Residual-Current Excitation in Plasmas Produced by Few-Cycle Laser Pulses

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Along with the generation of extreme-ultraviolet and soft x-ray radiation, gas ionization by an intense few-cycle laser pulse can also induce the generation of low-frequency terahertz waves. The latter is caused by the excitation of a residual quasi-dc current in the produced plasma by the electric field of the laser pulse. We describe this phenomenon using the quantum-mechanical approach based on solving the 3D time-dependent Schrödinger equation. We calculate the dependences of the residual-current density on the carrier-envelope phase, duration, and intensity of the few-cycle laser pulse, and find optimal conditions for high-efficiency realization of the studied phenomenon.

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The ionization-induced phenomena caused by intense few-cycle laser pulses are traditionally considered in the context of the generation of extreme-ultraviolet and soft x-ray radiation, and, in particular, attosecond pulses. These phenomena are studied in many papers, which focus on finding high harmonics of polarization currents excited in the process of the interaction of atoms and molecules with intense ionizing laser fields. The main physical mechanism of high-harmonic generation is recombination of the photoelectron with the parent ion, when the former comes back under the electric field of the laser pulse (see reviews [1], also Refs. [2–5], and references therein).

Several recent papers have described observations in both experiments and numerical simulations of a new phenomenon, namely, ionization-induced conversion of few-cycle laser pulses into low-frequency radiation, more specifically, into terahertz (THz) waves [6–8]. The essence of this phenomenon can be explained as follows. When a laser pulse ionizes atoms or molecules, the newly freed electrons acquire the constant drift velocity along with the oscillatory one. The quantity and direction of the former are determined by the phase of the electric field at the moment of ionization. As a result, after the passage of the laser pulse, some residual-current density (RCD) remains in the produced plasma, and its value can be rather high if the laser pulse is very short. This dc RCD is an initial impetus to plasma polarization and excitation of transverse dipole oscillations which can have THz frequencies, if the plasma is sufficiently dense. The energy stored in these oscillations is proportional to RCD squared and is radiated through plasma boundaries into the environment [7,8]. This phenomenon was proposed both for efficient generation of high-power THz radiation [7] and as a comparatively simple way to determine the carrier-envelope phase (CEP) of few-cycle laser pulses by detecting the THz waves emitted by the plasma [6,8] [and not by measuring the parameter of the asymmetry in the distribution of photoelectrons along the electric field, as it is done conventionally (see the review [9] and Refs. [10,11])].

It is evident that the key problem of describing theoretically the phenomenon of the conversion of few-cycle laser pulses into low-frequency (THz) radiation is to find RCD. By analogy with the problems of finding high harmonics [1–5] and asymmetries in spatial distribution of photoelectrons [9–11], here one has to develop both the *ab initio* quantum-mechanical models, which are based on solving the time-dependent Schrödinger equation (TDSE), and the semiclassical models, which are less accurate, but much simpler. The quantum-mechanical approach allows one to calculate RCD precisely in a wide range of laser pulse parameters, as well as to determine the domain of applicability of simple semiclassical models. However, up to now, no quantum-mechanical approaches to description of the phenomenon of RCD excitation and to calculation of RCD have been developed.

In this Letter, we study consistently the ionization-induced excitation of RCD in the plasmas produced by few-cycle laser pulses within the framework of the semiclassical and quantum-mechanical approaches. We find the dependences of RCD on CEP, duration, and intensity of the laser pulse, and determine the optimal parameters of the laser pulse, for which the efficiency of RCD generation is maximal. The results yielded by the quantum-mechanical and the semiclassical models are compared and the domain of applicability of the semiclassical model is determined.

Consider hydrogen atoms affected by a few-cycle laser pulse with the electric field aligned with the z axis and having the Gaussian envelope with respect to time t,

$$\begin{aligned} E(t) &= \frac{1}{\omega_L}\frac{\partial a}{\partial t}, \\ a(t) &= E_0 \sin(\omega_L t + \varphi)\exp\left(-(2\ln 2)\frac{t^2}{\tau_p^2}\right). \end{aligned} \tag{1}$$

Here, E_0 is the envelope maximum, ω_L is the carrier frequency, τ_p is the pulse duration (intensity FWHM), and φ is CEP. We will focus on fairly low values of the intensity and neglect the influence of magnetic field of the laser pulse on the studied processes. We will also neglect the influence of the produced plasma on the laser field described by Eq. (1) under the assumption that the maximum possible plasma frequency $\omega_{pm} = \sqrt{4\pi e^2 N_g/m} \ll \tau_p^{-1} \ll \omega_L$. Here, N_g is the gas density before the start of the ionization process and e and m are the electron charge and mass, respectively.

The semiclassical approach to calculating RCD is based on the following assumptions: (i) the electron passes over from the ground state in the atom to the free state during a time, which is much shorter than the characteristic time of changes in the instantaneous value of the electric-field strength; (ii) at the ionization moment, electrons have zero velocity (or their velocity distribution is isotropic), and further, they move as classical particles under the electric field of the laser pulse. The semiclassical approach consists in solving the equation for the free-electron density $N(t)$ and the linear classical equation for the electron-current density $j(t)$ in plasma. These equations were used earlier in Refs. [7,8] to simulate numerically the processes of converting few-cycle laser pulses into THz radiation. The value of RCD j_{RCD} is found by solving these equations for $t \longrightarrow \infty$,

$$j_{\mathrm{RCD}} = \frac{e^2}{m} \int_{-\infty}^{+\infty} N(t)E(t)dt, \qquad (2)$$

where

$$N(t) = N_g\left[1 - \exp\left(-\int_{-\infty}^{t} w(|E(t')|)dt'\right)\right]. \qquad (3)$$

Here, $w(|E|)$ is the probability of tunneling ionization per unit time. We use the accurate dependence $w(|E|)$, which was found in Ref. [12] by solving numerically the quantum-mechanical problem about ionization of a hydrogen atom from the ground state in the static electric field E.

Unlike the semiclassical method, the quantum-mechanical approach to calculating RCD allows one to consider all possible stages of the electron dynamics, including transitions between bound states in the atom, transitions to the free state (ionization) with arbitrary initial velocities, motion under the action of the electric field $E(t)$ of the laser pulse and the Coulomb field of the nucleus, and recombination with the parent ion. It is based on solving the 3D TDSE for the electron wave function $\psi(\mathbf{r}, t)$,

$$i\hbar\frac{\partial\psi(\mathbf{r}, t)}{\partial t} = \left(-\frac{\hbar^2}{2m}\nabla^2 - \frac{e^2}{r} - eE(t)z\right)\psi(\mathbf{r}, t). \qquad (4)$$

The initial condition corresponds to the ground state of the electron in the atom. The electron-current density contains only the z component $j(t)$, which is determined by infinite-volume integration,

$$j(t) = \frac{eN_g}{m}\bar{p}_z(t) = -\frac{i\hbar e N_g}{m}\int \psi^* \frac{\partial\psi}{\partial z} d^3\mathbf{r}. \qquad (5)$$

Here, $\bar{p}_z(t)$ is the average value of the longitudinal momentum. Note that $\bar{p}_z(t)$ is the time integral of the polarization response of the atom, whose time dependence is usually calculated for the description of generation of high harmonics and attosecond pulses [1–5].

Equation (4) is solved numerically by the split-step method, which allows one to calculate the wave function with high accuracy in a wide range of laser pulse parameters [5,13,14]. The time dependence $\bar{p}_z(t)$ is found by using the Ehrenfest theorem, which makes it possible to reduce the calculation time significantly [3,5]. To prevent reflections of the wave function, absorbing conditions [4] are set at the boundaries of the calculation area, and their specific form is taken from Ref. [14].

After the passage of the laser pulse, at $t \gg \tau_p$, the current density is the sum of the dc RCD j_{RCD} of free electrons and the ac current density $j_b(t)$ of the electrons staying in the bound states in the atoms. The latter oscillates fast in time at the frequencies determined by the energies of the transitions between stationary bound states. The value of j_b is found numerically from Eq. (5) by integrating in the vicinity of the nucleus. Thus, j_{RCD} is found by subtracting j_b from the total current density j.

In what follows, we calculate RCD using both the quantum-mechanical method and the semiclassical one. In both cases, we normalize j_{RCD} with regard to the maximum possible oscillatory-current density $j_{\mathrm{osc}} = eN_g V_{\mathrm{osc}} = e^2 N_g E_0/(m\omega_L)$ in a pulse with the envelope maximum E_0. The normalized RCD $j_{\mathrm{norm}} = j_{\mathrm{RCD}}/j_{\mathrm{osc}}$ is the dimensionless excitation factor which does not depend on the gas density N_g. The square of the normalized RCD characterizes the efficiency of conversion of the laser pulse energy into the energy of low-frequency plasma oscillations [7].

The results of numerical calculations for the central wavelength 800 nm are presented in Figs. 1–3. Figure 1 shows the dependences of the normalized RCD $j_{\mathrm{norm}}(\varphi) = -j_{\mathrm{norm}}(\varphi + \pi)$ on CEP φ for the two values of FWHM, which correspond to the single-cycle [$\tau_p = 2.67$ fs [Fig. 1(a)]] and two-cycle [$\tau_p = 5.34$ fs [Fig. 1(b)]] laser pulses, at different fixed values of the intensity $I = (c/8\pi)E_0^2$, where c is the speed of light. We can see that RCD is a sign-alternating function of CEP, which reaches its absolute maximum at some optimal values of CEP φ_{opt}. The results obtained from the semiclassical model always look like smooth curves with one extremum (at $0 \le \varphi < \pi$). They agree well with the results obtained from the quantum-mechanical model for sufficiently high intensities and, correspondingly, small values of the Keldysh parameter $\gamma = \sqrt{I_p/2U_p} \ll 1$ [here, $I_p = 13.6$ eV is the ionization potential of the hydrogen atom and $U_p = (eE_0)^2/(4m\omega_L^2)$ is the maximum ponderomotive potential [15]].

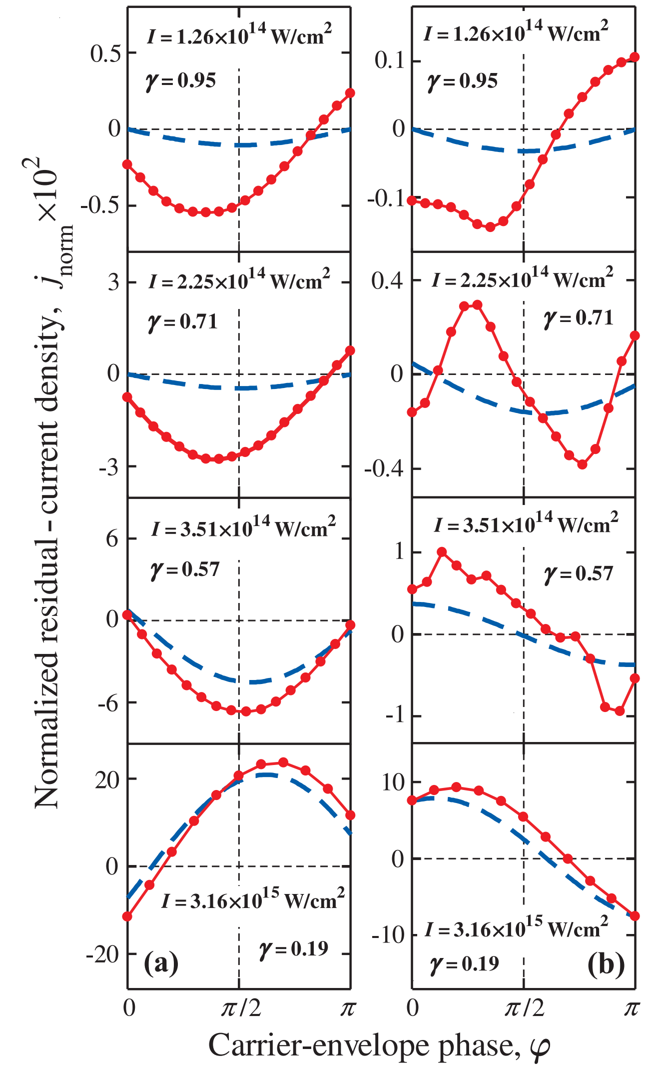

FIG. 1 (color online). Normalized residual-current density $j_{\mathrm{norm}} = j_{\mathrm{RCD}}/j_{\mathrm{osc}}$ as a function of the carrier-envelope phase φ for (a) single-cycle ($\tau_p = 2.67$ fs) and (b) two-cycle ($\tau_p = 5.34$ fs) laser pulses, central wavelength 800 nm, and different values of the laser intensity $I = (c/8\pi)E_0^2$. Here, $j_{\mathrm{osc}} = e^2 N_g E_0/(m\omega_L)$ is the maximum oscillatory-current density and E_0 is the maximum of the laser pulse envelope. The figures show also the corresponding values of the Keldysh parameter γ. The results obtained from the semiclassical model are shown as dashed curves, and the results obtained from the quantum-mechanical model are shown as dots connected with solid lines.

The results obtained from the quantum-mechanical and semiclassical models start to differ significantly at sufficiently small intensities, when the Keldysh parameter approaches unity. Under these conditions, the role of electron scattering on and recombination with the parent ion grows, since the cross sections of these processes increase with decreasing electron velocity [2]. The electron wave packet can scatter repeatedly on the ion under the electric field of the laser pulse. It leads to a complicated dependence of RCD on CEP, which is found from the quantum-mechanical model and is shown in Fig. 1(b). Note that for such laser intensities, similar complicated (nonsinusoidal) CEP dependences were obtained in Ref. [10] in calculations of the asymmetry parameters in the spatial distribution of photoelectrons. The influence of scattering and recombination is much weaker in the case of a shorter (single-cycle) laser pulse [Fig. 1(a)], in which CEP dependences are simpler.

Another important difference between the semiclassical and quantum-mechanical approaches is the maximum value of RCD. When the Keldysh parameter is close to unity, the contribution of multiphoton ionization becomes significant. However, it is not allowed for by the semiclassical model based on Eqs. (2) and (3). The rate of multiphoton ionization is a power function of the electric field amplitude, unlike the rate of tunneling ionization, which is exponentially small at low intensities [15,16]. Besides, a significant part of photoelectrons produced in the multiphoton ionization regime can have initial velocities, which are comparable with the maximum possible oscillatory velocities in the laser-pulse field or even exceed them [16]. Because of this, for low laser intensities, the maximum RCD values obtained from the semiclassical model turn out to be strongly underrated as compared with the *ab initio* results.

Thus, the results obtained from the quantum-mechanical and semiclassical models can differ considerably, which

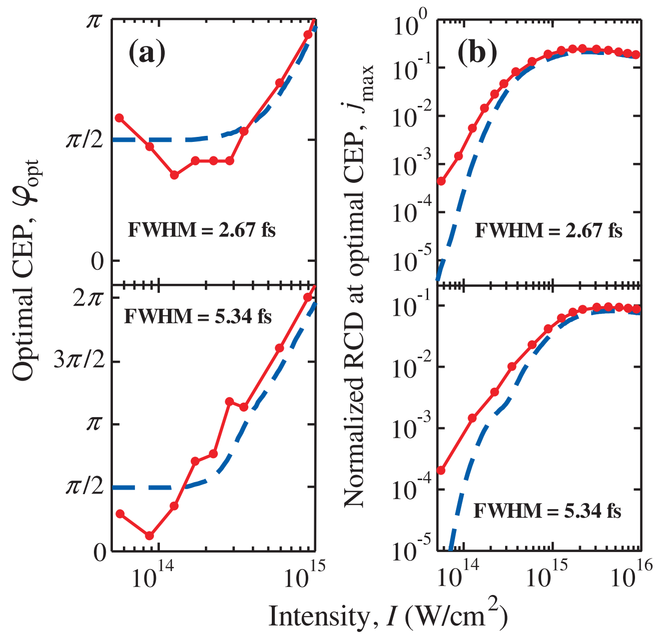

FIG. 2 (color online). (a) Optimal carrier-envelope phase φ_{opt} and (b) the corresponding absolute value of normalized RCD $j_{\max} = |j_{\mathrm{norm}}(\varphi_{\mathrm{opt}})|$ as functions of the intensity I for single-cycle ($\tau_p = 2.67$ fs) and two-cycle ($\tau_p = 5.34$ fs) laser pulses. The results obtained from the semiclassical model are shown as dashed curves, and the results obtained from the quantum-mechanical model are shown as dots connected with solid lines.

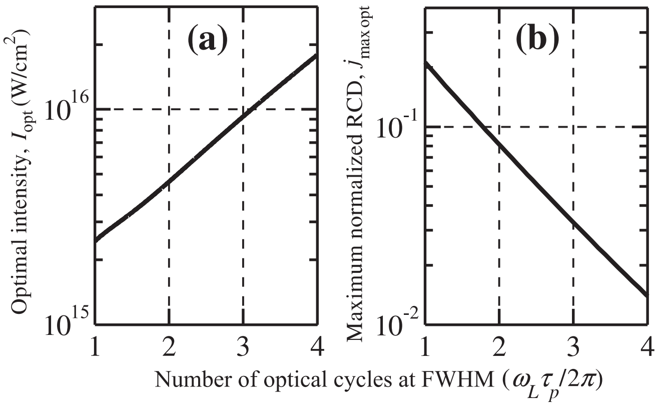

FIG. 3. (a) Optimal laser intensity I_{opt} for efficient excitation of RCD and (b) the corresponding maximum normalized RCD $j_{max\ opt} = j_{max}(I_{opt})$ as functions of the number of optical cycles at FWHM.

can be of fundamental importance. This is seen also in Fig. 2, which shows the dependences of the optimal CEP φ_{opt} [Fig. 2(a)] and the corresponding absolute value of the normalized RCD $j_{max} = |j_{norm}(\varphi_{opt})|$ [Fig. 2(b)] on the intensity I. Specifically, the semiclassical model predicts that with decrease in the laser intensity, the optimal CEP always tends to $\pi/2$ (this regularity was also found in Ref. [8] using the semiclassical model). However, as seen from Fig. 2, it contradicts the *ab initio* results, since no such regularity is observed in them (which may be very important for further experiments proposed in Refs. [6,8] to determine CEP of few-cycle laser pulses).

As the laser intensity grows, the value of j_{max} increases and, as seen from Fig. 2, reaches its maximum $j_{max\ opt}$ at some sufficiently high intensity I_{opt}, when the results obtained from the semiclassical and quantum-mechanical models coincide with very high accuracy (which may be used in the future, e.g., to calculate the optimal regimes of RCD excitation and generation of THz waves during ionization of other gases). Figure 3 shows the dependences of I_{opt} [Fig. 3(a)] and $j_{max\ opt}$ [Fig. 3(b)] on the number of optical cycles at FWHM. Actually, the square of $j_{max\ opt}$ determines the maximum possible efficiency of conversion of the laser pulse energy into the energy of low-frequency (THz) radiation for a given pulse duration. This efficiency can be very high (~10%) for a single-cycle pulse and, as seen from Fig. 3, it decreases exponentially when the pulse duration grows. The optimal intensity, which is equal to 2.2×10^{15} W/cm^2 for a single-cycle pulse, grows exponentially with the increasing duration and reaches approximately 10^{16} W/cm^2 for the pulse containing 3–4 optical cycles at FWHM.

To conclude, the ionization-induced excitation of the residual current in the plasmas produced by few-cycle laser pulses was studied on the basis of the *ab initio* quantum-mechanical approach for the first time. The residual current is the initial impetus to plasma polarization and excitation of low-frequency plasma oscillations which can emit THz waves [6–8]. The value of the residual-current density is strongly CEP dependent, which can be used to determine CEP of few-cycle laser pulses by detecting the low-frequency (THz) radiation [6,8]. Under optimal conditions, the efficiency of RCD excitation grows exponentially with decrease in the pulse duration and reaches values allowing optical-to-THz conversion with very high efficiency. The phenomenon of RCD excitation can be studied on the basis of the same theoretical models and realized efficiently under the same experimental conditions as ionization-induced generation of high harmonics and attosecond pulses.

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