Variation of harmonic generation from He interacting with short laser pulses of 5 eV as a function of pulse rise time and intensity

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We have applied the state-specific expansion approach (SSEA) to the solution of the time-dependent Schrödinger equation describing the interaction of He with laser pulses of $\hbar\omega=5$ eV, which have a shape with a sharp rise and peak intensities in the range $5\times10^{14}-3.46\times10^{15}$ W/cm^2. The calculations include the effects of the state-specific self-consistent field, the electron correlation, and the $n=2$ intrashell doubly excited states. The results show that, provided the pulses have fast rise times, neutral He can generate harmonics of higher order than previously thought before the He^+ contribution takes over. The computed spectra agree with the observations of Sarukura *et al.* [Phys. Rev. A **43**, 1669 (1991)]. In particular, the SSEA results reveal ''anomalously'' high peaks for the 13th and 21st harmonics, in agreement with experiment.

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I. INTRODUCTION

The interaction of short laser pulses of very high intensity with atoms leads to high-order harmonic generation (HOHG). A number of experiments [1–10] and calculations performed on noble gases [8,11–17] during the 1990s have produced useful results and addressed the problem of understanding at the atomic level (i.e., apart from collective effects and phase matching) the origin and features of the harmonic spectra appearing during the interaction of noble gases with laser pulses of intensities in the range $10^{14}-10^{17}$ W/cm^2 and photon energies in the range 1–5 eV, and their dependence on peak intensity I_0 and laser frequency ω. A related aim has been the possibility of creating harmonic spectra of high order using short and intense driving pulses from which very short pulses of very short wavelength can be extracted. Obviously, the shorter the source wavelength the quicker very short wavelengths from harmonics are reached. For example, Preston *et al.* [8] reported the observation of the 37th harmonic of the KrF laser (248.6 nm) from its interaction with He corresponding to an emitted photon wavelength of 67 Å.

In the works of [1–8,11–16], the question of the degree of participation of singly or even doubly [6] ionized atoms in the production of the high-order harmonics was raised and discussed. The emphasis of the related analysis was on the significance of the saturation intensity or effective intensity *vis à vis* the peak intensity. It has generally been accepted that due to the ionization that takes place during the rise of the intensity of the laser pulse, the neutral atom contributes only to the low-order part of the harmonic spectrum (for laser sources in the range 2–5 eV), while the high-order part is due to the ion (e.g., [8,11,12,14]). A dissenting view by Moiseyev and Weinhold appeared recently [16]. In earlier works, Sarukura *et al.* [1] and Sanpera *et al.* [14] they concluded that ''the intensity of 3.5×10^{15} W/cm^2 can be regarded as the field intensity where the transition from neutral He to He^+ occurs.'' They then proceeded with calculations of He-field interactions with $\hbar\omega=5$ eV and intensity up to 3.46×10^{15} W/cm^2 using a Floquet-type formalism with the assumption of an ac-field rather than of a pulse. They employed only two Floquet blocks: those of ^1S and $^1P^o$ symmetries. Their results led them to the conclusion that it is the neutral atom rather than the ion that produces harmonics of high order, and that the reason is the effective role of electron correlation in He, which keeps the two electrons apart. However, we took issue with this approach to the problem [17] since not only is the pulse temporal shape not accounted for, but, as was demonstrated via systematic calculations with electron correlation included [17], for such intensities the function space must also include state symmetries higher than $^1P^o$ for the overall calculation of the time-dependent wave function $\Psi(t)$ and the concomitant properties to reach convergence.

In this paper, we reexamine the issue of the capacity of neutral noble gases to produce harmonics of high order by paying attention to the effect of the rise time of the pulse. We note that Watson *et al.* [15] have found evidence in their numerical simulation, using a model for representing He, that the harmonic response depends critically on the shape of the pulse. In addition, Balcou *et al.* [4] experimentally demonstrated that the shorter the driving laser pulse the higher the order of the observed harmonics.

By solving *ab initio* the time-dependent Schrödinger equation (TDSE) for He interacting with laser pulses of 5 eV and peak intensities in the range $5\times10^{14}-3.46\times10^{15}$ W/cm^2, we found that the rise time, which in principle can be controlled experimentally to some extent, drastically influences the level of contribution of the neutral atom to the overall high-order harmonics spectrum.

II. THEORY

The solution of the TDSE and the calculation of the harmonic spectra of He were achieved by the state-specific ex-

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TABLE I. Probability $[P_b(t)]$, at two time instants, that the He atom interacting with laser pulses of temporal shape given by Eq. (1) with $T_r=4T$ and frequency $\hbar\omega=5$ ev, remains in its discrete spectrum, as a function of peak intensity I_0.

I_0 (W/cm^2)	$P_b(t)$ $t=4.25T$	$P_b(t)$ $t=13.75T$
5×10^{14}	0.994	0.943
6×10^{14}	0.990	0.940
7×10^{14}	0.987	0.747
8×10^{14}	0.984	0.538
9×10^{14}	0.978	0.693
1×10^{15}	0.971	0.641
2×10^{15}	0.908	0.306
3.46×10^{15}	0.531	0.015

pansion approach (SSEA) (e.g., [17–19]). Since the framework and the computational details of the SSEA have been presented in [17–19] and in references therein, here we only present the data related to this calculation and the analysis and significance of the results.

We consider that the He atom interacts with laser pulses of the following temporal shape:

$$f(t)=\begin{cases} t/T_r, & 0\leqslant t\leqslant T_r \\ 1, & T_r\leqslant t\leqslant T_r+T_f. \end{cases} \tag{1}$$

T_r is the linear rise time of the field strength, while during the time T_f the atom is exposed to the constant peak intensity. T_r was varied in the range 4–10 T (cycles) in search of the possible effect of the fast rise time of the pulse on the production of high-order harmonics. The choice of the upper limit for intensity of 3.46×10^{15} W/cm^2 was made in accordance with the observations and arguments given in [1,14,16] and in accordance with calculations indicating that the ''saturation intensity'' for this system seems to be around 9×10^{14} W/cm^2 [18,20].

The time-dependent wave function $\Psi(\mathbf{r},t)$, which describes the response of the He atom to the short intense laser pulse, was expanded in terms of state-specific wave functions for discrete, scattering, and doubly excited autoionizing states, each containing all the significant electronic structure, electron correlation, and normalization effects. Specifically, the two-electron basis set over which $\Psi(\mathbf{r},t)$ was allowed to evolve under the influence of the atom-laser pulse interaction consisted of the field-free bound states $1s^2$,$1snl$, the energy-normalized scattering states $1s\varepsilon l$, and the autoionizing states $2s^2$, $2p^2\,{}^1S$, $2s2p\,{}^1P$, and $2p^2\,{}^1D$. As in our previous work [17], the ground-state wave function $\varphi(1s^2)$ was computed as a ten-term expansion in the multiconfiguration Hartree-Fock (MCHF) scheme. The set of bound states $1snl$ went up to $n=20$ and $l=14$, with the $1sns$ ($n\leqslant7$) functions calculated at the state-specific MCHF level. The scattering states $1s\varepsilon l$ ($l\leqslant15$) were calculated numerically via the fixed-core Hartree-Fock method for energies in the range $0\leqslant\varepsilon\leqslant4.03$ a.u. with step $\Delta\varepsilon=0.004$ a.u. The energy cutoff, $\varepsilon=4.03$ a.u., was decided after the usual convergence tests showed that the harmonic spectrum remained unchanged with increasing energy range. These convergence results are shown explicitly in Sec. III. Finally, the $n=2$ intrashell autoionizing states were obtained from state-specific MCHF calculations according to previous theory ([21], and references therein).

TABLE II. Maximum order of emitted harmonics ($N_{\max}$) as a function of peak intensity I_0, by a He atom subjected to the intense field of the laser pulse given in Table I.

I_0 (W/cm^2)	$N_{\max}$
5×10^{14}	45
6×10^{14}	47
7×10^{14}	51
8×10^{14}	51
9×10^{14}	57
1×10^{15}	53
2×10^{15}	41
3.46×10^{15}	27

The resulting system of coupled integro-differential equations consisted of 16 327 equations. The solution of the TDSE was achieved with time step $\Delta t=T/128$. The induced-dipole moment, whose fast-Fourier transformation (FFT) gives the HOHG spectra, was calculated with the dipole operator in the velocity form with time step equal to $T/512$, which is sufficient to obtain convergent results. (Calculations with time steps equal to $T/1024$ provided identical HOHG spectra.) Tests of the sensitivity of the results of the FFT to the time interval showed that they remain essentially the same for three trial intervals, $(7T,10T)$, $(11T,14T)$, and $(7T,14T)$. The reported HOHG spectra were obtained for the last one.

III. RESULTS AND DISCUSSION

A physically meaningful parameter concerning the production mechanism of high-order harmonics is the time-dependent ionization probability or its complement, the probability of the atom remaining in the discrete spectrum. In Table I, the probability that the He atom is in the discrete spectrum for two time instants $(t=4.25T,13.75T)$ is presented for various peak intensities and $T_r=4T$. According to these results, which express the effect of the fast rise time, the atom essentially remains in its bound state at the beginning of its exposure to the peak intensity, except for the case of the maximum peak intensity $I_0=3.46\times10^{15}$ W/cm^2. The ionization during T_f increases. Nevertheless, at the end of the interaction the He atom survives in its neutral form, with its bound states appreciably populated. As the peak intensity increases, two opposing factors, with regard to the production of high-order harmonics, increase. These factors are the ionization and the polarization of He. The result of their essential interplay is shown in Table II, where the maximum order of the computed harmonics as a function of the peak intensities is given. These results show that for peak intensity $I_0=9\times10^{14}$ W/cm^2, the HOHG is more efficient. In fact, not

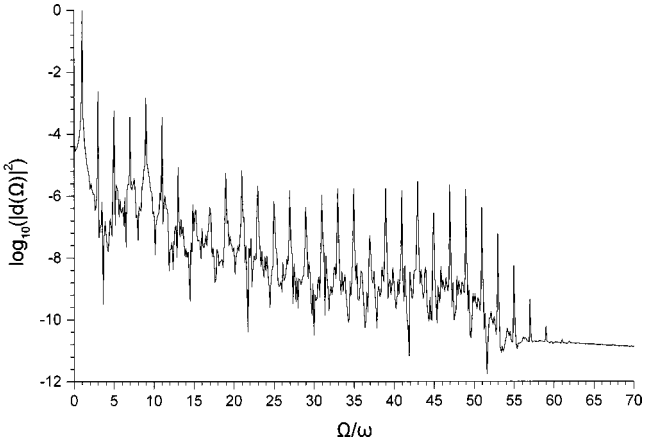

FIG. 1. Harmonic generation spectrum of He interacting with a laser pulse of $\hbar\omega=5$ eV, peak intensity $I_0=9\times10^{14}\,\mathrm{W/cm^2}$, and with pulse shape given by Eq. (1) with $T_r=4T$.

only is it here that the maximum harmonic order is observed but, also, the intensities of the harmonics are high and well above the background (Fig. 1). An increase of the laser peak intensity to $I_0=2\times10^{15}\,\mathrm{W/cm^2}$, where ionization is enhanced considerably, results in the decrease of the number of the harmonics from 57 to 41, while the intensity level from the 31st to the 41st harmonic is very low. For $I_0=3.46\times10^{15}\,\mathrm{W/cm^2}$, the number of harmonics decreases to 27 while the harmonic peaks are broadened with the two last ones being just above the background (Fig. 2).

We point out that we have already published SSEA results showing the extent to which ground-state correlation and doubly excited states affect the generation of harmonic spectra of He for $\hbar\omega=5$ eV as a function of intensity [17]. Specifically, it was found that the influence of ground-state correlation changes the spectra quantitatively as intensity increases. Nevertheless, for the range of intensities studied (6×10^{14}–$3.46\times10^{15}\,\mathrm{W/cm^2}$), the qualitative features of the harmonic spectra were not affected by ground-state correlation ([17], Fig. 4). On the other hand, the quantitative as well as qualitative effects of the lowest doubly excited states were insignificant for these laser parameters.

An interesting feature of the computed HOHG spectra is

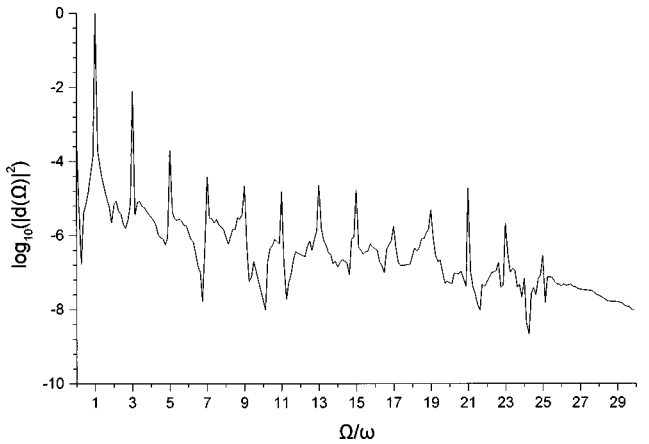

FIG. 2. As in Fig. 1 but for $I_0=3.46\times10^{15}\,\mathrm{W/cm^2}$.

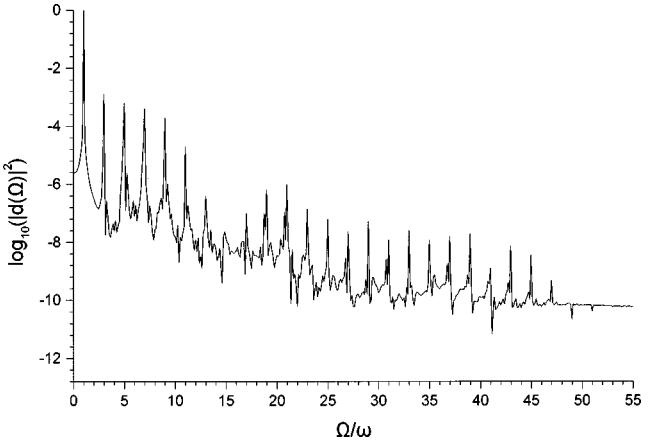

FIG. 3. As in Fig. 1 but for $I_0=6\times10^{14}\,\mathrm{W/cm^2}$.

that for the laser intensities used in this work, the 21st harmonic is of higher intensity compared to the 19th and 17th ones. For $I_0=3.46\times10^{15}\,\mathrm{W/cm^2}$, the 13th harmonic is also anomalous. These features are in agreement with the experimental finding that ''in He, the 13th and the 21st harmonics appeared slightly higher than the average shot'' ([1], p. 1670). Since such measurements are supposed to be influenced by a number of parameters, this agreement should be tested by additional experiments.

The increase of the linear rise time T_r results in the dramatic decrease of the number of the produced harmonics. For example, for $I_0=6\times10^{14}\,\mathrm{W/cm^2}$, by expanding the duration of the rise time to $T_r>8T$ we obtain a HOHG spectrum where the maximum harmonic order is 13, (for $T_r=4T$ the harmonics found are 45; Table II and Fig. 3), in agreement with previous calculations [12]. This dependence of the HOHG spectrum on the temporal pulse shape can be attributed to the interplay between the time-dependent ionization on the one hand and the polarization of the atomic state on the other. The slower the rise time of the pulse, the larger the ionization at the end of the turn on time. In fact, for $I_0=2\times10^{15}\,\mathrm{W/cm^2}$, by increasing T_r to $6T$ we obtain a HOHG spectrum with decreased intensity of the high-order harmonics.

The HOHG spectrum in Fig. 2 is in very good qualitative agreement with the experimental findings of Sarukura *et al.* [1], even for the 13th and 21st anomalous harmonics. This means that although consideration of the He^+ contribution is necessary for the complete interpretation of the HOHG spectra produced by the 5-eV laser pulses, the contribution of neutral He in the case of pulses with sharp rises is significant and in harmony with observation [1]. Of course, a more definitive answer with regard to the dynamics of the simultaneous He^+ contribution to the HOHG spectrum could be given if the ionic discrete and continuum states were included in the total time-dependent wave function $\Psi(\mathbf{r},t)$. In principle, the SSEA method allows this generalization, although this is not computationally economic at the present time.

Finally, we discuss the relation of our results to the predictions of a model [11,13] which consecutively combines the steps of ionization, propagation of the free electron in the

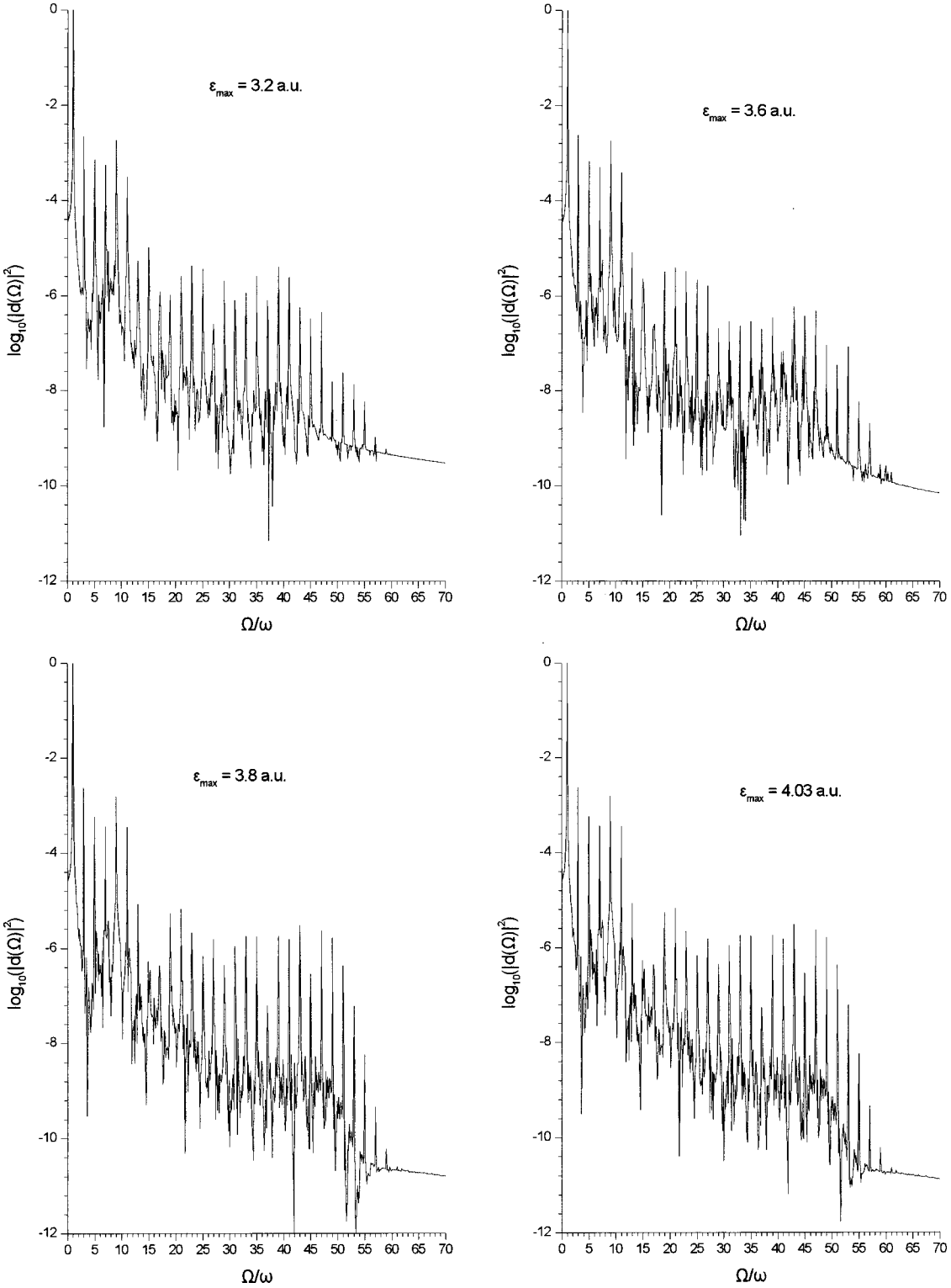

FIG. 4. As in Fig. 1 for four different energy ranges; i.e., for $\varepsilon_{\max}=3.2$ a.u., 3.6 a.u., 3.8 a.u., 4.03 a.u. These results are typical of the convergence characteristics of the SSEA calculations of high-order harmonic spectra.

strong laser field, and generation of high-order harmonics due to the collision of the returning electron with the core. This model predicts that the maximum order of the produced harmonics is given by

$$N=(I_p+3.17U_p)/\omega, \tag{2}$$

where I_p is the ionization potential and U_p is the ponderomotive energy.

The HOHG spectrum shown in Fig. 1 contains two groups of harmonics. The first group contains a plateau of the low-order harmonics, up to the 9th. At the 11th harmonic an abrupt decrease of the harmonic intensity appears at about

two or three orders of magnitude, in harmony with the ''cutoff'' rule of Eq. (2), according to which, for this case, N is equal to eight. The second group contains a plateau with the higher-order harmonics, up to the 51st. The production of harmonics beyond the order predicted by Eq. (2) has also been observed, with or without the distinct features of a plateau, in previous theoretical studies of HOHG [11,12,14,17,19,22,23]. The origin of the harmonics beyond the cutoff limit of Eq. (2) probably has to do with the fact that when the pulse rise time becomes shorter, the discrete states are occupied with appreciable probability (Table I). Thus parallel to the intense emission of the first plateau, whose main energy feature is approximated by Eq. (2), additional harmonics of lower intensity appear, depending on intensity and on pulse shape.

The above picture is supported by the HOHG spectrum obtained with $I_0=3.46\times10^{15}\,\mathrm{W/cm^2}$ (Fig. 2), where, during the first cycles after the fast rise time, the He atom is almost completely ionized (Table I). For this intensity, the 23rd harmonic is the last one well above the background. This means that the cutoff rule of Eq. (2), which for $I_0=3.46\times10^{15}\,\mathrm{W/cm^2}$, gives $N\approx18$, is only in approximate agreement with our findings. The same is true for $I_0=6\times10^{14}\,\mathrm{W/cm^2}$, for which we obtained a HOHG spectrum (Fig. 3) where the cutoff is as in Fig. 1 and the second plateau ends at the 45th harmonic. This peak intensity is supposed to represent the saturation intensity for pulses with rise times larger than four cycles, where the cutoff rule is supposed to be valid. Equation (2) gives $N=7$. Xu *et al.* [12] have reported $N=13$ in agreement with our findings [17], which were obtained with a pulse rise time equal to eight cycles.

A comment on the convergence of our calculations is needed here. Since the number of the state-specific bound and autoionizing states that are found to contribute to a particular property at the end of a SSEA calculation is always relatively small, regardless of the spectrum of the atom or molecule under consideration, the tests of convergence in our work focus on $\varepsilon_{\max}$, i.e., on the energy range of the continuous spectrum, and on the number of the energy-normalized scattering states in this range. For example, in a previous application to the most difficult case treated thus far in the literature, namely the HOHG spectrum of hydrogen for $\lambda=1064\,\mathrm{nm}$ and peak intensity $2\times10^{14}\,\mathrm{W/cm^2}$ [19], we reported on the results of such convergence tests (p. 4791 of [19]). An additional example taken from the present calculations is shown in Fig. 4, where four HOHG spectra are given for four different values of $\varepsilon_{\max}$ (3.2 a.u., 3.6 a.u., 3.8 a.u., and 4.03 a.u., the last being our final result). It is seen that the SSEA calculations have converged very well and that no artifacts are present. We note that we give the spectrum (the 1st harmonic peak is normalized to one) with the background included, and not just the peaks, as is often done in other works. With regard to the energy step $\Delta\varepsilon$, the calculations showed that no changes occur for values smaller than 0.004 a.u.

IV. CONCLUSION

The results and discussion presented in this work indicate that it is practical to influence the mechanisms producing HOHG by changing the temporal shape of the ultrashort intense laser pulse so as to affect its rise time. In the present case of He interacting with laser pulses of $\hbar\omega=5$ eV with peak intensities in the range 5×10^{14}–$3.46\times10^{15}\,\mathrm{W/cm^2}$, we found that the fast rise time of the pulse allows, with appreciable probability, the exposure of the neutral atom to very high peak intensities. Consequently, when the pulse is ''right'' it is neutral He rather than He^+ that is responsible for a much larger portion of the HOHG spectrum than previously thought [11,12,14,15]. In fact, this condition seems to have been met in experiment, since even the anomalous details of our calculated He HOHG spectrum (13th and 21st harmonics) agree with the pioneering measurements [1]. Finally, we point out that the dependence of the interaction on the pulse shape and on intensity influences the overall form of the harmonic spectrum, with reference point a first cutoff whose position is given approximately by Eq. (2), a relation first reported and discussed by Krause *et al.* [11] and Corcum [13].

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