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Analytical calculation of free-electron current density at low-order harmonics of ionizing elliptically polarized laser pulse in the presence of a static electric field

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The paper presents the derivation of an analytical expression for the current density of free electrons excited during tunneling gas ionization by an elliptically polarized pulse in the presence of a static electric field. The analytical calculation of the spectral components of the current density at the low-order odd and even harmonics of the laser pulse is in good agreement with the results of the numerical simulation.

Keywords: laser pulse, ionization, plasma, low-order harmonics generation, detection of terahertz and mid-infrared radiation.

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Recently, low-order harmonics generation (LOHG) (with photon energy equal and lower than the ionization potential) of optical radiation during the interaction of laser pulses with various media has been of great interest [1]. The interest in LOHG is mainly due to the capability of short pulses generation in the UV band [1–6]. Also, the interest in LOHG is associated with the problems involving the combined action of optical and lower-frequency radiation on matter. They include detection of terahertz (THz) or mid-IR radiation using even harmonics generation of laser pulse [7,8]. Since the amplitudes of the even harmonics are linear in the lower-frequency field strength at the laser pulse arrival moment, the dependences of even harmonics intensities on the gating pulse delay time reproduce the time profile of the lower-frequency field squared, and the addition of an external bias field helps to retrieve the field direction [7–9]. Currently, this method using gating-pulse second harmonic generation due to the cubic nonlinearity is used to measure THz pulses, in particular, in the time-domain THz spectroscopy [9–14].

As was shown in our recent study [8], the use of even Brunel harmonics (the low-order harmonics caused by free electron current excitation during tunneling ionization of atoms and molecules) ensures much higher time resolution of detection compared with the use of the cubic (Kerr) nonlinearity. This is ensured by the much shorter duration of Brunel harmonics pulses compared with the laser pulse duration [5,6,8]. It was shown that a strong noise signal, which arises when linearly polarized pulses are used, is associated with the population of atom or molecule excited states and can exceed the even Brunel harmonics intensities. This noise signal is removed when using elliptically polarized laser pulses [8].

In this paper, we present the analytical approach to investigating even and odd Brunel harmonics generated during gas ionization by elliptically polarized pulses in the presence of a static field. Results provided by this approach are compared with the numerical calculation results.

We assume that the electric field acting on atoms or molecules consists of the static field $\mathbf{E}_\mathrm{S}$ and elliptically polarized laser pulse with frequency ω_0 in optical or IR range:

$$\mathbf{E}(t) = \mathbf{E}_\mathrm{S} + \mathbf{E}_\mathrm{L}(t), \quad \mathbf{E}_\mathrm{S} = \hat{\mathbf{x}} E_\mathrm{S}, \tag{1}$$

$$\mathbf{E}_\mathrm{L}(t) = A(t)\mathrm{Re}\left[e^{i\omega_0 t}\hat{\mathbf{e}}\right], \quad \hat{\mathbf{e}} = \hat{\mathbf{x}} + i\varepsilon\hat{\mathbf{y}}, \tag{2}$$

where ε is the ellipticity, $A(t) = E_0 f(t) \geq 0$ is the slowly varying bell-shaped envelope with peak E_0. The approximating of the electric field of the detected lower-frequency pulse by the static field $\mathbf{E}_\mathrm{S}$ is possible at a sufficiently large period of the detected field, much larger than the duration of the generated harmonics pulses. At the same time, the detected pulse duration (equal to or lower than picosecond) is much lower than the time scale, at which avalanche growth of the ion concentration is possible due to collisional ionization at gas pressures equal to one atmosphere and lower [15]. This allows considering high field strengths $\mathbf{E}_\mathrm{S}$ up to several MV/cm and higher (but at the same time insufficient for tunneling ionization of atoms or molecules by field $\mathbf{E}_\mathrm{S}$). The intensity of the laser field $\mathbf{E}_\mathrm{L}(t)$ is assumed as equal to or higher than the threshold value for gas ionization, which is about $10^{14} - 10^{15}$ W/cm^2 depending on the gas type and pulse duration and can be obtained both during sharp and moderate focusing of the laser pulse.

Free-electron current density $\mathbf{j}(t)$ is obtained from the solution of the classical equations of the hydrodynamics of cold collisionless plasma with a variable number of particles:

$$\frac{\partial \mathbf{j}}{\partial t} = \frac{e^2 N}{m}\mathbf{E}, \quad \frac{\partial N}{\partial t} = (N_g - N)w(E), \tag{3}$$

with zero initial conditions at $t \to -\infty$ [16]. Here, N is the free electron density, N_g is the initial gas density, e and m are electron charge and mass, respectively, $w(E)$ is the probability of tunneling ionization per unit time in the electric field $E = |\mathbf{E}|$.

For calculation of the current density harmonics, let us expand the ionization probability per unit time in a Taylor series in low field strength E_S assuming that ellipticity ε is rather low:

$$w(E) \approx w(|E_{Lx}|) + E_S w'(|E_{Lx}|)\mathrm{sign}[E_{Lx}], \tag{4}$$

where $E_{Lx} = A\cos\omega_0 t$ is the laser field strength projection on the x axis, the prime means a derivative with respect to the argument, and it is assumed that $E_S \ll 2A/n_1(A)$, where $n_1(A) = w''(A)A/w'(A)$. Due to the laser field periodicity, $w(E)$ is presented as an infinite sum of quasi-harmonic components on frequencies that are multiples to laser pulse frequency, $w(E) \approx \mathrm{Re}\sum_{k=0}^{\infty} w_k(t)e^{ik\omega_0 t}$. Slow harmonic amplitudes for $k \geq 1$ are determined by $w(E)$ as

$$w_k(t) = \frac{2}{T}\int_{t-T/2}^{t+T/2} w[E(t')]e^{-ik\omega_0 t'}dt', \tag{5}$$

where $T = 2\pi/\omega_0$ is the field period. For analytical calculation of $w_k(t)$, assume that w is a sharp function of its argument, and near the times of maximum/minimum field $t = kT/2$, where k is integer, is approximated as:

$$w(|E_{Lx}|) \approx w(A)e^{n_0(A)[(-1)^k\cos(\omega_0 t)-1]}, \tag{6}$$

$$w'(|E_{Lx}|) \approx w'(A)e^{n_1(A)[(-1)^k\cos(\omega_0 t)-1]}, \tag{7}$$

where $n_0(A) = w'(A)A/w(A)$. Using these expressions, we obtain

$$w_k \approx 4E_S w'(A)e^{-n_1}I_k(n_1), \quad \text{odd } k, \tag{8}$$

$$w_k \approx 4w(A)e^{-n_0}I_k(n_0), \quad \text{even } k. \tag{9}$$

Here, $I_k(\xi)$ is the modified Bessel function of the first kind, and $n_{0,1}$ are taken at argument A. Due to the sharp dependence of ionization probability on field strength, $n_{0,1} \gg 1$ and, thus, we can use asymptotics of the modified Bessel function $I_k(\xi) \approx e^{\xi - k^2/2\xi}/\sqrt{2\pi\xi}$ at $\xi \gg 1$. Thus, in addition to even harmonics arising at $E_S = 0$, ionization probability in the presence of the static field also contains odd harmonics which are linear in E_S. Concentration of free electrons $N(t)$ is also presented as a quasi-harmonic series $N(t) \approx \mathrm{Re}\sum_{k=0}^{\infty} N_k(t)e^{ik\omega_0 t}$, where complex amplitudes of harmonics with numbers $k \geq 1$ are equal to

$$N_{k\geq 1} \approx -i(N_g - N_0)\frac{w_k}{k\omega_0}. \tag{10}$$

Free-electron concentration $N_0(t)$ averaged over the field period satisfies the equation

$$\frac{\partial N_0}{\partial t} = (N_g - N_0)w_0(t). \tag{11}$$

In (10), (11), it is assumed that field period T is much smaller than the ionization duration τ_i (characteristic growth time of N_0) defined as $\tau_i = [-N_0'(t_0)/N_0'''(t_0)]^{1/2}$, where t_0 is the time at which $\partial N_0/\partial t$ takes the maximum value. In case when the final degree of ionization is low, $|t_0| \ll \tau$ and $\tau_i \approx \tau/\sqrt{n_0(E_0)}$, where $\tau = [-f(0)/f''(0)]^{1/2}$ is the pulse duration [5,17–19]. Thus, the ionization duration defining the Brunel harmonics duration is much smaller than the laser pulse duration. At high laser pulse intensities, when the ionization degree is high, $|t_0|$ increases, while the ionization duration (in the case of the Gaussian envelope of the laser pulse) decreases [19–21].

Electric field mixing with quasi-harmonic components of concentration results in the appearance of quasi-harmonic components in the derivative of the current density:

$$\frac{\partial \mathbf{j}}{\partial t} \approx \mathrm{Re}\sum_{k=0}^{\infty}\mathbf{F}_k e^{ik\omega_0 t}, \tag{12}$$

$$\mathbf{F}_{k\geq 2}(t) = (e^2/m)[(N_{k-1}\hat{\mathbf{e}} + N_{k+1}\hat{\mathbf{e}}^*)A/2 + N_k\mathbf{E}_S]. \tag{13}$$

For even k, the ratio of the second term ($N_k\mathbf{E}_S$) in equation (13) to the first term is of the order of $1/n_0(A) \ll 1$, i.e. the second term is negligible. For odd k, the second term in equation (13) is quadratic in E_S and also can be neglected. As a result, we obtain

$$\mathbf{F}_{k\geq 2}(t) = (e^2 A/m)(C_+\hat{\mathbf{x}} + i\varepsilon C_-\hat{\mathbf{y}}), \tag{14}$$

$$C_\pm = N_{k-1} \pm N_{k+1} = -i\frac{\alpha_k D_\pm}{\omega_0}\frac{\partial N_0}{\partial t}, \tag{15}$$

$$D_\pm = \frac{e^{-(k-1)^2/2n_0}}{k-1} \pm \frac{e^{-(k+1)^2/2n_0}}{k+1}, \tag{16}$$

$$\alpha_k = \begin{cases} 1, & \text{odd } k, \\ n_0(A)E_S/A, & \text{even } k. \end{cases} \tag{17}$$

For the last expression, $n_1 \approx n_0$ [5] was assumed. For $k \ll 2n_0$,

$$D_+ \approx \frac{2ke^{-k^2/2n_0}}{k^2-1}, \quad D_- \approx \frac{2(k^2/n_0+1)e^{-k^2/2n_0}}{k^2-1}.$$

The derived analytical expressions show that the generated low-order harmonics have the phase shift by $\pi/2$ with respect to the laser field and elliptical polarization with the ellipticity $\varepsilon_{k\geq 2} \approx \varepsilon D_-/D_+$. It should be noted that the last expression is true at any (including those comparable with unity) degrees of gas ionization. Thus, Brunel harmonics ellipticity (a) is lower than laser pulse ellipticity, and (b) monotonically decreases with the increase in harmonics number up to $k \approx \sqrt{n_0(E_0)}$ and then grows tending asymptotically to $\varepsilon k/n_0(E_0)$. Amplitudes of even/odd

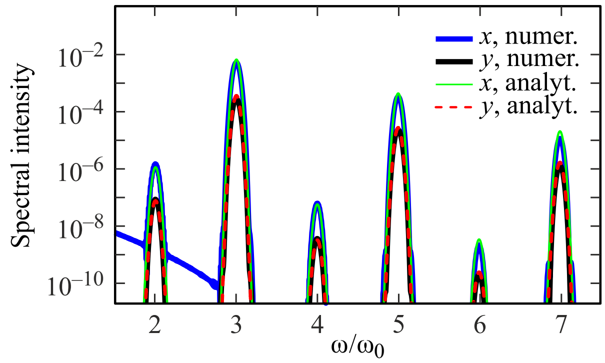

Figure 1. The square of the spectrum of x- and y-components $\partial \mathbf{j}/\partial t$ normalized to N_g^2 (in atomic units) excited during helium ionization by a 50 fs, 800 nm laser pulse with peak intensity 10^{15} W/cm^2 and ellipticity $\varepsilon = 0.4$ in the presence of static electric field $E_S = 500$ kV/cm. Thick solid lines denote the numerical solution of equations (3), thin solid and dashed lines denote analytical formula (19) for x- and y-components, respectively.

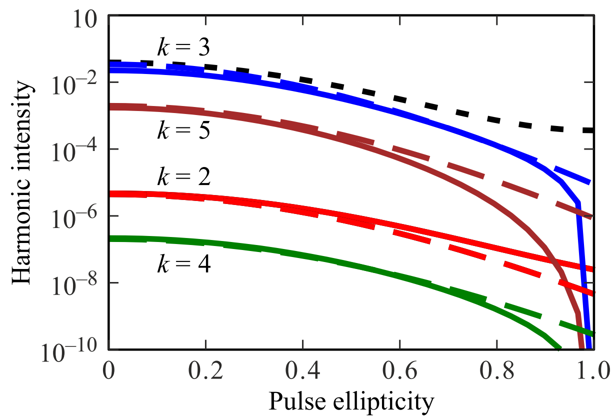

Figure 2. Spectral intensity of $k = 2$ — 5 harmonics of $\partial \mathbf{j}/\partial t$ normalized to N_g^2 (in atomic units) as function of laser-pulse ellipticity ε. Solid lines denote numerical calculation, dashed lines denote analytical formula (19). The dotted line shows the numerically calculated square of the final degree of gas ionization.

harmonics of $\partial \mathbf{j}/\partial t$ at low k decrease inversely proportional to the harmonic number. At fixed peak intensity of laser pulse $I = (c/8\pi)(1+\varepsilon^2)E_0^2$ (where c is the speed of light in vacuum), harmonic amplitudes are maximum at zero ellipticity and decrease with increase in ε in proportion to the factor $w(E_0) \approx w(E_{\max})(1 - n_0(E_{\max})\varepsilon^2/2)$, where $E_{\max} = (8\pi I/c)^{1/2}$. This factor defines the maximum concentration growth rate $\partial N_0/\partial t$ in equation (14) for complex amplitudes of Brunel harmonics. Thus, the characteristic scale of harmonics amplitudes decrease with an increase in ellipticity is $\varepsilon \sim 1/\sqrt{n_0(E_{\max})}$.

In order to check that the obtained analytical formulas have high accuracy, we compare them with the numerical solution of equations (3) for the helium atom. We specify the probability of helium atom ionization per unit time as

$$w(E) = \alpha\omega_a \left(E_a/E\right)^{\delta} \exp\left(-\beta_1 E_a/E - \beta_2 E/E_a\right), \quad (18)$$

where $\omega_a = 4.13 \cdot 10^{16}$ s^{-1} and $E_a = 5.14 \cdot 10^9$ V/cm - atomic units of frequency and field, respectively, $\alpha = 9.2$, $\beta_1 = 1.6$, $\beta_2 = 3.2$, and $\delta = 0.49$ [8,22]. In all calculations, $E_S = 500$ kV/cm, wavelength $\lambda = 2\pi c/\omega_0 = 800$ nm, envelope $f(t) = e^{-t^2/2\tau^2}$ with intensity full-width at half-maximum duration $\tau_p = (2\sqrt{\ln 2})\tau = 50$ fs and with peak intensity $I = 10^{15}$ W/cm^2.

Figure 1 shows the numerically calculated squared Fourier spectrum of x- and y components of the current density derivative, $S_{x,y}(\omega) = |\int(\partial j_{x,y}/\partial t)e^{-i\omega t}dt|^2$ for the laser pulse ellipticity $\varepsilon = 0.4$. The obtained result is compared with the analytical result for low ionization degree:

$$S_{x,y}(\omega > 0) = \frac{j_{\text{osc}}^2 w^2(E_0)\tau^2}{n_0^2} \sum_{k=2}^{\infty} \alpha_k^2 G_{x,y} e^{-\frac{(\omega - k\omega_0)^2\tau^2}{n_0}}, \quad (19)$$

where $j_{\text{osc}} = e^2 N_g E_0/m\omega_0$, $G_x = D_+^2$, $G_y = D_-^2$; $D_\pm$ are set using (16). As shown in Figure 1, at these parameters, the analytical formula agrees with high accuracy with the results of numerical calculation.

Figure 2 shows dependences of harmonics intensities $S_x(\omega) + S_y(\omega)$ on the laser-pulse ellipticity ε. Harmonic intensities are maximum at zero ellipticity and decrease with ellipticity increase approximately in the same manner as final ionization degree $N(\infty)/N_g$ squared (shown by the dotted line in the figure). The numerically calculated harmonic intensities tend to zero sharply when ellipticity approaches unity, excluding the second harmonic intensity, which is approximately by two orders of magnitude lower at $\varepsilon = 1$ than at $\varepsilon = 0$ (approximately the same ratio is also observed for the final ionization degree squared). Harmonic intensities calculated numerically using formula (19) agree with the numerical calculation for a wide ellipticity range. Significant deviations are observed only when ε approaches unity. For example, in the case of the third harmonic generation, the analytical and numerical calculations almost totally coincide at ellipticity $\varepsilon < 0.9$. The same good agreement is achieved for the second and fifth harmonics at $\varepsilon < 0.6$ and for the fourth harmonic at $\varepsilon < 0.8$. Thus, the developed analytical model is highly accurate and may be used both for estimating the free-electron current contribution to LOHG mechanisms and determining the conditions for effective LOHG, as well as for developing and optimizing methods for THz and mid-IR radiation detection.

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Conflict of interest

The authors declare that they have no conflict of interest.

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