Photoemission and Ionization of He^+ under Simultaneous Irradiation of Fundamental Laser and High-Order Harmonic Pulses

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We theoretically study the response of He^+ exposed simultaneously to an intense Ti:sapphire laser and its 27th or 13th harmonic pulses. High-order harmonic emission from He^+ is enhanced by many orders of magnitude compared with the case of the fundamental pulse alone. Moreover, while an individual 10 fs laser (wavelength $\lambda_F = 800$ nm) or its 27th harmonic pulse with a peak intensity of 3×10^{14} and 10^{13} W/cm^2, respectively, ionizes no more than 5×10^{-6} of He^+, their combined pulse leads to a surprisingly high He^{2+} yield of 17%.

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The recent progress in the high-order harmonic generation (HHG) technique has enabled the production of high-power coherent soft x-ray and extreme ultraviolet (xuv) pulses [1–4]. Takahashi *et al.* [1] have recently succeeded in the generation of 27th harmonic pulses (wavelength $\lambda_H = 29.6$ nm) of a Ti:sapphire laser with a pulse width smaller than 30 fs and an output energy of 0.33 μJ per pulse. The same authors [2] have subsequently reported the generation of 11th, 13th, and 15th harmonic pulses ($\lambda_H = 72.7$, 62.3, and 54 nm) with an output energy as high as 7, 4.7, and 1 μJ, respectively, while Hergott *et al.* [3] has generated 15th harmonic pulses ($\lambda_H = 53.3$ nm) of a Ti:sapphire laser with an energy of 1.9 μJ per pulse. Yoshitomi *et al.* [4] have recently generated 5th harmonic pulses ($\lambda_H = 49.7$ nm) of a KrF excimer laser with an energy of 1.2 μJ. When these harmonic pulses are focused to an area of 10 μm^2 using a commercially available mirror, its average intensity may reach 10^{14} W/cm^2 in the soft x-ray and 10^{15} W/cm^2 in the xuv domain. With such intense light sources at hand, study of nonlinear optical processes in the xuv and soft x-ray ranges, limited thus far to longer wavelength [5–7], might become possible. In this context, Ishikawa and Midorikawa [8] have proposed two-photon ionization of He^+ by Ti:sapphire 27th harmonic (H27) pulses as a candidate for the experimental observation of a nonlinear optical effect in the soft x-ray domain, and Nakajima and Nikolopoulos [9] have theoretically shown that two-photon double ionization of He may be observed with the same harmonic.

In this Letter, we study how a He^+ ion behaves when subject to a fundamental laser pulse and an intense 27th or 13th harmonic pulse at the same time. There exist studies on the behavior of a hydrogen atom irradiated simultaneously by a fundamental and its harmonic (H3 [10], H7 [11], and H11 [12]) pulses [10–12]. It should be noted, however, that due to its high ionization potential (54.4 eV), He^+ may be applied to generate harmonics of higher orders than possible today and is not ionized by a single H27 and H13 photon in contrast to a hydrogen atom. Moreover, He^+ ions are experimentally more friendly since they can easily be produced, e.g., through single-photon ionization of He by 27th harmonics. In the present study, we are interested especially in the effects of the simultaneous irradiation on harmonic photoemission and ionization. The fundamental laser pulse can hardly ionize He^+ as we will see later. Although thanks to high ionization potential the harmonic spectrum from this ion would have higher cutoff energy than in the case of commonly used rare-gas atoms [1–4], the conversion efficiency is extremely low due to the small ionization probability. It is expected, however, that the addition of a Ti:sapphire 27th or 13th harmonic facilitates ionization and photoemission, either through two-color frequency mixing or by assisting transition to the $2p$ or $2s$ levels. Our results based on numerical solution of the time-dependent Schrödinger equation show in fact that the combination of fundamental laser and its 27th or 13th harmonic pulses dramatically enhance both high-order harmonic generation and ionization by more than 10 orders of magnitude.

To study the interaction of a He^+ ion with a combined laser and harmonic pulse, we solve the time-dependent Schrödinger equation in the length gauge,

$$i\frac{\partial \Phi(\mathbf{r}, t)}{\partial t} = \left[-\frac{1}{2}\nabla^2 - \frac{2}{r} - zE(t) \right]\Phi(\mathbf{r}, t), \qquad (1)$$

where $E(t)$ is the electric field of the pulse. Here we have assumed that the field is linearly polarized in the z direction. Equation (1) is numerically integrated using the alternating direction implicit (Peaceman-Rachford) method [13]. 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 [14]. 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 [14] 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. The electric field $E(t)$ is assumed to be given by

$$E(t) = F_F(t)\sin(\omega t) + F_H(t)\sin(n\omega t + \phi), \qquad (2)$$

with $F_F(t)$ and $F_H(t)$ being the pulse envelope of the fundamental and harmonic pulse, respectively, chosen to be Gaussian with a duration (full width at half maximum) of 10 fs, ω the angular frequency of the fundamental pulse, n the harmonic order, and ϕ the relative phase. In typical calculations, we use a grid with a maximum radius of 125–250 a.u. and maximum number of partial waves $L_{\max} = 100$–200. The grid spacing is 0.125 a.u., and the time step is 1/65536 of an optical cycle t_L of the Ti:sapphire laser light. The fundamental wavelength is 800 nm unless otherwise stated. Since we have found that the results are not sensitive to ϕ, we set $\phi = 0$ hereafter.

In Fig. 1, we show the harmonic photoemission spectrum from He^+ for the case of simultaneous fundamental and 27th harmonic (H27) irradiation. The peak fundamental intensity I_F is 3×10^{14} W/cm^2, and the peak H27 intensity I_{H27} is 10^{13} W/cm^2. The spectrum is obtained from the Fourier transform of the dipole acceleration. For comparison, we also show the spectra obtained when only the fundamental pulse of the same intensity is applied to a He^+ ion and a hydrogen atom. The general feature of HHG can be explained by the classical model, usually called the *simpleman's theory* [15]: An electron is lifted to the continuum with no kinetic energy, the subsequent motion is governed classically by an oscillating electric field, and a harmonic is emitted upon recombination. As a result, the highest harmonic (the cutoff) has an energy E_c given by [15]

$$E_c = I_p + 3.17U_p, \qquad (3)$$

where I_p and U_p are the ionization potential and the ponderomotive energy, respectively. This equation tells us that a way of getting higher harmonics is to use an atom or an ion with larger I_p. For the case of Fig. 1, the cutoff energy is calculated from Eq. (3) to be 70 eV (H45) for H and 111 eV (H73) for He^+. The cutoff positions in Fig. 1 agree with these values, and harmonics of much higher orders are generated from He^+ than from H. We note, however, that the harmonic intensity from He^+ when only the laser is applied is extremely low compared with the case of H. This is because the large ionization potential hinders ionization, the first step of the simpleman's theory. The situation changes completely if the H27 pulse is simultaneously applied to He^+. From Fig. 1, we can see that the conversion efficiency is enhanced by about 17 orders of magnitude. Moreover, the advantage of high cutoff is preserved. Figure 2 shows the harmonic photoemission spectrum from He^+ for the case of simultaneous fundamental and H13 irradiation. Again the harmonic intensity is enhanced by more than 10 orders of magnitude compared to the case of the laser pulse alone. We have varied the fundamental wavelength between 750 and 850 nm and found similar enhancement over the entire range.

The effects found in Figs. 1 and 2, by far more dramatic than previously reported moderate enhancement [10,16], can be qualitatively understood as follows. The H27 photon energy (41.85 eV) is close to the $1s$-$2p$ transition energy of 40.8 eV, and the H13 photon (20.15 eV) is nearly two-photon resonant with the $1s$-$2s$ transition. Moreover, the $2p$ and $2s$ levels are broadened due to the laser-induced dynamic Stark effect. As a consequence, the

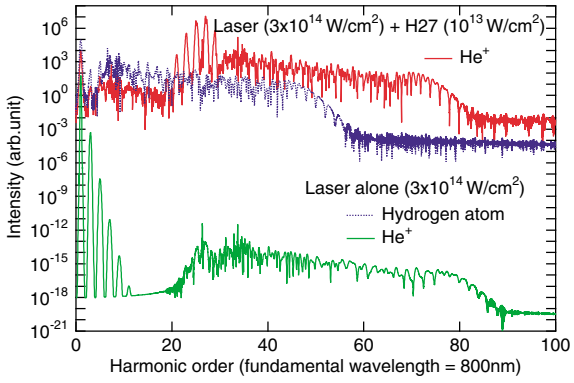

FIG. 1 (color online). Upper solid curve: Harmonic spectrum from He^+ exposed to a Gaussian combined fundamental and its 27th harmonic pulse with a duration (FWHM) of 10 fs, the former with a peak intensity of 3×10^{14} W/cm^2 and the latter 10^{13} W/cm^2. The fundamental wavelength is 800 nm. Lower solid and dotted curves: Harmonic spectrum from He^+ and a hydrogen atom, respectively, exposed to the fundamental pulse alone. Nearly straight lines beyond the cutoff energy are due to numerical noise.

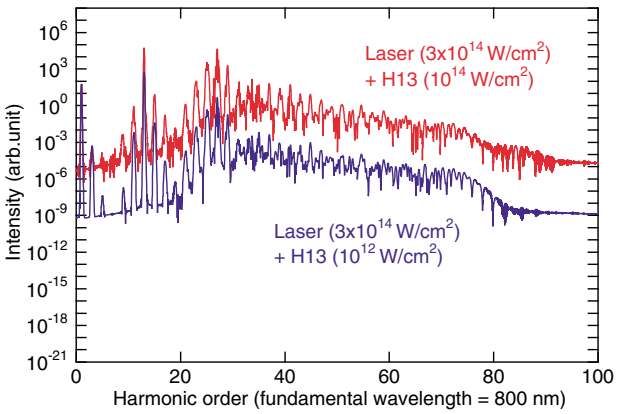

FIG. 2 (color online). Harmonic spectrum from He^+ exposed to a Gaussian combined fundamental and its 13th harmonic pulse with a duration (FWHM) of 10 fs, the former with a peak intensity of 3×10^{14} W/cm^2, and the latter 10^{14} W/cm^2 (upper curve) and 10^{12} W/cm^2 (lower curve). The fundamental wavelength is 800 nm. Note that the horizontal axis is of the same scale as in Fig. 1.

H27 and H13 promote transition to a virtual state near these levels. Depending on the laser wavelength, resonant excitation of $2s$ or $2p$ levels, in which fundamental photons may be involved in addition to harmonic photons, also takes place. In fact, the $2s$ level is excited through two-color two-photon transition for the case of Fig. 1 at $\lambda_F = 800$ nm, as we will see below in Fig. 5, and about 8% of electron population is left in the $2s$ level after the pulse. Since this level lies only 13.6 eV below the ionization threshold, the electron can now be lifted to the continuum by the laser pulse much more easily. Thus, the HHG efficiency is largely increased. We have found that the harmonic spectrum from the superposition of the $1s$ (92%) and $2s$ (8%) states subject to the laser pulse alone is strikingly similar to the one in Fig. 1. Hence, the effect may also be interpreted as harmonic generation from a coherent superposition of states [17]. In this sense, the H27 addition provides a means to prepare such a superposition, which was not explained in Ref. [17]. On the other hand, at a different fundamental wavelength, e.g., at $\lambda_F = 785$ nm, there is practically no real excitation. Nevertheless, the photoemission enhancement (not shown) is still dramatic. In this case, the H27 pulse promotes transition to a virtual state near the $2p$ level. Again, the electron can easily be lifted from this state to the continuum by the fundamental pulse, and the HHG efficiency is largely augmented. This may also be viewed as two-color frequency mixing enhanced by the presence of a near-resonant intermediate level. In general, both mechanisms of harmonic generation from a coherent superposition of states and two-color frequency mixing coexist, and their relative importance depends on fundamental wavelength. A similar discussion applies to the case of the H13 addition. The comparison of the two curves in Fig. 2 reveals that the harmonic spectrum is proportional to I_{H13}^2, where I_{H13} denotes the H13 peak intensity. We have also confirmed that the photoemission intensity is proportional to I_{H27} for the case of H27. These observations are compatible with the discussion above.

Let us now examine ionization probability. Table I summarizes the He^{2+} yield. As can be expected from the discussion in the preceding paragraph, the ionization probability by the combined pulse is by many orders of magnitude higher than that by the fundamental laser pulse alone. Especially dramatic enhancement is found in the case of the combined fundamental and H27 pulses: The He^{2+} yield is increased by orders of magnitude also with respect to the case of the H27 irradiation alone. This reflects the fact that field ionization from the $2s$ state by the fundamental pulse is much more efficient than two-photon ionization from the ground state by the H27 pulse. In Fig. 3, we plot the He^{2+} yield as a function of I_{H27} for a fixed fundamental intensity of 3×10^{14} W/cm^2. The ionization probability is linear in I_{H27} except for the saturation at $I_{\mathrm{H27}} > 10^{13}$ W/cm^2. This is compatible with our view that an H27 photon promotes $1s$-$2s$ two-color two-photon transition, followed by field ionization.

Figure 4 shows the dependence of the He^{2+} yield on the peak intensity I_F of the fundamental pulse. Surprisingly, the yield is not monotonically increasing in I_F. In order to clarify the origin of this counterintuitive behavior, we have calculated the dependence of the He^{2+} yield on the fundamental wavelength λ_F for different values of fundamental intensity. The results are shown in Fig. 5. The curve for $I_F = 5 \times 10^{13}$ W/cm^2 peaks around $\lambda_F = 820$ nm, for which the H27 photon energy is exactly resonant with the $1s$-$2p$ transition. At $I_F = 8 \times 10^{13}$ W/cm^2, we can see a second peak around $\lambda_F = 793$ nm, for which $26\hbar\omega$ $(= 27\hbar\omega - \hbar\omega)$ coincides with the $1s$-$2s$ transition energy. This indicates that at this wavelength the $2s$ level is resonantly populated through two-color two-photon excitation. With increasing laser intensity, the peaks are shifted to longer wavelengths due to the laser-induced dynamic Stark effect, and at $I_F = 3 \times 10^{14}$ W/cm^2 the second peak, now higher than the first one, is located around 800 nm, which leads to high ionization yield at this wavelength. At $I_F = 3 \times 10^{14}$ W/cm^2, we can also see a third peak around $\lambda_F = 770$ nm corresponding to two-color $2p$ excitation

TABLE I. The He^{2+} yield for various combinations of a Gaussian fundamental and its 27th or 13th harmonic pulses with a duration (FWHM) of 10 fs and a peak intensity listed in the table. $\lambda_F = 800$ nm.

I_F (W/cm^2)	I_{H27} (W/cm^2)	I_{H13} (W/cm^2)	He^{2+} yield
3×10^{14}	⋯	⋯	2.29×10^{-15}
⋯	10^{13}	⋯	4.79×10^{-6}
3×10^{14}	10^{13}	⋯	0.173
⋯	⋯	10^{14}	1.25×10^{-4}
3×10^{14}	⋯	10^{14}	2.04×10^{-4}

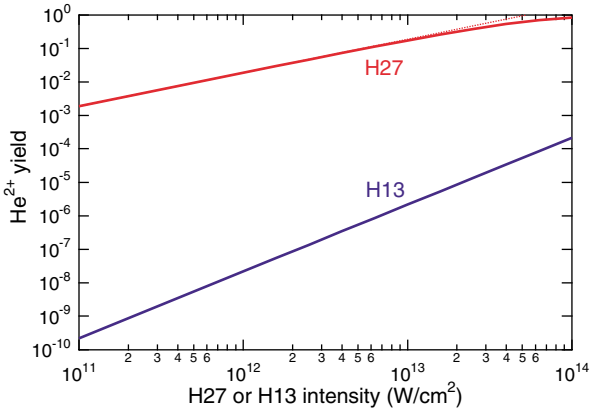

FIG. 3 (color online). The He^{2+} yield as a function of peak intensity of the 27th (upper curve) and 13th (lower curve) harmonic pulse when He^+ is exposed to a Gaussian combined fundamental and its 27th or 13th harmonic pulse with a duration (FWHM) of 10 fs. The wavelength λ_F and the peak intensity I_F of the fundamental pulse are 800 nm and 3×10^{14} W/cm^2, respectively.

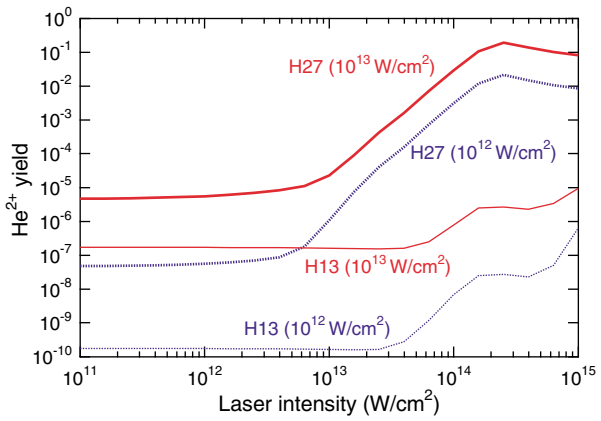

FIG. 4 (color online). The He^{2+} yield as a function of peak intensity I_F of the fundamental laser pulse when He^+ is exposed to a Gaussian combined fundamental and its 27th or 13th harmonic pulse with a duration (FWHM) of 10 fs. The fundamental wavelength λ_F is 800 nm. The peak intensity of each harmonic pulse is indicated in the figure along with its order.

involving one H27 and two fundamental photons. The dynamic Stark effect induces not only the peak shift but also the peak broadening, which results in the decrease of peak heights at higher fundamental intensity, leading to the decrease of the ionization yield at $I_F > 3 \times 10^{14}$ W/cm^2 in Fig. 4. Thus, the role of the fundamental laser pulse is threefold: to lift the electron in an excited (real or virtual) level to the continuum through optical-field ionization, to assist $1s$-$2s$, $2p$ transitions through two-color excitation, and to induce dynamic Stark shift and broadening. The interplay of these three leads to a complicated behavior seen in Fig. 4.

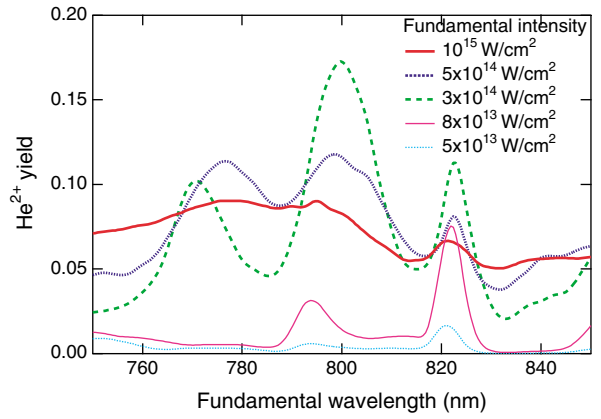

FIG. 5 (color online). The He^{2+} yield as a function of fundamental wavelength when He^+ is exposed to a Gaussian combined fundamental and its 27th harmonic pulse with a duration (FWHM) of 10 fs. The peak intensity I_{H27} of the 27th harmonic pulse is fixed at 10^{13} W/cm^2, and we plot the results for five different values of fundamental peak intensity I_F indicated in the figure. Note that the wavelength of the 27th harmonic varies with that of the fundamental pulse.

In conclusion, we have investigated the ionization and photoemission behavior of a He^+ ion under a combined field of an ultrashort intense fundamental laser and its high-order harmonic pulses. Specifically, we have considered the 27th and 13th harmonics, for which especially high intensity can now be achieved with the HHG technique. The response of He^+ is totally different from that in the case of the fundamental pulse alone or the harmonic pulse alone. The harmonic pulse plays an essential role in transition to $2p$ and $2s$ states or to a nearby virtual state, and the fundamental pulse is important in optical-field ionization from these levels as well as in $1s$-$2p$, $2s$ transitions through two-color excitation; their copresence leads to dramatic enhancement of both harmonic photoemission intensity and He^{2+} yield. The efficient H27- or H13-assisted harmonic generation might open a way to develop an intense light source of an even shorter wavelength than available today.

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