Two-photon ionization of He$^+$ as a nonlinear optical effect in the soft-x-ray region

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We present numerical simulations of two-photon ionization of He$^+$ by 27th harmonic pulses of a Ti:sapphire laser. This process is chosen as a candidate for the experimental observation of a nonlinear optical effect in the soft-x-ray domain. We solve the time-dependent Schrödinger equation and evaluate the ionization probability as the number of electrons absorbed by the mask function at the outer radial boundary. Our model can address questions concerning possible saturation and quantum interference in ionization at high intensity and ultrashort pulse duration with no ambiguity. According to our results, in spite of saturation of ionization found at intensity higher than $10^{13}$ W/cm$^2$, the ionization probability by a 30 fs harmonic pulse with a peak intensity of $(2-5) \times 10^{13}$ W/cm$^2$, attainable with the latest progress in high-order harmonic generation, should be sufficiently high to put the second-order nonlinear optical process in the soft-x-ray region within experimental reach, along with desirable properties such as its nearly quadratic dependence on intensity and approximate linearity in pulse width. Our simulations also show that the variation of the yield with the pulse width of the 27th harmonic is no longer linear for a pulse width shorter than 5 fs, while the dependence on intensity is still quadratic. Our analysis on the ionization of He$^+$ by a double pulse in such an ultrashort pulse regime shows that the yield is not simply twice as large as that by a single pulse, but exhibits an oscillation of quantum origin with the interval and the phase difference between the two pulses. A simple model of the oscillation is discussed.

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I. INTRODUCTION

Nonlinear optics [1] is a field in physics with a long history that studies the nonlinear interaction of intense light with matter. Optical phenomena are called nonlinear when the material response to an applied optical field depends on the field strength in a nonlinear fashion. Although theoretical work [2] existed as early as 1931, it was not until the observation of multiphoton transitions between Zeeman sublevels of an atom in the radiofrequency domain [3] that nonlinear optical effects were experimentally demonstrated. Since the discovery of second-harmonic generation [4] and two-photon excitation [5] in 1961, remarkable advances in laser technology have enabled a wide variety of nonlinear optical phenomena [1], including sum- and difference-frequency generation, optical parametric oscillation, intensity-dependent refractive index, multiphoton ionization, and high-order harmonic generation (HHG). Due to considerable progress in the development of x-ray lasers, synchrotron radiation sources, free-electron lasers, and high-order harmonic sources, high-intensity pulses in vacuum and extreme ultraviolet (VUV-XUV) and soft-x-ray regions have now become available.

Among these light sources, the simplicity of HHG is advantageous in that it involves only table-top devices in short pulse duration down to sub-fs [6,7] and in high intensity. Using this type of radiation, two-photon and three-photon ionization [8–10] of rare-gas atoms in VUV and XUV regions was realized. Takahashi et al. [11] recently succeeded in the generation of 23rd to 31st harmonic pulses ($\lambda = 25.8 \text{ nm} \approx 34.8 \text{ nm}$) of a Ti:sapphire laser with a pulse width of 30 fs and an output energy as high as 300 nJ (or $4.5 \times 10^{10}$ photons) per pulse. When such a soft-x-ray pulse is focused to an area of 10 $\mu$m$^2$ using a commercially available soft-x-ray mirror, its average intensity may reach $10^{14}$ W/cm$^2$. Now it is time to tackle the problem of inducing nonlinear effects in the soft-x-ray domain. Chen et al. [12] proposed second-harmonic generation in the x-ray region using nonlinear optical crystals such as $\beta$-BaB$_2$O$_4$, LiB$_2$O$_5$, and CsB$_3$O$_5$. However, to our knowledge, its experimental demonstration has not been reported yet. Main obstacles may include high x-ray absorption by these crystals and difficulty in estimating their second-order nonlinear susceptibility.

In the present study, we perform numerical experiments of two-photon ionization of He$^+$ by the 27th harmonic ($\lambda = 29.6 \text{ eV}$) of a Ti:sapphire laser as a nonlinear optical effect in the soft-x-ray region. Why He$^+$? Because He$^+$ is easy to prepare from He, e.g., by means of optical field ionization using an intense laser or single-photon ionization with high-order harmonics. Since the photon energy $\hbar \omega_X = 41.85 \text{ eV}$ is not resonant with the $1s-2p$ transition energy $\hbar \omega_{21} = 40.8 \text{ eV}$ but close to it, high yield (defined as the ratio of the number of produced He$^+$ ions to that of initially present He$^+$ ions) is expected. Moreover, this simple hydrogenlike system allows us to evaluate yield precisely. In fact, the cross section $\sigma$ of this process calculated analytically by second-order perturbation theory is $2.9 \times 10^{-52} \text{ cm}^2 \text{s}$ [13]. This value is only one order of magnitude lower than the two-photon ionization cross section of Xe or Kr by the fifth harmonic (VUV) [10] and that of neutral He by XUV [14]. The number of photons per pulse at the 27th harmonic (up to $8 \times 10^{10}$), which is much larger than in the VUV and XUV...
ranges (up to a few $10^9$ [10]), can well compensate for this difference.

Why numerics, if we know the analytical cross section? The two-photon ionization of He$^+$ is equivalent to that of atomic hydrogen except for a factor stemming from the difference in nuclear charge. The history of analytical calculation of the latter is nearly as long as that of nonlinear optics experiments. A large amount of effort [15] has been devoted to the refinement of the evaluation of transition matrix elements since the first attempt by Zernik [16], and the cross section is tabulated or graphed in the literature [13,17–19]. Strictly speaking, however, these analytical values are valid only at the long-pulse and low-intensity limit. In order for this intensity domain, although the ionization rate begins to saturate at $I_{\text{sat}}$, the numerical result is slightly smaller than the analytical, after each time step the wave function is multiplied by a factor stemming from the difference in nuclear charge.

To study the interaction of a He$^+$ ion initially in the ground state ($1s$) with a soft-x-ray pulse, we solve the time-dependent Schrödinger equation in the length gauge, where $E_X(t)$ is the electric field of the pulse. Here we have assumed that the field is linearly polarized in the $z$ direction. Equation (2.1) is numerically integrated using the alternating direction-implicit (Peaceman-Rachford) method [26]. The numerical scheme is now well documented [26] and not difficult to implement. To reduce the difference between the discretized and analytical wave function, we scale the Coulomb potential by a few percent at the first grid point [27]. To prevent reflection of the wave function from the grid boundary, after each time step the wave function is multiplied by a $\cos^{1/8}$ mask function [27] that varies from 1 to 0 over a width of 2/9 of the maximum radius at the outer radial boundary. The ionization yield is evaluated as the decrease of the norm of the wave function on the grid, or equivalently, the number of electrons absorbed by the mask function. In typical calculations, we use a grid with a maximum radius of 125 a.u. and maximum number of partial waves $L_{\text{max}}=2$. The grid spacing is 0.125 a.u., and the time step is $1/65536$ of an optical cycle $\tau_2$ of the Ti:sapphire laser light, whose wavelength is 800 nm.

III. TWO-PHOTON IONIZATION PROBABILITY

In this section, we examine the two-photon ionization probability of He$^+$ by a short, intense 27th-harmonic pulse. As was already mentioned, for applications such as pulse duration measurements by autocorrelation, the ionization probability should be not only sufficiently high but also quadratic in intensity and linear in pulse duration. We consider a single pulse whose electric field is given by

$$E_X(t) = F_X(t) \sin(\omega_X t), \quad (3.1)$$

with $F_X$ being the pulse envelope, chosen to be Gaussian.

A. Dependence on intensity

Figure 1 shows the yield of He$^{2+}$ obtained by a 27th-harmonic pulse with a duration (FWHM) of 30 fs as a function of peak intensity $I_{\text{max}}$. The solid line is the numerical results. The ionization probability is $3.3 \times 10^{-4}$ at $I_{\text{max}} = 5 \times 10^{13}$ W/cm$^2$ and $5.6 \times 10^{-5}$ at $I_{\text{max}} = 2 \times 10^{13}$ W/cm$^2$, which is sufficiently high to be observed experimentally. The dotted line is calculated by the analytical formula $I_{\text{max}} \pi r(t)^2 \pi dt$, where $r(t)$ denotes the pulse intensity. Below $I_{\text{max}} = 10^{13}$ W/cm$^2$ both lines virtually overlap. On the other hand, the numerical result is slightly smaller than the analyti-
harmonic. In order to compare Eq. 3.1, we have performed a simulation with a constant proportional to intensity $I$ and the ratio $G/I$, where $G$ is the intensity limit and to the continuum, theory, the two-photon ionization rate $W$ is given by

$$W = \Gamma_{2p} \left( 1 - \frac{d}{\Omega_R} \right),$$

(3.2)

where $\Gamma_{2p}$ is the energy width of the $2p$ state for ionization to the continuum, $\Omega_R = \sqrt{\omega^2 + d^2}/4$ is the Rabi frequency, $d = \omega_X - \omega_21$ is the detuning, and $V$ is the dipole matrix element between $1s$ and $2p$ states. Since $\Gamma_{2p}$ and $V$ are proportional to intensity $I$, the rate $W$ is proportional to $I^2$ at the low intensity limit and to $I$ at the high intensity limit [25]. Especially at the low intensity limit, $W$ has the following form:

$$W_{low} = 2\Gamma_{2p} \left( \frac{V}{d} \right)^2,$$

(3.3)

and the ratio $R$ of $W$ to $W_{low}$ is given by

$$R = \frac{2}{z} \left( 1 - \frac{1}{\sqrt{1+z}} \right),$$

(3.4)

where $z = 4V^2/d^2$. $z = I/(3.77 \times 10^{14}$ W/cm$^2$) for the 27th harmonic. In order to compare Eq. (3.4) with a numerical result, we have performed a simulation with a constant intensity of $5 \times 10^{13}$ W/cm$^2$ preceded by a Gaussian ramp which peaks at $t = 133.4$ fs with a FWHM of 90 fs, and we obtained the ionization rate $R_{sim} = 1.45 \times 10^{10}$ s$^{-1}$. This value is 0.90 of that calculated from the quadratic relation $\sigma I^2 = 1.61 \times 10^{10}$ s$^{-1}$. This ratio agrees well with the value 0.91 obtained from Eq. (3.4). Thus, the theory by Haberland et al. [29], though, strictly speaking, valid only for the long pulse limit, is useful to understand the saturation of the ionization probability as in Fig. 1.

We plot the yield of He$^{2+}$ as a function of photon energy $\hbar \omega_X$, of a Gaussian pulse with a duration of 30 fs. The solid and dotted lines correspond to peak intensity $I_{max} = 5 \times 10^{13}$ W/cm$^2$ and $2 \times 10^{13}$ W/cm$^2$, respectively.

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We plot the yield of He$^{2+}$ as a function of photon energy $\hbar \omega_X$ for $I_{max} = 2 \times 10^{13}$ W/cm$^2$ and $5 \times 10^{13}$ W/cm$^2$ in Fig. 2. We also plot their ratio in Fig. 3. As can be seen from these two figures, if the soft-x-ray pulse were closer to the resonance (40.8 eV) than the 27th harmonic is, an approximately quadratic dependence on intensity, one of the desirable properties, would be no longer guaranteed. On the other hand, if it were farther away from the resonance, the ionization probability would be too low. As for the 27th harmonic of a Ti:sapphire laser pulse, the ionization probability is high enough and, at the same time, approximately quadratic in $I_{max}$ even at $I_{max}$

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The Gaussian 27th harmonic pulse on its pulse duration function of potential of He increases when the pulse duration decreases. The uncertainty to the uncertainty principle, the width of photon energy in- and then decreases. This can be understood as follows: due to the spectrum shifts from 29.3 eV at found that the peak position of the photoelectron energy result, the ionization probability is augmented. In fact, we have so short, the 2 harmonic is situated at a very convenient photon energy.

In order to examine the dependence of the yield on intensity for such an ultrashort pulse, we plot the ratio of the yield at \( I_{\text{max}} = 5 \times 10^{13} \text{ W/cm}^2 \) to that at \( I_{\text{max}} = 2 \times 10^{13} \text{ W/cm}^2 \) as a function of pulse width \( t_0 \) (FWHM) of a Gaussian pulse. The value expected for quadratic yield-intensity relation is 6.25. Another point to notice is that the yield is nearly perfectly quadratic in \( I_{\text{max}} \) for \( t_0 < 1 \text{ fs} \), for which the bandwidth is again so large that most of it is sufficiently detuned for the quadratic relation.

In Fig. 6, we plot the ratio of the ionization probability by a 30 fs pulse to that by a 15 fs pulse as a function of photon energy. The peak intensity is \( 5 \times 10^{13} \text{ W/cm}^2 \). The first autocorrelation experiments in the soft-x-ray range might be performed with such values of pulse width. The yield is approximately linear in pulse duration as long as \( h\omega_X \) is not too close to the resonance.

**IV. IONIZATION BY DOUBLE PULSE**

In this section, we study the two-photon ionization of He\(^+\) by a 27th-harmonic double pulse in detail, especially in the case in which the duration of each pulse is 1 fs. We consider the electric field given by

\[
E_X(t) = F_1(t)\sin(\omega_X t + \phi_1) + F_2(t)\sin(\omega_X t + \phi_2),
\]

(4.1)

\[\text{FIG. 4. The He}^{2+} \text{ yield divided by the pulse duration (FWHM) } t_0 \text{ of a Gaussian 27th harmonic pulse with a peak intensity of } 5 \times 10^{13} \text{ W/cm}^2 \text{ (solid line) and } 2 \times 10^{13} \text{ W/cm}^2 \text{ (dotted line) as a function of } t_0. \text{ Note that this quantity would be constant for the case of linear dependence of the yield on pulse duration.}
\]

\[\text{FIG. 5. The ratio of the He}^{2+} \text{ yield at } I_{\text{max}} = 5 \times 10^{13} \text{ W/cm}^2 \text{ to that at } I_{\text{max}} = 2 \times 10^{13} \text{ W/cm}^2 \text{ as a function of pulse width } t_0 \text{ (FWHM) of a Gaussian pulse. The value expected for quadratic yield-intensity relation is 6.25.}
\]

\[\text{FIG. 6. The ratio of the He}^{2+} \text{ yield by a Gaussian 27th harmonic pulse at } I_{\text{max}} = 5 \times 10^{13} \text{ W/cm}^2 \text{ with a duration of 30 fs to the yield by a pulse with a duration of 15 fs.}
\]
with $F_{1,2}$ and $\phi_{1,2}$ being the envelope, chosen to be Gaussian, and the phase of each pulse, respectively. The breakdown of the linear dependence of the ionization yield on intensity has a remarkable consequence that the yield of He$^{2+}$ produced by a pulse train is not proportional to the number of pulses contained in the train. An example is illustrated in Fig. 7. The solid line is the temporal evolution of the population of He$^{2+}$ produced by two consecutive 1-fs 27th-harmonic pulses, each of which has a peak intensity of $5 \times 10^{13}$ W/cm$^2$, and the interval between the two pulse peaks is 3 fs.

Let us now, however, look at this situation from a completely different point of view. The breakdown of linearity is a quantum effect which cannot be observed using longer pulses. The progress of the pulse compression technique may enable the observation of such quantum effects. It is, therefore, important to study how the final yield of He$^{2+}$ by double pulse irradiation depends on the interval between the two pulses and on their relative phase.

Figure 8 shows the He$^{2+}$ yield by a 1-fs 27th-harmonic double pulse as a function of $\phi_2$. Each pulse has a peak intensity of $5 \times 10^{13}$ W/cm$^2$ at $t=2t_L$ and $2t_L+4.148$ fs, respectively. The solid line is for $\phi_1=0$ and the dashed line is for $\phi_1=\pi/2$. We also plot the yield by a single pulse as a function of $\phi_1$, corresponding to the energy difference $\hbar \omega_{21}$ = 3/2 a.u. between 2p and 1s. Its period $T_2$ is 0.1013 fs. The relation between the phase of the oscillating dipole moment and that of the second soft-x-ray pulse causes an oscillation in the He$^{2+}$ yield as seen in Fig. 8.

In order to confirm this further, we have performed simulations by varying the interval $\Delta t$ between the two pulse peaks. $\phi_1$ is fixed to zero and $\phi_2=-\omega_2 \Delta t$. Note that by such a choice of $\phi_1$ and $\phi_2$, not only the intensity of the second pulse but also the electric field itself is shifted by $\Delta t$ with respect to the first pulse. This is a situation realized in usual autocorrelation experiments. The simulation result is plotted in Fig. 9. It is clear that the yield oscillates with the same period as the dipole moment ($T_2$).

Let us now investigate how the center and the amplitude of the oscillation as in Figs. 8 and 9 depend on pulse inten-

FIG. 7. Temporal evolution of the He$^{2+}$ population by two consecutive 27th harmonic pulses (solid line) and 23rd harmonic pulses (dotted line), respectively. The width (FWHM) of each pulse is 1 fs, the peak intensity is $5 \times 10^{13}$ W/cm$^2$, and the interval between the two pulse peaks is 3 fs.

FIG. 8. He$^{2+}$ yield by a 27th harmonic double pulse whose electric field is of the form Eq. (4.1) as a function of $\phi_2$. Each pulse with a duration of 1 fs has a peak intensity of $5 \times 10^{13}$ W/cm$^2$ at $t=2t_L$ and $2t_L+4.148$ fs, respectively. Solid curve: $\phi_1=0$. Dotted curve: $\phi_1=\pi/2$. Dashed line: He$^{2+}$ yield by a single pulse as a function of $\phi_1$.

FIG. 9. He$^{2+}$ yield by a 27th harmonic double pulse whose center and amplitude of the oscillation as functions of the interval $\Delta t$ between the two pulse peaks. Each pulse has a Gaussian temporal profile with a duration of 1 fs and a peak intensity of $5 \times 10^{13}$ W/cm$^2$. The first pulse peaks at $t=2t_L$. $\phi_1=0$ and $\phi_2=-\omega_2 \Delta t$. 

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sity. We have performed simulations by varying the peak intensity of the second pulse while fixing that of the first pulse to $5 \times 10^{13}$ W/cm$^2$, and we obtained the center and the amplitude, as defined in Fig. 8, of the oscillation of the number of He$^{2+}$ produced by the second pulse. Their dependence on the peak intensity of the second pulse is shown in Fig. 10. The production of He$^{2+}$ by the second pulse proceeds in two ways: one-photon ionization from $2p$ and two-photon ionization from $1s$. By taking their interference into account, the transition amplitude of the entire process has the following form:

$$\sqrt{(\alpha_0 I_1)(\alpha_1 I_2)} + \sqrt{(1-\alpha_0 I_1)(\alpha_2 I_2^*)e^{i(\Delta \phi + \phi_0)}},$$

(4.2)

where $I_{1,2}$ is the peak intensity of the first and the second pulse, respectively. $\alpha_0 I_1$ describes the excitation of the $2p$ level by the first pulse, $\alpha_1 I_2$ the ionization from $2p$ to the continuum, and $\alpha_2 I_2^*$ the two-photon ionization from $1s$. Then we can write the number of He$^{2+}$ produced by the second pulse as

$$a I_2 + b I_2^* + c I_2^{3/2} \cos(\Delta \phi + \phi_0),$$

(4.3)

where

$$a = \alpha_0 \alpha_1 I_1,$$

(4.4)

$$b = (1-\alpha_0 I_1) \alpha_2 = \alpha_2,$$

(4.5)

$$c = 2 \sqrt{ab}.$$  

(4.6)

The simulation result in Fig. 10 is very well fitted by a function of the form in Eq. (4.3) as is shown by dotted curves, though a small discrepancy is found for the highest value of $I_2$. The coefficients $a$, $b$, and $c$ obtained by the fitting are listed in Table I along with their values for the case of $I_1$.

TABLE I. The parameters $a$, $b$, and $c$ obtained by fitting the numerically obtained values of center and amplitude of the oscillation of the He$^{2+}$ yield by the second pulse with Eq. (4.3). The intensity is in W/cm$^2$, $c_{\text{th}}$ is the value of $c$ calculated from $a$ and $b$ with Eq. (4.6). The numbers in brackets indicate powers of 10.

<table>
<thead>
<tr>
<th>Intensity</th>
<th>$a$</th>
<th>$b$</th>
<th>$c$</th>
<th>$c_{\text{th}}$</th>
</tr>
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$=2 \times 10^{13}$ W/cm$^2$. From Table I, $a$ is proportional to $I_1$ while $b$ is approximately constant as is expected from Eqs. (4.4) and (4.5), respectively. The relation Eq. (4.6) is also roughly satisfied. Hence, the oscillation of the He$^{2+}$ yield can be explained essentially by the simple phenomenological model discussed above.

V. CONCLUSIONS

Using numerical simulations, we have examined the suitability of two-photon ionization of He$^+$ by a 27th harmonic pulse of a Ti:sapphire laser as a system for the experimental observation of a nonlinear optical effect in the soft-x-ray region. We have considered the pulse parameters accessible with a state-of-the-art high-order harmonic generation technique, namely, intensity up to $5 \times 10^{13}$ W/cm$^2$ and pulse width as short as $1–30$ fs. Our simulation results have revealed that such an ultrashort intense harmonic pulse should be sufficient to render realistic the observation of the second-order nonlinear optical process. Although the saturation effect reduces the ionization probability by about 10% at $I_{\text{max}}=5 \times 10^{13}$ W/cm$^2$, the variation of the He$^{2+}$ yield with intensity is nearly quadratic even in this high-intensity region. Moreover, the ionization is approximately proportional to the pulse width at $t_0>5$ fs. These properties are desirable for the experimental demonstration of the nonlinear effect.

Our analysis of two-photon ionization by a harmonic double pulse with a duration of 1 fs has shown that the interference between the direct two-photon process from the ground state and the single-photon process from the 2p state populated by the first pulse induces oscillation in the He$^{2+}$ yield with the interval and the phase difference between the two pulses. The excitation to the 2p level is possible due to the large bandwidth associated with the ultrashort pulse width. The oscillation period is precisely predictable from the 1s-2p transition energy. The detection of such an oscillation may serve as a unique test of quantum mechanics.

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