Enhancement of bichromatic high-order-harmonic generation with a high-frequency field

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Using a high-frequency field superposed to a linearly polarized bichromatic laser field composed by a wave with frequency ω and a wave with frequency 2ω, we show it is possible to enhance the intensity of a group of high harmonics in orders of magnitude. These harmonics have frequencies about 30% higher than the monochromatic-cutoff frequency, and, within the three-step-model framework, correspond to a set of electron trajectories for which tunneling ionization is strongly suppressed. Particular features in the observed enhancement suggest that the high-frequency field provides an additional mechanism for the electron to reach the continuum. This interpretation is supported by a time-frequency analysis of the harmonic yield. The additional high-frequency field permits the control of this group of harmonics leaving all other sets of harmonics practically unchanged, which is an advantage over schemes involving only bichromatic fields.

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Within the last few years, the perspective of obtaining efficient laser sources in the extreme-ultraviolet regime has led to the proposal of several schemes for controlling the harmonic spectrum of an atom subject to a strong laser field ($I \sim 10^{14}$ W/cm^2), using for instance additional static fields [1], ultrashort pulses [2], bichromatic driving fields [3–8], or additional confining potentials [9]. These schemes are based on the "three-step model" [10], which describes very well the spectral features observed in experiments. These features are the "plateau," where harmonics of roughly the same intensities exist, and the "cutoff," where the harmonic signal suddenly decreases in orders of magnitude. According to this model, the generation of high-order harmonics in strong-laser fields corresponds to a dynamical process in which an electron leaves an atom at an instant t_0 via tunneling ionization, this electron in the continuum is then accelerated by the driving fields, and, if it comes back and recombines with the parent ion at a time t_1, a high-energy photon will be generated. The harmonic energy is given by $\Omega = |\varepsilon_0| + E_{\mathrm{kin}}(t_0,t_1)$, with $E_{\mathrm{kin}}(t_0,t_1)$ and $|\varepsilon_0|$ being, respectively, the kinetic energy of the electron upon return and the ionization potential of its parent ion. The cutoff corresponds to the maximum electron kinetic energy. By manipulating the different steps of the dynamical process, one can, in principle, control high-order-harmonic generation.

If the purpose is to increase the cutoff energy, very efficient schemes exist. They involve either additional static fields [1], or atoms placed in confining potentials [9]. In both cases one introduces an additional force that modifies the propagation of the electron after injection, resulting in an increased cutoff energy. However, such schemes may require extremely high static fields [11], or appropriate solid-state materials, whose existence is still under investigation [12].

Another scheme for manipulating high-order harmonics involves bichromatic driving fields [3–8]. By varying the frequencies, the relative phase, and the intensities of the two driving waves, one can modify the propagation of the electron in the continuum, and even the "first step," i.e., the tunneling ionization. In contrast with the previously discussed schemes, high-order-harmonic generation with bichromatic driving fields is already experimentally feasible [7,8].

In the bichromatic case, however, no simple expression for the cutoff energy exists [3–6], so that it is not straightforward to predict whether the plateau can be extended. As a general feature, high-harmonic spectra from atoms in bichromatic fields exhibit a plateau with a relatively complex structure, which may have several cutoffs. These cutoffs are given by local maxima of $E_{\mathrm{kin}}(t_0,t_1)$, which are, however, unequally important for the resulting spectra. Thus, it may happen that the absolute maximum of $E_{\mathrm{kin}}(t_0,t_1)$ leads to a much less pronounced decrease in the harmonic signal than a local maximum at a lower energy position.

A very illustrative example is a bichromatic field consisting of a wave with frequency ω and its second harmonic [4–6]. The addition of the second driving wave causes a splitting in the monochromatic-field cutoff energy $\varepsilon_{\max} = |\varepsilon_0| + 3.17U_p$, U_p being the ponderomotive energy. As a direct consequence, there is a double-plateau structure in the harmonic spectra, with an upper and a lower cutoff, whose energies are higher and lower than $\varepsilon_{\max}$, respectively. The upper cutoff corresponds to the absolute maximum for $E_{\mathrm{kin}}(t_0,t_1)$. However, it appears in the spectrum only as a small shoulder due to the relative low-harmonic intensities in the upper-cutoff energy range. The lower cutoff, on the other hand, is related to a decrease of orders of magnitude in the harmonic yield, being therefore the one effectively measured in experiments (see, e.g., [5,8] for a more complete discussion). One can explain the intensity difference in this double-plateau structure in terms of the width of the effective potential barrier through which the electron tunnels. This barrier is given by $V_{\mathrm{eff}} = V(x) - xE(t_0)$, where $E(t_0)$ is the field at the electron emission time and $V(x)$ the atomic potential. For the upper cutoff, the atomic potential is not as much distorted by the field as for the lower cutoff. This results in a considerably wider effective potential barrier and, consequently, much weaker harmonics.

In this paper we consider again the case of a linearly polarized $\omega - 2\omega$ field, as in [4,5]. However, this time our aim is to increase the intensities of the upper-cutoff harmonics close to the intensities of those belonging to the lower cutoff, effectively extending the cutoff energy beyond $\varepsilon_{\max}=|\varepsilon_0|+3.17U_p$. For this purpose, one must provide an additional mechanism for the electronic wave packet to reach the continuum, thus compensating the weak tunneling ionization. We demonstrate that this can be done with an additional driving wave. This third wave has a relatively high frequency but low intensity compared to the bichromatic field. The nature of the third wave is such that it alters the electron injection into the continuum and, at the same time, it does not appreciably modify the ponderomotive energy and the cutoff. This field configuration is similar to that used in [13], where the influence of the background field on x-ray-atom scattering processes was investigated.

We restrict ourselves to a one-dimensional model, which still describes high-order-harmonic generation with linearly polarized fields well in qualitative terms. We solve the time-dependent Schrödinger equation

$$i\frac{d}{dt}|\psi(t)\rangle=\left[\frac{p^2}{2}+V(x)-pA(t)\right]|\psi(t)\rangle \tag{1}$$

numerically, for an atom initially in the ground state, with binding potential $V(x)$ subject to a laser field $E(t)=-dA(t)/dt$. Atomic units are used throughout. The external laser field is taken as

$$E(t)=E_{01}\sin(\omega_1 t)+\sum_{i=2}^{3}E_{0i}\sin(\omega_i t+\phi_{1i}), \tag{2}$$

with E_{0i}, ω_i, and ϕ_{1i} being the field amplitudes, frequencies and the phases with respect to the first driving wave, respectively. In this paper, we choose $\omega_1=\omega$, $\omega_2=2\omega$, $E_{03}\ll E_{01},E_{02}$ and $\omega_3\gg\omega$. Unless stated otherwise, the relative phase ϕ_{13} is set to zero. The binding potential is taken as

$$V_G(x)=-\alpha\exp(-x^2/\beta^2), \tag{3}$$

which is a widely used expression for modeling short-range potentials. The harmonic spectra are calculated from the dipole acceleration $\ddot{x}=\langle\psi(t)|-dV(x)/dx+E(t)|\psi(t)\rangle$ [14]. Spectral and time-frequency analysis is subsequently done on $\ddot{x}(t)$. This latter method has been extensively used to extract the main contributions to high-order-harmonic generation within a cycle of the driving field [4,15]. These contributions give the electron return times corresponding to a particular set of harmonics (see [4,15] for a more complete discussion).

Time-resolved spectra are computed by performing a Fourier transform with a temporally restricted window function. For an arbitrary function $f(t')$, this transform is

$$\mathcal{F}(t,\Omega,\sigma)=\int_{-\infty}^{+\infty}dt' f(t')W(t,t',\Omega,\sigma). \tag{4}$$

We consider a Gabor transform, which has a Gaussian window function

$$W(t,t',\Omega,\sigma)=\exp[-(t-t')^2/\sigma^2]\exp[\mathrm{i}\Omega t']. \tag{5}$$

The usual Fourier transform $\mathcal{F}(\Omega)$, for which all the temporal information is lost, is recovered for $\sigma\to\infty$. The temporal width σ corresponds to a frequency bandwidth $\sigma_\Omega=2/\sigma$.

The kinetic energy of the electron upon return is taken as

$$E_{\mathrm{kin}}(t_0,t_1)=\frac{1}{2}[A(t_1)-A(t_0)]^2. \tag{6}$$

The ponderomotive energy is given by

$$U_p=\tfrac{1}{2}\int_0^T A^2(t)dt=\sum_{i=1}^{3}\frac{E_{0i}^2}{4\omega_i^2}. \tag{7}$$

To first approximation, if the third driving wave is much weaker than the others and its frequency is much higher than ω, the contribution from the additional field to the ponderomotive energy (7) and to the kinetic energy (6) can be neglected. More specifically, for the parameters used in this paper, we observed, after solving the classical equations of motion of an electron in a field (2), that for $E_{03}/E_{02}\leqslant 1$ and $\omega_3>5\omega$ the influence of the third driving wave on these two latter quantities was not significant.

As a starting point, we shall discuss the existence of the enhancement in question. We will restrict ourselves to varying the relative phase ϕ_{12} between the two low-frequency driving waves and the high-frequency field parameters. The bichromatic field strengths are similar to these in [5], namely $E_{01}=0.1$ a.u., $E_{02}=0.032$ a.u., which give the intensity ratio $I_{2\omega}/I_\omega=0.1$. For these field parameters, the high-order-harmonic spectrum displays a clear double-plateau structure, with a lower and an upper cutoff (c.f. Fig. 1 and Refs. [4,6]). The lowest frequency is taken as $\omega=0.057$ a.u., which is typically used in experiments. The ground-state energy was taken as $|\varepsilon_0|=0.57$ a.u., which roughly corresponds to the argon ionization potential and, unless stated otherwise, we took $\alpha=1.15$ a.u. and $\beta=1$ a.u. in Eq. (3). This gives a model atom with a single bound state.

The influence of the third driving wave on the harmonic spectra is shown in Fig. 1, for relative phases $\phi_{12}=0$ and $\phi_{12}=0.3\pi$ and several field strengths E_{03}. The relative phase ϕ_{13} was set to zero in both cases, and ω_3 was chosen to be ten times ω_1. Apart from the strong enhancement around ω_3 (near harmonic order ten), the main differences between the case with a third wave and the bichromatic case ($E_{03}=0$) are displayed in the so-called "upper-cutoff" harmonics, which lie, for the phases in question, roughly at $\varepsilon_u=|\varepsilon_0|+3.8U_p$, with the corresponding harmonic orders near $N=62$. The intensity of this group of harmonics can be changed significantly when the field strength of the third wave is increased. Whereas for $\phi_{12}=0$ these changes are quite irregular, for $\phi_{12}=0.3\pi$ they appear as very pronounced enhancements, which reach three orders of magnitude. They make the intensities of the upper-cutoff and lower-cutoff harmonics comparable, effectively extending the harmonic production region. This occurs already for a third wave whose intensity is only a few percent of that of the bichromatic field.

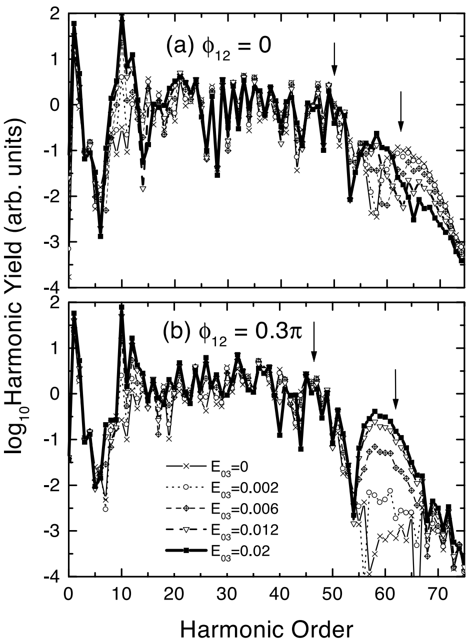

FIG. 1. Harmonic spectra for an atom subject to the three-color field (2), with strong-field amplitudes $E_{01}=0.1$ a.u., $E_{02}=0.032$ a.u., high-frequency field strengths $0\leqslant E_{03}\leqslant 0.02$ a.u., frequencies $\omega_1=0.057$ a.u., $\omega_2=0.114$ a.u. and $\omega_3=0.57$ a.u., and relative phase $\phi_{13}=0$, for $\phi_{12}=0$ [part (a)] and $\phi_{12}=0.3\pi$ [part (b)]. The lower cutoff (near harmonic order 50) and the upper cutoff (near harmonic order 62) are marked with arrows. The lower-cutoff energy depends more sensitively on ϕ_{12} than the upper-cutoff energy [5].

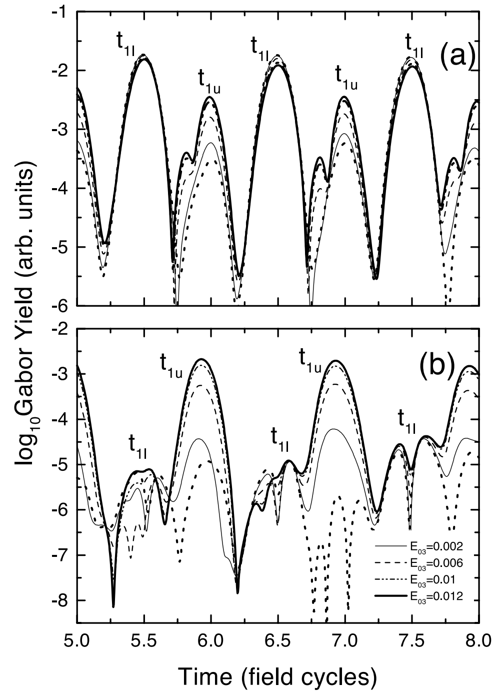

FIG. 2. Time-resolved spectra for the lower [part (a)] and upper [part (b)] cutoff harmonics, for the same bichromatic-field parameters as in Fig. 1, $\phi_{12}=0.3\pi$ and $\phi_{13}=0$, and several high-frequency field amplitudes E_{03}. The thick dotted lines correspond to $E_{03}=0$. The temporal width was chosen as $\sigma=0.1T$, such that the frequency width σ_ω is of roughly three harmonics. The precise center of the window function has been chosen as $\Omega_l=48\omega$ [part (a)] and $\Omega_u=58\omega$ [part (b)]. The return times t_{1l} and t_{1u} are indicated in the figure.

A particularly interesting feature of the scheme is that it affects mainly the group of harmonics in the upper cutoff, leaving other groups of harmonics practically unaffected. This is in contrast with the purely bichromatic case, for which a change in the relative phase ϕ_{12} leads to changes in the whole plateau structure [4,5]. In analyzing the enhancement effects, we recall that the presence of the high-frequency third wave does not modify the ponderomotive energy and the classical trajectories. Thus, the three-step model leads us to the conclusion that a high-frequency induced process is injecting the electron into the continuum. This physical picture also explains why the remaining harmonic intensities are not influenced by the high-frequency wave. Let us consider an electron leaving the atom at the emission time t_{0l}, so that, at its recombination time t_{1l}, the lower-cutoff harmonics are generated. At the lower-cutoff emission time t_{0l}, tunneling is so pronounced that, even in the presence of the third driving wave, this mechanism is still the dominant path for the electron to reach the continuum. Thus, the high-frequency induced process is in comparison negligible. A similar argument explains the differences between Figs. 1(a) and 1(b). For $\phi_{12}=0$, there is still enough tunneling to compete with the high-frequency induced process, such that the quantum interference between both processes leads to irregular intensity variations. For $\phi_{12}=0.3\pi$, on the other hand, tunneling is strongly suppressed, such that the electron reaches the continuum mainly due to the high-frequency induced process [5].

This interpretation is supported by the time-frequency analysis. In Fig. 2, we display the time-frequency yield for groups of harmonics centered at the frequencies $\Omega_l=48\omega$ [Fig. 2(a)] and $\Omega_u=58\omega$ [Fig. 2(b)], for several field strengths E_{03} and a relative phase $\phi_{12}=0.3\pi$. The frequency Ω_l corresponds to the lower-cutoff energy and Ω_u was chosen at the most prominent harmonic frequency in the upper cutoff. These time-resolved spectra are periodic within a cycle $T=2\pi/\omega$ of the driving field, exhibiting peaks at times $t_{1l}=1.4T$ and $t_{1u}=0.9T$. These are the electron return times related to the lower- and the upper-cutoff harmonics, respectively, and have been computed in [5], together with the corresponding emission times (c.f. Fig. 1 in there). For a given set of harmonics, if the high-frequency field is enhancing the injection of the electron wave packet at the emission time t_0, the corresponding peak at t_1 in the time-resolved spectra should get more prominent as E_{03} is increased, and occurs only for the upper-cutoff return time t_{1u}. This can be observed in both parts of the figure.

In the following we understand how the observed enhancements depend on the relative phase ϕ_{13} between the lowest- and highest-frequency fields, on the field strength E_{03}, on the frequency ω_3, and whether the atomic potential has any influence on it.

The behavior with respect to ϕ_{13} gives further information about the nature of the effect due to the third wave. In

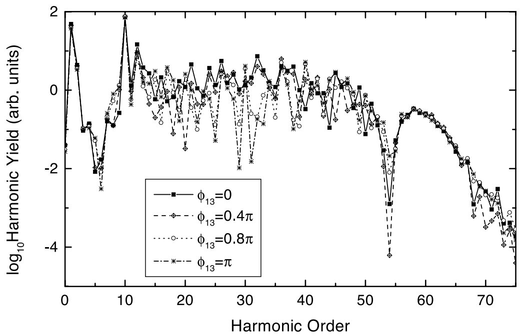

FIG. 3. Harmonic spectra for the same bichromatic-field parameters as in the previous figures, $\phi_{12}=0.3\pi$, field strength $E_{03}=0.015$ a.u., and several relative phases ϕ_{13}.

case this wave enhances tunneling ionization by changing the effective potential barrier, or increases the harmonic intensities by distorting the electron motion in the continuum, this influence will be different for different phases ϕ_{13}. We verified that the upper-cutoff harmonics remain unchanged when ϕ_{13} is varied. This result suggests that the third wave is playing a very different role than the bichromatic field. These results are displayed in Fig. 3.

Another very important issue is the dependence of the enhancements on the field strength E_{03}. A closer inspection of Figs. 1(b) and 2(b) suggests that there is a saturation intensity for the upper-cutoff harmonics (in the figure, close to $E_{03}=0.01$ a.u.). Furthermore, these harmonics are unequally enhanced, the maximum enhancement occurring, for the example in question, at the harmonic order $N=58$. This maximum can be slightly displaced, depending on the frequency ω_3. In Fig. 4, we show explicitly the behavior of this maximally enhanced harmonic with respect to the field strength E_{03}, for several frequencies ω_3. This figure shows that the enhancement effect increases quickly in the weak-field region but it eventually saturates when reaching the strong-field region, which means that upper-cutoff harmonics cannot be indefinitely enhanced by increasing the strength of the high-frequency wave.

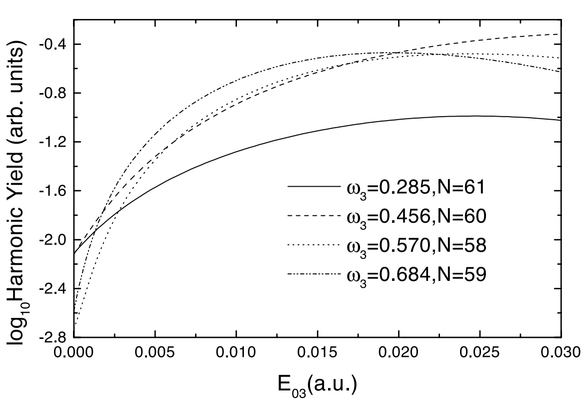

FIG. 4. Harmonic yields as functions of the high-frequency field strength E_{03}, for the same field as in the previous figures, with $\phi_{12}=0.3\pi$ and 0.285 a.u.$\leqslant\omega_3\leqslant$0.684 a.u. The harmonic orders displayed in the figure correspond to the most efficient enhancements obtained.

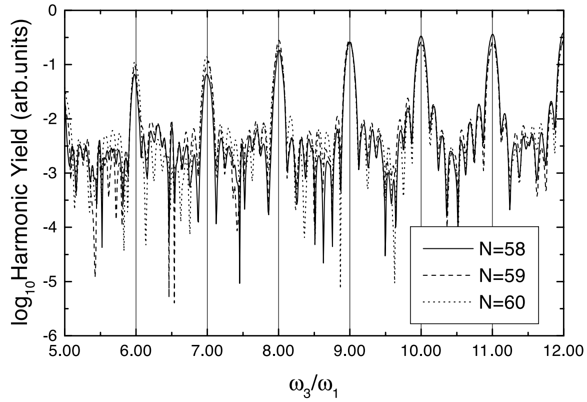

FIG. 5. Harmonic yields of neighboring upper-cutoff harmonics as functions of the frequency ratio ω_3/ω_1, for a driving field as in the previous figures and $E_{03}=0.015$ a.u. The ionization threshold is at $\omega_3=10\omega_1$.

In Fig. 5 we show the dependence of the enhanced harmonics with respect to the frequency ω_3, for $5\omega_1\leqslant\omega_3\leqslant12\omega_1$. The figure displays major peaks at integer frequency ratios ω_3/ω_1 and subpeaks between the integers. The enhancements are especially strong at the integers, where the frequencies of the three driving waves are commensurate. The strong enhancements at these particular frequencies can be understood as a consequence of the high-order-harmonic generation process: since the production of harmonics takes place within many cycles of the driving field, one expects it to be most efficient when this field is periodic [16]. Figure 5 also demonstrates that the observed enhancements are not related to threshold effects, since they occur when the frequency of the third wave is either below or above the threshold.

The investigations presented above are from our numerical calculations for the short-range potential (3), with α and β chosen in such a way that it has a single bound state. To rule out the possibility that the effect is due to an artifact introduced by this model potential, we performed similar calculations for several potentials with different values for depth α and width β. We also studied the soft-core potential $V_C=-\alpha[x^2/\beta^2+1]^{-1/2}$. We observed that the enhancements are present in all situations, being however stronger for short-range potentials. This is related to the fact that tunneling ionization is more efficiently suppressed in the short-range case, such that it does not compete with the high-frequency induced process. For the Gaussian potential used, we obtain very pronounced enhancements for $\beta\leqslant2$ a.u., which is well within the experimental range.

In conclusion, we have shown that, with an additional high-frequency field, we are able to enhance a group of very high-order harmonics of a bichromatic driving field consisting of a wave of frequency ω and its second harmonic. These harmonics have energies up to $|\varepsilon_0|+4U_p$, which is about 30% higher than the monochromatic cutoff frequency $\varepsilon_{\max}=|\varepsilon_0|+3.17U_p$. They correspond to a set of electron trajectories for which tunneling is not very pronounced in the bichromatic case. The high-frequency field provides an additional mechanism for the electron to reach the continuum, resulting in the enhancement of this group of harmonics.

This physical interpretation is supported by several characteristics of the enhancements, such as, for instance, the fact that the remaining groups of harmonics are not altered by the high-frequency field. For an electron leaving at a time t_0 such that the lower-cutoff and plateau harmonics are generated, tunneling ionization is the dominant process. This is true even in the presence of the third wave, so that the high-frequency induced process plays practically no role. For the upper-cutoff harmonics, on the other hand, tunneling is relatively weak, so that the high-frequency induced process may compete with it or even be far more prominent. This latter effect is observed in the behavior of the enhancements with respect to the relative phase ϕ_{12} between the two low-frequency waves. The time-resolved spectra of the lower- and upper-cutoff harmonics confirm this physical picture. As the high-frequency field is increased, the time-frequency yield at the return times t_{1l} corresponding to the lower cutoff remains pratically unaltered, whereas the yield at the upper-cutoff return time t_{1u} is enhanced in orders of magnitude. Thus, the third field is increasing the electron injection in the continuum at the corresponding electron-emission times t_{0u}. Finally, since the enhancements are independent of the phase ϕ_{13} between the highest- and lowest-frequency fields, they cannot be attributed to mechanisms such as tunneling ionization or a distortion in the electron propagation.

The enhancements are particularly strong when the relative phase between the two strong driving waves is chosen such that tunneling ionization is strongly suppressed. Such a case is provided by taking $\phi_{12}=0.3\pi$ [5], for which an appreciable enhancement is already obtained with high-frequency fields whose intensities are only a few percent of that of the bichromatic field. This phase control is already experimentally feasible [8]. Another prerequisite for pronounced enhancement effects concerns the frequency of the third wave, which must be an integer multiple of the fundamental frequency of the bichromatic field. Furthermore, our theoretical studies show that the enhancements are always present for very different potentials, but they also suggest that a short-range potential gives stronger enhancements and therefore is a more appropriate choice for a possible experimental consideration.

It is also worth mentioning that, in principle, a similar enhancement scheme is applicable to any sets of harmonics corresponding to a set of electron trajectories for which tunneling ionization is strongly suppressed. For the specific $\omega - 2\omega$ configuration considered in this paper, this condition is satisfied by the upper-cutoff harmonics.

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