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Producing High Intense Attosecond Pulse Train by Interaction of Three-Color Pulse and Overdense Plasma

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Amplifying the attosecond pulse by the chirp pulse amplification method is impossible. Furthermore, the intensity of attosecond pulse is low in the interaction of laser pulse and underdense plasma. This motivates us to propose using a multi-color pulse to produce the high intense attosecond pulse. In the present study, the relativistic interaction of a three-color linearly-polarized laser-pulse with highly overdense plasma is studied. We show that the combination of ω_1, ω_2 and ω_3 frequencies decreases the instance full width at half maximum reflected attosecond pulse train from the overdense plasma surface. Moreover, we show that the three-color pulse increases the intensity of generated harmonics, which is explained by the relativistic oscillating mirror model. The obtained results demonstrate that if the three-color laser pulse interacts with overdense plasma, it will enhance two orders of magnitude of intensity of ultra short attosecond pulses in comparison with monochromatic pulse.

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In recent decades, much more attention has been paid to studying the attosecond pulses and the methods that can produce them[1−6] because the attosecond laser pulse permitted observation of control of electron wave packets,[3] probing of nuclear dynamics[4] and electronic dynamics,[5] attosecond time resolved spectroscopy[6] and tomographic imaging of molecular orbital.[7] There are different methods to create attosecond pulses, and among them high order harmonic generation (HOHG) seems to be the most effective and reliable technique and was carried out by interaction of intense laser pulse with gas or solid targets.[8,9] HOHG by interaction of ultra intense laser pulse with overdense plasma in the relativistic regime has become interesting in recent decades.[10] Harmonic generation mechanism from solid surfaces is coherent wake emission (CWE) at lower intensities, relativistic oscillating mirror (ROM) at relativistic intensities and coherent synchrotron emission (CSE) for particular conditions at relativistic intensities.[11−13] ROM allows us to model some crucial qualitative aspects of the relativistic laser-plasma interaction.[14]

The attosecond pulse with intensity significantly higher than that obtained from strong field laser-atom interaction is needed to study the ultra-fast processes in atoms, molecules and solids.[15] The best source to provide them is the high harmonics produced at the plasma surface in the relativistic regime.[16,17] Because this procedure is highly nonlinear optical process sensitive to a slight change in a dichromatic laser field,[18,19] other techniques were proposed to obtain better results and an extremely short attosecond pulse.[20,21] However, many applications remain unexplored largely because of the low harmonic intensity, and it seems that investigating the generation of attosecond pulses using multicolored fields will be very interesting.

In this Letter, we study the interaction of three-color ultra intense laser pulses and overdense plasma in the relativistic regime. In strong-field physics, shaping laser fields by combining a fundamental and its second harmonic has been successfully used for a long time,[22] and recently even three-color fields have been created and used.[23,24] However, HOHG in plasma interaction, multi-color drivers has so far not been used experimentally and even theoretically, which have barely been considered. Here we give a general outline of investigations about this new HOHG mechanism, because it is a part of the effort to increase harmonic generation efficiency, especially to shorter wavelengths and producing more intense attosecond pulses.

High-order harmonic generation from a solid surface is an interesting example of relativistic non-linear plasma physics. Incident and reflected waves make electric and magnetic fields and these fields cause the electrons near the overdense plasma boundary to be driven. However, in the plasma it is different and the electromagnetic fields decay exponentially over a distance given by the skin depth. Motion of an electron near the boundary is modelled mathematically as

$$\frac{d(m\gamma(t)\boldsymbol{v})}{dt} = \boldsymbol{F}_{\rm p} - e(\boldsymbol{E} + \boldsymbol{v}\times\boldsymbol{B}), \tag{1}$$

where m and e are mass and charge of electron, $\boldsymbol{E}$ and $\boldsymbol{B}$ show the electric and magnetic fields, respectively, $\boldsymbol{v}$ shows the electron velocity, and $\gamma(t)$ is the Lorentz factor of the electrons. If we show the normal vector on plasma by $\hat{x}$ then $\boldsymbol{F}_{\rm p} = -m\omega_{\rm p}^2 x\hat{x}$, which is obtained from the electron ion charge separation,[25] where $\omega_{\rm p}$ is the plasma frequency. Substituting $\boldsymbol{F}_{\rm p}$ into Eq. (1) and using the derivative of $\gamma(t)$ gives

$$m\gamma(t)\frac{d\boldsymbol{v}}{dt} = -m\omega_{\rm p}^2\boldsymbol{x} - e(\boldsymbol{E}+\boldsymbol{v}\times\boldsymbol{B}) + \frac{e}{c^2}(\boldsymbol{E}\cdot\boldsymbol{v})\boldsymbol{v}. \tag{2}$$

Assume a trichromatic driving field, which reads

$$\begin{aligned}\boldsymbol{E} =& [E_1\cos(\omega_1 t) + E_2\cos(\omega_2 t+\alpha)\\ &+ E_3\cos(\omega_3 t+\beta)]\hat{j},\\ \boldsymbol{B} =& [B_1\cos(\omega_1 t) + B_2\cos(\omega_2 t+\alpha)\\ &+ B_3\cos(\omega_3 t+\beta)]\hat{k}.\end{aligned}$$

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As for a highly reflective plasma, the amplitude of the reflected wave is nearly equal to that of the incident wave, the total field $\boldsymbol{B}_{\rm t}$ at the plasma surface is approximately two times larger than the magnetic field of the incident wave ($B_{\rm t} = 2E/c$). The electric field is given by $\boldsymbol{E}_{\rm t} = \zeta(\boldsymbol{B}_{\rm t} \times \hat{x})$, where $\zeta = -c\omega/\omega_{\rm p}$ is the plasma impedance, c is the velocity of light, and ω is the incident wave frequency. Substituting these fields in Eq. (2) gives

$$\ddot{x} = \gamma^{-1}\Big(- \omega_{\rm p}^2 x - F_1\frac{\dot{y}}{c} + F_2\frac{\dot{x}\dot{y}}{c^2}\Big), \tag{3}$$

$$\ddot{y} = \gamma^{-1}\Big(- F_2 + F_1\frac{\dot{x}}{c} + F_2\frac{\dot{y}^2}{c^2}\Big), \tag{4}$$

$$\ddot{z} = \gamma^{-1}\Big(F_2\frac{\dot{y}\dot{z}}{c^2}\Big), \tag{5}$$

where

$$\gamma = \Big(1 - \frac{\dot{x}^2 + \dot{y}^2 + \dot{z}^2}{c^2}\Big)^{-1/2},$$

$$F_1 = \frac{2e}{m}[E_1\cos(\omega_1 t) + E_2\cos(\omega_2 t + \alpha) + E_3\cos(\omega_3 t + \beta)],$$

$$F_2 = \frac{2e}{m}\Big[\frac{\omega_1 E_1}{\omega_{\rm p}}\sin(\omega_1 t) + \frac{\omega_2 E_2}{\omega_{\rm p}}\sin(\omega_2 t + \alpha) + \frac{\omega_3 E_3}{\omega_{\rm p}}\sin(\omega_3 t + \beta)\Big].$$

Considering $\dot{z}(0) = 0$ gives $\ddot{z} = 0$. Therefore, we should only solve the coupled differential Eqs. (3) and (4). In non-relativistic intensities $\gamma = 1$ and $x = \dot{x} = 0$, then

$$\begin{aligned}\ddot{x} = -\frac{4e^2}{m^2 c\omega_{\rm p}}\Big[&\frac{E_1^2 + E_2^2 + E_3^2}{2} + \frac{E_1^2}{2}\cos(2\omega_1 t) \\ &+ \frac{E_2^2}{2}\cos(2\omega_2 t + 2\alpha) + \frac{E_3^2}{2}\cos(2\omega_3 t + 2\beta) \\ &+ E_1E_2\cos((\omega_2 + \omega_1)t + \alpha) \\ &+ E_1E_2\cos((\omega_2 - \omega_1)t + \alpha) \\ &+ E_1E_3\cos((\omega_3 + \omega_1)t + \beta) \\ &+ E_1E_3\cos((\omega_3 - \omega_1)t + \beta) \\ &+ E_2E_3\cos((\omega_3 + \omega_2)t + \beta + \alpha) \\ &+ E_2E_3\cos((\omega_3 - \omega_2)t + \beta - \alpha)\Big].\end{aligned} \tag{6}$$

Thus the surface electron oscillating frequencies are $2\omega_1$, $2\omega_2$, $2\omega_3$, $\omega_2 + \omega_1$, $\omega_2 - \omega_1$, $\omega_3 + \omega_1$, $\omega_3 - \omega_1$, $\omega_3 + \omega_2$ and $\omega_3 - \omega_2$ and these alternative changes of electron density cause a phase modulation in the reflected wave.

We solve the coupled differential equations of motion by the fourth order Runge–Kutta numerical method. Figure 1 is plotted for the interaction of three-color pulse with mixing of frequencies ω, 2ω and 3ω to overdense plasma for different values of phase difference (α, β). In numerical solving more oscillating frequencies will certainly be found that come from the second order of approximation. In other words, when high intense laser pulse is incident on overdense plasma due to the relativistic non-linear terms, more frequencies will appear. Zero phase delay indeed maximizes the achievable instantaneous intensity. Therefore, in the following zero phases delay is chosen.

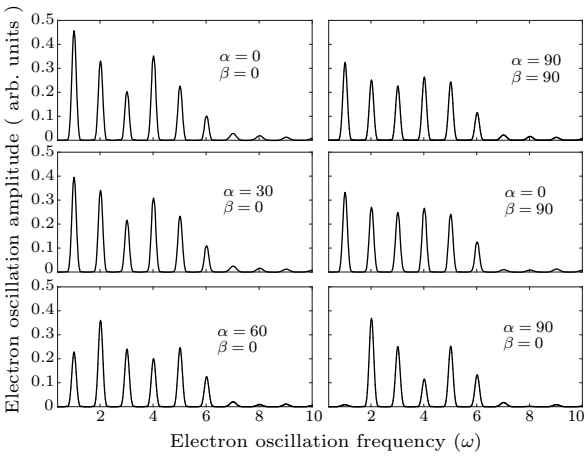

Fig. 1. The dependence of $x(\omega)$ obtained by applying the Fourier transform on $x(t)$ to phase delay in the interaction of three-color pulse and overdense plasma. The amplitude of electron oscillation is decreased with larger phase delay.

Displacement of the reflecting surface in the x-direction as $x(t) = x_0\sin(\omega_m t)$ causes the phase shift of the reflected wave as

$$\phi(t) = \phi_0\sin(\omega_m t). \tag{7}$$

The electric field $E_{\rm r}$ of the reflected wave is defined as[26]

$$\begin{aligned}E_{\rm r} &\propto \exp(-i\omega t)\exp(i\phi(t)) \\ &= \exp(-i\omega t)\sum_{n=-\infty}^{\infty} J_n(\phi_0)\exp(-in\omega_m t),\end{aligned} \tag{8}$$

where J_n is the Bessel function. Therefore, when the electron oscillating frequency is ω_m, the reflected wave frequency can be obtained as $\omega_{\rm r} = \omega + n\omega_m$. In the three-color pulse interaction $\phi(t)$ should be written in terms of all possible plasma surface frequencies $2\omega_1$, $2\omega_2$, $2\omega_3$, $\omega_2+\omega_1$, $\omega_2-\omega_1$, $\omega_3+\omega_1$, $\omega_3-\omega_1$, $\omega_3+\omega_2$ and $\omega_3 - \omega_2$. Hence, the high harmonics in the reflected wave will be produced in higher intensity.

The trichromatic driving fields in oblique p-polarized can be considered as

$$\begin{aligned}\boldsymbol{E} = &-[E_1\cos(\omega_1 t) + E_2\cos(\omega_2 t) \\ &+ E_3\cos(\omega_3 t)]\sin(\theta)\hat{i} + [E_1\cos(\omega_1 t) \\ &+ E_2\cos(\omega_2 t) + E_3\cos(\omega_3 t)]\cos(\theta)\hat{j}, \\ \boldsymbol{B} = &[B_1\cos(\omega_1 t) + B_2\cos(\omega_2 t) + B_3\cos(\omega_3 t)]\hat{k},\end{aligned} \tag{9}$$

and the angle of θ is related to the x-direction. Equation (2) of motion becomes

$$\begin{aligned}\ddot{x} = \gamma^{-1}\Big[&- \omega_{\rm p}^2 x - F_1\frac{\dot{y}}{c} - F_2\Big(\sin(\theta) \\ &- \sin(\theta)\frac{\dot{x}^2}{c^2} - \cos(\theta)\frac{\dot{x}\dot{y}}{c^2}\Big)\Big],\end{aligned} \tag{10}$$

$$\ddot{y} = \gamma^{-1}\Big[F_1\frac{\dot{x}}{c} - F_2\Big(\cos(\theta)+\sin(\theta)\frac{\dot{x}\dot{y}}{c^2} - \cos(\theta)\frac{\dot{y}^2}{c^2}\Big)\Big], \tag{11}$$

$$\ddot{z} = \gamma^{-1}\Big[F_2\Big(-\sin(\theta)\frac{\dot{x}\dot{z}}{c^2} + \cos(\theta)\frac{\dot{y}\dot{z}}{c^2}\Big)\Big]. \tag{12}$$

As the normal incidence, the coupled differential Eqs. (10) and (11) should be solved. If $\theta = 0$, then these coupled equations will be exactly the same as the normal incident equations. These equations in non-relativistic intensities can be solved as the normal incidence are solved,

$$\begin{aligned}\ddot{x} = &-\frac{4e^2\cos(\theta)}{m^2c\omega_{\rm p}}\Big[\frac{E_1^2+E_2^2+E_3^2}{2} + \frac{E_1^2}{2}\cos(2\omega_1 t)\\ &+ \frac{E_2^2}{2}\cos(2\omega_2 t) + \frac{E_3^2}{2}\cos(2\omega_3 t)\\ &+ E_1E_2\cos((\omega_2+\omega_1)t) + E_1E_2\cos((\omega_2-\omega_1)t)\\ &+ E_1E_3\cos((\omega_3+\omega_1)t) + E_1E_3\cos((\omega_3-\omega_1)t)\\ &+ E_2E_3\cos((\omega_3+\omega_2)t) + E_2E_3\cos((\omega_3-\omega_2)t)\Big]\\ &- \frac{2e\sin(\theta)}{m\omega_{\rm p}}[\omega_1E_1\sin(\omega_1 t) + \omega_2E_2\sin(\omega_2 t)\\ &+ \omega_3E_3\sin(\omega_3 t)].\end{aligned} \tag{13}$$

Hence, surface electron oscillating frequencies are ω_1, ω_2, ω_3, $2\omega_1$, $2\omega_2$, $2\omega_3$, $\omega_2+\omega_1$, $\omega_2-\omega_1$, $\omega_3+\omega_1$, $\omega_3-\omega_1$, $\omega_3+\omega_2$ and $\omega_3-\omega_2$. Equation (6) has 9 oscillating components whereas in oblique incident $E_{\rm r}$ combines 12 oscillating parts. Therefore, in an oblique incident much higher harmonics are generated.

Due to the incidence of relativistically intense laser pulse onto an overdense plasma, more frequencies for electron oscillating will appear. Thus the relativistic nonlinear mixing of these oscillations can produce harmonics. The process of harmonic generation in overdense plasma has the relativistic origin that depends on electric fields' amplitude. As shown by Eq. (13), the electron oscillation amplitude that oscillates with frequencies ω_1, ω_2 and ω_3 is related to the electrical field. However, the relativity of the electron oscillation amplitude that oscillates with frequencies $2\omega_1$, $2\omega_2$, $2\omega_3$, $\omega_2+\omega_1$, $\omega_3+\omega_1$ and $\omega_3+\omega_2$ to the electrical field is of second order. Therefore, the effect of these frequencies increases in a higher intense pulse. Therefore, the intensity of the higher order of harmonics is increased and the way to the generation of high order harmonics with higher intensities by three-color oblique incident radiation is opened.

Particle-in-cell (PIC) simulation is the most powerful tool for studying the high intense laser plasma interaction.[27] To analyze the behavior of HOHG, we simulate the interaction of laser pulse and plasma by $1D3V$ (one spatial, three velocity dimensions) PIC code. This allows one to simulate the interaction between a laser pulse and a plasma in any angle and polarizations.

To evaluate the HOHG in normal incidence, a three-color Gaussian pulse with a zero phases delay is used as the input pulse with frequencies of ω ($\lambda = 1\,\mu$m), 2ω and 3ω. The pulse duration is chosen 20 fs, and the intensity of ω, 2ω and 3ω pulses is considered $I = 10^{22}\,{\rm Wcm}^{-2}$. Total intensity is the same as the intensity of a monochromatic pulse with a frequency of ω.

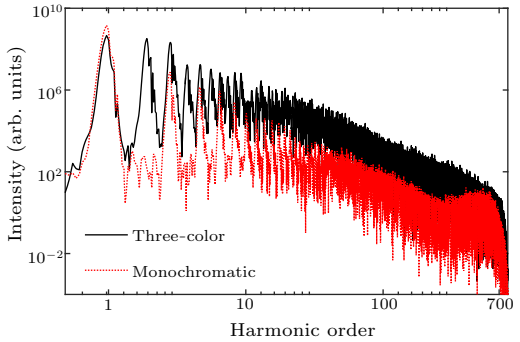

Fig. 2. Harmonic spectrum obtained by PIC simulation for three-color ω, 2ω and 3ω p-polarized at normal incident, compared with the ω spectrum in the same energy. Here $I = 3\times10^{22}\,{\rm Wcm}^{-2}$, $\lambda = 1\,\mu$m and $\omega/\omega_{\rm p} = 0.04$ are considered, and zero phases delay is chosen.

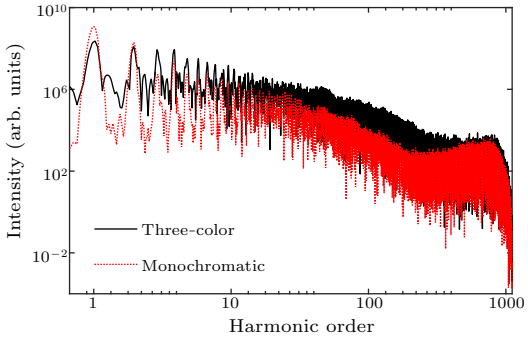

Fig. 3. Harmonic spectrum obtained by PIC simulation for three-color ω, 2ω and 3ω p-polarizations at 45° incidence angle, compared with the ω spectrum. The energy of the three-color pulse is considered to be the same as the monochromatic pulse. Here $I = 3\times10^{21}\,{\rm Wcm}^{-2}$, $\lambda = 1\,\mu$m and $\omega/\omega_{\rm p} = 0.04$ are considered, and zero phases delay is chosen.

The plasma is considered fully overdense ($\omega_{\rm p} = 25\omega$). The spectrum is evaluated by applying the Fourier transformation on the reflected pulse. Figure 2 is dedicated to show the three-color harmonic spectra obtained from the PIC simulation. A comparison between the obtained spectra and the monochromatic pulse is also shown in Fig. 2. In this simulation we use the same peak intensity of the monochromatic pulse as the combined three-color pulse. It is clear that using a monochromatic pulse we have only odd harmonics, while both odd and even harmonics are created in interactions of the three-color pulse with overdense plasma. It is exactly what we expect with the ROM model. Also, based on ROM in the three-color pulse, the relativistic mirror oscillation is composed of six Fourier components (ω, 2ω, 3ω, 4ω, 5ω and 6ω), where each can play a role in harmonic generation. To generate arbitrary harmonics, different choices for combining these six frequencies can be applied. For example to generate the harmonic of (10ω), we can mix $2\times(5\omega)$, $(6\omega)+(4\omega)$, $(6\omega)+2\times(2\omega)$, $2\times(6\omega)-(2\omega)$, $2\times(4\omega)+(2\omega)$, $(\omega)+3\times(3\omega)$, $3\times(4\omega)-(2\omega)$, $5\times(2\omega)$, and so on. This result agrees with the PIC simulation.

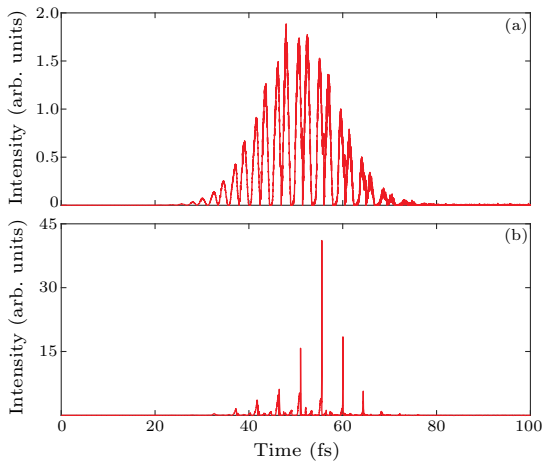

Fig. 4. Attosecond pulse train: temporal structure of the reflected radiation (a) for monochromatic pulse $I = 3\times10^{21}\,\mathrm{Wcm}^{-2}$, in which there is no attosecond pulse without filtering, (b) for three-color pulse at 45° incidence angle with $I = 10^{21}\,\mathrm{Wcm}^{-2}$ for each frequency. The FWHM duration of attosecond pulses is about 15 As.

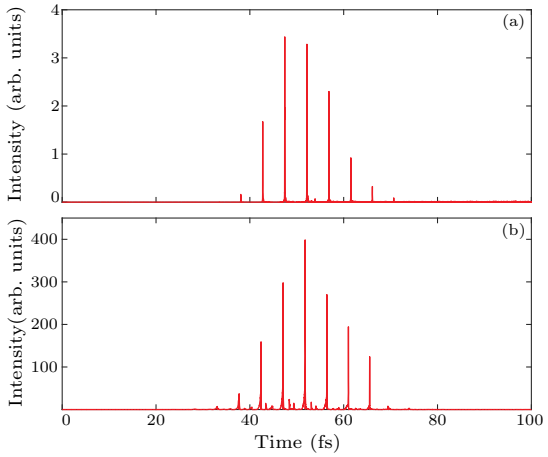

Fig. 5. Attosecond pulse train: temporal structure of the reflected radiation (a) for monochromatic pulse, $I = 3\times10^{22}\,\mathrm{Wcm}^{-2}$, attosecond pulse train seen after spectral filtering ($m > 30$), where the FWHM duration is about 10 As, (b) for three-color pulse at 45° incidence angle that the FWHM duration of attosecond pulses is about 10 As as seen for the same duration, where the intensity of the attosecond pulse produced by the three-color pulse is 115 times greater than the monochromatic pulse.

The simulation for oblique incidence of the three-color pulse on the overdense plasma has the same results as the normal incidence. The pulse is incident on the target with p-polarization at the incidence angle of 45°. Figure 3 shows one of the typical harmonic spectra obtained from the PIC simulation. The spectrum of the obtained high harmonics indicates that the generated harmonics for the three-color pulse are more intense than those for the monochromatic pulse, specially in high orders. This is consistent with the ROM model prediction. In Fig. 4 the peak intensity of the three-color pulse is considered as $I = 3.0\times10^{21}\,\mathrm{Wcm}^{-2}$. Each of the three frequencies ω, 2ω and 3ω has the same intensity. The FWHM duration of reflected attosecond pulse is about 15 As. The intensity and duration of the attosecond pulses are the same as those produced with the two-color pulse with peak intensity $I = 2.74\times10^{22}\,\mathrm{Wcm}^{-2}$.[22] In other words, in the three-color pulse, the incident pulse with 9.13 times lower intensity than that in the two-color pulse gives the same attosecond pulse. The lower intensities are more available practically. In the case of three-color oblique incidence with peak intensity $I = 3\times10^{22}\,\mathrm{Wcm}^{-2}$ (Fig. 5) the attosecond pulse train with the FWHM duration of about 10 As is produced. However, for the monochromatic pulse, the proper filtering ($m > 30$) is needed for the same attosecond pulse duration. The intensity of the attosecond pulse produced by the three-color pulse is 115 times greater than the pulse produced by the monochromatic pulse in the same duration.

In summary, we have shown that using the multi-color incident pulse can increase the intensities of the higher order of harmonics. This result is confirmed by ROM modeling and PIC simulation. We have also shown that due to the increase of the intensities of higher order of harmonics by combining the frequencies, the instance FWHM reflected attosecond pulse will decrease. The PIC simulation confirms this obtained result. Our finding shows that the interaction of high intense multi-color incident pulse and solid could be an excellent source of high intense attosecond pulses. The results demonstrate the ability of our proposed harmonic generation process to generate attosecond pulses even in practically available intensities. Three-color HOHG holds the promise to generate attosecond pulses with unprecedented intensities, which will open up a new regime of attosecond physics.

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