Generation of few- and subcycle radiation at combination frequencies of ultrashort multicolor ionizing laser pulse

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The ionization of a medium by strong femtosecond two-color pulses attracts steadily growing interest associated with accompanying generation of secondary electromagnetic radiation. The generation is particularly manifested around various combination frequencies (CFs) of the two-color pump in different frequency ranges, from terahertz to the deep-ultraviolet. In the last two decades the most attention was given to the two-color laser-plasma generation of terahertz waves where the associated combination frequency is small enough or zero. The considerable success was achieved in obtaining strong and broadband terahertz radiation [1, 2] as well as in exploring various ways to tailor the two-color scheme. These ways include manipulating polarizations and frequencies of the one-color components of the ionizing pump, pump focusing and properties of the ionized medium: state of matter, pressure, and kind of ionized particles [1–6]. Recently, an advance was made from terahertz to the neighboring mid-infrared range with incommensurate two-color pulses employed, opening a way to generate few- and subcycle pulses tunable over the whole mid-infrared range [3, 7–9]. Some works also consider the higher CFs in visible and even ultraviolet range where the generation of tunable high-quality extremely short pulses was demonstrated [9–11]. However, such ionization-induced wavemixing leading to generation at higher CFs remains mostly unexplored as compared to the terahertz generation.

In this work, we present an analytical approach that should facilitate study of the ionization-induced wavemixing. This approach allows one to calculate radiating currents generated by ionizing two-color femtosecond pulse at CFs for arbitrary polarizations and ratios between intensities of one-color pump components. The method is based on the decomposition of ionization rate over quasimonochromatic components at various CFs and describes the excitation of electron current at any CF in a unified way (regardless of the particular frequency range). Using this ionization rate decomposition method, one can find the slow envelope of the current component at some CF as dependent on parameters of the ionizing two-color pump: intensities, polarizations, durations, and chirps of its one-color components as well as the phase and group shifts between one-color components. Knowing these dependences, one can simplify electrodynamical program codes or estimate analytically the conversion efficiency in some focusing models (for example, for short microplasmas from tightly focused pumps or for long quasi-uniform plasma columns from axicon-focused pumps).

Let us consider the two-color ionizing field $\mathbf{E}(t)=\mathbf{E}_0(t)+\mathbf{E}_1(t)$, where $\mathbf{E}_{0,1}(t)= =\operatorname{Re}\mathbf{A}_{0,1}(t)e^{-i\omega_{0,1}t}$ are the one-color components of the two-color pump, $\mathbf{A}_{0,1}(t)$ and $\omega_{0,1}$ are their slow complex amplitudes and frequencies, respectively. In the tunnel ionization regime, electrons transfer from bound to continuum state quickly enough, and one can introduce the ionization probability per unit time, w, which is determined by the instantaneous field value. For the strong-field ionization of a common gas of non-aligned molecules, the ionization probability w does not depend on the direction of the electric field and can be written as a function of the field magnitude, $w=w(|\mathbf{E}|)$. The electric field squared $\mathbf{E}^2$ can be represented as superposition of a slow component and components at frequencies $2\omega_{0,1}$ and $|\omega_0\pm\omega_1|$,

$$\mathbf{E}^2=P[1+q_0\cos(2\zeta-2\varphi_0)+q_1\cos(2\eta-2\varphi_1)+q_+\cos(\zeta+\eta-\varphi_+)+q_-\cos(\zeta-\eta-\varphi_-)],$$

where $P=\left(|\mathbf{A}_0|^2+|\mathbf{A}_1|^2\right)/2=4\pi I/c$ is proportional to the total intensity I, c is the speed of light, $q_{0,1}=\left|\mathbf{A}_{0,1}^2\right|/2P$, $q_+=|\mathbf{A}_0\cdot\mathbf{A}_1|/P$, $q_-=|\mathbf{A}_0\cdot\mathbf{A}_1^*|/P$, $\varphi_{0,1}=\left(\arg\mathbf{A}_{0,1}^2\right)/2$, $\varphi_+= =\arg(\mathbf{A}_0\cdot\mathbf{A}_1)$, $\varphi_-=\arg\left(\mathbf{A}_0\cdot\mathbf{A}_1^*\right)$ are the parameters characterizing phase and polarization structure of the field (the 'mutual polarization'), and $\zeta=\omega_0 t$, $\eta=\omega_1 t$.

The ionization probability $w(|\mathbf{E}|)$ is 2π-periodic with respect to ζ and η. This allows one to decompose w into the double Fourier series,

$$w[|\mathbf{E}(t)|]=\sum_{\alpha,\beta=-\infty}^{\infty}w_{\alpha\beta}(t)e^{-i\omega_{\alpha\beta}t}, \tag{1}$$

where $\omega_{\alpha\beta}=\alpha\omega_0+\beta\omega_1$ are the CFs and $w_{\alpha\beta}$ are the time-dependent Fourier coefficients; $w_{\alpha\beta}\equiv 0$ for odd $\alpha+\beta$ and

$$w_{\alpha\beta}(t)=e^{i[\alpha\varphi_0(t)+\beta\varphi_1(t)]}W_{\alpha\beta}[P(t),q_0(t),q_1(t),q_+(t),q_-(t),\psi_+(t),\psi_-(t)],$$

$$W_{\alpha\beta}[P,q_0,q_1,q_+,q_-,\psi_+,\psi_-]=\frac{1}{4\pi^2}\int_{-\pi}^{\pi}\int_{-\pi}^{\pi}e^{i\frac{(\alpha+\beta)u+(\alpha-\beta)v}{2}}\times$$

$$\times w\left(\sqrt{P[1+q_0\cos(u+v)+q_1\cos(u-v)+q_+\cos(u-\psi_+)+q_-\cos(v-\psi_-)]}\right)du\,dv,$$

for even $\alpha+\beta$, $\psi_\pm=\varphi_\pm-\varphi_0\mp\varphi_1$. Despite the cumbersome look, the above integrals can be evaluated to closed-form expressions for common steep dependences $w(|\mathbf{E}|)$ in virtually any reasonable particular case. Here, we give formulas for the most important cases of (i) parallel linear polarizations, $\mathbf{A}_{0,1} = M_{0,1} e^{i\varphi_{0,1}} \hat{\mathbf{x}}$ and

$$W_{\alpha\beta} \approx \frac{1}{2} w(M_0 + M_1) e^{-n} I_\alpha \left(\frac{nM_0}{M_0 + M_1} \right) I_\beta \left(\frac{nM_0}{M_0 + M_1} \right),$$

(ii) orthogonal linear polarizations, $\mathbf{A}_0 = M_0 e^{i\varphi_0} \hat{\mathbf{x}}$, $\mathbf{A}_1 = M_1 e^{i\varphi_1} \hat{\mathbf{y}}$ and

$$W_{\alpha\beta} \approx w\left(\sqrt{M_0^2 + M_1^2}\right) e^{-\frac{n}{4}} I_{\frac{\alpha}{2}} \left(\frac{nM_0^2}{4(M_0^2 + M_1^2)} \right) I_{\frac{\beta}{2}} \left(\frac{nM_1^2}{4(M_0^2 + M_1^2)} \right)$$

for even indices, $W_{\alpha\beta} \equiv 0$ for odd ones, and (iii) coplanar circular polarizations, $\mathbf{A}_0 = M_0(\hat{\mathbf{x}} + i\hat{\mathbf{y}})$, $\mathbf{A}_1 = M_1 e^{i\psi_\mp}(\hat{\mathbf{x}} \pm i\hat{\mathbf{y}})$ and

$$W_{\alpha,\mp\alpha} \approx e^{i\alpha\psi_\mp} w(M_0 + M_1) \exp\left(-\frac{nM_0 M_1}{(M_0 + M_1)^2} \right) I_\alpha \left(\frac{nM_0 M_1}{(M_0 + M_1)^2} \right),$$

$W_{\alpha\beta} \equiv 0$ for $\alpha \pm \beta \neq 0$ with the upper and lower sings corresponding to the co- and counter-rotating polarizations, respectively. In these formulas, $I_k(z)$ denotes the modified Bessel function; $\hat{\mathbf{x}}$ and $\hat{\mathbf{y}}$ are the orthogonal unit vectors; $M_{0,1}$ are the field amplitudes; and $n = w'(E)E/w(E) >> 1$ is characteristic nonlinearity order (number of mixed waves) with $E = M_0 + M_1$ for the first and third cases, and $E = \sqrt{M_0^2 + M_1^2}$ for the second one.

Note that expansion (1) is exact and does not impose any constraints on the complex amplitudes $\mathbf{A}_{0,1}(t)$ (aside from obvious finiteness). That said, this expansion fits naturally with slow enough amplitudes $\mathbf{A}_{0,1}(t)$ in the time scale of the ionization duration τ_i (i.e., characteristic time of plasma creation) when the coefficients $w_{\alpha\beta}(t)$ depend smoothly on time and each term in sum (1) presents a harmonic at some CF. In this case, the expansion (1) over various CFs can be used to analyze spectral composition of plasma density $N(t)$ and free-electron current density $\mathbf{j}(t)$. To that end, one should substitute the expansion (1) into commonly used equations

$$\frac{\partial N}{\partial t} = (N_m - N)w, \qquad \frac{\partial \mathbf{j}}{\partial t} = \frac{Nq_e^2}{m_e}\mathbf{E}, \tag{2}$$

where N_m is the initial density of neutral particles, q_e is the elementary charge, and m_e is the electron mass. In accordance with Eqs. (2), the electron current density can be also decomposed as a superposition of harmonics at CFs,

$$\frac{\partial \mathbf{j}}{\partial t} = \frac{1}{2} \sum_{\substack{a,b=-\infty \\ a+b \text{ is odd}}}^{\infty} \mathbf{F}_{ab}(t) e^{-i\omega_{ab} t},$$

where $\mathbf{F}_{ab}$ are the complex amplitudes of these harmonics. For a and b with $|\omega_{a\pm1,b}|$, $|\omega_{a\pm1,b}| >> 1/\tau_i$, one can express $\mathbf{F}_{ab}$ using $W_{\alpha\beta}$, $\mathbf{F}_{ab} \approx \left(iq_e^2/m_e\right) e^{i(a\varphi_0 + b\varphi_1)} \left(\partial\overline{N}/\partial t\right) \mathbf{G}_{ab}$,

$$\mathbf{G}_{ab} = \frac{1}{\overline{w}} \left[\frac{\omega_{ab} U_{ab}^{(+)} - \omega_0 U_{ab}^{(-)}}{\omega_{ab}^2 - \omega_0^2} \mathbf{M}_0 + i \frac{\omega_{ab} U_{ab}^{(-)} - \omega_0 U_{ab}^{(+)}}{\omega_{ab}^2 - \omega_0^2} \mathbf{m}_0 + \right.$$

$$\left. + \frac{\omega_{ab} V_{ab}^{(+)} - \omega_1 V_{ab}^{(-)}}{\omega_{ab}^2 - \omega_1^2} \mathbf{M}_1 + i \frac{\omega_{ab} V_{ab}^{(-)} - \omega_1 V_{ab}^{(+)}}{\omega_{ab}^2 - \omega_1^2} \mathbf{m}_1 \right],$$

where $\mathbf{M}_{0,1} = \mathrm{Re}\left(\mathbf{A}_{0,1} e^{-i\varphi_{0,1}}\right)$ and $\mathbf{m}_{0,1} = \mathrm{Im}\left(\mathbf{A}_{0,1} e^{-i\varphi_{0,1}}\right)$ are the major and minor semiaxis vectors of polarization ellipses, $\overline{N}(t) = N_m\left[1 - \exp\left(-\int_{-\infty}^{t} \overline{w}(t')dt'\right)\right]$ and $\overline{w}(t) \equiv W_{00}(t)$ are the time-averaged plasma density and ionization probability, $U_{ab}^{(\pm)} = W_{a+1,b} \pm W_{a-1,b}$, $V_{ab}^{(\pm)} = = W_{a,b+1} \pm W_{a,b-1}$. The resulting expressions for $\mathbf{F}_{ab}$ and $\mathbf{G}_{ab}$ can be now used for analyzing generation of CFs in various situations. For short radiating plasmas (microplasmas), which can be obtained from tightly focused pumps, one can use dipole approximation from plasma object of subwavelength size and critical density and obtain the efficiency estimate $\eta_{ab} \sim \sigma_f^2 |\mathbf{G}_{ab}(t_i)|^2 / 4\pi\tau\tau_i P(t_i)$ for pump conversion into CF ω_{ab}, where t_i is the time moment when the $\partial\overline{N}/\partial t$ is at maximum, $\sigma_f \approx \left(\sqrt{2\pi}\tau_i/N_m\right)\left[\partial\overline{N}(t_i)/\partial t\right]$ is the final ionization degree (for τ_i defines as in [9]), τ is the pump duration. The resulting efficiency estimate is rather convenient for comparison of different pump frequencies, polarizations, and intensities.

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