Simultaneous Extension and Enhancement of the HHG Plateau by Using Combined Lasers Irradiating on a United Two-Atom System *

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We present the high-order harmonic generation spectrum from a united two-atom system simultaneously exposed to a fundamental Ti:sapphire and its 33rd harmonic (H33) lasers by numerically solving a one-dimensional time-dependent Schrödinger equation. At a suitable inter-nuclear separation, the harmonic plateau is widened from $I_p+3.2U_p$ in the single-atom case up to $I_p+5.6U_p$ in the united-atom case. Furthermore, the plateau is heightened in excess of six orders of magnitude by using the combined lasers compared with the case of the fundamental laser alone. Such a scheme can step by step accomplish the following processes: through a single H33 photon resonance transition an appropriate amount of electrons are first populated to an excited state, in which the electrons are easily ionized and subsequently accelerated by the fundamental laser; then they can recombine with the neighbouring atom, so that high-efficiency emissions of the harmonics beyond $I_p + 3.2U_p$ are successfully realized.

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High-order harmonic generation (HHG) is a promising way to generate coherent radiations in the "water window" (4.4–2.3 nm) and to produce attosecond extreme ultraviolet pulses.[1] It has become an attractive research area over the past few years theoretically and experimentally. When intense lasers interact with atoms, molecules, clusters, and solids, HHG can be generated as a consequence of highly nonlinear dynamics.[2−5] According to the three-step model,[6] in the simplest case of an atom subject to a strong laser field, HHG corresponds to the following processes: the electron is ionized and accelerated by the laser field, and can recombine with the parent atom by emitting a high-energy photon. The maximal photon energy is $I_p+3.2U_p$, where I_p is the ionization potential and U_p is the ponderomotive energy.

For more sufficient applications of HHG, researchers are interested in simultaneously widening and heightening the HHG plateau on a large scale. It is well known, for example, that He^+ is a suitable candidate for extending the plateau due to its higher ionization potential. Unfortunately, the relevant small ionization yield leads to a poor harmonic efficiency. Recently, Ishikawa[7] has theoretically demonstrated that an intense Ti:sapphire laser and its 27th harmonic pulses simultaneously interacting with He^+ could dramatically enhance the ionization and then the harmonic efficiency by many orders of magnitude compared with the case of the fundamental-frequency pulse alone. However, to widen the HHG plateau such a scheme is limited due to only employing an atomic system with higher I_p, so it is expected that the plateau can be further widened in an imaginable enlarged system. In a recent theoretical study,[8] our numerical experiments showed that a large extension of the harmonic plateau could be obtained by a molecular ion subject to a monochromatic laser. When the inter-nuclear separation $R = \pi a_0/2$ (a_0 is the quiver radius), in the extended harmonics appear three distinct plateaus, whose cutoffs are $I_p+5.6U_p$, $I_p+7.0U_p$ and $I_p + 8.5U_p$, respectively. Here we are not interested in increasing emission efficiencies of those harmonics beyond $I_p + 5.6U_p$, because it is impossible to raise them to an absolute height for actual applications. It is noted that the generation of those harmonics involves the energetic electron reflection above the atomic potential-well, and that the probability of such a reflection is rather small. However, for the harmonics from $I_p + 3.2U_p$ to $I_p + 5.6U_p$, because they are produced accompanying the recombination process of ionized electrons directly with the neighbouring atoms (or ions), there is a potential possibility to enhance their efficiency to an applicable height. In this Letter, we show that this expectation is indeed realizable. The fundamental idea for realizing this purpose is to utilize a united two-atom model instead of a molecular ion model. The physical distinction of these two models is as follows: in the former the two atoms are identical, especially in each of them the electron is initially populated in its ground state; while in the latter[9] it is imaged that the system is initially made of a ground-state atom and an ion (or

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in the equivalent sense, a ground-state ion and another ion of higher ionization stage). Considering the stimulated property of all processes in laser fields,[10] in the former the ionized electron can efficiently recombine to the ground state of the non-parent atom due to its originally large population there, while in the latter the ionized electron can hardly recombine to the ground state of the non-parent atom because of its originally vacant population there. As far as the reality of this united two-atom model is concerned, we can consider a plasma expansion process (e.g., when an intense laser irradiates on clusters or solid surfaces), in which the atom-separation will reach the designed value in some periods. Noticing the great difference between the atomic speed and the electronic speed in plasmas, we suppose that all the electronic events occur at a definite inter-nuclear distance.

In this Letter, we show that Ishikawa's scheme can indeed be further developed. The idea is just to utilize the united two-atom model to replace the single atom model. Furthermore, we emphasize two points in the following. Firstly, in the usual situation (i.e., when ionization probability is not too large) it is the case that the larger the ionization, the higher the harmonic efficiency. Nevertheless, the validity of this law is limited. When the ground state population is more and more depleted, the reverse trend may set in: the larger the ionization, the lower the harmonic efficiency. The mechanism to cause this trend is in the recombination probability: the less the ground state population, the smaller the recombination probability because of the stimulated property of the recombination (SR) in laser fields.[10] Therefore, as far as the harmonic efficiency enhancement is concerned, it is necessary to raise the ionization to an appropriate level. Here we control the ionization by adjusting the intensity of the high-frequency laser. Secondly, we note that the high-frequency laser sources (photon energy of 51.16 eV, intensity of $10^{14}\,\mathrm{Wcm^{-2}}$) used here should be easily obtained, for example, with the aid of the VUV-FELs at HASYLAB,[11] where photon energies in excess of 200 eV, at high intensities ($10^{18}\mathrm{Wcm^{-2}}$ and up), should soon become available.

Our results are obtained by numerically solving the time-dependent Schrödinger equation (TDSE) for a united two-atom model subject to linearly polarized lasers. The numerical technique is the usual Crank–Nicholson time-propagation method.[12] We restrict ourselves to a one-dimensional (1D) model potential under the single active-electron approximation (SAEA). Furthermore, we suppose that the potential is short-range to simplify the computation and to guarantee the calculation accuracy benefited from the whole set of analytic eigenfunctions in this potential. Of course, this approximate model is rather poor in a viewpoint of conventional atomic and molecular physics, since, on the one hand, there is not any 1D atom in the world, and on the other hand, under the SAEA there remains only an invariant potential acting on an electron in the theoretical description for an arbitrary atom (i.e., all omitted other interactions, including the interaction between two electrons, are incorporated into the invariant potential). However, we observed previously that such a kind of numerical experiments played an important role in research of special regularities in a lot of strong-field phenomena. We believe that this simple model will continue to play its role, provided that the details omitted by it do not affect those vital processes.

In the length gauge and under the electric dipole approximation, the TDSE can be written as (atomic units are used throughout the Letter unless stated specially)

$$i\frac{\partial}{\partial t}\psi(x,t) = \Big[-\frac{1}{2}\frac{\partial^2}{\partial x^2} + V(x,R) - E(t)x\Big]\psi(x,t). \quad (1)$$

Here the laser field $E(t)$ is taken as

$$E(t) = E_f \sin(\omega t) + E_h \sin(33\omega t), \quad (2)$$

where E_f and E_h are respectively the field amplitudes of the fundamental Ti:sapphire and its 33rd harmonic (H33) lasers, ω is the angular frequency of the fundamental laser. The united two-atom potential $V(x,R)$ is composed of two modified Pöschl–Teller potentials[13]

$$V(x,R) = -\frac{U}{\cosh^2[\beta(x-R/2)]} - \frac{U}{\cosh^2[\beta(x+R/2)]}. \quad (3)$$

In the present study, we set $U = 3.5$, $\beta = 1.5$, thus in each atomic potential there are two bound states with eigenenergies $\varepsilon_0 = -2.0$ and $\varepsilon_1 = -0.125$, respectively. In order to ensure the remarkable extension of the plateau and the higher efficiency of the extended plateau, we set the inter-nuclear separation $R = \pi\alpha_0/2$ ($a_0 = E_f/\omega^2$). The atomic number density in the plasma derived from this separation is about $10^{20}\,\mathrm{cm^{-3}}$, such a condition may occur in some periods in the relevant plasma expansion process. The HHG spectrum of the system can be calculated by performing a Fourier transformation of the dipole acceleration

$$a(t) = \frac{d^2}{dt^2}\langle x\rangle = \Big\langle \psi(x,t)\Big| -\frac{\partial V(x,R)}{\partial x} + E(t)\Big|\psi(x,t)\Big\rangle. \quad (4)$$

Figure 1 shows the HHG spectrum from the united two-atom system exposed to the combined lasers. For comparison, we also present the HHG spectrum for an atom subject to the fundamental field alone. Here the intensity I_f of the fundamental field is $5.05 \times 10^{14}\,\mathrm{Wcm^{-2}}$, and that of the H33 laser is $2.02 \times 10^{13}\,\mathrm{Wcm^{-2}}$. It is noted that the HHG plateau

from the former is not only heightened by about six orders of magnitude but also widened from $I_p + 3.2U_p$ up to $I_p + 5.6U_p$ compared with that from the latter. This result shows that the aim for dramatically extending the width and simultaneously enhancing the height of the plateau is achieved here. Thereinafter we will give the explanation for the extension and enhancement of the plateau in turn.

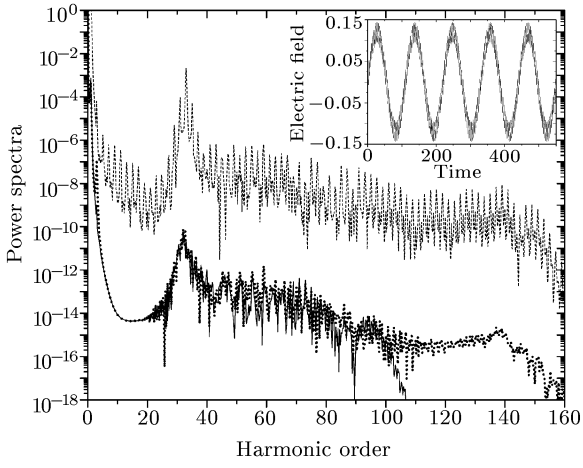


Fig. 1. HHG spectrum (dashed) from the united two-atom model subject to the fundamental Ti:sapphire and its 33rd harmonic lasers, the former with the intensity $5.05 \times 10^{14}\,\mathrm{Wcm^{-2}}$, and the latter $2.02 \times 10^{13}\,\mathrm{Wcm^{-2}}$. The inter-nuclear separation $R = 58$. The solid and dotted curves: HHG spectra from the atom and the united two-atom model respectively exposed to the fundamental laser alone. The inset shows the electric field of the combined lasers.

In Fig. 1, we also display the HHG spectrum from the united two-atom system irradiated by the fundamental laser alone. It is observed that the harmonic plateaus from the united two-atom model have the same cutoff $I_p + 5.6U_p$ whether it interacts with the combined lasers or only the fundamental laser. Actually, the relevant ponderomotive energy of the H33 laser is too small (compared with that of the fundamental laser) to influence the second and third steps in the three-step model. This means that the extension of the HHG plateau is essentially independent of the H33 laser in the case of our combined lasers. In order to more specifically test the above idea, we calculated the instants of the electron recombination with the neighbouring atom and the corresponding kinetic energies in both the cases. As is expected, one can see from Fig. 2 that the instant curves or the kinetic energy curves in both the cases are almost the same at the graphical level.

From the above discussion, it is clear that the H33 laser hardly affects the acceleration and recombination processes of the ionized electrons; so, what role does it play in the case of the combined lasers? When the united two-atom system interacts with the fundamental laser ($5.05 \times 10^{14}\,\mathrm{Wcm^{-2}}$) alone, the ionization yield is very small (1.16×10^{-6}) due to the high I_p of the ground-state atom. However, the situation completely changes owing to the addition of the H33 laser ($2.02 \times 0^{13}\,\mathrm{Wcm^{-2}}$): the ionization yield (0.39) is prominently increased (in the case of the H33 laser alone, the ionization yield is only 1.14×10^{-3}). Such an enhancement can be easily understood: the electrons are first resonantly excited to the excited state by a single H33 photon, and then much more easily ionized by the fundamental laser. From the mechanism of the enhancement, one can deduce that the ionization enhancement is close related to the intensity I_h of the H33 laser. In Fig. 3(a), we display the dependence of the ionization probability on I_h for the fixed I_f. This figure shows definite regularities in three different ranges of I_h: at the beginning the ionization yield slowly rises up to $2.02 \times 10^7\,\mathrm{Wcm^{-2}}$, then is proportional to I_h, lastly the saturation appears as I_h is higher than $2.02 \times 10^{13}\,\mathrm{Wcm^{-2}}$. The dependence of the ionization on I_h vividly reflects the competition between two major ionization channels (i.e., tunnel ionization by the fundamental field alone and the combined laser ionization described in detail above). In the first range, the tunnel ionization gradually loses the major role and the other

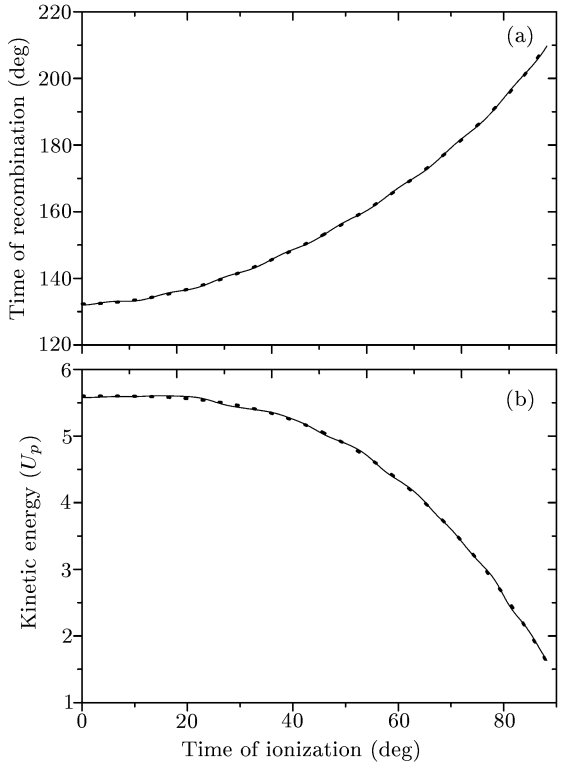


Fig. 2. (a) Variation of the instants of the electron recombination with the neighbouring atom as a function of the instants of ionization and (b) the corresponding kinetic energies of the electron at the recombination instants. The solid and dotted curves correspond to the cases of the combined lasers and the fundamental laser alone, respectively. The laser parameters are the same as that used in Fig. 1.

becomes more and more important. In the second and third ranges, the combined laser ionization becomes absolute ruler. Hence, it is clear that the role of the H33 laser is just to ensure the sufficient population of electrons in the excited state, where they can be effectively ionized by the fundamental laser.

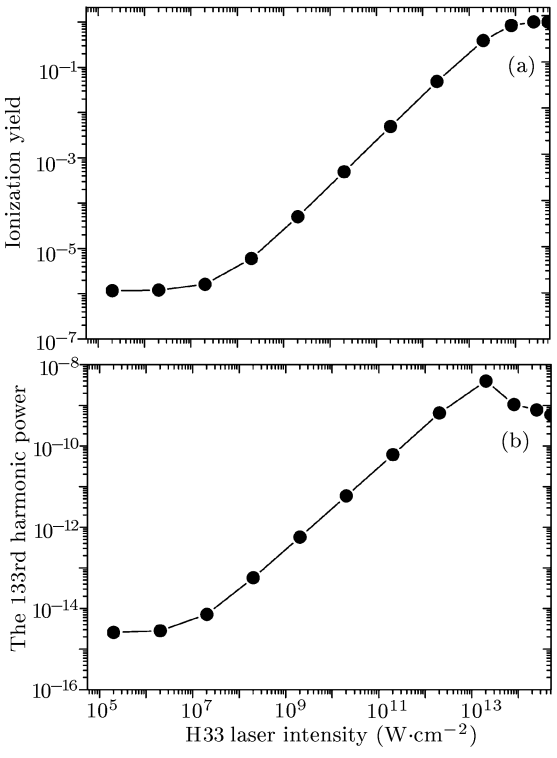


Fig. 3. (a) The ionization yields and (b) the 133rd harmonic efficiencies as a function of the intensity I_h. The intensity of the fundamental laser is $5.05 \times 10^{14}\,\mathrm{W cm^{-2}}$.

As far as the efficiency of the HHG plateau is concerned, it is closely correlated with the ionization yield. Consequently, one can naturally infer that the harmonic efficiency is also tightly related to I_h. In Fig. 3(b), taking the 133rd harmonic as an example, we examine dependence of the HHG efficiency on I_h. In the first and second ranges of I_h, the behaviour of the 133rd harmonic powers is identical to that of the ionization yields, which means that the larger the ionization, the higher the harmonic efficiency. However, the things are completely reversed in the third range (i.e., the larger the ionization, the lower the harmonic efficiency). Why does such a counter-intuitive behaviour appear? The reason for this lies in the fact that the ground state population is more and more depleted when the ionization is larger and larger. As a result, the recombination probability becomes smaller and smaller due to the property of SR, and so is the harmonic efficiency. Therefore, in the case of our combined lasers, the enhancement of the harmonic efficiency is caused by increase of the ionization, but it is necessary that the ionization is increased to an appropriate level by adjusting I_h.

In conclusion, we provide a method for simultaneously widening and heightening the HHG plateau on a large scale. By an intense fundamental laser and a weak H33 laser simultaneously interacting with a united two-atom system at the inter-nuclear separation $R = \pi a_0/2$, the HHG plateau is not only heightened by about six orders of magnitude but also widened from $I_p + 3.2U_p$ up to $I_p + 5.6U_p$ compared with the case of an isolated atom subject to the fundamental laser alone. Such a scheme first generates appropriate amount of ionized electrons through an H33 photon resonance transition to the excited state, in which the electrons are easily ionized and subsequently accelerated by the fundamental laser, and then can recombine with the neighbouring atom, so that high-efficiency emissions of the harmonics beyond $I_p + 3.2U_p$ can be achieved.

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