NONLINEAR OPTICAL PHENOMENA

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Generation of attosecond pulses of recombination radiation in two-colour laser fields

V.D. Taranukhin, Zheng Jiangang

***Abstract*. The possibility of generation of attosecond pulses of recombination radiation upon the above-threshold tunnel ionisation of atoms (ions) in two-colour laser fields is studied. The method of classical photoelectron trajectories in the continuum is used to determine the laser field parameters minimising the duration of recombination radiation. It is shown that pulses of duration of several tens of attoseconds can be generated in such fields. The principal possibility of generation of coherent electromagnetic pulses of duration $\tau_g \lesssim 1$ as is also shown.**

Keywords: *recombination radiation, high-order harmonic generation, attosecond pulses.*

1. Introduction

Modern lasers can generate coherent few-cycle optical radiation pulses ($\tau \sim 1 - 10$ fs). This enables monitoring and control of molecular processes and chemical reactions (femtochemistry). However, to 'trace' faster electronic processes, pulses of attosecond duration (1 as $= 10^{-18}$ s) are required. At present, the possibility of generation of attosecond pulses and their applications is extensively investigated (in the literature, the area of these investigations is referred to as 'attophysics') [1, 2]. The main direction of these studies is the use of high-order harmonic generation (HHG) in strong laser fields [1 – 3], because recombination radiation in this case has a high frequency, making it basically possible to obtain subfemtosecond pulses. The generation of short pulses upon HHG is possible when the phase-matching condition is fulfilled for a group of harmonics [4] or with the use of varied-polarisation pump radiation [5]. In both cases, the duration of recombination radiation $\tau_g \lesssim T = \lambda/c$ (where T and λ are the optical period and wavelength of the pump radiation and c is the velocity of light), which corresponds to a duration of the order of a femtosecond. To date, pulses of duration of several hundred attoseconds have been generated [2].

V.D. Taranukhin, Zheng Jiangang Department of Physics, M.V. Lomonosov Moscow State University, Vorob'evy gory, 119992 Moscow, Russia; e-mail: tvd@ssf.phys.msu.su

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In this paper, we study the possibility of obtaining substantially shorter pulses upon HHG by using a two-colour pump, which was earlier proposed [6] for the selection of the HHG spectrum and phase control of the instants of ionisation and recombination [7]. The phase changes at which recombination of photoelectron and a parent ion occurs only at specific parameters of the two-colour pump allow controlling the duration of recombination radiation.

The recombination radiation appearing upon HHG (upon tunnel atomic ionisation and subsequent parent ion – photoelectron recombination) is described by the wave equation with a source which is the second time derivative of the field-induced dipole moment $\boldsymbol{D}(t)$. The dipole moment $\boldsymbol{D}$ is calculated from the Schrödinger equation, which gives, in particular, the dependence of $\boldsymbol{D}$ on the time t, which determines the radiation duration. It was shown in Refs [7 – 9] that, under conditions characteristic of the tunnel ionisation regime, HHG can be described quasiclassically and the photoelectron evolution in the continuum can be described by classical mechanical trajectories. In this case, it is the duration of the interaction between the returning photoelectron and the parent ion (which is determined by the difference in the instant of return of different parts of the electron wave packet to the parent ion) that determines the duration of recombination radiation of the atom under study. In an extended medium, the propagation of laser radiation and its spatial inhomogeneity may also affect the duration of recombination radiation. These effects require special calculations [10, 11] and are not considered in this paper (which is devoted to the study of the possibility of generating ultimately short radiation pulses).

2. Electron wave-packet trajectories in a two-colour field

In the case of a single-frequency circularly polarised pump, no harmonic generation occurs [5], because the photoelectron produced after the ionisation of an atom goes away from the parent ion and never comes back to it: collisional electron trajectories are absent at any instants of ionisation. With a linearly polarised pump radiation, on the contrary, the wave packet moves in the direction of field polarisation and at any instant of ionisation (within an optical period) there is a collisional trajectory, resulting in a relatively long duration of recombination radiation ($\tau_g \sim T$). In the case of a special two-colour pump (which includes circularly polarised radiation as well), HHG is possible [7], with an opportunity to control the generation parameters, in particular, the duration of recombination radiation.

Consider the field of two-colour pump radiation, which is a combination of an elliptically polarised high-frequency (HF) field and a linearly polarised low-frequency (LF) field:

$$\boldsymbol{F} = F_0\left[\boldsymbol{e}_x(1-\alpha^2)^{1/2}\cos\omega t + \boldsymbol{e}_y\alpha\sin\omega t\right] + \boldsymbol{F}_{\rm dc}, \quad (1)$$

$$\boldsymbol{F}_{\rm dc} = -\beta F_0\left[\boldsymbol{e}_x(1-\gamma^2)^{1/2} + \boldsymbol{e}_y\gamma\right], \quad (2)$$

where F_0, ω, and α are the amplitude, frequency, and ellipticity of the HF pump component, respectively; $\boldsymbol{e}_x$ and $\boldsymbol{e}_y$ are the unit vectors; β and γ are parameters determining the relative amplitude and direction of the linearly polarised LF pump field component $\boldsymbol{F}_{\rm dc}$. As the LF component, we can use radiation of a CO_2 laser synchronised with the HF field. In this case, the CO_2-laser field is almost constant during the optical period of HF radiation with a wavelength $\lambda \sim 1$ μm. For simplicity we restrict ourselves to the case of collinear propagation of the LF and HF fields (along the z direction).

As noted above, the photoelectron evolution in the continuum (for a tunnel ionisation regime) can be described by classical trajectories of different parts of the electron wave packet. The solution of the classical equation of motion $m\ddot{\boldsymbol{r}} = e\boldsymbol{F}$ (where m and e are the electron mass and charge; and $\boldsymbol{r}$ is the electron radius vector) in the field (1) with zero initial electron coordinates [12] (it is assumed that the atom is located at the origin of the coordinates and the tunnel barrier width is small compared to the wavelength λ) gives the following trajectories:

$$x(t) = V_{0x}(t-t_0) - (1-\alpha^2)^{1/2}\left[\cos t - \cos t_0 + (t-t_0)\sin t_0 + \beta_1(t-t_0)^2\right], \quad (3)$$

$$y(t) = V_{0y}(t-t_0) - \alpha\left[\sin t - \sin t_0 - (t-t_0)\cos t_0 + \beta_2(t-t_0)^2\right], \quad (4)$$

where t_0 is the ionisation phase (the initial instant of the photoelectron evolution in the continuum) and $\beta_1 = (\beta/2)[(1-\gamma^2)/(1-\alpha^2)]^{1/2}$; $\beta_2 = (\beta/2)(\gamma/\alpha)$. Hereafter, we use dimensionless variables, with the time t normalised to $1/\omega$, the coordinates x and y to $eF_0/(m\omega^2)$, the velocities to $eF_0/(m\omega)$, and the energy normalised to the ponderomotive energy $U_{\rm p} = e^2F_0^2/(4m\omega^2)$ of the HF pump component. The initial velocities $V_{0x,0y}$ are different for different parts of the electron wave packet. For the central part of the packet, which begins to move in the continuum along the direction of the field $\boldsymbol{F}(t_0)$, we have $V_{0x} = V_{0y} = 0$ [12]. For the remaining parts of the packet, $V_{0x,0y} \neq 0$ due to spreading. Let us denote the angle between the initial direction of the total field $\boldsymbol{F}(t_0)$ and the x axis by φ (Fig. 1). Then, for different parts of the electron packet in the xy plane we obtain

$$\boldsymbol{V}_0 = \pm V_\Delta(\boldsymbol{e}_x\sin\varphi - \boldsymbol{e}_y\cos\varphi), \quad (5)$$

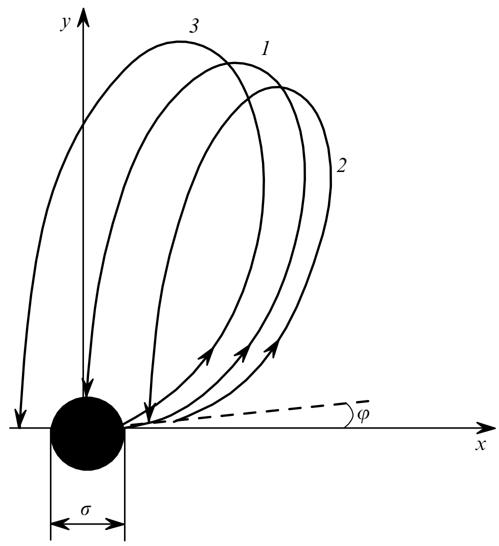

Figure 1. Trajectories of the central part of the electron wave packet in the continuum in the case of two-colour pump with the parameters $\alpha = 1/\sqrt{2}$, $\beta \approx -0.2$, $\gamma \approx -1$ for the instants of ionisation $t_0 = t_0^* = 0$ (*1*), $t_0 < t_0^*$ (*2*), and $t_0 > t_0^*$ (*3*), where t_0^* is the central value of the interval of the instants of ionisation δt_0. The arrows indicate the direction of photoelectron motion at the instant the photoelectron leaves the atom and at the instant it returns to the region of the parent ion; σ is the characteristic size of the parent ion.

where the parameter V_Δ varies from zero for the central part of the packet to the characteristic spreading velocity $V_{\rm sp}$ for the peripheral parts of the packet [13]. The velocity $V_{\rm sp}$ is determined by the width of the transverse tunnel-ionisation probability distribution [13] and depends on the instantaneous magnitude of the pump field $\boldsymbol{F}(t_0)$:

$$V_{\rm sp} \sim F^{1/2}(t_0)U_{\rm i}^{-1/4}, \quad (6)$$

where $U_{\rm i}$ is the atomic ionisation potential. For high intensities of the HF pump radiation component I, highly charged ions should be employed to preserve the regime of tunnel ionisation. For this reason, as the intensity I increases simultaneously with an increase in the ionisation potential (in going over to highly charged ions), the packet spreading velocity (6) changes only slightly. In the subsequent discussion, for the velocity $V_{\rm sp}$ we will use the value ~ 0.6 nm fs^{-1}, which is in good agreement with experimental data (for a pump radiation intensity $I \approx 10^{14} - 10^{15}$ W cm^{-2}) [5]. Note that the wave packet spread takes place along the z axis as well. However, this spread merely lowers the intensity of recombination radiation but does not affect its duration.

Therefore, the electron wave-packet evolution in the continuum is described by Eqns (3) and (4) with the initial velocities

$$V_{0x} = \mp\frac{V_\Delta\varkappa}{(1+\varkappa^2)^{1/2}}, \qquad V_{0y} = \pm\frac{V_\Delta}{(1+\varkappa^2)^{1/2}}, \quad (7)$$

where $\varkappa$, as follows from expression (5), is determined by the angle of electron escape from the atom φ (see Fig. 1): $\varkappa = \tan\varphi$. On the other hand, $\tan\varphi = F_y(t_0)/F_x(t_0)$, and from Eqns (1) and (2) there follows the dependence of $\varkappa$ on the two-colour pump parameters and the instant of ionisation t_0:

$$\varkappa = \frac{\alpha}{(1-\alpha^2)^{1/2}} \frac{\sin t_0 - 2\beta_2}{\cos t_0 - 2\beta_1}. \quad (8)$$

Collisional trajectories correspond to the case when both coordinates x and y of the central part of the packet tend to zero simultaneously at a certain instant $t_{\rm f}$ (Fig. 2). In this case, the 'collision' time is determined by the scatter in the instants of recombination of different parts of the packet $\delta t_{\rm f}$: the 'longitudinal' scatter $\delta t_{\rm f1}$, which is determined by the interval of ionisation instants δt_0 for which collisional trajectories exist (Fig. 1), and the 'transverse' scatter $\delta t_{\rm f2}$, which is determined by the dimension of the electron wave packet in the direction of its approach to the parent ion. For a conventional linearly polarised pump, both of these factors coincide and $\delta t_{\rm f} \sim T$ irrespective of the pump radiation intensity. For a two-colour pump (1), the trajectory of the electron wave packet can substantially shift perpendicular to the initial direction of the total field $\boldsymbol{F}(t_0)$. In this case, a significant reduction in the 'longitudinal' scatter $\delta t_{\rm f1}$ is possible, while the 'transverse' scatter $\delta t_{\rm f2}$ is determined by the spreading of the wave packet.

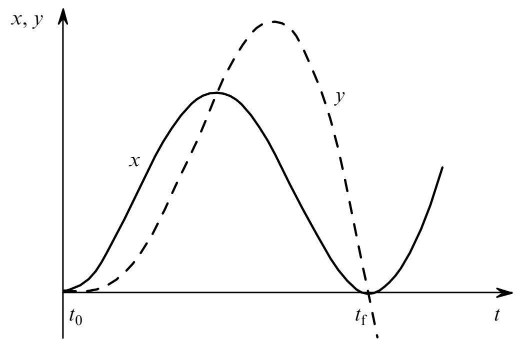

Figure 2. Dependences of the coordinates of the central part of electron wave packet on the time of its evolution in the continuum for a two-colour pump with the parameters $\alpha = 1/\sqrt{2}$, $\beta \approx -0.28$, $\gamma \approx -0.96$, and the instant of ionisation $t_0^* = 0.1\pi$.

It follows from Eqns (3) and (4) that, unlike single-frequency pump, collisional electron trajectories exist only for specific sets of the parameters α, β, γ, and the instant of ionisation t_0. Figure 1 shows the trajectories of the central part of the electron wave packet for different t_0. One can see that, for fixed pump parameters, a collision with the parent ion can occur only for a narrow interval of the instants of ionisation δt_0 near a certain value $t_0 = t_0^*$. When selecting the pump radiation parameters, it is desirable that the electron energy $\varepsilon_{\rm f}$ at the instant of recombination should be as high as possible (for higher-order harmonic generation), the time the electron spends in the continuum be as short as possible (for reducing the electron packet spread and increasing the recombination efficiency), the LF-to-HF field amplitude ratio be as low as possible (because lasers emitting at 1 and 10 μm have different powers), and that only one collisional trajectory should exist during the pump pulse (for generating a single pulse of recombination radiation).

An analysis of Eqns (3) and (4) shows, however, that there is no unambiguous solution of this problem: there exists many (in the general case, an infinite number) different sets of the parameters α, β, γ, and t_0^* at which collisional trajectories exist. Nevertheless, most important for the generation of the shortest pulses of recombination radiation is to minimise the time which the returning electron wave packet spends near the parent ion (of particular importance is minimisation of the parameter $\delta t_{\rm f}$). This is achieved by increasing the electron kinetic energy $\varepsilon_{\rm f}$ at the instant of recombination and decreasing the dimension of the recombining part of the packet. The energy $\varepsilon_{\rm f}$ is mainly determined by the intensity of pump radiation I. For a fixed intensity I, $\varepsilon_{\rm f}$ is maximised by selecting the parameters α, β, γ, and t_0^* that provide maximum velocity ${\rm d}r(t_{\rm f})/{\rm d}t$ of the the returning electron, where $r = (x^2+y^2)^{1/2}$ is the distance to the parent ion. The dimension of the recombining part of the packet is minimised by selecting the spatial orientation of the electron wave packet at the instant $t_{\rm f}$ (with respect to the direction of its approach to the parent ion) as well as by minimising the 'transverse' dimension of the packet determined by the rate of its spreading and the time of electron evolution in the continuum.

Furthermore, as noted above, it is required to minimise the interval δt_0 of the instants of ionisation for which collisional trajectories exist, because the dependence $t_{\rm f}(t_0)$ can make a substantial contribution to the scatter of the instants of recombination $\delta t_{\rm f}$ (like in the case of linearly polarised pump). A significant advantage of the two-colour pump (1), (2) is the possibility to substantially shorten the interval δt_0. This is explained by the fact that by selecting the pump parameters in a proper way, the electron wave packet at the instant of return to the nucleus can be spatially oriented so that its propagation direction will be perpendicular to the longitudinal axis of the packet. In this case, recombination (intersection with the parent ion domain) will take place only for a small 'longitudinal' part of the packet (corresponding to the short interval of the instants of ionisation $\delta t_0 \ll 2\pi$), which is realised when the derivative ${\rm d}r(t_{\rm f})/{\rm d}t_0$ is sufficiently large.

3. Optimisation of the two-colour pump parameters

Let us find the conditions that minimise the dimension of the recombining part of the electron wave packet and accordingly minimise the scatter $\delta t_{\rm f}$ of the instants of recombination. The instant of recombination $t_{\rm f}$ can be determined from Eqn (3) for the central part of the packet (for which $V_{0x} = 0$):

$$x_{\rm c}(t_{\rm f}) = 0. \quad (9)$$

Hereafter the subscript 'c' corresponds to the central part of the packet. In this case, ${\rm d}x_{\rm c}(t_{\rm f})/{\rm d}t = V_{{\rm c}x}(t_{\rm f}) \equiv 0$ (see Fig. 2), which fixes the directions of the x and y axes $[\boldsymbol{e}_y \| \boldsymbol{V}_{\rm c}(t_{\rm f})]$. The scatter of recombination times $\delta t_{\rm f1}$ of the 'longitudinal' part of the packet is minimised when the axis of the returning packet is perpendicular to the direction of approach to the parent ion (i.e., along the x axis) and when the derivative ${\rm d}r_{\rm c}(t_{\rm f})/{\rm d}t_0$ is maximised. The first condition implies that the vector $\boldsymbol{F}(t_0)$, which determines the initial packet orientation, should also be directed along the x axis, i.e., the following condition should be fulfilled:

$$\sin t_0 - 2\beta_2 = 0. \quad (10)$$

Note that the parameter $\varkappa$ (8) also vanishes in this case. The second condition minimises the interval of the instants of ionisation δt_0 (and accordingly the scatter of the instants of recombination of different 'longitudinal' parts of the packet): $\delta t_0 = \sigma/[\mathrm{d}r_c(t_f)/\mathrm{d}t_0]$, where σ is the effective dimension of the parent ion (Fig. 1). We take into account that the relation $\mathrm{d}x_c(t_f)/\mathrm{d}t_0 \equiv 0$ is also valid in case (9), and the maximum of the derivative $\mathrm{d}r_c(t_f)/\mathrm{d}t_0$ providing the fastest passage from collisional trajectories to the non-collisional ones upon changing the ionisation phase, is reached when the condition

$$\frac{\mathrm{d}y_c(t_f)}{\mathrm{d}t_0} \equiv \frac{\partial y_c(t_f)}{\partial t_0} + \frac{\partial y_c(t_f)}{\partial t_f}\frac{\partial t_f}{\partial t_0} = \infty \quad (11)$$

is fulfilled, where the function $t_f(t_0)$, which is required to calculate the derivative $\partial t_f/\partial t_0$, is implicitly defined by Eqn (9). One can easily verify that the condition $\partial t_f/\partial t_0 = \infty$, and simultaneously Eqn (11), are realised when the pump parameters α, β, γ and the instant of ionisation t_0 satisfy the equation:

$$\sin t_f - \sin t_0 = 2\beta_1(t_f - t_0). \quad (12)$$

For a small temporal scatter δt_{f1} of the 'longitudinal' parts of the electron wave packet, the contribution to the duration of recombination radiation arising from the scatter δt_{f2} of the instants of recombination of different 'transverse' parts of the packet becomes significant. This contribution is determined by the packet velocity $V_{cy}(t_f) = \mathrm{d}y_c(t_f)/\mathrm{d}t$ at the instant of recombination and the transverse packet dimension formed due to spreading. This dimension depends on the spreading velocity V_{sp} and the time $t_f - t_0$ of electron evolution in the continuum. Normally, this time is $t_f - t_0 \sim 2\pi$ and cannot be substantially shortened (otherwise the electron does not manage to acquire a sufficiently high kinetic energy ε_f by the instant of recombination). As for the velocity $V_{cy}(t_f)$, it can be increased by selecting the pump parameters. For instance, the condition for the $V_{cy}(t_f)$ velocity maximum depending on the instant of ionisation t_0 leads to a certain relation between the pump parameters, which, however, coincides with Eqn (10). Finally, we note that expressions (10) and (12) do not permit determining the unique optimal set of the parameters α, β, γ, and t_0^*. Nevertheless, these expressions significantly facilitate a computer-aided search for this set.

Therefore, the numerical solution of the equations $x_c(t_f) = 0$ and $y_c(t_f) = 0$ simultaneously with Eqns (10) and (12) determines the appropriate set of pump radiation parameters α, β, γ and the instant of ionisation t_0^*. Then, computer simulation of the electron trajectories for this set and for different parts of the electron packet (for different initial velocities $V_{0x,0y}$) allows us to determine the scatter in the instants of collision δt_f of the recombining parts of the electron wave packet with the parent ion. The maximum scatter in these instants gives an estimate of the duration τ_g of recombination radiation.

4. Results of numerical experiments

Below, we present the results of numerical experiments on the determination of the duration τ_g of recombination radiation during HHG upon a two-colour pump (1), (2) in accordance with the procedure described above. We assume that a HF pump component is the laser radiation at $\lambda = 01 - 0.8$ μm of intensity $I = 10^{16} - 10^{17}$ W cm^{-2}. Figures 3 and 4 show the evolution of the 'longitudinal' part of the electron wave packet upon a two-colour pump (1), (2) when conditions (10) and (12) are fulfilled and upon an ordinary linearly polarised pump ($\alpha = \beta = 0$). One can see that in both cases the packet lengthens considerably along the initial direction of the pump field (the maximum displacement of the peripheral part of the packet is of the order of the wavelength λ). For an ordinary pump, this results in a relatively long time δt_f of interaction between the returning packet and the parent ion and in a relatively long recombination time ($\tau_g \sim T$). For a two-colour pump with specially selected parameters (Fig. 3), the longitudinal packet elongation does not lead to a long recombination time, because the packet returns to the parent ion along the y axis rather than along the initial direction – the x axis. In this case, only a small fraction of the 'longitudinal' packet ($\sim\sigma$) recombines.

We give the results of a typical numerical experiment in this case (dimensionless quantities will be converted to the dimensional ones for the pump radiation with the intensity $I = 5 \times 10^{17}$ W cm^{-2} at $\lambda = 0.79$ μm). From the system of four equations $x_c(t_f) = 0$, $y_c(t_f) = 0$, (10), and (12), we determine the following set of parameters: $\alpha = 1/\sqrt{2}$ (which corresponds to circularly polarised HF radiation), $\beta \approx -0.29$, $\gamma \approx -0.94$, and the single (during an optical

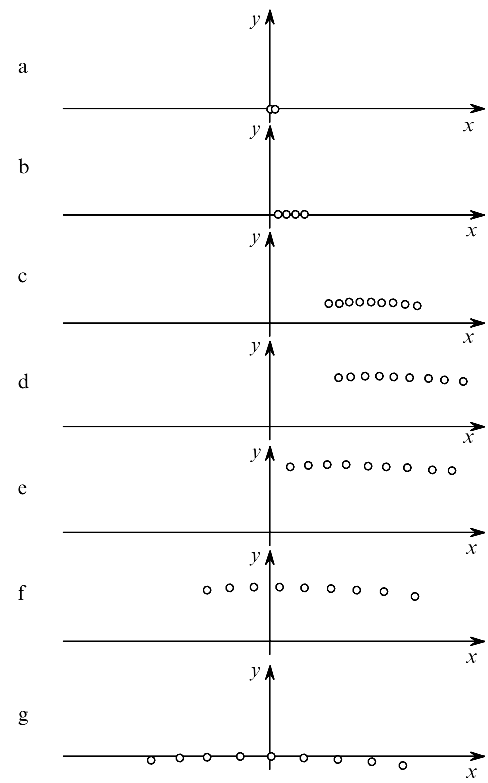

Figure 3. Longitudinal evolution of the electron wave packet in the continuum [shown are parts of the packet (○) that correspond to the instants of ionisation $t_0 = 0.1\pi \pm 0.02\pi n$, where $n = 0, 1, 2, 3, 4$; the transverse packet dimension $d_\perp \gg \sigma$ is not shown] for a two-colour pump with the parameters $\alpha = 1/\sqrt{2}$, $\beta \approx -0.28$, $\gamma \approx -0.96$ at the instants $(t - t_0)/T = 0$ (a), 0.1 (b), 0.3 (c), 0.45 (d), 0.6 (e), 0.75 (f), and 0.9 (g). The atom (with an effective size σ) is located at the origin of coordinates.

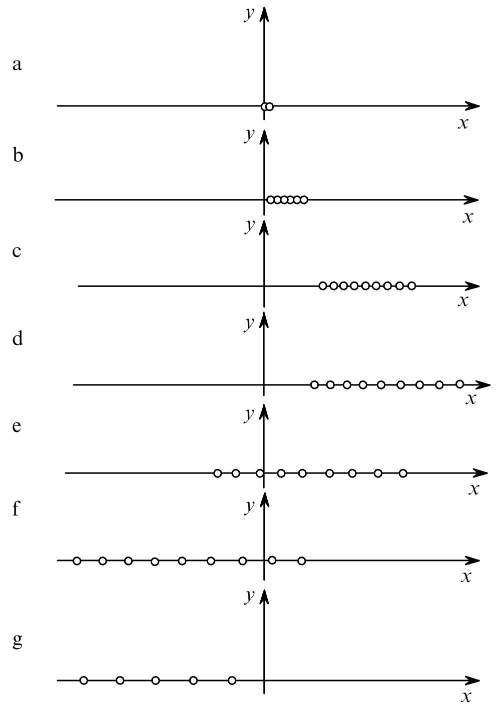

Figure 4. Same as in Fig. 3 for an ordinary linearly polarised pump ($\alpha = \beta = 0$).

cycle of the HF field) instant of ionisation $t_0^* \approx 0.13\pi$ whereby there exist collisional trajectories. The numerical simulation of the trajectories for the central part of the electron packet with these parameters gives an instant of recombination $t_f \approx 1.87\pi$ and a kinetic electron energy at the instant of recombination $\varepsilon_f \approx 4.5$ (or $\varepsilon_f \approx 130$ keV in dimensional units), which corresponds to the generation of harmonics with numbers $\sim\varepsilon_f/(\hbar\omega) \approx 10^5$. In this case, the range δt_0 of the instants of ionisation at which the recombination condition is still fulfilled is extremely short and is ~ 0.1 as, while the scatter of the instants of recombination of the 'longitudinal' parts of the packet δt_{f1} is in fact absent. In this case, the duration of recombination radiation is determined by the maximum scatter $\delta t_{f2} \approx 0.028$ of the instants of recombination of the 'transverse' parts of the packet, which corresponds to a dimensional duration of recombination radiation $\tau_g \approx 12$ as.

As the pump intensity I increases, the duration τ_g decreases (unlike the case of conventional linearly polarised pumping). In this case (within the framework of a rigorous approach), it is necessary to take into account the effect of the magnetic field of the pump radiation on the electron motion in the continuum [14]. This effect, however, does not significantly change the magnitude of τ_g, although the specific values of the required pump parameters may change.

The value of δt_{f2} obtained in the numerical simulation is in good agreement with the estimate:

$$\delta t_{f2} \approx d_\perp / V_{cy}(t_f), \quad d_\perp = \Delta V_y (t_f - t_0), \quad \Delta V_y = 2V_{sp}. \tag{13}$$

Here, $d_\perp$ is the packet transverse dimension formed due to its spreading; ΔV_y is the difference of the velocities of motion of the peripheral (along the y axis) parts of the packet. For the electron velocity $V_{cy}(t_f)$ at the instant of recombination comparable with the speed of light and a time of electron evolution in the continuum of the order of T, the estimate (13) for the HF radiation at $\lambda = 0.79$ μm gives $\delta t_{f2} \approx 20$ as. This value can be significantly decreased only by selecting the pump parameters so that the time of electron evolution in the continuum is appreciably shorter than the optical period T and the energy ε_f remains high enough. Note also that the obtained duration τ_g of the recombination radiation pulse is more than two orders of magnitude shorter than the duration of the optical HF radiation period T. However, this pulse contains many optical cycles of the recombination radiation (which are determined by the parameter $\varepsilon_f = 2\pi\hbar/T_f$, where T_f is the recombination radiation period) and is almost monochromatic in this sense.

Finally, note another feature of recombination radiation discovered in our numerical experiment: the energy ε_f of different recombining parts of the electron packet is different. Thus, under the conditions of the above numerical experiment, this energy increases in fact linearly during the successive recombination of different 'transverse' parts of the packet, the greatest energy variation being $\Delta\varepsilon_f/\varepsilon_f \approx 10^{-2}$. This results in the phase modulation (chirp) of the recombination radiation. The physical effect underlying this modulation is the spread of the electron wave packet. From Eqn (4), we can obtain the estimate:

$$\frac{\Delta\varepsilon_f}{4V_{cy}(t_f)} \approx \Delta V_{cy} - 2V_{sp}, \tag{14}$$

where $\Delta V_{cy} = [\partial V_{cy}(t_f)/\partial t_f]\delta t_{f2}$. It follows from expression (14) that the variation of the energy ε_f is caused by the difference in the initial velocities (which vary from $-V_{sp}$ to V_{sp}) of different 'transverse' parts of the packet [the second term in expression (14)] and by the consequential difference in the instants of their recombination: $\delta t_{f2} = f(V_{sp})$ (the first term). In the numerical experiment considered above, these terms have opposite signs, and therefore the degree of phase modulation of the pulse of recombination radiation is sensitive to the specific spreading velocity V_{sp}.

The presence of a chirp makes possible a compression of the recombination pulse. The compression coefficient k is estimated in the usual way [15]:

$$k \approx \frac{\tau_g \Delta\varepsilon_f}{\hbar}. \tag{15}$$

For the above numerical experiment, $k \approx 25$, making it basically possible to produce recombination radiation pulses of duration $\tau_g/k < 1$ as.

5. Conclusions

The approach proposed in our work, which uses a special two-colour pump for HHG, allows the generation of coherent pulses of electromagnetic radiation of duration $\tau_g \sim 10$ as and opens up the principal possibility to obtain subattosecond pulses.

Of importance is the efficiency of such generation. As in the case of an ordinary pump, this efficiency is determined by three factors: the probability of atomic ionisation, the velocity of spreading of the electron wave packet in the continuum, and the electron–ion recombination probability. The use of a two-colour pump (including circularly polarised radiation) does not substantially change the tunnel ionisation probability, because this probability is determined by the instantaneous magnitude of the field. The wave-packet spreading and the recombination probability are about the same as in the conventional HHG. However, the pump (1), (2) produces the selection of only a small fraction of all photoelectrons recombining with the parent ion, i.e., of a small 'longitudinal' fraction of the electron wave packet. Accordingly, the energy contained in the pulse of recombination radiation decreases.

Note that photoelectron selection takes place in the conventional HHG scheme as well – the selection of a narrow 'transverse' part of the wave packet of size $d_\perp = \sigma$ (Fig. 4). In this case, the energy $\varepsilon_{\rm f1}$ of recombination radiation is proportional to the longitudinal packet dimension $d_\parallel$. In the case of pump (1), (2), on the contrary, the selection of a narrow 'longitudinal' part of the wave packet of size $d_\parallel = \sigma$ occurs (Fig. 3) and the energy of recombination radiation is $\varepsilon_{\rm f2} \sim d_\perp$.

Therefore, the losses in 'longitudinal' packet energy in the case of a two-colour pump are partly compensated for by the acquisition of its 'transverse' energy. As this takes place, the ratio between the energies in the two cases is $\varepsilon_{\rm f2}/\varepsilon_{\rm f1} \sim d_\perp/d_\parallel$, or $\varepsilon_{\rm f2}/\varepsilon_{\rm f1} \sim \tau_\perp/\tau_\parallel$, where $\tau_{\parallel,\perp}$ are the durations of recombination radiation upon HHG in the case of a conventional and two-colour pump, respectively. By comparing the duration $\tau_\perp \approx 10$ as obtained in our work with the record short duration $\tau_\parallel \approx 250$ as, which was experimentally achieved in Ref. [2] by using an ordinary HHG scheme, we obtained the estimate $\varepsilon_{\rm f2}/\varepsilon_{\rm f1} \approx 0.04$ for the relative reduction in the energy of recombination radiation pulses of duration $\tau_{\rm g} \sim 10$ as.

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