Appl. Phys. B 78, 855–858 (2004)
DOI: 10.1007/s00340-004-1459-9

Applied Physics B
Lasers and Optics

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Enhanced photoemission and ionization of He^+ by simultaneous irradiation of fundamental laser and high-order harmonic pulses

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Received: 21 October 2003
Published online: 7 April 2004 •

ABSTRACT We simulate the response of He^+ exposed simultaneously to fundamental and 27th harmonic pulses from an intense Ti : sapphire laser. High-order harmonic emission from He^+ is enhanced by 17 orders of magnitude compared with the case of the fundamental pulse alone. Moreover, while an individual 10 fs laser with a fundamental wavelength of 800 nm and a peak intensity of 3×10^{14} W/cm^2, or its 27th harmonic pulse with a peak intensity of 10^{13} W/cm^2, ionizes no more than 5×10^{-6} of He^+, their combined pulses lead to a surprisingly high He^{2+} yield of 17%. The underlying mechanism is either harmonic generation from a coherent superposition of states or two-color frequency mixing, depending on the laser wavelength.

PACS 32.80.Rm; 42.50.Hz; 42.65.Ky

1 Introduction

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 [1–7]. 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 an energy of 0.33 μJ. 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 energies as high as 7, 4.7, and 1 μJ, respectively, while Hergott et al. [6] have generated 15th harmonic pulses ($\lambda_H = 53.3$ nm) of a Ti : sapphire laser with an energy of 1.9 μJ. Yoshitomi et al. [7] have recently generated 5th harmonic pulses ($\lambda_H = 50$ 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, their 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 relatively simple but intense light sources at hand, the study of nonlinear optical processes in the XUV and soft X-ray ranges, previously limited to longer wavelengths [8–10], is emerging. In this context, Ishikawa and Midorikawa [11] 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 [12] have theoretically shown that two-photon double ionization of He may be observed with the same harmonic.

In the present study, we investigate harmonic photoemission and ionization of He^+ subject to a combination of fundamental laser and its 27th harmonic pulses at the same time. The fundamental pulse can hardly ionize He^+, as we will see later. Although thanks to high ionization potential the harmonic spectrum would have a higher cut-off energy than in the case of rare-gas atoms [1, 2, 6, 7], the conversion efficiency is extremely low due to the small ionization probability. It is expected, however, that the addition of a Ti : sapphire 27th harmonic will facilitate ionization and photoemission, either through two-color frequency mixing or by assisting the transition to the $2p$ or $2s$ levels. Our results, based on numerical solution of the time-dependent Schrödinger equation (TDSE), show in fact that the combination of fundamental laser and its 27th harmonic pulses dramatically enhances both high-order harmonic generation and ionization by as much as 17 orders of magnitude [16].

2 Model

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,

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

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 [17]. 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:

$$E(t) = F_\mathrm{F}(t)\sin(\omega t) + F_\mathrm{H}(t)\sin(n\omega t + \varphi), \tag{2}$$

where $F_\mathrm{F}(t)$ and $F_\mathrm{H}(t)$ are the pulse envelopes of the fundamental and harmonic pulses, respectively, chosen to be Gaussian with a duration (full width at half maximum) of 10 fs, ω is the angular frequency of the fundamental pulse, n is the

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harmonic order, and φ is the relative phase. In typical calculations, we use a grid with a maximum radius of 125–250 a.u. and a 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 $\varphi = 0$ hereafter. We have also solved (1) using an explicit finite difference scheme and confirmed that the results remain virtually the same as those obtained with the Peaceman–Rachford method.

3 Photoemission

3.1 *Photoemission enhancement*

In Fig. 1 we show the harmonic photoemission spectrum from He^+ for the case of simultaneous fundamental and 27th harmonic (H27) irradiation. The spectrum is obtained from the Fourier transform of the dipole acceleration. We also show the spectra obtained when only the fundamental pulse is applied to a He^+ ion and a hydrogen atom. The general features of HHG can be explained by the classical model, usually called the "simpleman's theory" [19]: 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 cut-off) has an energy, E_c, given by [19]:

$$E_c = I_p + 3.17U_p, \tag{3}$$

where I_p and U_p are the ionization potential and the ponderomotive energy, respectively. This equation tells us that one way to obtain higher harmonics is to use a species with larger I_p. For the case of Fig. 1, the cut-off energy is calculated from (3) to be 70 eV (H45) for H and 111 eV (H73) for He^+. The cut-off 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

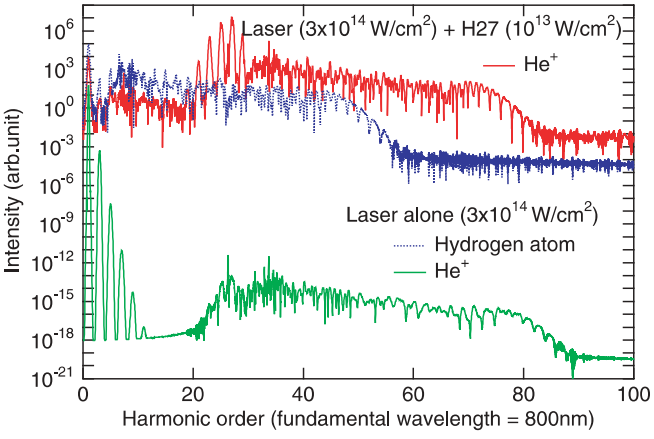

FIGURE 1 *Upper solid curve:* harmonic spectrum from He^+ exposed to a combined fundamental and its 27th harmonic pulse, the former with a peak intensity of 3×10^{14} W/cm^2 and the latter 10^{13} W/cm^2. The fundamental wavelength is 800 nm. *Lower solid and dotted curves:* harmonic spectra from He^+ and a hydrogen atom, respectively, exposed to the fundamental pulse alone

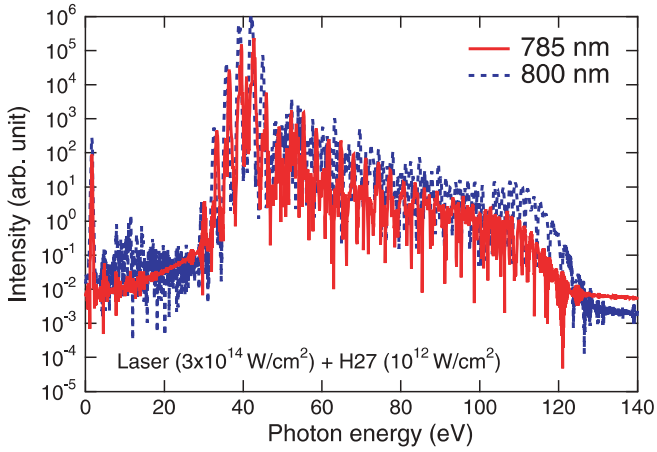

FIGURE 2 Harmonic spectra from He^+ exposed to a combined fundamental and its 27th harmonic pulse, the former with a peak intensity of 3×10^{14} W/cm^2 and the latter 10^{12} W/cm^2. The fundamental wavelengths are 785 nm (*solid curve*) and 800 nm (*dashed curve*), respectively

He^+ when only the fundamental laser is applied is extremely low compared with the case of H. The situation changes completely if the H27 pulse is added: the conversion efficiency is enhanced by about 17 orders of magnitude. Moreover, the advantage of high cut-off is preserved.

Figure 2 compares harmonic spectra from He^+ irradiated by a combined fundamental and H27 pulse with fundamental wavelength, λ_F, equal to 785 nm or 800 nm. This figure shows that the harmonic intensity does not depend much on fundamental wavelength. We have varied λ_F between 750 and 850 nm and found similar enhancement over the entire range.

3.2 *Enhancement mechanism*

The observed dramatic effect, by far more dramatic than previously reported moderate enhancements [13, 20, 21], can be understood as follows. The H27 photon energy (41.85 eV) is close to the $1s$–$2p$ transition energy of 40.8 eV. Moreover, the $2p$ level is broadened due to the laser-induced dynamic Stark effect. As a consequence, the H27 promotes transition to a virtual state near the $2p$ level. 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 discuss below in Fig. 5, and about 8% of the 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 greatly increased. 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, both in peak heights and positions (see Fig. 3). Hence the effect may also be interpreted as harmonic generation from a coherent superposition of states [22]. On the other hand, at a different fundamental wavelength, e.g., at $\lambda_F = 785$ nm, there is practically no real excitation: the populations of $2s$ and $2p$ levels after the pulse are only

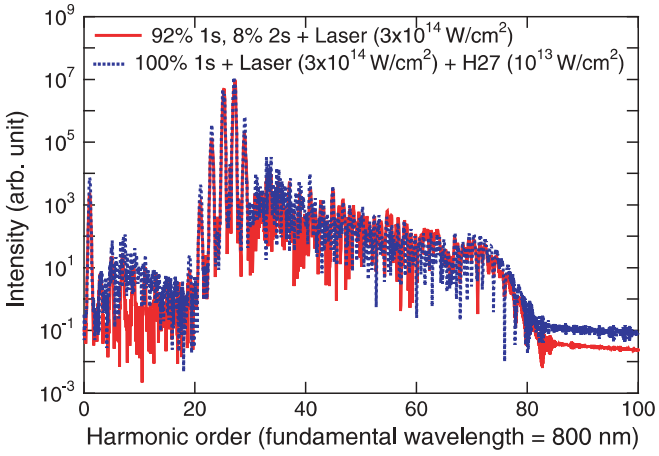

FIGURE 3 *Solid curve:* harmonic spectrum from the superposition of the 1*s* (92%) and 2*s* (8%) states of He^+ exposed to a fundamental laser pulse with a peak intensity of 3×10^{14} W/cm^2. *Dotted curve:* harmonic spectrum from ground-state He^+ exposed to a Gaussian combined 10 fs fundamental and its 27th harmonic pulse, 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. *The two curves* for the most part overlap each other

1.0×10^{-4} and 7.7×10^{-6}, respectively. Nevertheless the photoemission enhancement is still dramatic, and harmonic generation from the superposition of states cannot explain the spectrum. 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 greatly augmented. This may also be viewed as two-color frequency mixing enhanced by the presence of a near-resonant intermediate level.

4 Ionization

4.1 *Ionization enhancement*

Let us now examine ionization probability. Table 1 summarizes the He^{2+} yield for each case of the fundamental pulse alone, the harmonic pulse alone, and the combined pulse. As can be expected from the discussion in Sect. 3, the ionization probability from the combined pulse is more than 10 orders of magnitude higher than that from the fundamental laser pulse alone and is also increased by orders of magnitude with respect to the case of the H27 irradiation alone.

We have confirmed that the ionization probability is linear with respect to the H27 peak intensity, I_{H27}. This is compatible with our view that an H27 photon promotes a 1*s*–2*s* two-color, two-photon transition followed by field ionization.

I_F (W/cm^2)	I_{H27} (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

TABLE 1 The He^{2+} yield for various combinations of a Gaussian fundamental and its 27th harmonic pulses with a duration (full width at half maximum) of 10 fs and peak intensities listed in the table. $\lambda_F = 800$ nm

4.2 *Dependence on intensity and wavelength of the fundamental pulse*

Figure 4 shows the dependence of the He^{2+} yield on the peak intensity, I_F, of the fundamental pulse. Surprisingly, the yield does not monotonically increase with 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. 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 1*s*–2*p* 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 1*s*–2*s* transition energy. This indicates that the 2*s* level is resonantly populated through two-color, two-photon excitation. With increasing laser intensity, the peaks are shifted to longer wavelengths due to the dynamic Stark effect. 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 involving one H27 and two fundamental photons.The dynamic Stark effect induces not only the peak shift

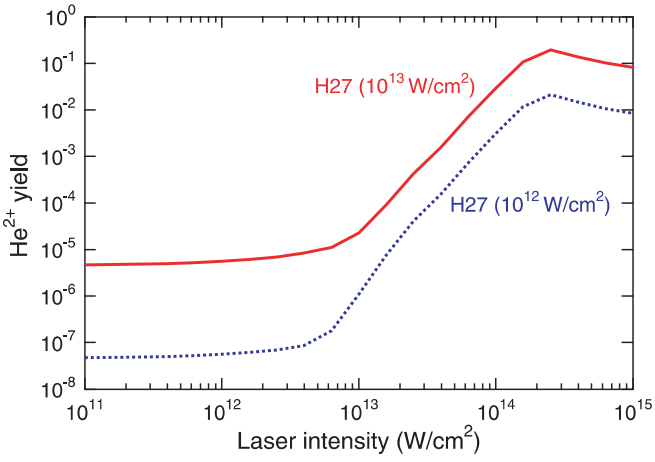

FIGURE 4 The He^{2+} yield as a function of peak intensity, I_F, of the fundamental laser pulse when He^+ is exposed to a combined fundamental and its 27th harmonic pulse. $\lambda_F = 800$ nm. The peak intensity of each harmonic pulse is noted in the figure

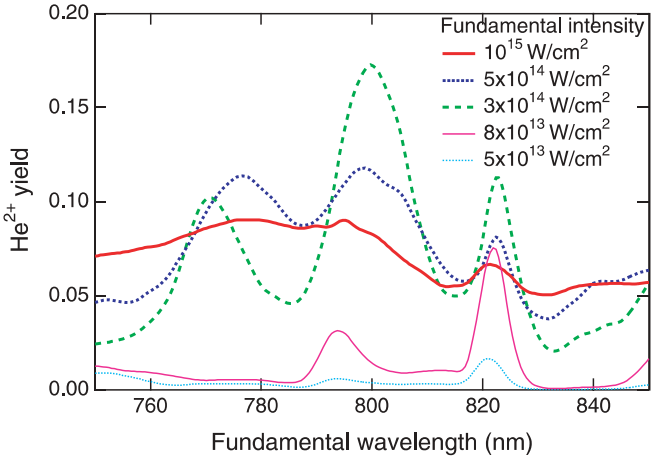

FIGURE 5 The He^{2+} yield as a function of fundamental wavelength when He^+ is exposed to a combined fundamental and its 27th harmonic pulse. The peak intensity, I_{H27}, of the 27th harmonic pulse is fixed at 10^{13} W/cm^2

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 the dynamic Stark effect. The interplay of these three leads to the complicated behavior in Fig. 4.

5 Conclusion

We have investigated the ionization and photoemission behavior of a He^+ ion under a combined field of ultrashort, intense fundamental laser and its 27th harmonic pulses. The response of He^+ is totally different from that in the case of the fundamental pulse alone or the harmonic pulse alone: both photoemission and ionization are enhanced by more than 10 orders of magnitude due to harmonic generation from a coherent superposition of states and two-color frequency mixing. The H27 plays an essential role in the transition to a real or virtual excited state, and the fundamental pulse is important in optical-field ionization from these levels as well as in $1s$–$2s$, $2p$ transitions through two-color excitation. The efficient H27-assisted harmonic generation might open a way to develop an intense light source of an even shorter wavelength than is available today.

ACKNOWLEDGEMENTS We would like to thank E.J. Takahashi and K. Midorikawa for helpful discussions on experimental feasibility. This work has been partially supported by Ministry of Education, Culture, Sports, Science and Technology of Japan Grant 15035203.

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