PHYSICAL REVIEW A, VOLUME 65, 013405

Ionization of alkali-metal atoms by ultrashort laser pulses

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(Received 28 May 2001; published 14 December 2001)

The ionization of alkali-metal atoms by ultrashort laser pulses, which perform less than two optical cycles, is investigated by means of a theoretical method based on Coulomb-Volkov (CV) states. This approach was shown to provide reliable predictions for the ionization of hydrogen atoms both at small and high laser intensities. In the present paper, it is extended to the ejection of electrons from the valence shell of alkali-metal atoms. CV predictions are shown to be in good agreement with the results given by a full numerical treatment of the time-dependent Schrödinger equation. Compared to hydrogen targets, the presence of the core results in a much more extended domain of application of the CV approach.

DOI: 10.1103/PhysRevA.65.013405 PACS number(s): 42.50.Hz, 32.80.Rm, 32.80.Fb

I. INTRODUCTION

Nowadays, intense laser pulses that contain less than two optical cycles (in the full width at half maximum) have been achieved [1]. Such pulses lead to a new kind of laser-atom interaction physics in which the monochromatic photon aspect of the laser field vanishes [2]. As a result, the ionization process is strongly affected. In previous papers [3,4], we showed that the electron-energy spectrum no longer exhibits equally spaced peaks connected to multiphoton absorption (above threshhold ionization). Even with energetic enough photons, the standard photoelectric peak does not show up in this case. Instead, the distribution exhibits a peak at an energy corresponding to a transfer from a pure classical field [2].

In principle, the new features of the ionization process can be predicted by a full numerical treatment of the time-dependent Schrödinger equation for a one-active electron system in an external electric field. However, in case of intense laser fields, this type of approach, hereafter referred to as TDSE, becomes very difficult [3,4]. Moreover, TDSE calculations appear unrealistic with two-active electron systems. Therefore, we developed an approach, which is specifically adapted to atom ionization by very short laser pulses whatever the intensity [3,4]: it is based on Coulomb-Volkov (CV) states [5]. For hydrogen targets, we have shown that it provides accurate predictions of ejected electron distributions as long as the two following conditions are simultaneously fulfilled: (i) the laser electric field makes less than two oscillations, (ii) the interaction time τ does not exceed $T/2$ where T is the initial orbital period. Now, the ionization of alkali-metal atoms is an interesting case because the orbital period of the valence electron in the ground state is much longer than the orbital period of H$(1s)$, thus meeting the two preceeding requirements with more realistic laser pulse durations. Further, the presence of a rare-gas-type core in alkali-metal atoms has a specific influence on the ejection of the valence electron: once it is in the continuum, the ejected electron experiences a nuclear Coulomb field that is weaker than in the initial s orbital, which penetrates deeply in the core. Thus, the condition (ii) that requires $\tau \leqslant T/2$ might appear too restrictive. Therefore, one may expect a wider range of application of the CV theory with alkali-metal atoms.

In Sec. II, the extension of TDSE and CV methods to alkali-metal targets is presented. The consistency of the physical assumptions is analyzed in Sec. III A. The physics of the ionization process is exhibited in Sec III B by comparing the energy distributions of the ejected electrons predicted by CV and TDSE approaches. It is shown that with alkali-metal atoms, good CV predictions may be made with pulse durations significantly longer than $T/2$, thus extending the domain of application of the CV theory slightly beyond the limits of the sudden approximation. In our previous papers, all studies have been made using fully symmetric pulses. In the present paper, the influence of the absolute phase of the laser field on the accuracy of CV predictions is investigated in Sec. III C. Conclusions are drawn in Sec. IV.

Atomic units are used throughout unless otherwise stated.

II. EXTENSION OF TDSE AND CV THEORIES

A. Model of laser pulses

We consider a symmetric linearly polarized laser pulse with a photon energy set to $\omega = 0.05$ a.u. in order to overlap the energy range of photons commonly generated by Ti:= sapphire lasers. The finite duration of the pulse is featured through a square-sine envelope. Under these conditions, the field expression is

$$\vec{F}(t) = \begin{cases} \vec{F}_0 \sin(\omega t + \phi_0) \sin^2\left(\dfrac{\pi t}{\tau}\right) & \text{if } 0 < t < \tau \\ \vec{0} & \text{elsewhere} \end{cases} \tag{2.1}$$

with

$$\phi_0 = \frac{\pi}{2} - \frac{\omega \tau}{2}. \tag{2.2}$$

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This value of ϕ_0 ensures the field to be symmetric with respect to $\tau/2$.

B. Model for valence electron orbitals of alkali-metal atoms

The full numerical resolution of the time-dependent Schrödinger equation for a one-active electron system in a laser field has been briefly described in our last papers [3,4] (see Ref. [6] for more details). Let us just recall that the radial part of the total wave function $\Psi(\vec{r},t)$ is expanded on a *B*-spline basis set. Substituting this form of $\Psi(\vec{r},t)$ in TDSE gives a set of coupled equations that is solved numerically. In the length gauge version of the dipole approximation, the time-dependent Schrödinger equation is given by

$$i\frac{\partial\Psi(\vec{r},t)}{\partial t}=\left(-\frac{\vec{\nabla}^2}{2}+V(r)+\vec{r}\cdot\vec{F}(t)\right)\Psi(\vec{r},t), \qquad (2.3)$$

where, in the frozen core approximation, $V(r)$ is the model potential experienced by the valence electron of alkali-metal atoms. In the present case, the form of $V(r)$ is

$$V(r)=-\frac{Z}{r}+\frac{Z_c}{r}-\frac{Z_c}{r}(1+\alpha r)e^{-2\alpha r}, \qquad (2.4)$$

where Z is the nuclear charge and Z_c is the number of core electrons. α is a parameter to be optimized in order to reproduce, as well as possible, the ground and a few excited levels of the valence electron. This type of potential was initially developed to represent an electron in the field of a $1s^2$ core [7]. TDSE calculations (not presented here) show that the orbital energies E_i of the valence electron for the ground states of sodium, potassium, and rubidium agree very well with the corresponding orbital energies given by Clementi and Roetti [8] by setting $\alpha=1.8972$, $\alpha=1.6730$, and $\alpha=1.9785$, respectively. Further, for each alkali-metal atom in its ground state, the TDSE wave function of the valence electron fits almost perfectly the corresponding orbital of Clementi and Roetti. Now, the orbital period of valence electrons T is estimated by the following formula:

$$T=\frac{2\pi\langle r\rangle}{\langle v\rangle}, \qquad (2.5)$$

where $\langle r\rangle$ is an average value of r that is obtained numerically. The average value of the velocity in the initial bound state $\langle v\rangle$ is approximated by $(-2E_i)^{1/2}$ as prescribed by the virial theorem. Then, estimated orbital periods T are $T_{\mathrm{Na}}\simeq 35.5$ a.u.$\simeq 0.88$ fs, $T_{\mathrm{K}}\simeq 48.6$ a.u.$\simeq 1.17$ fs and $T_{\mathrm{Rb}}\simeq 54.$ a.u.$\simeq 1.3$ fs.

C. Extension of the CV theory to alkali-metal atoms

If one can find a good analytical approximation for the solution of Eq. (2.3), its projection onto the final state should give an accurate prediction of the transition amplitude T_{fi}. Then, the probability to eject an electron with a momentum $\vec{k}$ is $|T_{fi}|^2$. Although the well-known Coulomb-Volkov states have been already used in perturbative conditions with very long laser pulses [9], we showed that, subject to the two conditions (i) and (ii) quoted in the introduction, they are good approximate solutions of Eq. (2.3) [5]. Further, we also showed that one may introduce CV bound states, which read

$$\chi_i^{+}(\vec{r},t)=\varphi_i(\vec{r},t)\exp\left[i\vec{A}(t)\cdot\vec{r}-\frac{i}{2}\int_0^t dt'A^2(t')\right], \qquad (2.6)$$

where $\varphi_i(\vec{r},t)$ is the initial unperturbed target bound state. $\vec{A}(t)$ is the vector potential given by $\vec{A}(t)=-\int_0^t dt'\vec{F}(t')$. In the case of the ionization of hydrogen targets, $\chi_i^{+}(\vec{r},t)$ is projected onto the ingoing continuum Coulomb wave function $\mathcal{C}_{\vec{k}}^{-}(\vec{r},t)$ given by

$$\mathcal{C}_{\vec{k}}^{-}(\vec{r},t)=\frac{1}{(2\pi)^{3/2}}e^{\pi\nu/2}\Gamma(1+i\nu)F(-i\nu,1,-ikr-i\vec{k}\cdot\vec{r})e^{i\vec{k}\cdot\vec{r}}e^{-i\epsilon t}, \qquad (2.7)$$

which describes exactly a free electron in the field of the proton. In the case of alkali-metal atoms, there is no simple analytical wave function $\varphi_f(\vec{r},t)$ describing the electron in the field of the core. It can be obtained only by a full numerical treatment of the stationary Schrödinger equation. Therefore, it appears interesting not to loose the power of the CV approach that is based on simple analytical expressions to be handled. Thus, one has to look for an approximate analytical final state. With intense laser fields, the electron is ejected at large radial distance r in a very short time. Under these conditions, the electron experiments rapidly the field of a remote positive charge, which may be considered as a quasipunctual charge. Hence, we will assume that the final electron wave function is approximatively given by the regular continuum Coulomb state (2.7), i.e.,

$$\varphi_f(\vec{r},t)\simeq\mathcal{C}_{\vec{k}}^{-}(\vec{r},t) \qquad (2.8)$$

The discrepancy between $\varphi_f(\vec{r},t)$ and $\mathcal{C}_{\vec{k}}^{-}(\vec{r},t)$ comes from the behavior of the electronic wave function in the region where the core influence is significant. As a result, we know that a non-Coulombic phase appears explicitly in the asymptotic form of $\varphi_f(\vec{r},t)$. Thus, our theory is based on the asumption that the non-Coulombic phase has no major influence on the continuum wave function in the context of intense laser fields.

III. RESULTS AND DISCUSSION

A. Consistency of the model

Our model is realistic as long as the non-Coulombic phase has little influence on the continuum wave function. Therefore, it is important to check under which conditions one may assume that it is the case before performing any calculation. To do that, the regular continuum Coulomb state (2.7) is compared to the numerical form of $\varphi_f(\vec{r},t)$ that is given by the structure calculation module of the TDSE code with the

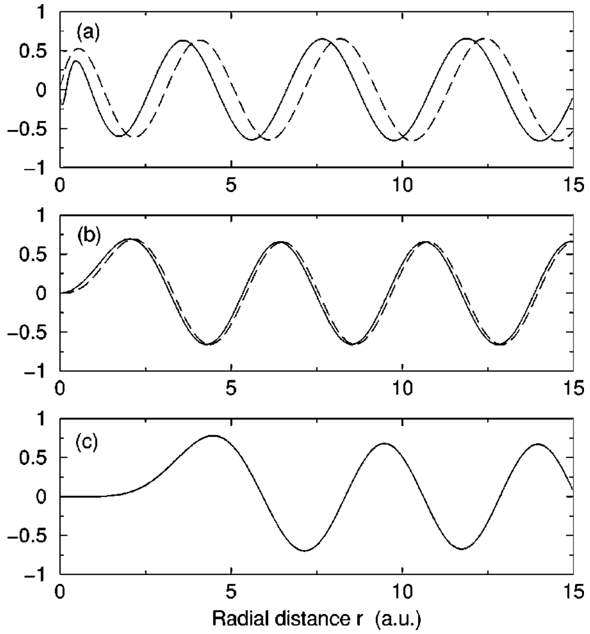

FIG. 1. Comparison between the radial continuum wave functions of sodium given by TDSE and the regular Coulomb wave function for an electron energy 1 a.u. and for various partial waves: $l=0$ [Fig. 1(a)], $l=2$ [Fig. 1(b)], and $l=5$ [Fig. 1(c)].

model potential that is defined in Sec. II B. The latter gives the radial part of the wave function called $R_{E_k,l}^{TDSE}(r)$ for a given angular momentum l and for a given energy E_k. Therefore, it is necessary to first expand the ingoing continuum Coulomb wave function $\mathcal{C}_{\vec{k}}^{-}(\vec{r},t)$ defined by Eq. (2.7) in partial waves in order to compare with the radial component $R_{E_k,l}(r)$ of a given l to $R_{E_k,l}^{TDSE}(r)$. According to Ref. [10] one has

$$\mathcal{C}_{\vec{k}}^{-}(\vec{r})=\sum_{l,m} R_{E_k,l}(r)Y_l^m(\theta,\varphi), \tag{3.1}$$

where the time-dependent phase factor is omitted and where

$$R_{E_k,l}(r)=\left(\frac{2k}{\pi}\right)^{1/2}\frac{e^{\pi\nu/2}}{(2l+1)!}|\Gamma(l+1-i\nu)|r(2kr)^l \times e^{-ikr}F_1(l+1+i\nu,2l+2,2ikr). \tag{3.2}$$

In Fig 1, we compare $R_{E_k,l}(r)$ with $R_{E_k,l}^{TDSE}(r)$ for sodium targets. The comparison is made for $E_k=1$ a.u. and for $l=0$, $l=2$, and $l=5$. Sodium appears to be the most difficult case among alkali-metal atoms, because it shows the largest overlap between the ground state and the whole continuum H-like spectrum. Since the non-Coulombic phase decreases smoothly with the energy, it is clear that $\varphi_f(\vec{r},t)$ tends to the pure Coulomb state when the ejected electron energy increases. An energy of 1 a.u. is small enough to make it a hard test. In Fig. 1, one notices that a good agreement is rapidly obtained when the value of l increases. It backs up the soundness of the replacement of $\varphi_f(\vec{r},t)$ by $\mathcal{C}_{\vec{k}}^{-}(\vec{r},t)$.

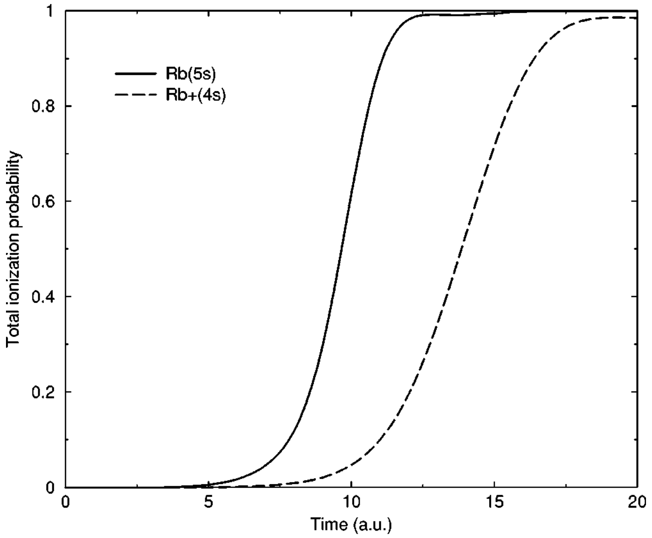

FIG. 2. Total ionization probabilities of electrons ejected from 4s and 5s orbitals of rubidium as functions of time. The laser parameters are: $I_0=3.51\times10^{18}$ W cm^{-2}, $\tau=5.3$ fs, and $\lambda=800$ nm.

Still, it is important to check to what extent this approximation of the continuum wave function may provide good predictions for the ionization process. In what follows, it is shown that the approximation (2.8) gives reliable predictions of the electronic spectra as long as the two required conditions quoted in the introduction are fulfilled.

Further, a second question arises that concerns the use of a model potential to describe the ionization of a valence electron by a laser pulse that may be intense enough to eject inner-shell electrons as well. In other words, can we consider that it is reasonable to assume that the alkali-metal core stays almost unchanged during the period of time required to fully ionize the valence shell. It is equivalent to check that the multiple ionization process is a sequential mechanism. In order to get significant indication that it might be the case, total ionization probabilities of electrons ejected from 4s and 5s orbitals of rubidium have been calculated when Rb targets are illuminated by 3.51×10^{18} W cm^{-2}, 5.3 fs, 800 nm laser pulses. Ionization probabilities predicted by CV calculations are reported in Fig. 2 as functions of time. For the shortest interaction time, perturbative conditions prevail. Therefore, a Schmidt orthogonalization procedure is used to make $|\mathcal{C}_{\vec{k}}^{-}(t)\rangle$ orthogonal to the initial bound state. Thus, one ensures the probability to be zero at $t=0$. In order to check the sequential mechanism with our CV approach, a very high laser intensity is necessary to keep calculations within the time range where CV applies. On Fig. 2, one notices that the ionization of 4s orbitals begins just a while before the full ionization of the 5s orbital. It is a good indication that the ionization of alkali-metal atoms by ultrashort and intense laser pulses is a sequential process. Thus, the frozen core approximation appears reliable as long as laser intensities and interaction times are not so large that the core and valence electrons are ionized at the same time. Under these conditions, a model potential may be used to describe the ionization of valence electrons by such laser pulses.

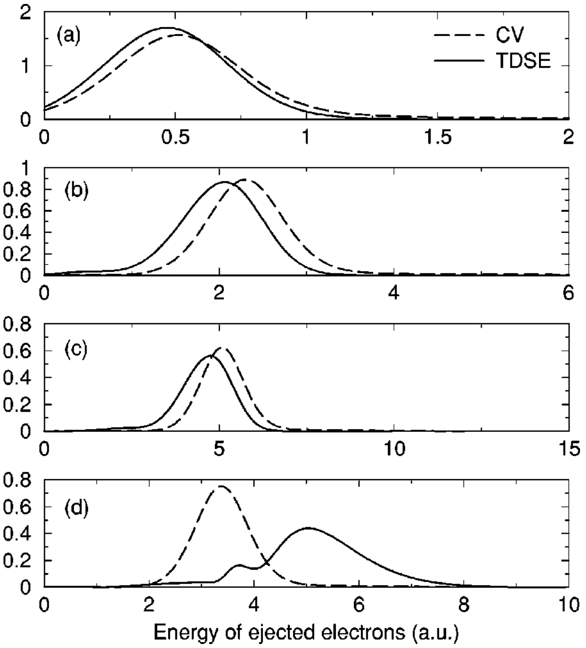

FIG. 3. Energy distributions of ejected electrons from Na(3s). In all cases, the laser field amplitude is $F_0=0.1$ a.u. and the photon energy is $\omega=0.05$ a.u.. The pulse durations are $\tau=25$ a.u. [Fig. 3(a)], $\tau=50$ a.u. [Fig. 3(b)], $\tau=100$ a.u. [Fig. 3(c)], and $\tau=150$ a.u. [Fig. 3(d)].

B. Energy spectra of the ejected electrons

From previous studies [3,4], the CV approach is known to apply as long as the interaction time τ is shorter than half the orbital period T of the valence electron. In order to check how the prescription could be extended to alkali-metal atoms, the ionization of Na(3s) by a laser pulse, whose field amplitude and photon energy are $F_0=0.1$ a.u. ($I_0=3.51\times10^{14}$ W/cm^2) and $\omega=0.05$ a.u. (Ti:sapphire), respectively, is investigated for increasing values of the interaction time τ. The energy distribution of ejected electrons is displayed in Fig. 3 for $\tau=25,50,100$ and 150 a.u. (3.6 fs). Although the orbital period of the unperturbed orbital 3s of sodium is $T_{\mathrm{Na}}=36.5$ a.u., a reasonably good agreement between CV and TDSE distributions is obtained up to $\tau=100$. According to our previous studies with atomic hydrogen targets [4], good predictions would be expected for $\tau<18$ a.u.. Actually, the small value of T_{Na} is mainly due to the nonzero probability of finding the valence electron of sodium close to the nucleus where it "sees" a nuclear charge equal to 11. However, once it is slightly shifted by the laser field, the electron stops rapidly experiencing the Coulomb field close to the nucleus. Thus, imposing $\tau<T_{\mathrm{Na}}/2$ appears too restrictive a rule to define the domain where our CV method applies in this case. Since CV results are reliable up to $\tau\simeq100$ a.u., the range of validity for sodium stretches out to $\tau\leqslant5T_{\mathrm{Na}}/2$ or so. It is a factor of 5 greater than expected. One may attribute this extension to a dynamical screening effect of the core since the same feature does not show up for H(3s) or H(4s) targets.

For the same parameters of laser pulses as previously, the energy distributions of electrons ejected from K(4s) and Rb(5s) are reported on Fig. 4 and 5, respectively. A look at these figures permits right away to draw conclusions similar to the ones of the last paragraph. Finally, whatever the alkali-metal target, the domain where the CV approach provides reliable data is $\tau\leqslant2T$ or so. Compared to the case of hydrogen targets, it is extended at least by a factor of 4.

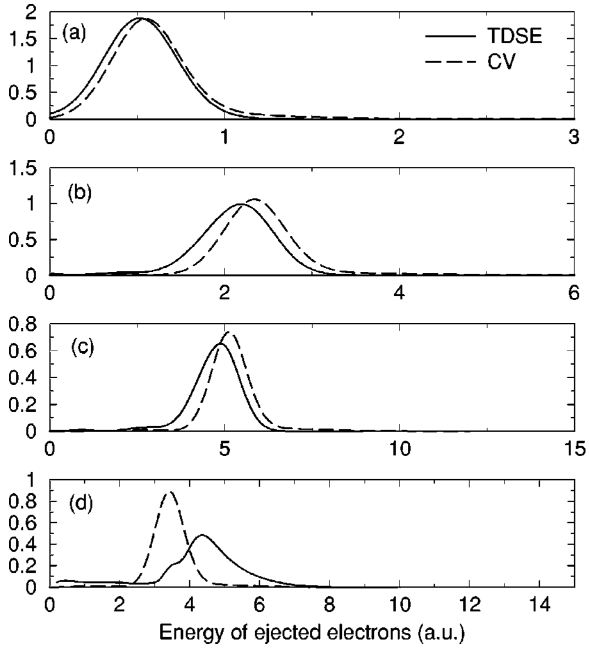

FIG. 4. Same as Fig. 3 for K(4s).

Therefore, it is interesting to investigate the influence of the core on the energy distributions of ejected electrons. To get a better insight, let us take a pulse duration at the limit of validity of CV. Thus, we choose $\tau=100$ a.u. and we compare the distributions for sodium, potassium, and rubidium

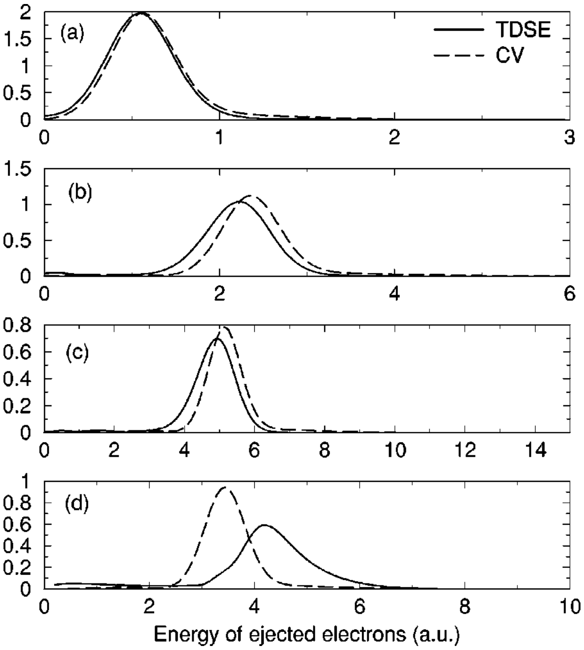

FIG. 5. Same as Fig. 3 for Rb(5s).

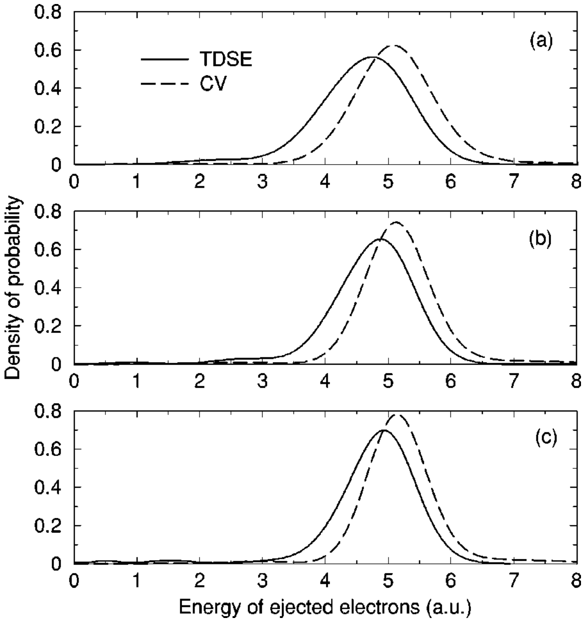

FIG. 6. Energy distributions of electrons ejected from Na(3*s*) [Fig. 6(a)], K(4*s*) [Fig. 6(b)], and Rb(5s) [Fig. 6(c)] by laser pulses such that $F_0=0.1$ a.u., $\tau=100$ a.u., and $\omega=0.05$ a.u.

targets (see Fig. 6). One might expect that the influence of the core increases with the atomic number because of an increasing electronic density. Actually, the core size of alkali-metal atoms is roughly the same whatever the atomic number. Further, for hydrogen targets in interaction with half-cycle pulses, it was shown in Ref. [2], that TDSE calculations predict a peak of energy distributions that is located at a smaller energy than CV ones. It is due to the dynamical influence of the nucleus during the interaction. The nuclear attraction slows down the ejected electron but this effect is not taken into account in our CV theory since the latter assumes $\partial\vec{p}(t)/\partial t=-\vec{F}(t)$. With alkali-metal atoms, the nuclear attraction is replaced by the interaction with the rare-gas-like core. Because of inner-shell electron screening, the nuclear attraction decreases relatively much faster than for hydrogen. Indeed, the discrepancy between CV and TDSE predictions still appears, but it requires a longer interaction time to become significant.

Now, a look at Fig 6 shows that discrepancy between TDSE and CV predictions shows up for ejection energies E_k much smaller than the energy of the maximum of the distribution, more especially for $E_k\leqslant 2$ a.u.. The discrepancy becomes more apparent when the atomic number increases. Again, this difference should be attributed to the influence of the core that is not considered in the CV theory. Let us consider valence-electron densities as functions of the distance to the nucleus. On the whole, they are comparable in size for the three alkali-metal atoms (although they do not have the same number of zeros). The three densities have small but non-negligible values inside the core. Although screening due to inner-shell electrons reduces the nuclear Coulomb field, the attraction of the nucleus is all the more effective at a given distance that the atomic number is large. Indeed, electrons ejected at small energies stay longer in the vicinity of the nucleus. Therefore, the attraction of the nucleus is all the more effective to reduce the velocity during the pulse that the atomic number is large. As a result, the energy distribution predicted by TDSE at small ejection energies is shifted towards the ionization threshold compared to CV predictions. Indeed, the discrepancy between CV and TDSE is larger for heavier alkali-metal atoms.

Now, let us consider the energy distributions of Figs. 3(d), 4(d), and 5(d). One notices that TDSE calculations predict electrons at a higher energy than CV ones. This is due to the fact that the electric field performs more than one oscillation. For $\tau=150$ a.u., it makes 1.2 oscillation, whereas it makes only 0.8 oscillation for $\tau=100$ a.u. Since one has roughly a half-cycle-like pulse in the latter case, the effect of the core is just to slow down the escaping electron. In the case of a 1.2 cycle laser pulse, the electron is moved back and forth by the laser electric field and a simple classical model shows that the net effect of the nuclear attraction is to increase the final electron velocity. Since the CV theory does not take into account this dynamical influence, it predicts a peak in the distribution that is located at smaller ejection energies than TDSE when the field performs one oscillation or so.

C. Influence of the absolute phase of the laser pulse

All calculations, which are presented here and in our previous papers, have been performed with an absolute phase of the pulse $\phi_0=\pi/2-\omega\tau/2$ such that the oscillations of the laser field are symmetric with respect to the midpoint $\tau/2$ of the pulse duration. With less than two oscillations, it ensures $\vec{A}(\tau)\neq\vec{0}$. Then, the CV state $\chi_i^+(\vec{r},t)$ is never reduced to $\varphi_i(\vec{r},t)$ [see Eq (2.6)] and the CV transition amplitude T_{fi}^{CV} is generally nonzero. However, when the pulse is not symmetric, it may happened accidentally that $\vec{A}(\tau)=\vec{0}$, in which case T_{fi}^{CV} is exactly zero although the exact value of T_{fi} (as given by TDSE) may well be different from zero.

Therefore, it is interesting to know to which extent CV predictions are reliable when ϕ is varied to pass from a symmetric pulse to an antisymmetric pulse. To do that, let us introduce

$$\phi'=\phi-\phi_0. \tag{3.3}$$

Then for $\phi'=0$, the pulse is symmetric and the field is maximum at $t=\tau/2$ whereas for $\phi'=\pi/2$ the pulse is antisymmetric and the field is zero at $t=\tau/2$.

For the sake of simplicity, our study is made for a hydrogen atom in the state 4*s* with a laser pulse whose characteristics are $\omega=0.055$ a.u. ($\lambda\simeq800$ nm), $\tau=114$ a.u. (1 cycle), and $F_0=0.1$ a.u. We use a dichotomy procedure to make ϕ' approaches slowly the value $\pi/2$ where CV fails. Again, CV predictions are compared to TDSE ones (see Fig. 7). It appears that the difference between CV and TDSE is noticeable only when ϕ' is close to $\pi/2$. Therefore, except for almost antisymmetric pulses, our CV approach applies (when τ does not exceed $2T$ and when the field does not perform more than two oscillations). Indeed the conclusion related to the laser phase ϕ' does not depend upon the particular target that is ionized.

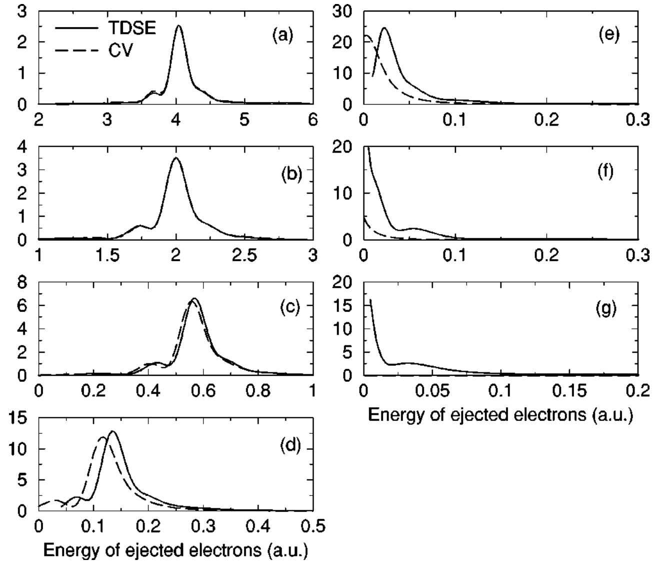

FIG. 7. Energy distributions of electrons ejected from H$(4s)$ for various value of the absolute laser phase ϕ' (see text). In all cases, the laser field amplitude is $F_0=0.1$ a.u., the pulse duration is $\tau=114$ a.u., and the photon energy is $\omega=0.055$ a.u. (1 cycle). The values of ϕ' are $\phi'=0$ [Fig. 7(a)], $\phi'=\pi/4$ [Fig. 7(b)], $\phi'=3\pi/8$ [Fig. 7(c)], $\phi'=7\pi/16$ [Fig. 7(d)], $\phi'=15\pi/32$ [Fig. 7(e)], $\phi'=31\pi/64$ [Fig. 7(f)], and $\phi'=\pi/2$ [Fig. 7(g)].

IV. CONCLUSION

The present CV approach can provide reliable predictions for the ionization of alkali-metal atoms by intense and ultrashort laser pulses of wavelength comparable to the Ti:Sapphire laser wavelength, provided that the laser field performs less than two optical cycles as in Nisoli *et al*'s experiments [1]. In the case of hydrogen targets, it was shown [3,4] that a second condition to get reliable predictions is $\tau \leqslant T/2$ where τ is the pulse duration and T is the initial orbital period of the valence electron. For alkali-metal targets, the present study shows that the CV theory gives accurate predictions of electron-energy spectra as long as $\tau \leqslant 2T$ and for an absolute phase ϕ' not too close to $\pi/2$. This extended range of application is due to the influence of the core. Therefore, one may envisage the use of the CV approach to investigate the ejection of valence electrons from complex atoms with realistic pulse durations.

Further, we showed that the rare-gas-like core has little influence on the energy distribution of electrons that are ejected from the valence shell. It is noteworthy that the CV theory allows to perform simple reliable calculations at high intensities where full numerical treatments are cumbersome; the higher the intensity, the better the CV predictions [3].

It is possible to extend the present CV approach to double ionization of two-active electron targets. In this case, full numerical quantum calculations are very hazardous with intense and very short laser pulses. Some preliminary results may be found in [11].

ACKNOWLEDGMENTS

We are indebted to Eric Cormier for helping us to adapt his one-electron TDSE code to the case of valence electrons of alkali-metal atoms. Helpful discussions with Henri Bachau are also acknowledged. Thanks are due to Professor Presnyakov for enlightening discussions in the framework of INTAS Contract No.99-01326. This work was partially supported by the Conseil Régional d'Aquitaine. One of us (G.D.) wishes to thank the Centre National de la Recherche Scientifique and the Conseil Régional d'Aquitaine.

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