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Amplification of high-order harmonics using weak perturbative high-frequency radiation

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The mechanism underlying the substantial amplification of the high-order harmonics $q\pm 2K$ (K integer) upon the addition of a weak seed extreme uv (XUV) field of harmonic frequency $q\omega$ to a strong IR field of frequency ω is analyzed in the framework of the quantum-mechanical Floquet formalism and the semiclassical recollision model. According to the Floquet analysis, the high-frequency field induces transitions between several Floquet states and leads to the appearance of new dipole cross terms. The semiclassical recollision model suggests that the origin of the enhancement lies in the time-dependent modulation of the ground electronic state induced by the XUV field.

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Focusing intense linearly polarized monochromatic IR laser pulses into a gas of atoms can lead to the emission of high-energy photons with frequencies extending into the extreme ultraviolet (XUV) and x-ray region by high-order harmonic generation (HHG). The HHG phenomenon stands as one of the most promising methods of producing short attosecond (as) pulses [1].

The contamination of the strong IR field with a second [2–4] or more [5–7] weak XUV fields has a dramatic effect on the dynamical behavior of the electrons, and had drawn a lot of attention in recent years. On the basis of the three-step (recollision) model [8–10], it had been argued that the role of the XUV field is to switch the initial step in the generation of high-order harmonics from tunnel ionization to the more efficient XUV single-photon ionization. This might explain the improved macroscopic HHG signal obtained in experiments: the XUV-assisted ionization increases the number of atoms that participate in the HHG process and improves phase matching [11].

The effect at the single-atom level, however, is less clear. It has been shown that the XUV photons control the timing of ionization, and preferentially select certain quantum paths of the electron [12]. While this effect may lead to the enhancement of the low-order harmonics in the plateau, it cannot account for the large enhancement in the cutoff and beyond (Fig. 1). A three-step-model classical analysis of HHG suggests that the contribution of the XUV field to the kinetic energy of the returning electron is negligible. The kinetic energy of a classical free electron of charge e and mass m, driven by a linearly polarized strong IR fundamental field of frequency ω, amplitude $\varepsilon_1^{\text{in}}$, and polarization $\mathbf{e_k}$ [$\mathbf{E}_1(t)=\mathbf{e_k}\varepsilon_1^{\text{in}}\cos(\omega t)$] is $E_k(t)=p^2(t)/2m$. $p(t)=(e\varepsilon_1^{\text{in}}/\omega)[\sin(\omega t)-\sin(\omega t_i)]$ is the momentum of the electron, and it has been assumed that the electron is freed at time t_i with zero momentum. The addition of a weak harmonic XUV field of frequency $q\omega$ (where q is a large integer) and amplitude $\varepsilon_q^{\text{in}}$ ($\varepsilon_q^{\text{in}}\ll\varepsilon_1^{\text{in}}$) with the same polarization [$\mathbf{E_q}(t)=\mathbf{e_k}\varepsilon_q^{\text{in}}\cos(q\omega t)$] adds a small correction to the momentum, which is proportional to $\varepsilon_q^{\text{in}}/q\omega$. As a result, the correction to the kinetic energy, which appears in the form of two additional terms, proportional to $\varepsilon_q^{\text{in}}/q\omega$ and $(\varepsilon_q^{\text{in}}/q\omega)^2$, is negligible. Thus, the additional XUV field will not affect the electron trajectories and will not contribute to their kinetic energy. For this reason the relative phase between the two fields does not play a role in the harmonic generation spectra (HGS), which is indeed verified in both classical analysis and quantum-mechanical simulations (a small q, however, will affect the dynamics differently [2,13]). In addition, assigning the electron a nonzero initial momentum to account for the photoelectric effect will not increase its kinetic energy upon recombination.

An illustrative time-dependent Schrödinger equation (TDSE) simulation, however (Fig. 1), shows an enhancement of the cutoff harmonics and the harmonics $q\pm 2K$ (K integer) upon addition of a weak XUV field to the strong IR field. Moreover, the HGS possesses certain symmetries: with respect to its center at harmonic q, the distribution of harmonics of the enhanced part of the spectrum (harmonics that have been produced only due to the addition of the XUV field) is symmetric with respect to q and remains almost invariant upon variation of q. This suggests that, despite the fact that the additional weak XUV field does not affect the electron trajectories, it does affect the recombination process. As will be shown later, the XUV field induces periodic modulations to the remaining ground electronic state, with the same frequency as the one of the XUV field. The returning electronic wave packet recombines with this modulated ground state to emit new harmonics. The purpose of this Rapid Communication is to reveal this mechanism which is responsible for the amplification phenomena due to the inclusion of the weak XUV field and to prove that the enhancement is a robust single-atom phenomenon. The mechanism could suggest new types of HHG experiments. It is not limited to the description of the self-occurring case in monochromatic HHG experiments where XUV radiation, generated by the leading edge of the IR pulse, copropagates with the IR field to form a bichromatic driver field in the last part of the medium (thus leading to the extension of the cutoff energy in real experiments as compared to single-atom calculations). In cases where the XUV field saturates, it might be useful to add it externally. For example, He is known to produce higher harmonics than Ar. Hence, the support of the HGS

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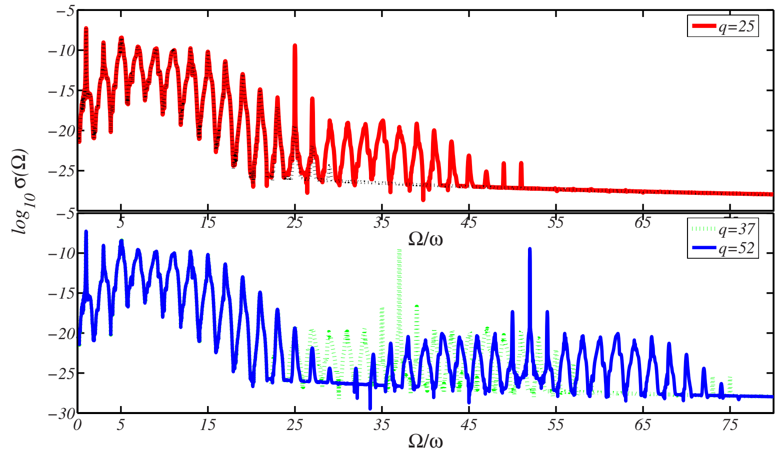

FIG. 1. (Color online) HGS obtained from a one-dimensional (1D) model Hamiltonian of Xe atom irradiated by a 50-oscillation sine-square pulse of bichromatic laser field composed of a *strong* laser field of frequency ω ($\lambda=800$ nm) and amplitude $\varepsilon_1^{\text{in}}$ ($I_1^{\text{in}}\simeq 4.3\times 10^{13}$ W/cm^2) and a *weak* field of frequency $q\omega$ and amplitude $\varepsilon_q^{\text{in}}$ ($I_q^{\text{in}}\simeq 3.5\times 10^{8}$ W/cm^2) for different values of q: $q=25$ [solid red (dark gray) line], 37 [dotted green (bright gray) line], and 52 [solid blue (dark gray) line]. HGS in the absence of the XUV field is shown in the dotted black (bright gray) line where the position of the cutoff is at the 15th harmonic. The harmonics above the 29th harmonic are enhanced in the addition of the XUV field, despite its small intensity. In addition, with respect to its center q, the distribution of the new harmonics in the HGS is symmetric [i.e., for $q=37$, $\sigma(33\omega)\simeq\sigma(41\omega)$, etc.] and upon variation of q it shifts but remains almost invariant.

obtained from Ar can be dramatically extended by illuminating the Ar with a high-order harmonic obtained from He (which is absent in the Ar HGS) in addition to the strong IR field.

In order to reveal the enhancement mechanism due to the inclusion of the weak XUV field, we study the dynamics of a single active electron in an atom described by the field-free Hamiltonian $H_0(\mathbf{r})$ subjected to a long pulse of the IR field $\mathbf{E}_1(t)$ in the length gauge and under the dipole approximation. The long pulse evolves the system adiabatically [14] from the initial ground state of the field-free Hamiltonian $|\phi_1(\mathbf{r})\rangle$ to a single resonance Floquet eigenstate $|\psi_1^{(0)}(\mathbf{r},t)\rangle$ of Eq. (1) which describes the entire dynamics of the system [15]. A formalism of *time-independent* perturbation theory is applicable since the time t may be treated as an additional coordinate [16,17]. In the following, all the parameters m, m', n, n', M, K denote integers ($\in\mathbb{Z}$).

$$H_F^{(0)}(\mathbf{r},t)|\psi_{j,m}^{(0)}(\mathbf{r},t)\rangle=\varepsilon_{j,m}^{(0)}|\psi_{j,m}^{(0)}(\mathbf{r},t)\rangle, \tag{1}$$

where $H_F^{(0)}(\mathbf{r},t)=H_0(\mathbf{r})-e\mathbf{r}\cdot\mathbf{E}_1(t)-i\hbar\partial/\partial t$ is the Floquet Hamiltonian. The indices (j,m) label the eigenstates j within any given Brillouin zone m and $\mathbf{r}$ describes the internal degrees of freedom. The Floquet eigenfunctions of this operator satisfy the c-product inner product [18,19] (written in the usual Dirac notation) $\langle\psi_{j,m}^{(0)}(\mathbf{r},t)|\psi_{j',m'}^{(0)}(\mathbf{r},t)\rangle_{\mathbf{r},t}=\delta_{jj'}\delta_{mm'}$ and form a complete set. Floquet eigenfunctions that lie within the mth Brillouin zone may be defined as $|\psi_{j,m}^{(0)}(\mathbf{r},t)\rangle\equiv|\psi_j^{(0)}(\mathbf{r},t)\rangle e^{i\omega mt}$ and $\langle\psi_{j,m}^{(0)}(\mathbf{r},t)|\equiv\langle\psi_j^{(0)}(\mathbf{r},t)|e^{-i\omega mt}$ with energies $\varepsilon_{j,m}^{(0)}\equiv\varepsilon_j^{(0)}+m\hbar\omega$. The bra and ket Floquet eigenfunctions are periodic with period $T\equiv 2\pi/\omega$ and can therefore be decomposed as a Fourier sum $|\psi_j^{(0)}(\mathbf{r},t)\rangle=\Sigma_n|\varphi_{j,n}^{(0)}(\mathbf{r})\rangle e^{i\omega nt}$ and $\langle\psi_j^{(0)}(\mathbf{r},t)|=\Sigma_n\langle\varphi_{j,n}^{(0)*}(\mathbf{r})|e^{-i\omega nt}$. Note that the Fourier components of the bra state are not complex conjugated [18].

In order to calculate the HGS one may assume the Larmor approximation [20] and analyze the time-dependent acceleration expectation value $\mathbf{a}_1^{(0)}(t)\equiv(\partial^2/\partial t^2)\langle\psi_1^{(0)}(\mathbf{r},t)|\mathbf{r}|\psi_1^{(0)}(\mathbf{r},t)\rangle_{\mathbf{r}}$ which is proportional to the emitted field. The acceleration in energy space is given by the Fourier transform $\mathbf{a}_1^{(0)}(\Omega)=(1/T)\int_0^T dt\,\mathbf{a}_1^{(0)}(t)e^{-i\Omega t}$. Exploring only frequencies that are integer multiples of ω [$\Omega=M\omega$ ($M\in\mathbb{Z}$)], and using the property $(1/T)\int_0^T dt\,e^{-i\omega nt}=\delta_{n,0}$, the expression obtained is $\mathbf{a}_1^{(0)}(M\omega)=-\omega^2M^2\Sigma_n\langle\varphi_{1,n}^{(0)*}(\mathbf{r})|\mathbf{r}|\varphi_{1,n+M}^{(0)}(\mathbf{r})\rangle_{\mathbf{r}}$. It can be shown to be nonvanishing only for integer odd values of M, which is a well-known feature of monochromatic HHG [21].

Suppose the weak XUV field $\mathbf{E}_{\mathbf{q}}(t)$ is added. A new Floquet problem is obtained, which could be described by the Floquet Hamiltonian $H_F^{\text{new}}(\mathbf{r},t)\equiv H_F^{(0)}(\mathbf{r},t)+V(\mathbf{r},t)$, where the additional term $V(\mathbf{r},t)=-e\mathbf{r}\cdot\mathbf{E}_{\mathbf{q}}(t)$ could be treated as a perturbation. Time-independent first-order perturbation theory may be used to get an approximate solution for the Floquet Hamiltonian $H_F^{\text{new}}(\mathbf{r},t)$ as

$$|\psi_1^{\text{new}}(\mathbf{r},t)\rangle=|\psi_1^{(0)}(\mathbf{r},t)\rangle+\sum_{(j',m')\neq(1,0)}c_1^{j',m'}(q)|\psi_{j'}^{(0)}(\mathbf{r},t)\rangle e^{i\omega m't}, \tag{2}$$

where the coefficients $c_1^{j',m'}(q)$ are given by

$$c_1^{j',m'}(q)=-\frac{1}{2}e\varepsilon_q^{\text{in}}\mathbf{e}_{\mathbf{k}}\cdot\sum_n\frac{\langle\varphi_{j',n}^{(0)*}(\mathbf{r})|\mathbf{r}|\varphi_{1,n+m'-q}^{(0)}(\mathbf{r})\rangle_{\mathbf{r}}+\langle\varphi_{j',n}^{(0)*}(\mathbf{r})|\mathbf{r}|\varphi_{1,n+m'+q}^{(0)}(\mathbf{r})\rangle_{\mathbf{r}}}{\varepsilon_1^{(0)}-\varepsilon_{j'}^{(0)}-m'\hbar\omega}. \tag{3}$$

Using this solution the time-dependent acceleration expectation value $\mathbf{a}_1^{\text{new}}(t) \equiv (\partial^2/\partial t^2)\langle \psi_1^{\text{new}}(\mathbf{r},t)|\mathbf{r}|\psi_1^{\text{new}}(\mathbf{r},t)\rangle_{\mathbf{r}}$ can be calculated. Keeping terms up to first order in $\varepsilon_q^{\text{in}}$, the following expression for the acceleration in the frequency domain is obtained:

$$\mathbf{a}_1^{\text{new}}(M\omega) = \mathbf{a}_1^{(0)}(M\omega) - \omega^2 M^2 \sum_{(j',m')\neq(1,0)} \sum_n [c_1^{j',m'}(q)\langle \varphi_{1,n}^{(0)*}(\mathbf{r})|\mathbf{r}|\varphi_{j',n-m'+M}^{(0)}(\mathbf{r})\rangle_{\mathbf{r}} + c_1^{j',m'*}(q)\langle \varphi_{j',n-m'-M}^{(0)*}(\mathbf{r})|\mathbf{r}|\varphi_{1,n}^{(0)}(\mathbf{r})\rangle_{\mathbf{r}}]. \tag{4}$$

This is the expression for the emitted HHG field. The HGS $[\sigma(M\omega) \equiv |\mathbf{a}_1^{\text{new}}(M\omega)|^2)$ has the same features as those presented in Fig. 1 (see [22]). The weak perturbative XUV field shifts the HGS beyond the cutoff obtained by the IR field alone. In the Floquet formalism presented here the origin of the HHG enhancement phenomenon lies in the interferences between the ground and excited Floquet states. The HGS is modified due to the new dipole cross terms introduced by the weak XUV field.

The features in the HGS could also be explained in terms of the recollision model. It was shown that the additional weak XUV field does not affect the electron trajectories, i.e., it does not modify the kinetic energy of the recolliding electron. According to the findings of the numerical simulation it must, however, affect the recombination process. To see this we turn to the semiclassical recollision model [8], where the electronic wave function at the event of recombination could be described as a sum of the following continuum and bound parts. Under the strong field approximation the returning continuum part in the direction of the polarization $\mathbf{e_k}$ (which we take as the x direction from now on for simplicity) is a superposition of plane waves $\psi_c^{\parallel}(x,t) = (1/\sqrt{2\pi})\int_{-\infty}^{\infty} dk \tilde{\psi}_c(k,t) e^{i[kx-(E_k/\hbar)t]}$ where $\mathbf{k}=k\mathbf{e_x}$ $(k=|\mathbf{k}|)$ is the momentum of the electron, $E_k \equiv \hbar^2 k^2/2m$ is the usual dispersion relation, and $\tilde{\psi}_c(k,t)$ are expansion coefficients which weakly depend on time. It is assumed that the continuum wave packet $\psi_c(\mathbf{r},t)$ is separable in the x coordinate and the two other lateral coordinates such that $\psi_c(\mathbf{r},t) = \psi_c^{\parallel}(x,t)\psi_c^{\perp}(y,z,t)$. It is assumed that the ground state is only slightly depleted during the tunnel ionization and that due to the ac Stark effect the electron adiabatically follows the instantaneous ground state of the potential, which is periodically modified by the IR and XUV fields. Since the ac Stark corrections to the instantaneous energy and wave function are small for normal field intensities, the instantaneous ground state could be approximated as $\psi_b(\mathbf{r},t) \cong \phi_1[x + \varepsilon_1^{\text{out}}\cos(\omega t) + \varepsilon_q^{\text{out}}\cos(q\omega t), y, z] e^{+(i/\hbar)I_p t}$ [where $I_p > 0$ and $\phi_1(\mathbf{r})$ are the field-free ground state eigenvalue and eigenstate, respectively]. Note that $\psi_b(\mathbf{r},t)$ approximately describes the resonance Floquet state $|\psi_1^{\text{new}}(\mathbf{r},t)\rangle$. The quiver amplitudes $\varepsilon_1^{\text{out}}$, $\varepsilon_q^{\text{out}}$ of the spatial oscillations of the ground state are of the order of $\varepsilon_q^{\text{out}} = \varepsilon_q^{\text{in}}/q^2\omega^2$, i.e., a tiny fraction of a Bohr radius for normal laser intensities and/or large values of q. The bound part may therefore be expanded in a Taylor series as $\psi_b(\mathbf{r},t) \cong e^{+(i/\hbar)I_p t}\{\phi_1(\mathbf{r}) + [\varepsilon_1^{\text{out}}\cos(\omega t) + \varepsilon_q^{\text{out}}\cos(q\omega t)](\partial/\partial x)\phi_1(\mathbf{r})\}$. Using the total wave function at the event of recombination $\Psi(\mathbf{r},t) = \psi_b(\mathbf{r},t) + \psi_c(\mathbf{r},t)$, the time-dependent acceleration expectation value $\mathbf{a}(t) \equiv (1/m)\langle \Psi(\mathbf{r},t)|-\nabla V_0(\mathbf{r})|\Psi(\mathbf{r},t)\rangle_{\mathbf{r}}$ could be calculated, where $V_0(\mathbf{r})$ is the field-free potential. The dominant terms that are responsible for the emission of radiation at frequencies other than the incident frequencies ω and $q\omega$ are the bound-continuum terms $\mathbf{a}(t) \simeq 2\,\text{Re}\langle \psi_b(\mathbf{r},t)| -\nabla V_0(\mathbf{r})|\psi_c(\mathbf{r},t)\rangle_{\mathbf{r}}$. After some algebra it can be realized that the acceleration is composed of oscillating terms of the form

$$\begin{aligned}\mathbf{a}(t) \simeq -2\,\text{Re}\,\frac{1}{\sqrt{2\pi}}\int_{-\infty}^{\infty} dk [&\tilde{\tilde{\psi}}_{\text{IR}}(k) e^{-(i/\hbar)(E_k+I_p)t} \\ &+ \tilde{\tilde{\psi}}_{\text{XUV}}(k)\varepsilon_1^{\text{out}} e^{-(i/\hbar)(E_k+I_p\pm\hbar\omega)t} \\ &+ \tilde{\tilde{\psi}}_{\text{XUV}}(k)\varepsilon_q^{\text{out}} e^{-(i/\hbar)(E_k+I_p\pm q\hbar\omega)t}],\end{aligned} \tag{5}$$

where $\tilde{\tilde{\psi}}_{\text{IR}}(k) \equiv (1/m)\tilde{\psi}_c(k)\int_{-\infty}^{\infty} d^3r\, \phi_1(\mathbf{r})\nabla V_0(\mathbf{r})\psi_c^{\perp}(y,z,t)e^{ikx}$ and $\tilde{\tilde{\psi}}_{\text{XUV}}(k) \equiv (1/2m)\tilde{\psi}_c(k)\int_{-\infty}^{\infty} d^3r[\partial\phi_1(\mathbf{r})/\partial x]\nabla V_0(\mathbf{r})\psi_c^{\perp}(y, z,t)e^{ikx}$, and the $\pm$ sign in each of the last two terms stands for summation over two terms each. The emitted field in a single recollision event is composed of a continuum of these frequencies.

It is therefore seen that, despite their small magnitude, the periodic time-dependent modulations to the ground electronic state induced by the XUV weak field of frequency $q\omega$ are responsible for the appearance of the new harmonics around q in the HGS via recombination with the returning electronic wave packet. Each electron trajectory (plane wave) with kinetic energy E_k recombines with the nucleus to emit, with equal probabilities, one of three possible photons with energies I_p+E_k, $q\hbar\omega+I_p+E_k$, or $q\hbar\omega-(I_p+E_k)$. The HGS in the presence of the IR field alone $\hbar\Omega = I_p+E_k$ is now shifted by the energy of the XUV photon $\hbar q\omega$, and new harmonics are also formed, such that their distribution about the center q is symmetric. Also, with respect to the center q, the distribution of the XUV-formed harmonics is invariant to a change in the energy of the XUV photon $\hbar q\omega$, since these harmonics are created from the same set of electron trajectories, which are characteristic of the IR field alone. When each single recollision event is repeated every half cycle of the IR field, integer harmonics $q\pm 2K$ are obtained in the HGS. To see this, note that in two consecutive recollision events at times t_r and $t_r+T/2$ the following symmetry holds: $\tilde{\psi}_c(k,t_r+T/2) = \tilde{\psi}_c(-k,t_r)$. Consequently, since $V_0(\mathbf{r})$ and $\phi_1(\mathbf{r})$ are symmetric functions for atoms [and $\partial\phi_1(\mathbf{r})/\partial x$ is antisymmetric], the following symmetry holds: $\tilde{\tilde{\psi}}_{\text{IR}}(k,t_r+T/2) = -\tilde{\tilde{\psi}}_{\text{IR}}(-k,t_r)$. The acceleration that results from the IR field therefore switches signs between subsequent recollision events, which is the origin of the odd-selection rules. However, the behavior of the coefficients resulting from the

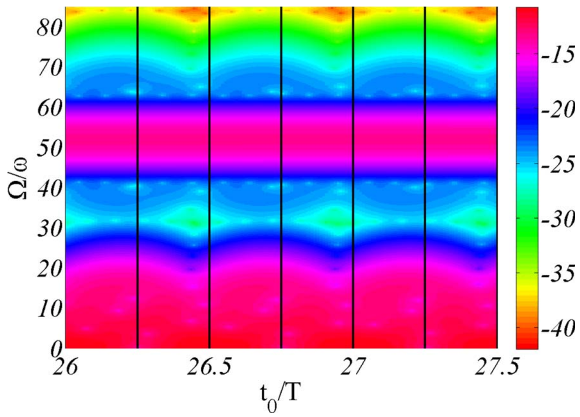

FIG. 2. (Color online) Top view [pink (dark gray) color—high intensity, yellow (bright gray) color—low intensity] of the absolute square of the Gabor-transformed acceleration expectation value $[(1/50T)\int_0^{50T} a(t) e^{-(t-t_0)^2/\tau^2} e^{-i\Omega t}$, $\tau=0.1T]$ of the quantum-mechanical simulation described in Fig. 1 for $q=52$, as a function of t_0 and Ω.

addition of the XUV field is different, $\tilde{\tilde{\psi}}_{\mathrm{XUV}}(k,t_r+T/2) = +\tilde{\tilde{\psi}}_{\mathrm{XUV}}(-k,t_r)$. The acceleration that results from the additional XUV field does not switch signs between subsequent recollision events and therefore yields even harmonics around q.

The above suggestion could be verified by plotting the time-frequency distribution of high-order harmonics (Fig. 2) obtained from the time-dependent acceleration expectation value whose spectra is given in Fig. 1 for $q=52$. In accordance with the classical recollision model, different harmonics are emitted repeatedly every half cycle, with the IR cutoff harmonic (the 15th harmonic) emitted at times $\sim 0.2T + K(0.5T)$. At those instants, the 38th and 66th harmonics, which are produced by the most energetic IR trajectory, are also emitted. Each electron trajectory in general, which under the IR field alone produces a harmonic Ω, generates upon the addition of the XUV field two duplicated new harmonics with energies $q\hbar\omega+\hbar\Omega$, $q\hbar\omega-\hbar\Omega$, and similar properties. For example, the harmonics of orders 38–48 and 56–66 have a "plateau" character (constant intensity), like the plateau harmonics 5–15. Moreover, as Eq. (5) predicts, the intensity ratio of the enhanced-plateau harmonics and the IR-plateau harmonics should be (for any of the values of q given in Fig. 1) $(\varepsilon_q^{\mathrm{out}})^2 = (\varepsilon_q^{\mathrm{in}}/q^2\omega^2)^2 \simeq 10^{-10}$, in agreement with the results of Fig. 1.

In conclusion, we have shown that the addition of a weak XUV harmonic field to a strong IR field leads to the extension of the cutoff in the HGS. The results of the quantum analytical expressions, quantum numerical simulations, and classical arguments suggest that the enhancement is a single-atom phenomenon. The seed XUV field modulates the ground state and affects the recombination process of all returning trajectories, and leads to the generation of new harmonics with structure well related to the HGS in the presence of the IR field alone. This amplification mechanism for the generation of high-order harmonics might be used to enhance the yield of harmonics in HHG experiments.

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