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Broadband UV Supercontinuum Generation by Three-Color Ionizing Laser Pulses

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Abstract: We theoretically investigated the properties of the ultraviolet secondary radiation generated under the action of three-color ionizing pulses obtained from parametric light generators. We show that the combination components in the spectrum of electron currents generated under the action of such three-color pulses can form a strictly exponentially decreasing sequence. For moderately short pump pulse durations (30 fs at 800 nm main field) with the use of this effect, it is possible to generate a smooth broadband supercontinuum in the 800–100 nm range.

Keywords: supercontinuum; ultraviolet; ionization; nonlinear optics; wavemixing; TDSE

1. Introduction

Gas ionization in strong optical laser fields can be accompanied by secondary radiation generation in a variety of frequency ranges: terahertz and mid-IR [1–7]; generation of Brunel harmonics (from the third to the ninth for a quasi-monochromatic field) [8–11]; and high harmonic generation (up to the 50th and even higher) [12,13].

The control over the parameters of the generated radiation is possible by changing the parameters of the laser pump [3–5]. When using multicolor ionizing pulses, the task of controlling the output parameters, on the one hand, becomes more complicated, but at the same time, it becomes possible to optimize the process, for example, to increase the efficiency of secondary radiation [14,15].

One of the most well-studied problems of laser-plasma nonlinear optics is the generation of terahertz radiation under the action of two-color pulses. Such pulses usually consist of a strong field with a small addition of its second harmonic [1–3], although other possibilities, including utilizing three-color pulses, have been actively studied recently [4–7]. At the same time, the generation of Brunel harmonics under the action of non-monochromatic pulses has historically received less attention. However, in a recent experiment, it was shown that when using few-cycle two-color pulses with the addition of a second harmonic, the secondary radiation can become a dense sequence of spectral components, forming a supercontinuum [10]. Radiation with such a broad ultraviolet (UV) spectrum is in demand in many areas related to high-precision measurements, detection of the composition of various media and tissues, and more [10,16–19].

In this work, we investigate the effect of alignment of the sequence of spectral components of secondary radiation, which occurs under the action of three-color pulses. We show that this alignment is not very sensitive to the intensity ratios in the ionizing pulse. We also demonstrate that, at moderately short (up to 30 fs for a 800 nm pump) durations of ionizing pulses, the generation of an aligned (smoothly decreasing) filled supercontinuum in the range of 800–100 nm is possible.

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2. Equations

We studied the spectra of secondary radiation produced under the action of three-color pulses with component parameters typical for optical parametric amplifiers (OPA) [4,10,12],

$$\mathbf{E}(t) = \mathbf{x}\big(E_0\cos(\omega_0 t) + E_+\cos([\omega_0/2+\Delta\omega]t+\phi_+) + E_-\cos([\omega_0/2-\Delta\omega]t+\phi_-)\big)f(t), \quad (1)$$

where $E_{0,\pm} = \sqrt{8\pi I_{0,\pm}/c}$, $I_{0,\pm}$ are peak intensities, c is the speed of light, $f(t)$ is bell-shaped envelope, and $\phi_\pm$ are phase shifts between the main and additional waves carriers. We use $f(t) = \cos^2(\pi t/2T)$ for $|t| \le T = \pi\tau_p/4\cos^{-1}(2^{-1/4})$, where τ_p is the full-width intensity at half-maximum (FWHM) duration, and set $\mathbf{E}(t) = 0$ for $|t| > T$. We also use the notation $\omega_\pm = \omega_0/2 \pm \Delta\omega$ later in the article. Such a field as given by Equation (1) can be obtained by passing a strong pulse with central frequency ω_0 (main field) through OPA, which generates two additional waves with frequencies $\omega_\pm$ tunable near the half-harmonic of the main field. We consider here He atoms as a nonlinear medium. Analysis of secondary radiation is performed by solving the three-dimensional time-dependent Schrödinger Equation (3D TDSE) in the single-active electron approximation

$$i\hbar\frac{\partial}{\partial t}\psi(\mathbf{r},t) = \left[-\frac{\hbar^2}{2m}\nabla^2 - e\mathbf{r}\mathbf{E}(t) + V(r)\right]\psi(\mathbf{r},t), \quad (2)$$

where $\psi(\mathbf{t},t)$ is the electron wavefunction, $\hbar$ is the reduced Plank constant, e and m are the electron charge and mass, respectively, and $V(r)$ is the parent ion potential. The potential $V(r)$ for He atom is found based on the density functional theory [20], which has binding energies in good agreement with the experimental data. The numerical 3D TDSE solution was performed similarly to [20] using the expansion of the wavefunction in spherical harmonics with maximum orbital momentum $l_{\max} = 256$ and with radial boundary at $r_{\max} = 200$ atomic units. The generated radiation is characterized by Fourier spectrum $\mathbf{a}_\omega = \int \mathbf{a}(t)e^{i\omega t}dt$ of the atom dipole acceleration $\mathbf{a}(t)$, which determines the time derivative of the electron current density, $\partial\mathbf{j}/\partial t$:

$$\frac{\partial\mathbf{j}}{\partial t} = eN_g\mathbf{a}, \quad \mathbf{a} = \frac{1}{m}[e\mathbf{E} - \langle\psi|\nabla V|\psi\rangle], \quad (3)$$

where N_g is the initial gas density.

3. Results and Discussion

Figure 1 shows the spectra of $d\mathbf{j}/dt$ at various values of the frequency detuning $\Delta\omega$. These spectra consist of sets of peaks, corresponding to different combinational frequencies of the form

$$\omega_n = P_0\omega_0 + P_+\omega_+ + P_-\omega_-, \quad (4)$$

where $P_{0,+,-}$ are integers with $P_n = |P_0| + |P_+| + |P_-|$ being a small odd number (the larger P_n, the less pronounced the corresponding peak) [6,9,11,21]. The amplitudes of combination components can be approximately calculated using the mathematical approaches described in the articles [9,11]. It is noticeable that the spectra on subplots (a) and (c) differ from cases (b) and (d). This is due to the special values of frequency detuning $\Delta\omega = \omega_0/k$ (where k are specific even natural numbers), at which the set of frequencies in a three-color pulse becomes a set of harmonics: (a) $\omega_- = \omega$, $\omega_+ = 2\omega$ and $\omega_0 = 3\omega$; (c) $\omega_- = 2\omega$, $\omega_+ = 3\omega$ and $\omega_0 = 5\omega$. In these cases, almost every peak in the spectrum of $d\mathbf{j}/dt$ corresponds to several sets of frequency combinations, for example, in case (c), the frequency $2\omega_0$ is obtained with $(P_-, P_+, P_0) = (1,1,1)$, $(5,0,0)$ and $(-1,4,0)$. In other words, such values of $\Delta\omega$ correspond to the merging of several spectral peaks. And if, at random values of $\Delta\omega$, such as presented on (b) and (d), the amplitudes of various peaks look disorganized, then when the peaks merge, their amplitudes line up in a visible sequence.

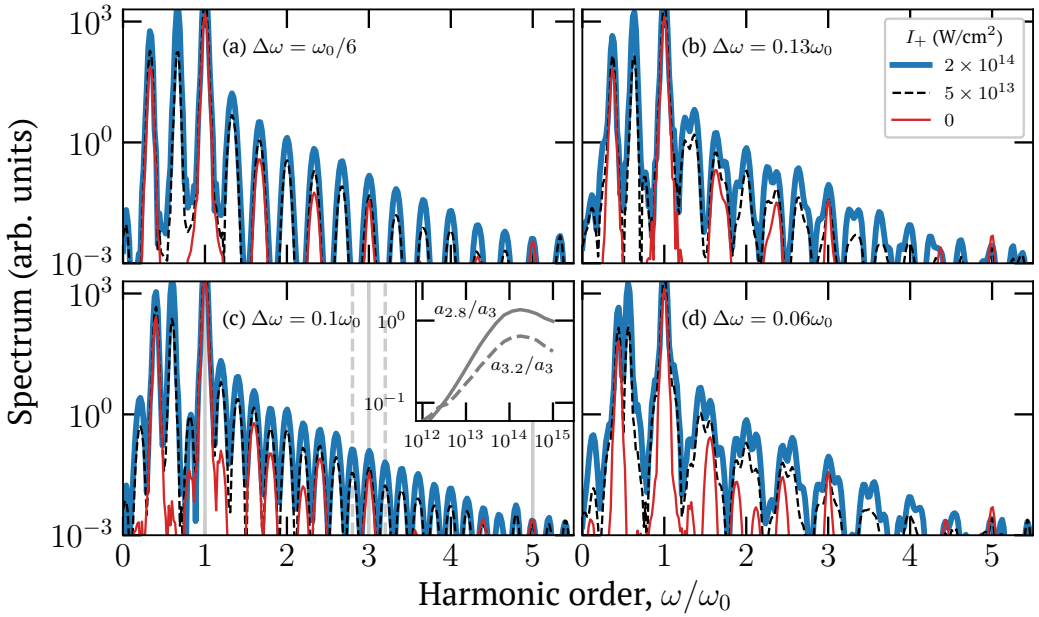

Figure 1. $d\mathbf{j}/dt$ squared spectrum from three-color ionizing pulses with ω_0 corresponding to $\lambda_0 = 800$ nm, $\omega_\pm = \omega_0/2 \pm \Delta\omega$ with four different values of $\Delta\omega = \omega_0/6$ (**a**), $0.13\omega_0$ (**b**), $0.1\omega_0$ (**c**), and $0.06\omega_0$ (**d**). Intensities $I_0 = 10^{15}$ W/cm^2 and $I_- = 10^{14}$ W/cm^2 are fixed, for blue lines $I_+ = 2 \times 10^{14}$ W/cm^2, for black lines $I_+ = 5 \times 10^{13}$ W/cm^2 and for red lines $I_+ = 0$. All the phases are equal to 0, pulse duration $\tau_p = 50$ fs. The inset to subplot (**c**) shows how the ratios between the amplitudes of different peaks depend on I_+: $a_{2.8}/a_3$ (solid line) and $a_{3.2}/a_3$ (dashed line), where a_n is the amplitude of the $d\mathbf{j}/dt$ squared spectrum at $\omega = n\omega_0$. I_0 and I_- are fixed at the same values as on the other curves.

This alignment looks unexpected, first of all, because the intensities of the ionizing pulse components were not selected specially. What is more, from Figure 1 we see that the effect weakly depends on the intensities of the components: when the intensity I_+ changes by the factor of four (solid blue and dashed black lines), the alignment does not break. More generally, it remains for any intensities $I_{0,\pm}$ if they differ by no more than an order of magnitude. A specific example of the dependence of the ratios between the amplitudes of two "satellite" peaks near the third harmonic and its amplitude is shown in the inset to the subplot (c). The peak centered at $2.8\omega_0$ is absent at $I_+ = 0$ and the peak at $3.2\omega_0$ is sufficiently smaller than at $3\omega_0$. However, at $I_+ \approx 4 \times 10^{12}$ W/cm^2, the peak at 2.8 overtakes the one at 3.2 and the alignment stabilizes near $I_+ \approx 5 \times 10^{13}$ W/cm$^2 = I_-/2$.

This behavior can be qualitatively explained using the standard semiclassical (photocurrent) model [9,11,22]. First, note that if electric field is a sum of harmonics $|\mathbf{E}|^2 = \sum I_s e^{is\omega t}$, where I_s are slow complex amplitudes, then $|\mathbf{E}|^4 = \sum[\sum I_k I_{s-k}]e^{is\omega t}$ and so on. In the work [11], we showed that the ionization probability, which is the source of nonlinearity in this system, can be approximated with a high order even polynomial. When a field module is raised to a high power, its spectrum undergoes numerous convolutions with itself. The same happens when summing up a set of random variables with given probability distributions. So, the appearance of order here is of the same nature as the appearance of the Gaussian envelope of the probability distribution of the sum of the set of random variables (central limit theorem [23]). In sparse spectra, the convolution operation generates new sum and difference frequencies, but if the spectrum is already a dense sequence of harmonics (cases (a) and (c)), then the convolution operation only transforms the shape of this sequence, ordering it.

Smooth Supercontinuum Generation

When reducing the duration of the ionizing pulse to approximately 30 fs, the components of the generated spectrum begin to merge (Figure 2), and for the case $\Delta\omega = \omega_0/10$, in which the frequency interval between adjacent peaks is less, at a duration of 20 fs we obtain a very smooth supercontinuum. With $\phi_\pm = 0$, subplots (a) and (b), the spectrum of $d\mathbf{j}/dt$ can become almost a straight line. At the same time, by controlling the phases $\phi_\pm$ of the components, we can obtain quasi-step structures, each step of which is a more conventional supercontinuum; examples are shown in Figure 2 (c, d). The relative phase of each distinguished component (4) is given by $\phi_n = P_-\phi_- + P_+\phi_+$, and when the peaks overlap, only at $\phi_\pm = 0$ do they all add up efficiently and line up at the same time. In our opinion, the case $\phi_\pm = 0$ is preferable for the usage of this supercontinuum for measurements, for example, of absorption in a medium and solving other typical problems, due to the best alignment [10,18,24,25]. We also note that in the two-color experiment [10], the correct phase shift between the components was also carried out to obtain the best effect.

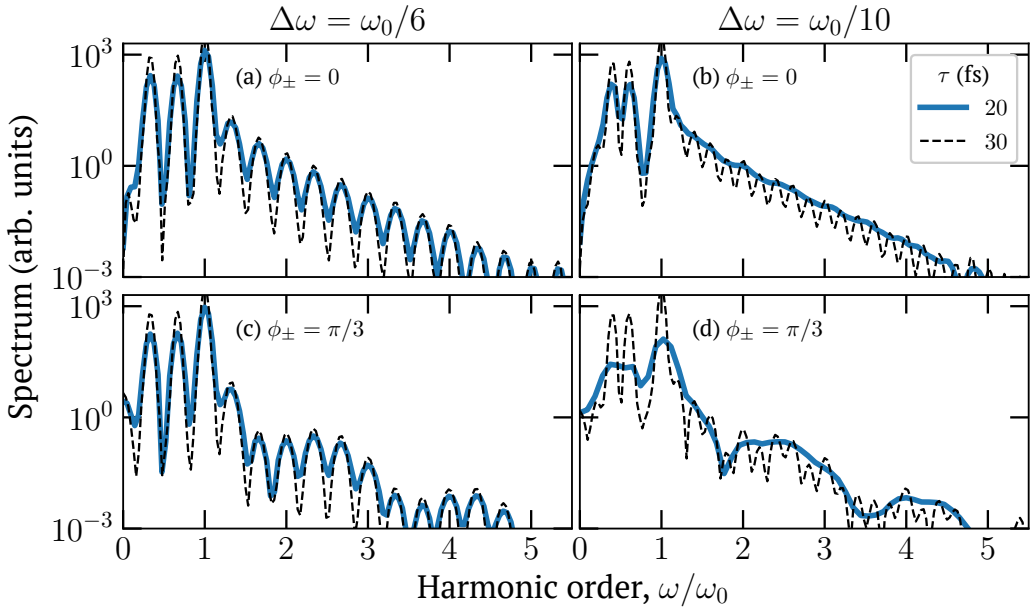

Figure 2. $d\mathbf{j}/dt$ squared spectrum from three-color ionizing pulses with ω_0 corresponding to $\lambda_0 = 800$ nm, $\omega_\pm = \omega_0/2 \pm \Delta\omega$ with two different values of $\Delta\omega = \omega_0/6$ (**a**,**c**) and $\omega_0/10$ (**b**,**d**) and two different values of phases $\varphi_+ = \varphi_- = 0$ (**a**,**b**) and $\varphi_+ = \varphi_- = \pi/3$ (**c**,**d**). Intensities $I_0 = 10^{15}$ W/cm^2 and $I_+ = I_- = 2 \times 10^{14}$ W/cm^2. Blue lines correspond to pulse duration $\tau_p = 20$ fs, dashed black lines to $\tau_p = 30$ fs.

On the Figure 3, we compare the spectra from three-, two- and one-color fields. From the result for the one-color field, one can obtain a visual representation of the spectrum width of the field itself (the spectrum of the first harmonic repeats the spectrum of the field of the ionizing pulse), the components in the two- and three-color fields have the same spectral width. It is clearly seen here that the usual two-color field is not enough to achieve a smooth supercontinuum even at 20 fs duration for a 800 nm main field. Here we have the number of cycles of the main field nearly equal to the one in the experiment [10], where there were pulses with 80 fs duration and 3.9 µm central wavelength. With further shortening of the duration, it becomes difficult to efficiently generate additional fields of the ionizing pulse (second or half harmonic). Thus, the three-color generation method seems to be a very effective solution for the UV-supercontinuum generation. Additionally, since both additional waves in the proposed three-color pulse can be generated in the same OPA with the desired frequency detuning [4], the use of a three-color pulse, in fact, does not require much complication of the circuit. Such table-top schemes for generating an

ultrabroadband smooth supercontinuum may possibly find application in modern biology and medicine [16,19].

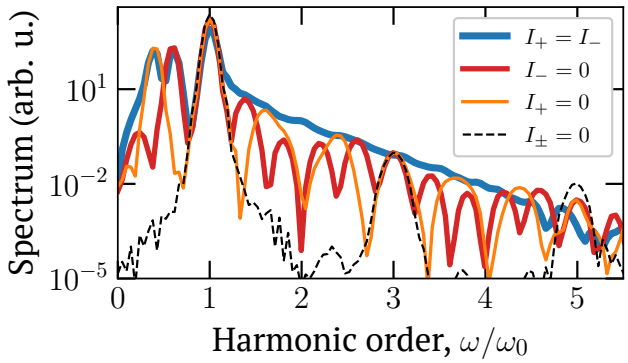

Figure 3. $d\mathbf{j}/dt$ squared spectrum from one-color (black dashed line), two-color (red and orange lines) and three-color (blue line) ionizing pulses with ω_0 corresponding to $\lambda_0 = 800$ nm, $\omega_\pm = \omega_0/2 \pm \Delta\omega$ with $\Delta\omega = \omega_0/10$. The pulse intensities are chosen so that the degree of gas ionization is approximately the same: $I_0 = 10^{15}$ W/cm^2, $I_+ = I_- = 2 \times 10^{14}$ W/cm^2 (blue line); $I_0 = 1.5 \times 10^{15}$ W/cm^2, $I_\pm = 2 \times 10^{14}$ W/cm^2, $I_\mp = 0$ (red and orange lines, respectively); $I_0 = 2 \times 10^{15}$ W/cm^2, $I_+ = I_- = 0$ (black dashed line). Pulse duration $\tau_p = 20$ fs, phases $\varphi_\pm = 0$.

Compared to other methods for generating ultraviolet supercontinuum, the one proposed in this paper has a number of advantages. Firstly, no special environment is required for generation. Our calculations were carried out for atomic helium, but similar generation is also possible using other atoms and molecules, as was demonstrated in the two-color experiment [10]. The same applies to the choice of the frequency of the main field of the ionizing pulse: provided that the tunneling regime of ionization is maintained, other frequencies can be used, and the range of the generated supercontinuum itself will shift proportionally, and this range can be very wide. Moreover, it is not even necessary to adjust certain values of the intensities of the additional field components, as the generation of a smooth supercontinuum occurs at any comparable intensities if the components are correctly phased. In turn, phase control allows customizing the type of generated spectrum to obtain wide nearly-horizontal areas. Using the other methods for generation, as a rule, requires a strictly defined pump frequency and special properties of a specific working medium to generate such a supercontinuum [18,26]. All of the above indicates the serious prospects of the method proposed in this article.

4. Conclusions

We have studied the spectra of Brunel combination components arising under the action of three-color ionizing pulses generated in optical parametric amplifiers. We have found an effect of alignment, due to which all combination components line up in a strictly geometrically decreasing sequence for certain values of the frequency detuning of the additional components of the three-color pulse, detuned from the half frequency of the main field. This effect can be used to generate an ultrabroadband smooth supercontinuum in the range of 800–100 nm with a pump duration of about 30 fs. Fine tuning of the intensities of the pump components is not required for generation of a supercontinuum, and an almost arbitrary gas is suitable as a working medium.

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Data Availability Statement: The data underlying the results presented in this paper are not publicly available at this time, but may be obtained from the authors upon reasonable request.

Conflicts of Interest: The authors declare no conflicts of interest.

Abbreviations

The following abbreviations are used in this manuscript:

FWHM	full width at half maximum
OPA	optical parametric amplifier
TDSE	time-dependent Schrödinger equation
UV	ultraviolet

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