behaves like $W_{g \rightarrow e}(\omega)$ which is much smaller than w_e and w_0 ; the indirect tunneling ionization is less effective than the direct tunneling ionization. In this high-intensity range, the potential barrier is highly transparent, which leads to indistinguishably high ionization probability for the g and e states (note that the energy gap ω_{eg} is quite small). Thus, we have found that the indirect tunneling ionization is more effective than the direct tunneling ionization in the low laser intensity region.

Figure 4(b) demonstrates that if the photon frequency ω is in resonance with the intermediate bound state (0.5 eV), $W_{g \rightarrow e}(\omega)$ is quite large, and w_e and w_0 are smaller due to the fact that the potential barrier is not transparent enough for the electron to penetrate through it. Therefore, the resonant ionization rate w_{si} behaves like w_e and is more enhanced than the direct ionization rate w_0 . In addition, we can see that when the photon frequency ω is smaller or larger than the energy gap ω_{eg} (0.5 eV), w_{si} behaves like $W_{g \rightarrow e}(\omega)$ due to the fact that in the off-resonance case $W_{g \rightarrow e}(\omega)$ is much smaller than w_e .

V. CONCLUDING REMARKS

In the present paper, we have generalized and improved the derivation of photoionization rate of hydrogenlike atoms proposed by Keldysh. We have performed the contour integration of Eq. (2.11) in a different way from Keldysh or many other authors [29,45,57]. In most of the cases, the saddle-point method has been used, while we adopted the residue theorem for the contour integration. In addition, we have taken into account the \vec{p} dependence of the preexponential factor in Eq. (2.15). Our formulas are more accurate than those derived by Keldysh's theory. Numerical calculations have shown that the photoionization rate for the hydrogen atoms should be more enhanced than Keldysh's, which is in accord with the experimental results [41].

Based on our derivational method, we have extended the Keldysh theory to the photoionization processes of the randomly oriented diatomic molecules: N₂. The photoionization rates calculated by our formula for the randomly oriented N₂ molecules were in a good agreement with those estimated by the ADK formula [21].

As was mentioned in Introduction, because of the long-range Coulomb potential between the remaining ionic core and the ionizing electron for neutral atoms, and positive ions and molecules, it is questionable to apply Keldysh's assumption to such systems. In the Keldysh theory, in the exact expression for the amplitude of the wave function of the continuum state,

$$c_{\vec{p}}(T) = \frac{i}{\hbar} \int_0^T dt \cos(\omega t) e^{-(1/\hbar)E_{g^*}t} \langle \psi_{\vec{p}}(\vec{r}, t) | \vec{d} \cdot \vec{F} | \psi_g(\vec{r}) \rangle - \frac{i}{\hbar} \int_0^T dt \int \frac{d^3 p'}{(2\pi\hbar)^3} c_{\vec{p}'}(t) \langle \psi_{\vec{p}}(\vec{r}, t) | U(\vec{r}) | \psi_{\vec{p}'}(\vec{r}, t) \rangle, \quad (5.1)$$

the second term of the right-hand side was totally neglected. In order to incorporate the effect of Coulomb potential, we could use Eq. (2.2) and substitute $c_{\vec{p}}(t)$ into the second term on the right-hand side of Eq. (5.1). In spite of the approximation adopted by Keldysh, that is, the replacement of the final ionized state by the Volkov function [Eq. (2.4)], it is expected that we can obtain accurate analytical photoionization rate expressions up to second order including the effect of Coulomb potential. This idea has been pointed out by others [23,24,58], but analytical or qualitative formulas have not yet been obtained. The work in this direction is now under way.

ACKNOWLEDGMENTS

The authors wish to thank Academia Sinica, the National Science Council of ROC, and DFG for supporting this work.

APPENDIX A: COMPACT FORMS OF $L(\vec{p})$ DEFINED BY EQ. (2.15)

To facilitate the integration with respect to \vec{p} in w_0 of Eq. (2.7), we shall expand the functions included in Eq. (2.15) in

the power series of p up to second order inclusively. This means that the kinetic momentum of the ionizing electron is assumed to be low.

Using the following expression for u_s obtained from Eq. (2.9):

$$u_s = \gamma \left(i - \frac{p}{\sqrt{2mI_0}} \cos \theta + \frac{ip^2 \sin^2 \theta}{4mI_0} + \dots \right), \quad (A1)$$

it is easy to show that

$$j(u_s) = i\tilde{I}_0 \left(\sinh^{-1} \gamma - \frac{\gamma\sqrt{1+\gamma^2}}{1+2\gamma^2} \right) + \frac{eF \cos \theta}{m\omega} \times (1 - \sqrt{1+\gamma^2})p + \frac{i}{2m} \left(\sinh^{-1} \gamma - \frac{\gamma \cos^2 \theta}{\sqrt{1+\gamma^2}} \right) p^2, \quad (A2)$$

where

$$\gamma = \frac{\omega\sqrt{2mI_0}}{eF} \quad (\text{Keldysh parameter}) \quad (A3)$$

and

$$\vec{p} \cdot \vec{F} = pF \cos \theta. \quad (\text{A4})$$

Similarly, from Eq. (2.16), we find

$$\cos \theta_{pF} = 1 + \frac{p^2 \sin^2 \theta}{4mI_0} + \dots. \quad (\text{A5})$$

From Eq. (A1), we obtain

$$\sqrt{1 - \omega_s^2} = \sqrt{1 + \gamma^2} \left\{ 1 + \frac{i\gamma^2 p \cos \theta}{\sqrt{2mI_0}(1 + \gamma^2)} + \frac{\gamma^2 p^2 (1 - 2 \cos^2 \theta + \gamma^2 \sin^2 \theta)}{4mI_0(1 + \gamma^2)^2} \right\}. \quad (\text{A6})$$

Using the Taylor expansion of $\exp\{f(p)\}$ for an arbitrary function $f(p)$ around $p=0$,

$$\exp\{f(p)\} = \exp\{f(0)\} + f'(0)\exp\{f(0)\}p + \frac{f''(0) + f'(0)^2}{2}\exp\{f(0)\}p^2, \quad (\text{A7})$$

we find

$$\cos \theta_{pF} = \exp\left(\frac{p^2 \sin^2 \theta}{4mI_0}\right) \quad (\text{A8})$$

and

$$\sqrt{1 - u_s^2} = \sqrt{1 + \gamma^2} \exp\left\{ \frac{i\gamma^2 p \cos \theta}{\sqrt{2mI_0}(1 + \gamma^2)} + \frac{\gamma^2 p^2 (1 + \gamma^2 - 2 \cos^2 \theta)}{4mI_0(1 + \gamma^2)^2} \right\}. \quad (\text{A9})$$

Inserting Eqs. (A2), (A8), and (A9) into Eq. (2.15) yields

$$L(\vec{p}) = \frac{4\hbar\omega I_0 \sqrt{\pi a}}{eF \sqrt{1 + \gamma^2}} \exp[A + iB(\theta)p + C(\theta)p^2], \quad (\text{A10})$$

where

$$A = -\frac{\tilde{I}_0}{\hbar\omega} \left(\sinh^{-1} \gamma - \frac{\gamma\sqrt{1 + \gamma^2}}{1 + 2\gamma^2} \right), \quad (\text{A11})$$

$$B(\theta) = \frac{eF \cos \theta}{m\hbar\omega^2} (1 - \sqrt{1 + \gamma^2}) - \frac{\gamma^2 \cos \theta}{\sqrt{2mI_0}(1 + \gamma^2)}, \quad (\text{A12})$$

and

$$C(\theta) = \frac{1}{4mI_0} \left[-\frac{1 + 2\gamma^2}{1 + \gamma^2} + \frac{\gamma^4 + 4\gamma^2 + 1}{(1 + \gamma^2)^2} \cos^2 \theta \right] - \frac{1}{2m\hbar\omega} \left(\sinh^{-1} \gamma - \frac{\gamma \cos^2 \theta}{\sqrt{1 + \gamma^2}} \right). \quad (\text{A13})$$

The above procedure can be easily extended to the four cases of Eqs. (B3)–(B6) in Appendix B.

APPENDIX B: DEFINITION OF $N(\gamma, \omega, I_0, \tilde{I}_0, B, C)$ IN EQS. (2.17) AND (3.13)

As shown in the text, we have taken into account the \vec{p} dependence of the preexponential factors in Eq. (2.17), which was disregarded in Ref. [18]. In order to examine the influence of the \vec{p} dependence on the photoionization rate, we show some formulas depending on various treatments of the preexponential factors.

The preexponential factor $N(\gamma, \omega, I_0, \tilde{I}_0, B, C)$ in Eq. (2.17) is given by

$$N(\gamma, \omega, I_0, \tilde{I}_0, B, C) = \frac{1}{\sqrt{B}} \sum_{n=0}^{\infty} \exp \left[-2 \left(\left\langle \frac{\tilde{I}_0}{\hbar\omega} + 1 \right\rangle - \frac{\tilde{I}_0}{\hbar\omega} + n \right) \left\{ \sinh^{-1} \gamma - \frac{\gamma}{\sqrt{1 + \gamma^2}} + C \right\} \right] \times \Theta \left[\left\{ \frac{2\gamma}{\sqrt{1 + \gamma^2}} B \left(\left\langle \frac{\tilde{I}_0}{\hbar\omega} + 1 \right\rangle - \frac{\tilde{I}_0}{\hbar\omega} + n \right) \right\}^{1/2} \right], \quad (\text{B1})$$

where the symbol $\langle x \rangle$ denotes the integer part of the number x , $\Theta(x)$ is the Dawson's integral,

$$\Theta(x) = \exp(-x^2) \int_0^x \exp(y^2) dy. \quad (\text{B2})$$

Let us now consider the four cases: (i) $\sqrt{1 - u_s^2}$ and $\cos \theta_{pF}$ depend on \vec{p} , (ii) $\sqrt{1 - u_s^2}$ depends on \vec{p} and $\cos \theta_{pF} = 1$, (iii) $\sqrt{1 - u_s^2} = \sqrt{1 + \gamma^2}$ and $\cos \theta_{pF}$ depends on \vec{p} , and (iv) $\cos \theta_{pF} = 1$ and $\sqrt{1 - u_s^2} = \sqrt{1 + \gamma^2}$. It follows that

$$(i) \quad B = 1 + \frac{\hbar\omega(\gamma^4 + 4\gamma^2 + 1)}{2I_0\gamma(1 + \gamma^2)^{3/2}} \quad \text{and} \quad C = \frac{\hbar\omega\gamma^2(\gamma^2 - 1)}{2I_0(1 + \gamma^2)^2}, \quad (\text{B3})$$

$$(ii) \quad B = 1 + \frac{\hbar\omega\gamma}{I_0(1 + \gamma^2)^{3/2}} \quad \text{and} \quad C = \frac{\hbar\omega\gamma^2(\gamma^2 - 1)}{2I_0(1 + \gamma^2)^2}, \quad (\text{B4})$$

$$(iii) \quad B = 1 + \frac{\hbar\omega\sqrt{1 + \gamma^2}}{2I_0\gamma} \quad \text{and} \quad C = 0, \quad (\text{B5})$$

$$(iv) \quad B = 1 \quad \text{and} \quad C = 0. \quad (\text{B6})$$

In the case (iv), our formula is four times larger than that of Keldysh for the atomic system. This discrepancy stems from the different methods for the contour integration in Eq. (2.11).

The numerical results estimated by Eq. (2.17) for the above four cases and that by Eq. (3.13) for the case (iv) are compared and discussed in Fig. 1 and Fig. 3 of Sec. IV, respectively.

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- [1] N. B. Delone and V. P. Krainov, *Multiphoton Processes in Atoms* (Springer-Verlag, Berlin, 1994); S. L. Chin and P. Lambropoulos, *Multiphoton Ionization of Atoms* (Academic, New York, 1984).
- [2] M. Gavrilu, *Atoms in Intense Laser Fields* (Academic, New York, 1992).
- [3] H. R. Reiss, *Prog. Quantum Electron.* **16**, 1 (1992).
- [4] A. Giusti-Suzor, X. He, and O. Atabek, *Phys. Rev. Lett.* **64**, 515 (1990); A. Zavriyev, P. H. Bucksbaum, H. G. Muller, and D. W. Schumacher, *Phys. Rev. A* **42**, 5500 (1990).
- [5] G. Jolicard and O. Atabek, *Phys. Rev. A* **46**, 5845 (1992).
- [6] A. Giusti-Suzor and F. H. Mies, *Phys. Rev. Lett.* **68**, 3869 (1992); G. H. Yao and S.-I. Chu, *Chem. Phys. Lett.* **197**, 413 (1992).
- [7] A. D. Bandrauk, *Molecules in Laser Fields* (Marcel Dekker, New York, 1994).
- [8] A. Hishikawa, A. Iwamae, and K. Yamanouchi, *J. Chem. Phys.* **111**, 8871 (1999).
- [9] A. Hishikawa, A. Iwamae, K. Hoshina, M. Kono, and K. Yamanouchi, *Chem. Phys.* **231**, 315 (1999).
- [10] J. Kou, V. Zhakhovskii, S. Sakabe, K. Nishihara, S. Shimizu, S. Bulanov, Y. Izawa, Y. Kato, and N. Nakashima, *J. Chem. Phys.* **112**, 5012 (2000).
- [11] E. Constant, H. Stapefeld, and P. B. Corkum, *Phys. Rev. Lett.* **76**, 4140 (1996).
- [12] T. Seideman, M. Y. Ivanov, and P. B. Corkum, *Phys. Rev. Lett.* **75**, 2819 (1995).
- [13] C. Cornaggia, M. Schmit, and D. Normand, *Phys. Rev. A* **51**, 1431 (1995).
- [14] C. Cornaggia, *Phys. Rev. A* **52**, R4328 (1995).
- [15] C. Cornaggia, *Phys. Rev. A* **54**, R2555 (1996).
- [16] C. Cornaggia, F. Salin, and C. Leblanc, *J. Phys. B* **29**, L749 (1996).
- [17] A. Hishikawa, A. Iwamae, and K. Yamanouchi, *Phys. Rev. Lett.* **83**, 1127 (1999).
- [18] L. V. Keldysh, *Zh. Eksp. Teor. Fiz.* **47**, 1945 (1964) [*Sov. Phys. JETP* **20**, 1307 (1965)].
- [19] L. D. Landau and E. M. Lifshitz, *Quantum Mechanics: Non-Relativistic Theory*, 3rd ed. (Pergamon, Oxford, 1991).
- [20] A. M. Perelomov, V. S. Popov, and M. V. Terent'ev, *Zh. Eksp. Teor. Fiz.* **50**, 1393 (1966) [*Sov. Phys. JETP* **23**, 924 (1966)].
- [21] M. V. Ammosov, N. B. Delone, and V. P. Krainov, *Zh. Eksp. Teor. Fiz.* **91**, 2008 (1986) [*Sov. Phys. JETP* **64**, 1191 (1987)].
- [22] F. H. M. Faisal, *J. Phys. B* **6**, L89 (1973).
- [23] H. R. Reiss, *Phys. Rev. A* **22**, 1786 (1980).
- [24] F. Trombetta, S. Basile, and G. Ferrante, *Phys. Rev. A* **40**, 2774 (1989).
- [25] M. H. Mittleman, *Phys. Rev. A* **40**, 463 (1989); Y. Abranyos and M. H. Mittleman, *ibid.* **42**, 4284 (1990).
- [26] F. A. Ilkov, J. E. Decker, and S. L. Chin, *J. Phys. B* **25**, 4005 (1992).
- [27] L. A. Lompre, G. Mainfray, C. Manus, S. Repoux, and J. Thebault, *Phys. Rev. Lett.* **36**, 949 (1976).
- [28] G. Gibson, T. S. Luk, and C. K. Rhodes, *Phys. Rev. A* **41**, 5049 (1990).
- [29] V. A. Pazdzersky and V. A. Yurovsky, *Phys. Rev. A* **51**, 632 (1995); V. A. Pazdzersky, V. I. Usachenko, and A. A. Chernov, *J. Phys. B* **34**, 363 (2001).
- [30] M. Jain and N. Tzoar, *Phys. Rev. A* **18**, 538 (1978).
- [31] R. Cavaliere, G. Ferrante, and C. Leone, *J. Phys. B* **13**, 4495 (1980).
- [32] P. Kalman, *Phys. Rev. A* **38**, 5458 (1988).
- [33] G. Duchateau, E. Cormier, and R. Gayet, *Eur. Phys. J. D* **11**, 191 (2000).
- [34] G. Duchateau, E. Cormier, H. Bachau, and R. Gayet, *Phys. Rev. A* **63**, 053411 (2001).
- [35] J. Bauer, *Phys. Rev. A* **55**, 3261 (1997).
- [36] J. Z. Kaminski, A. Jaron, and F. Ehlötzky, *Phys. Rev. A* **53**, 1756 (1996).
- [37] H. R. Reiss and V. P. Krainov, *Phys. Rev. A* **50**, R910 (1994).
- [38] C. Cornaggia and Ph. Hering, *Phys. Rev. A* **62**, 023403 (2000).
- [39] M. J. Dewitt and R. J. Levis, *J. Chem. Phys.* **108**, 7739 (1998).
- [40] W. Gordon, *Z. Phys.* **40**, 117 (1926); D. M. Volkov, *ibid.* **94**, 250 (1935).
- [41] G. A. Kyrala and T. D. Nichols, *Phys. Rev. A* **44**, R1450 (1991).
- [42] I. Caelli, R. Moccia, and A. Rizzo, *Phys. Rev. A* **57**, 1895 (1998).
- [43] R. R. Lucchese, G. Raseev, and V. McKoy, *Phys. Rev. A* **25**, 2572 (1982).
- [44] G. S. Ondrey, C. Rose, D. Proch, and K. L. Kompa, *J. Chem. Phys.* **95**, 7823 (1991).
- [45] G. F. Gribakin and M. Yu. Kuchiev, *Phys. Rev. A* **55**, 3760 (1997).
- [46] H. R. Reiss and V. P. Krainov, *Phys. Rev. A* **50**, R910 (1994); J. Bauer, *J. Phys. B* **34**, 1343 (2001).
- [47] H. Siegbahn, L. Asplund, and P. Kelfve, *Chem. Phys. Lett.* **35**, 330 (1975); I. N. Levine, *Quantum Chemistry* (Prentice-Hall, Englewood Cliffs, NJ, 1991).
- [48] K. J. LaGattuta, *Phys. Rev. A* **41**, 5110 (1990).
- [49] P. H. Bucksbaum, A. Zavriyev, H. G. Muller, and D. W. Schumacher, *Phys. Rev. Lett.* **64**, 1883 (1990).
- [50] K. Codling, L. J. Fransiski, and P. A. Hatherly, *J. Phys. B* **22**, L321 (1989); K. Codling, L. J. Fransiski, and P. A. Hatherly, *Science* **246**, 1029 (1989); K. Codling, P. A. Hatherly, L. J. Fransiski, A. J. Langley, and W. Shaikh, *J. Phys. B* **23**, L291 (1990).
- [51] M. J. DeWitt and R. J. Levis, *J. Chem. Phys.* **110**, 11 368 (1999).
- [52] C. Guo, M. Li, J. P. Nibarger, and G. N. Gibson, *Phys. Rev. A* **58**, R4271 (1998).
- [53] C. Guo and G. N. Gibson, *Phys. Rev. A* **63**, 040701(R) (2001).

- [54] J. P. Nibarger, S. V. Menon, and G. N. Gibson, *Phys. Rev. A* **63**, 053406 (2001).
- [55] M. J. DeWitt, B. S. Prall, and R. J. Levis, *J. Chem. Phys.* **113**, 1553 (2000).
- [56] I. N. Levine, *Molecular Spectroscopy* (Wiley, New York, 1975), p. 316.
- [57] H. S. Brandi, L. Davidovich, and N. Zagury, *Phys. Rev. A* **24**, 2044 (1981); G. L. Yudin and M. Yu. Ivanov, *ibid.* **64**, 013409 (2001); S. V. Popruzhenko and S. P. Goreslavskii, *J. Phys. B* **34**, L239 (2001).
- [58] W. Becker, L. Davidovich, and J. K. McIver, *Phys. Rev. A* **49**, 1131 (1994).