

Quantum-mechanical calculations of residual current density excited during gas ionisation by an intense two-colour laser pulse

N.V. Vvedenskii, A.A. Romanov, A.A. Silaev

Abstract. By solving analytically and numerically the three-dimensional time-dependent Schrödinger equation, we have studied the excitation of a residual current density during gas ionisation by a two-colour laser pulse containing a field at the fundamental frequency and an additional field at the doubled frequency. We have found the dependences of the residual current density on the phase shift between the components of the field and on the intensity of the fundamental harmonic. It is shown that the strong-field approximation taking into account the interaction of freed electrons with the parent ion yields a good quantitative agreement with the results of direct numerical simulation.

Keywords: imaginary-time method, two-colour laser pulse, ionisation, plasma, residual current density, terahertz radiation.

1. Introduction

Currently, much attention is focused on the excitation of a residual current density (RCD) in plasma produced through gas ionisation by intense femtosecond laser pulses. The RCD is responsible for plasma polarisation and excitation of oscillations whose frequencies, at a sufficient plasma density, lie in the terahertz range, which can lead to the generation of broadband terahertz pulses with a high enough peak power [1–12].

Various mechanisms for accelerating free electrons to generate RCD in plasma are being considered at present. When multicycle laser pulses are used, the RCD can be excited due to gas ionisation by two-colour pulses with a strong fundamental field and a weak-amplitude field at the doubled or halved frequency [1–5, 10]. In this case, the maximum (with respect to the phase shift between the field components) RCD is a linear or quadratic function of the amplitude of the additional field in a wide range of intensities of its components. Furthermore, the RCD can also be efficiently excited by using few-cycle (with the duration of the order of the field period) laser pulses [2, 7, 8, 13–15] or a spatially asymmetric ionised medium [16].

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The RCD excited via gas ionisation by a two-colour laser pulse has been previously calculated analytically [10, 17, 18] and numerically [8, 19, 20] with the help of semi-classical and quantum-mechanical approaches. The semi-classical approach is based on the solution of the hydrodynamic equation for the electron current density and the equation for the density of free electrons with a quasi-static probability of tunnelling ionisation per unit time [7]. The quantum-mechanical approach is based on the solution of the three-dimensional time-dependent Schrödinger equation for the electron wave function [13]. The range of applicability of the semi-classical approach is limited by the laser pulse parameters corresponding to the tunnelling ionisation regime, at which the Keldysh parameter $\gamma = \sqrt{I_p}/(2U_p)$ [21] is much less than unity (here I_p is the ionisation potential of an atom, and U_p is the ponderomotive energy of an electron in the laser field) [13–15]. For $\gamma \geq 1$, the electron detachment from the ion occurs at a time of the order of the field period and greater, and to adequately calculate the RCD it is necessary to apply the quantum-mechanical approach.

Previous works, which employed quantum-mechanical numerical calculations of the RCD excited through gas ionisation by a two-colour laser pulse, were aimed at finding the optimal phase shift between the components of the laser pulse fields [19, 20]. In addition, based on the approximate solution of the time-dependent Schrödinger equation, an optimal ratio between the amplitudes of harmonics was found by neglecting the depletion of the ground state of the atoms [18]. Nevertheless, this depletion has a significant impact on the RCD value already at moderately low intensities corresponding to the ionisation threshold.

The aim of this work is to study the dependence of the RCD excitation efficiency on the intensity of the main field of a two-colour laser pulse at a fixed ratio of the intensities of the main and additional fields. The calculations employ the semi-classical approach and the exact numerical solution of the three-dimensional time-dependent Schrödinger equation, which takes into account the depletion of the ground state of the atoms. Based on the approximate semi-analytical solution of the time-dependent Schrödinger equation by the imaginary-time method [22], we interpret differences in the results of the quantum-mechanical and semi-classical calculations at the Keldysh parameter $\gamma \geq 1$.

2. Statement of the problem

Let the electric field \mathbf{E} of an ionising two-colour laser pulse be linearly polarised along the z axis. In order to ensure equality of the time integral of the electric field to zero, we will set $\mathbf{E}(t)$ through the vector potential

$$\begin{aligned} E(t) &= -\frac{1}{c} \frac{dA}{dt}, \quad A(t) = -\hat{z} \frac{cE_0}{\omega_0} a(t), \\ a(t) &= f(t) \left[\sin(\omega_0 t) + \frac{\alpha}{2} \sin(2\omega_0 t + \varphi) \right]. \end{aligned} \quad (1)$$

Here \hat{z} is the unit vector directed along the z axis; E_0 is the maximum amplitude of the main field; α is a small ratio of the amplitudes of the main and additional fields; ω_0 is the frequency of the main field; φ is the phase shift between the components of the additional and main fields; $f(t)$ is the pulse envelope; and c is the velocity of light in vacuum. We assume that the laser pulse envelope has a Gaussian shape

$$f(t) = \exp(-2 \ln 2 t^2 / \tau_p^2), \quad (2)$$

and the pulse duration τ_p is much larger than the field period.

To neglect the collisions of electrons with neighbouring atoms, we assume that the concentration of the gas is sufficiently low. Also, we do not take into account the polarisation response of the plasma, assuming that the maximum plasma density is much smaller than the critical density and the plasma frequency is much smaller than the inverse duration of the laser pulse. For simplicity, we use atomic units in which $|e| = \hbar = m = 1$, where \hbar is the reduced Planck constant, and e and m are the electron charge and mass.

The quantum-mechanical approach to the RCD calculation is based on solving the time-dependent Schrödinger equation for the electron wave function ψ :

$$i \frac{\partial \psi}{\partial t} = \left(-\frac{1}{2} \nabla^2 + U(r) + zE_z \right) \psi. \quad (3)$$

Here E_z is the projection of the electric field strength on the z axis; and $U(r)$ is the potential of interaction with the parent ion. For simplicity, we assume that the gas consists of hydrogen atoms. In this case, the potential of the ion is a Coulomb potential, $U(r) = -1/r$. The RCD directed along the z axis is expressed as

$$j_{\text{RCD}} = -N_g \langle \psi_f | \hat{p}_z | \psi_f \rangle \Big|_{t \rightarrow \infty}, \quad (4)$$

where N_g is the initial concentration of gas atoms; $\hat{p}_z = -i(\partial/\partial z)$ is the z component of the momentum operator; and ψ_f is the wave function of free electrons. The time-dependent Schrödinger equation is solved by the split-step method with a Hankel transform and a fast Fourier transform with respect to spatial variables according to the method described in [15, 23, 24].

The semi-classical approach is based on the classic equation for the electron current density $j(t)$ directed along the z axis,

$$\frac{\partial j}{\partial t} = NE_z, \quad (5)$$

where the concentration of free electrons $N(t)$ is obtained from the equation

$$\frac{\partial N}{\partial t} = (N_g - N)w(|E_z|). \quad (6)$$

Here w is the ionisation probability per unit time, which is determined from the solution of the stationary Schrödinger

equation for an atom in a constant electric field and is a function of an instantaneous value of the electric field. In this paper we assume that the function $w(E)$ is given by the formula corresponding to the process of tunnelling ionisation of the hydrogen atom [25]:

$$w(E) = (4/E) \exp(-2/(3E)). \quad (7)$$

Note that the range of applicability of this formula is limited by sufficiently low laser pulse intensities, corresponding to the tunnelling regime in which the upper limit of the potential barrier is higher than the energy of the ground state of the atom. When this condition is not satisfied (in the above-barrier ionisation regime), it is more appropriate to use an empirical formula for the tunnelling ionisation rate [26]. Application of formula (7) is attributed to a more convenient comparison of the results of the semi-classical approach and the below-described semi-analytical method of the solution of the time-dependent Schrödinger equation. Using this method, the period-averaged ionisation probability per unit time at a small Keldysh parameter coincides with the time-averaged formula (7) [22, 27].

The residual current density j_{RCD} is found as a solution of equation (5) at $t \rightarrow \infty$,

$$j_{\text{RCD}} = \int_{-\infty}^{\infty} NE_z dt. \quad (8)$$

Then we normalise the RCD to the maximum possible oscillatory current density in the field of the fundamental harmonic, $j_{\text{osc}} = E_0 N_g / \omega_0$. The obtained normalised current density

$$j_{\text{norm}} = j_{\text{RCD}} / j_{\text{osc}} \quad (9)$$

is independent of N_g , and its square characterises the conversion efficiency of the laser pulse energy to the energy of low frequency terahertz radiation [7].

3. Calculation of the residual current density by the imaginary-time method

The high accuracy of the results obtained on the basis of numerical solution of the time-dependent Schrödinger equation requires much computing time. In addition, the numerical results often do not have a clear physical interpretation. Therefore, of great interest is the development of analytical or semi-analytical methods for finding the RCD, based on an approximate solution of the time-dependent Schrödinger equation. In this section we describe the imaginary-time method used for this purpose [22, 27].

Assume that the pulse duration is so great that the concentration of free electrons increases for a large number of periods of the electric field. This makes it possible to present the RCD as an integral:

$$j_{\text{RCD}} = \int_{-\infty}^{\infty} \frac{\partial \bar{j}}{\partial t} dt, \quad (10)$$

where $\partial \bar{j} / \partial t$ is the time derivative of the low-frequency (averaged over the field period) current density \bar{j} . The value of $\partial \bar{j} / \partial t$ is

$$\frac{\partial \bar{j}}{\partial t} = -\bar{N}_n(t) \int p_z \bar{W}(\mathbf{p}, t) d^3 p, \quad (11)$$

where $\bar{W}(\mathbf{p}, t)$ is the period-averaged momentum distribution of the ionisation probability per unit time; and $\bar{N}_n(t)$ is the averaged concentration of neutral atoms. This value is equal to the initial gas concentration N_g and satisfies the approximate equation

$$\frac{\partial \bar{N}_n}{\partial t} = -\bar{N}_n(t) \int \bar{W}(\mathbf{p}, t) d^3p. \quad (12)$$

Since the characteristic time of variation of $\bar{W}(\mathbf{p}, t)$ is much larger than the field period, in calculating $\bar{W}(\mathbf{p}, t)$ we can assume that the electric field has a constant envelope [17] and that the depletion of the ground state of an atom for the period of the laser pulse field is negligible. The latter assumption is valid even at a very high peak intensity, because in this case, the depletion of the ground state of the atom takes place mainly at the leading edge of the laser pulse at an intensity that is close to the breakdown threshold. Within the framework of the strong-field approximation [21], in which we neglect the interaction of photoelectrons with the parent ion, $\bar{W}(\mathbf{p}, t)$ is represented as the sum of the probabilities of n -photon processes:

$$\bar{W}(\mathbf{p}, t) = \frac{\omega_0}{2\pi} |L(\mathbf{p})|^2 \sum_{n=n^*}^{\infty} \delta\left(\frac{\Delta E}{\omega_0} - n\right). \quad (13)$$

Here $n^* = \langle \tilde{I}_p/\omega_0 + 1 \rangle$ is the minimum possible number of absorbed photons (brackets denote the integer part); $\tilde{I}_p = I_p + U_p$; $U_p = U_{p0}(1 + \alpha^2/4)$ is the ponderomotive energy of an electron in a two-colour laser field; $U_{p0} = E_f^2/(4\omega_0^2)$ is the ponderomotive energy of an electron in the field of the fundamental harmonic; $E_f(t) = E_0 f(t)$ is the amplitude of the electric field of the laser pulse; and $\Delta E = p^2/2 + \tilde{I}_p$ is the energy spent on ionisation and acceleration of the electron.

The function $L(\mathbf{p})$ in formula (13) describes the envelope of the momentum distribution of the ionisation probability per unit time. Assuming that the photon energy is much less than the ionisation energy (i.e. $n_0 = I_p/\omega_0 \gg 1$), the function $L(\mathbf{p})$ is written as

$$L(\mathbf{p}) = (I_p/2)^{1/4} \sum_s [S^s(\mathbf{p}, t_s)]^{-1/2} \exp(iS(\mathbf{p}, t_s)). \quad (14)$$

Here

$$S(\mathbf{p}, t) = \int_0^t \left[\frac{(\mathbf{p} + \mathbf{A}(t')/c)^2}{2} + I_p \right] dt' \quad (15)$$

is the part of the action of a free electron, which does not depend on the coordinates; t_s are the stationary points $S(\mathbf{p}, t)$; and s is an index numbering the stationary points. The t_s values satisfy the equation

$$\partial S(\mathbf{p}, t)/\partial t|_{t_s} = 0 \quad (16)$$

and have a positive imaginary part and a real part which lies in the range $[0, 2\pi/\omega_0]$.

The strong-field approximation does not take into account the electron interaction with the parent ion at the stage of motion of an electron in the continuum. This leads to significant differences in the magnitude and the character of the function of the photoelectron momentum distribution from the results of the numerical solution of the three-dimensional Schrödinger equation. Interaction of photoelectrons with the parent ion can be taken into account on the basis of the imag-

inary-time method which is used for the correction of the electron action and trajectories in the laser field [28, 29]. Let us express action S through complex trajectories of the electrons:

$$S(\mathbf{p}, t_s) = c_0 + \int_{t_s}^{t_{\max}} \left(\frac{\mathbf{p}_s^2}{2} - \mathbf{E}r_s - I_p \right) dt - (\mathbf{p}_s r_s)|_{t_s}^{t_{\max}}, \quad (17)$$

where c_0 is a constant independent of s ; and t_{\max} is the time after the pulse propagation. Trajectories r_s correspond to the motion of electrons under the action of an external electric field of the laser pulse and satisfy Newton's equations

$$\frac{\partial r_s}{\partial t} = \mathbf{p}_s, \quad \frac{\partial \mathbf{p}_s}{\partial t} = -\mathbf{E}(t) \quad (18)$$

with the initial conditions

$$r_s(t_s) = 0, \quad \mathbf{p}_s^2(t_s) = -2I_p, \quad (19)$$

which determine the most probable trajectories, minimising action S . Account for the electron interaction with the parent ion leads to an additional term

$$S_C = \int_{t_s}^{t_{\max}} \frac{dt}{r_s(t)} \quad (20)$$

in the action, and to the correction of the equation for the trajectory of the electron motion:

$$\frac{\partial \mathbf{p}_s}{\partial t} = -\mathbf{E}(t) - \frac{1}{r_s^3} \mathbf{r}_s. \quad (21)$$

Allowance for the electron interaction with the parent ion leads to the additional condition (that is automatically fulfilled for complex trajectories, not taking into account the Coulomb interaction):

$$\mathbf{p}_s(t_{\max}) = \mathbf{p}. \quad (22)$$

The solution of equations (18) with initial conditions (19) for $t \rightarrow t_s$ corresponds to $r_s^2 \approx -2I_p(t - t_s)^2$. This leads to the fact that the correction to action (20) has a logarithmic divergence. Regularisation of the action is based on the following idea [27]. When an electron is located near a nucleus, its motion is determined primarily by the interaction with the parent ion rather than with the laser field. In this case, the action of the electron is determined by the asymptotic behaviour of the atomic wave function at large distances from the nucleus. After the regularisation procedure, the correction to the action takes the form

$$S_C = -iv \ln[2iI_p(t_{\max} - t_s)] + \int_{t_s}^{t_{\max}} \left(\frac{1}{r_s(t)} + \frac{iv}{t - t_s} \right) dt, \quad (23)$$

where $v = 1/\sqrt{2I_p}$ is the effective principal quantum number. As a result, taking into account corrections associated with the interaction of electrons with the parent ion,

$$L(\mathbf{p}) = (I_p/2)^{1/4} \sum_s [S^s(t_s)]^{-1/2} \exp[iS(\mathbf{p}, t_s) + iS_C(\mathbf{p}, t_s)]. \quad (24)$$

To find the complex trajectories satisfying (21) with conditions (19), (22), an iterative approach is used [28]. In the first step, the initial momentum \mathbf{p}_{s0} is used, and the trajectory of the electron and its final momentum \mathbf{p}_s are calculated. In

the second step, the initial momentum is corrected based on the comparison of the obtained final momentum $\mathbf{p}_{s'}$ with the desired value of the momentum \mathbf{p} . The procedure is repeated to achieve a high convergence of calculations of electron trajectories.

4. Results of calculations

The RCD value strongly depends on such parameters of a two-colour laser pulse as the phase shift φ between the pulse components, their intensities and the wavelength of the main field $\lambda_0 = 2\pi c/\omega_0$. The model developed in this paper, based on the imaginary-time method, allows one to find the dependences of the RCD on these parameters. The results obtained show that the dependence of the RCD on the phase shift φ is close to sinusoidal, i.e. it vanishes at a nonoptimal phase and is maximal at an optimal phase φ_{opt} . The RCD corresponding to the optimal phase depends linearly on the amplitude of the additional fields in accordance with the results of semi-classical calculations [2, 30].

Let us now study the dependence of the RCD on the main field intensity $I_0 = cE_0^2/(8\pi)$. Figure 1 shows the dependences of the normalised maximum current density $j_{\text{max}} = |j_{\text{norm}}(\varphi_{\text{opt}})|$ corresponding to the optimal phase shift φ_{opt} on I_0 at a fixed ratio of the amplitudes of the main and additional fields, $\alpha = 0.2$.

One can see from Fig. 1 that with increasing intensity, j_{max} increases sharply, reaches a maximum at an intensity of $\sim 2 \times 10^{14} \text{ W cm}^{-2}$ and gradually decreases at higher intensities, according to the law $\propto I_0^{-1/2}$. The maximum normalised RCD j_{max} corresponding to the optimal intensity depends weakly on the laser wavelength and is equal to ~ 0.2 both in quantum-mechanical and semi-classical calculations. At high intensities corresponding to the tunnelling ionisation regime ($\gamma \ll 1$), the results of semi-classical and quantum-mechanical calculations are in good agreement with each other. At the same time, when $\gamma \geq 1$, the semi-classical approach greatly underestimates the value of j_{max} .

To explain the differences in the results of semi-classical and quantum-mechanical calculations for $\gamma \geq 1$, we consider the semi-analytical solution of the time-dependent Schrödinger equation by the imaginary-time method. The solid curve in Fig. 1 shows the function $j_{\text{max}}(I_0)$ found on the basis of this method. It can be seen that this curve quantitatively coincides with the result of a direct numerical calculation in the entire range of the values of the Keldysh parameter γ . In the multiphoton ionisation regime ($\gamma \gg 1$), the dependence $j_{\text{max}}(I_0)$ is close to a power function with an exponent increasing with the wavelength of the laser pulse.

The power dependence $j_{\text{max}}(I_0)$ in the multiphoton ionisation regime is associated with a corresponding power-law dependence of the ionisation probability per unit time on the laser pulse intensity. It follows from Fig. 2, in which the solid and dotted curves show the dependence of the period-averaged ionisation probability \bar{w} per unit time, obtained by the imaginary-time method, on the intensity I of the main field of a two-colour laser pulse under the assumption that the pulse envelope is constant. In the calculations the ratio of the amplitudes of the main and additional fields is $\alpha = 0.2$, and the wavelength of the main field is $\lambda_0 = 800$ or 1200 nm. The phase shift φ is set so as to maximise the absolute value of the RCD. It can be seen that the curves $\bar{w}(I)$ for different wavelengths coincide with each other in the region of tunnelling ionisation and differ at $\gamma \geq 1$. In this region the curves $\bar{w}(I)$

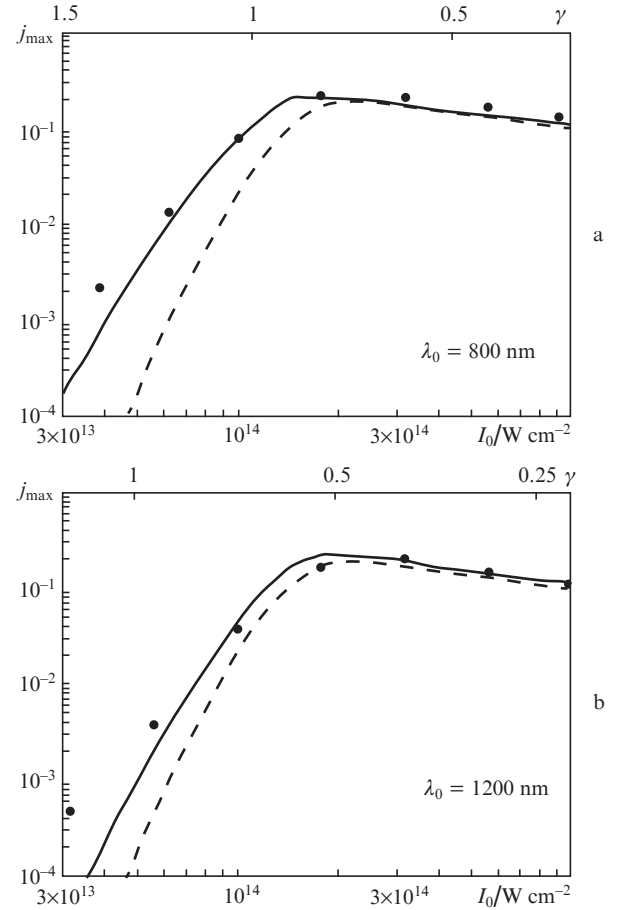


Figure 1. Dependences of the maximum normalised residual current density $j_{\text{max}} = |j_{\text{norm}}(\varphi_{\text{opt}})|$ corresponding to the optimal shift phase φ_{opt} on the peak intensity I_0 of the fundamental harmonic for the wavelength of the main field $\lambda_0 =$ (a) 800 nm and (b) 1200 nm at a laser pulse duration $\tau_p = 50$ fs and amplitude ratio of the additional and main fields $\alpha = 0.2$. The dashed curves are the results of the semi-classical approach, solid curves are the calculations based on the imaginary-time method and points are the numerical solution of the time-dependent Schrödinger equation.

are close to power functions with an exponent that increases with increasing wavelength. Also in Fig. 2 the dashed curve shows the averaged tunnelling formula (7). It coincides with the quantum-mechanical calculations only in a narrow region corresponding to small values of the Keldysh parameter. In the region $\gamma \geq 1$, the tunnelling formula significantly underestimates the ionisation probability; as a result, the low-frequency current density found on the basis of the semi-classical approach is low compared with the results of quantum-mechanical calculations. It can be seen from Fig. 3, which shows the dependences of the maximum (with respect to the phase shift φ) time derivative of the low-frequency current density $|\partial \bar{j} / \partial t|$ on the intensity I , calculated on the basis of the imaginary-time method and semi-classical approach at the same parameters of the laser pulse, as in Fig. 2.

Let us now briefly consider the dependence of the optimal phase shift corresponding to the maximum RCD on the laser pulse intensity. Previously, these dependences have been investigated numerically in [19, 20]. It has been shown that the optimal phase shift φ_{opt} obtained on the basis of the semi-classical approach is approximately equal to $\pi/2$, regardless of the intensity of the fundamental harmonic. At the same time,

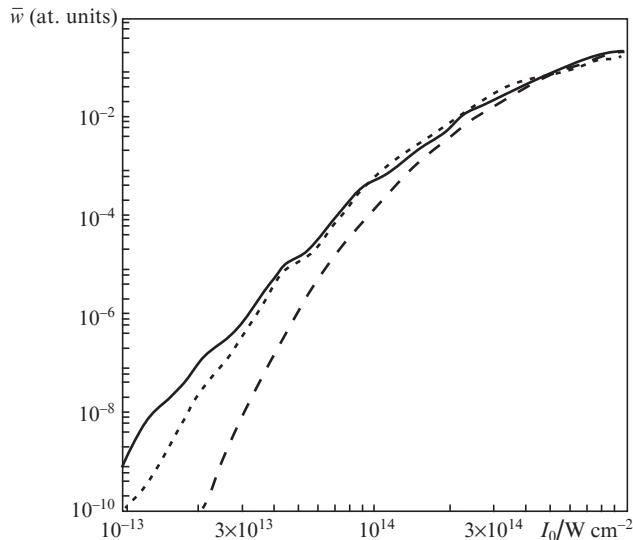


Figure 2. Dependences of the period-averaged ionisation probability \bar{w} per unit time on the intensity I of the main field of the laser pulse at $\alpha = 0.2$. The dashed curve is the averaged tunnelling formula (7), dotted curve is the calculation based on the imaginary-time method for $\lambda_0 = 800$ nm and solid curve is the same for $\lambda_0 = 1200$ nm.

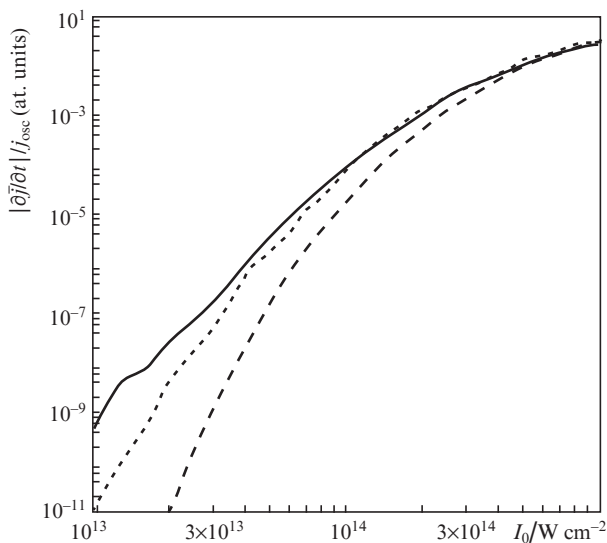


Figure 3. Dependences of the maximum derivative of the low-frequency current density $|\partial\bar{j}/\partial t|$ normalised to the oscillatory current density j_{osc} on the intensity I of the main field of the laser pulse at $\alpha = 0.2$. The dashed curve is the calculation based on the semi-classical approach, dotted curve is the calculation based on the imaginary-time method for $\lambda_0 = 800$ nm and solid curve is the same for $\lambda_0 = 1200$ nm.

the value of φ_{opt} found by solving numerically the time-dependent Schrödinger equation increases with decreasing laser pulse amplitude and tends to constant values that are close to π and $\pi/2$ at small and large intensities, respectively. It is also confirmed in Fig. 4, which shows the obtained dependences $\varphi_{\text{opt}}(I_0)$ at a wavelength of the main field $\lambda_0 = 1200$ nm. An approximate solution of the time-dependent Schrödinger equation on the basis of the imaginary-time method yields a good qualitative agreement with the results of direct numerical simulation. A detailed analysis of the

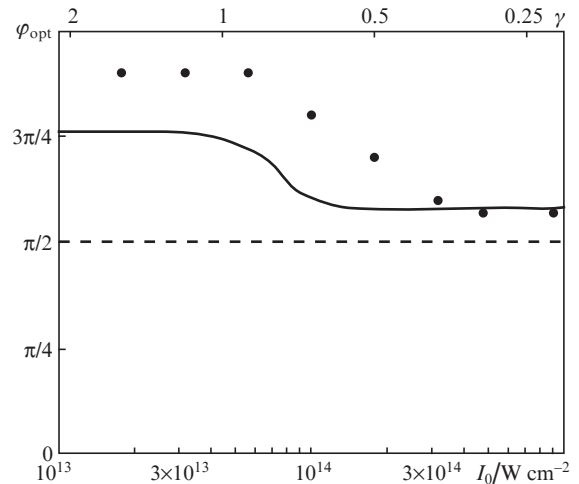


Figure 4. Dependences of the optimal phase shift φ_{opt} between field components of the laser pulse on the peak intensity I_0 of the main field at $\lambda_0 = 1200$ nm, $\tau_p = 50$ fs and $\alpha = 0.2$. The dashed curve is the result obtained on the basis of the semi-classical approach, solid curve is the calculation based on the imaginary-time method and points are the numerical solution of the time-dependent Schrödinger equation.

electron motion equations in a two-colour laser field shows that in taking into account the interaction of electrons with the parent ion, the trajectories to the ion are strongly distorted. This leads to a change in the optimal phase shift when changing the intensity I_0 of the main field near the values corresponding to $\gamma \sim 1$. For large values of intensity corresponding to $\gamma \ll 1$, the distortion of the trajectories is negligible, and the optimal phase shift is constant and close to the value obtained on the basis of the semi-classical approach.

5. Conclusions

Having solved numerically and semi-analytically the time-dependent Schrödinger equation, we have investigated the excitation of the residual current density during gas ionisation by a two-colour laser pulse. We have found the dependences of the optimal phase shift and the corresponding maximum RCD on the intensity of the main field. It is shown that the imaginary-time method, which takes into account the interaction of free electrons with the parent ion, allows one to calculate the RCD with high accuracy in a wide range of intensities and wavelengths corresponding to both tunnelling and multiphoton ionisation regimes. Calculations of the concentration of free electrons and RCD by the imaginary-time method can be used to interpret the results of numerical calculations and to find the optimal parameters of laser pulses in order to implement the ionisation mechanisms of generation of terahertz radiation.

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