Generation of high-order harmonics in plasmas of multicharged atomic ions produced by an intense laser pulse

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The yield of high-order harmonics has been derived for relativistic plasmas of multicharged atomic ions produced by an intense, linearly polarized laser pulse. Harmonics of the laser field are excited at the elastic electron-ion collisions in plasmas. In the nonrelativistic case only odd harmonics can be excited. The ultrarelativistic limit is considered when both even and odd harmonics are excited with the comparable efficiencies. Drift electron velocities are assumed to be small compared to the quiver electron velocities. It is shown that the harmonic yields do not depend on the instantaneous energy distribution of electrons produced at the multiple ionization on the leading edge of the laser pulse. The harmonic conductivity is a tensor with odd components along the laser polarization and with even components along the direction of propagation of the laser pulse. This conductivity tensor has been derived analytically in the ultrarelativistic limit. Possible observation of even harmonics is discussed with respect to the Weibel effect in laser plasmas.

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When an intense laser pulse interacts with an atomic medium (gases, solids, or clusters), multicharged atomic ions are produced due to the tunneling or barrier-suppression field ionization [1]. Most of the produced free electrons are ejected from atomic ions with drift velocities which are usually nonrelativistic; it is true, in particular, for electrons ejected at the initial part of the leading edge of the intense laser pulse. Indeed, the Keldysh parameter

$$\gamma=\frac{\omega\sqrt{2E_Z}}{F}\ll 1 \tag{1}$$

is very small for tunneling and barrier-suppression regimes of multiple ionization [2]. Here F and ω are the field strength amplitude and the field frequency of the laser pulse, respectively, and E_Z is the ionization potential of the considered multicharged atomic ion (Z is its charge multiplicity). The atomic units (a.u.) are used throughout the paper for all quantities, where $e=m_e=\hbar=1$. The typical electron drift velocities along the field strength ($V_{||}$) and in the transverse direction ($V_\perp$) are given by the simple expressions [3,4]:

$$V_{||}=\sqrt{\frac{3\omega}{2\gamma^3}},\quad V_\perp=\left(\frac{F}{2\sqrt{2E_Z}}\right)^{1/2}. \tag{2}$$

The total velocity of the free electron is the vector sum of this drift velocity and the quiver velocity in the linearly polarized laser field $V_F=V\sin\omega t$, where $V=F/\omega$ (in the nonrelativistic case). The most interesting case is realized both in nonrelativistic and relativistic cases when the quiver velocity is much larger than the drift velocity, i.e., $V\gg V_{||}>V_\perp$. This case was investigated by Silin [5–7] in the nonrelativistic limit. Odd harmonics of the laser radiation are irradiated at the elastic collisions of nonrelativistic electrons with multicharged atomic ions in the presence of the laser field. The harmonic conductivity is given by the simple analytic expression [5]

$$\sigma_{2n+1}=\left(\frac{\omega_p}{\omega}\right)^2\frac{4Z^2n_i\Lambda}{V^3}\ln\left(\frac{V}{(2n+1)V_\perp}\right). \tag{3}$$

Here $\omega_p=\sqrt{4\pi n_e}$ is the plasma frequency and n_e is the electron number density; n_i is the number density of the atomic ions with the charge multiplicity Z. Finally, $\Lambda=\ln(V^2/[(2n+1)\omega])$ is the quantum Coulomb logarithm for the considered case [5], which appears due to small effective scattering angles of an electron. It is seen that the conductivity decreases very slowly (only in logarithm term) with n. This is true at $n<n_{\max}$. Here $n_{\max}=eV^2/(8V_{||}^2)$ is the maximum number (cutoff) of the harmonic for nonrelativistic ionization [5]. The conductivity decreases very quickly (exponentially) with increasing of n when $n>n_{\max}$.

The Maxwell equation for the electric field $F_x^{(2n+1)}$ the nonrelativistic harmonics is of the form

$$\Delta F_x^{(2n+1)}-\frac{1}{c^2}\frac{\partial^2F_x^{(2n+1)}}{\partial t^2}=\frac{4\pi}{c^2}(2n+1)\omega\sigma_{2n+1}F\times\sin(2n+1)\varphi. \tag{4}$$

Here $\varphi=\omega(t-x/c)$ is the phase of the laser field. Hence, the relative efficiency of harmonic generation is determined by the ratio

$$\eta^{(2n+1)}=\left\langle\frac{F_x^{(2n+1)}}{F\cos\varphi}\right\rangle^2=\left|\frac{4\pi(2n+1)\omega\sigma_{2n+1}}{4n(n+1)\omega^2+\omega_p^2}\right|^2. \tag{5}$$

It is seen that first the harmonic efficiency increases with n [because of the numerator of Eq. (5)], but then it decreases [because of the denominator of Eq. (5)], while σ_{2n+1} depends on n only in logarithm.

As an example, let us consider the multiple ionization of titanium foils by a superintense femtosecond laser pulse. We choose the value of $Z=12$. The ionization potential of this Ti atomic ion is $E_Z=10.71$ a.u. The barrier-suppression field

TABLE I. The efficiency of excitation of harmonics at the irradiation of Ti metal foil by a nonrelativistic laser pulse.

$2n+1$	1	3	5	7	9
$\eta^{(2n+1)}$	0.015	0.075	0.136	0.182	0.208
$2n+1$	11	13	15	17	19
$\eta^{(2n+1)}$	0.215	0.208	0.192	0.172	0.150

strength is $F_{BSI}=2.4$ a.u. Thus, we can use the nonrelativistic approach. The laser frequency is $\omega=1.5$ eV$=0.055$ a.u. (Ti:sapphire laser). The Keldysh parameter is $\gamma=0.106\ll 1$. The longitudinal and transverse velocities are derived according to Eq. (2): $V_{\|}=8.30$ a.u. and $V_{\perp}=0.51$ a.u. The quiver velocity is $V=F_{BSI}/\omega=43.4$ a.u.$\ll c$. Thus, the inequalities $V_{\perp}\ll V_{\|}\ll V$ are fulfilled that confirm the validity of Eqs. (3) and (5).

The number density of Ti metal is $n_i=4.64\times 10^{22}$ cm^{-3} $=0.0069$ a.u. Hence, the plasma frequency is $\omega_p=\sqrt{4\pi Z n_i}$ $=1.02$ a.u. It is seen that $\omega_p\gg\omega$, i.e., such an ionization takes place only in skin layer on the surface of a Ti metal foil [8]. The frequency of electron-ion collisions is $\nu_{ei}(V)$ $=0.0042$ a.u.$\ll\omega$. Here we used the value of the Coulomb logarithm: $\Lambda=10$. The harmonic conductivity is derived according to Eq. (3): $\sigma^{(2n+1)}=0.187-0.042\ln(2n+1)$ a.u. The maximum value of harmonic cutoff is $(2n+1)_{\max}=19$. In Table I, the relative efficiency of harmonics is presented as a function of its number $(2n+1)$. It is derived according to Eqs. (3) and (5).

Of course, the total efficiency of all harmonics should not exceed 1 according to the energy conservation law. In order to take this law correctly into account, we should write the rate equations for harmonics field and the initial laser field. However, Table I allows us to conclude about the high efficiency of harmonics generation even in the nonrelativistic case.

The goal of this Brief Report is to consider the relativistic quiver motion of an electron, which occurs at the irradiation by an intense laser pulse. In this case, odd harmonics are irradiated along the laser field polarization while even harmonics are irradiated along the direction of the laser light propagation. Even harmonics are produced by the magnetic field of the laser pulse which is an axial vector, while odd harmonics are produced by the electric field which is a polar vector. Therefore, even harmonics cannot be observed in the nonrelativistic limit where the magnetic force of the laser pulse can be neglected. Even harmonics were observed recently in strongly relativistic regime experimentally [9]. However, the comparison of theoretical and experimental results is hard, since in experiments harmonics can be excited also at the collisions of free electrons with the diffuse nonuniformity of the target surface instead of electron collisions with atomic ions.

In the nonrelativistic case, we cannot neglect the drift electron velocity, since this results in divergent expression for the harmonic conductivity. This is seen, in particular, from Eq. (3). Oppositely, in relativistic case such a neglection is possible, since an electron does not stop during its motion along the eightfold relativistic trajectory. Thus, the relativistic electron current density is given by the well known Pauli formula [10]:

$$d\mathbf{j}=-n_e\mathbf{V}_F n_i V_F\sigma dt. \tag{6}$$

Here $\sigma=4\pi Z^2\Lambda/(P_F V_F)^2$ is the Mott cross section for relativistic scattering of an electron on the multicharged atomic ion in the case of small scattering angles, $\mathbf{V}_F(t)$ is the quiver relativistic velocity, and $P_F(t)$ is the quiver relativistic momentum. The quantity $n_i V_F\sigma dt$ is the (dimensionless) number of collisions of an electron with multicharged atomic ions during the time interval dt. It is seen that the drift velocity cancels from Eq. (6), unlike the nonrelativistic limit. The electron spin is also insignificant in relativistic case due to the electron scattering on small angles. This simplifies the general expression for the Mott cross section. Besides this, the anisotropic drift velocity distribution of the ejected electrons is not needed for analysis, unlike the nonrelativistic case.

In order to simplify the problem, we consider first the ultrarelativistic limit $F/\omega c\gg 1$ (c is the light speed). Let us direct the electric field strength of the linearly polarized laser radiation along the axis Y, the magnetic field strength along the axis Z, and the direction of the laser pulse propagation along the axis X. Then the ultrarelativistic quiver velocities are given by simple expressions [10]:

$$V_{Fx}=c\frac{\cos 2\varphi}{2+\cos 2\varphi},\quad V_{Fy}=c\frac{2\sqrt{2}\cos\varphi}{2+\cos 2\varphi}. \tag{7}$$

Here $\varphi=\omega(t-x/c)$ is the phase of the laser field. The ultrarelativistic electron momentum is

$$P_F=\frac{F}{\omega}\sqrt{\frac{1}{8}\cos^2 2\varphi+\cos^2\varphi}. \tag{8}$$

Substituting Eqs. (7) and (8) into Eq. (6), one obtains the harmonic current

$$d\mathbf{j}=-A\frac{\mathbf{i}_x\cos 2\varphi+\mathbf{i}_y 2\sqrt{2}\cos\varphi}{[\cos^2\varphi+(1/8)\cos^2 2\varphi]^{3/2}}(2+\cos 2\varphi)Fd\varphi. \tag{9}$$

Here the following notation is introduced:

$$A=\frac{\pi n_e n_i Z^2\Lambda\omega}{F^3\sqrt{2}}.$$

Expanding Eq. (9) into Fourier series and integrating over field phase, one obtains the tensor of nonlinear harmonic conductivity:

$$\sigma_x^{(2n)}=-A\sum_{n=0}^{\infty}C_{2n}\sin(2n\varphi),$$

$$\sigma_y^{(2n+1)}=-A\sum_{n=0}^{\infty}C_{2n+1}\sin[(2n+1)\varphi]. \tag{10}$$

Here the universal Fourier coefficients are given by the definite integrals

TABLE II. Coefficients of harmonic conductivity, Eq. (10) in the ultrarelativistic case.

C_1	C_2	C_3	C_4	C_5	C_6	C_7
12.32	5.209	−2.495	−1.239	0.625	0.318	−0.167
C_8	C_9	C_{10}	C_{11}	C_{12}	C_{13}	C_{14}
−0.093	0.043	0.022	−0.011	−0.006	0.003	0.001

$$C_{2n}=\frac{1}{n\pi}\int_0^{\pi}\frac{(2+\cos 2\varphi)\cos(2n\varphi)\cos 2\varphi d\varphi}{[\cos^2\varphi+(1/8)\cos^2 2\varphi]^{3/2}} \quad (11)$$

and

$$C_{2n+1}=\frac{4\sqrt{2}}{(2n+1)\pi}\times\int_0^{\pi}\frac{(2+\cos 2\varphi)\cos[(2n+1)\varphi]\cos\varphi d\varphi}{[\cos^2\varphi+(1/8)\cos^2 2\varphi]^{3/2}}. \quad (12)$$

Results of numerical derivation of these coefficients are presented in Table II. It is seen that the harmonic conductivity is a tensor with odd components along the the laser polarization and with even components along the direction of propagation of laser pulse. If $F/\omega c \sim 1$, these coefficients are the functions of the dimensionless parameter $F/\omega c$.

Thus, we conclude that even harmonics are excited efficiently in the ultrarelativistic case. It is seen from Table II that the number of even and odd harmonics is of the order of 10. The absolute intensity of both even and odd harmonics is comparable with the nonrelativistic case (neglecting the logarithmic factor). This is seen from comparison of Eqs. (9) and (3). Of course, we should keep in mind that the electron number density n_e and charge multiplicity of atomic ions Z increase with the growing laser field that diminishes this difference in harmonic yields.

Analogous derivations in the general relativistic case allows us to obtain the Fourier coefficients $C_{2n+1}(a)$ and $C_{2n}(a)$ as functions of the dimensionless relativistic parameter $(0<a<1)$:

$$a=\left(1+2\frac{c^2\omega^2}{F^2}\right)^{-1/2}.$$

The above considered ultrarelativistic case corresponds to the limit $a\to 1$. The nonrelativistic case is $a\to 0$. They are determined by expressions:

$$C_{2n}(a)=\frac{a}{n\pi}\int_0^{\pi}\frac{(2+a^2\cos 2\varphi)\cos(2n\varphi)\cos 2\varphi d\varphi}{[\cos^2\varphi+(a^2/8)\cos^2 2\varphi]^{3/2}} \quad (13)$$

and

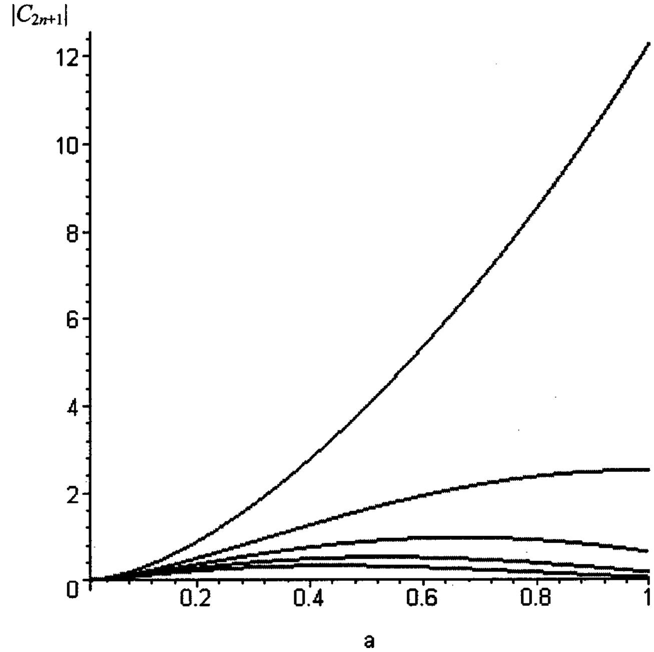

FIG. 1. Odd Fourier coefficients $|C_{2n+1}|$ as functions of the dimensionless field parameter $a=(1+2c^2\omega^2/F^2)^{-1/2}$ for the harmonic numbers $2n+1=1$, 3, 5, 7, and 9 (from above to below) derived according to Eq. (14).

$$C_{2n+1}(a)=\frac{4\sqrt{2}}{(2n+1)\pi}\times\int_0^{\pi}\frac{(2+a^2\cos 2\varphi)\cos[(2n+1)\varphi]\cos\varphi d\varphi}{[\cos^2\varphi+(a^2/8)\cos^2 2\varphi]^{3/2}}. \quad (14)$$

Absolute values of these coefficients are shown in Figs. 1 and 2, respectively. When $a=1$, these coefficients coincide with the values cited in Table II, as it should do. It is seen that the harmonic efficiency decreases with its number both for odd and even harmonics. Of course, the harmonic efficiency decreases also with the growing of the field strength F because of the factor $A\sim F^{-5}$ in the harmonic conductivity, Eq. (10).

Experimental harmonics generation was reported in Ref. [11] also for Ar clusters. It is demonstrated that a medium of intermediate-sized clusters of a few thousand atoms of inert gas is much better at generating the higher harmonics than a medium of isolated gas atoms of the same density. The enhancement factor for the 3rd–9th harmonics is about 5. Also, the dependence of the efficiency of generation of harmonics on the intensity of laser radiation is much more articulate for clusters than for isolated atoms. The highest harmonic number for clusters is higher than that for the isolated atoms. Our analytic approach is in qualitative agreement with the experimental data. However, exact comparison is impossible, since harmonics also can be generated at the electron reflection from the cluster surface. It allows us to derive both the harmonic conductivity and the harmonic fields generated by an intense laser pulse at the irradiation of solids and clusters. Of

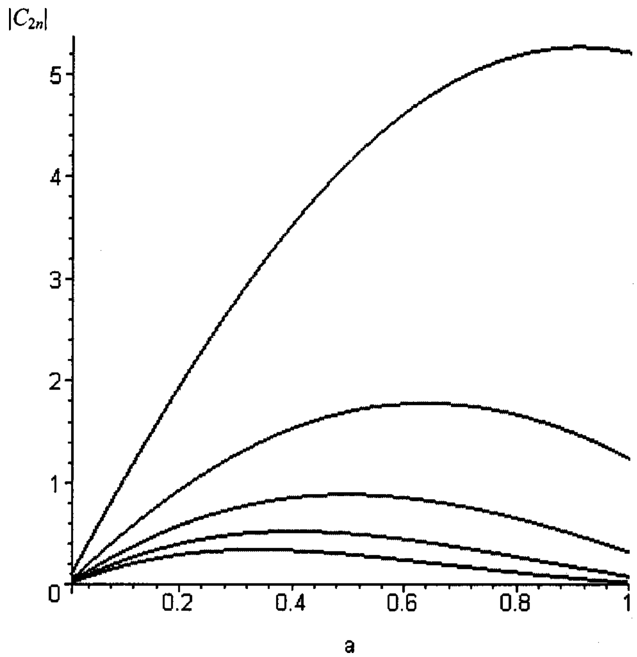

FIG. 2. Even Fourier coefficients $|C_{2n}|$ as functions of the dimensionless field parameter $a=(1+2c^2\omega^2/F^2)^{-1/2}$ for the harmonic numbers $2n=2$, 4, 6, 8, and 10 (from above to below) derived according to Eq. (13).

course, atomic clusters can be more preferable compared to solid foils due to relatively better penetration of laser radiation through a cluster beam. However, it is hard to compare theoretical predictions with the experimental data [9,11].

In conclusion, we should make the important comments with respect to the observation of even harmonics in experiments. Even harmonics produce the alternating electric currents which are directed along the laser propagation vector. However, these harmonics cannot be observed directly, since it follows from Maxwell equations that the corresponding transverse harmonic electromagnetic field vanishes in the wave region [12]. In order to observe even harmonics experimentally, we should take into account some additional fields in the laser plasma. First of all, this is the magnetic Weibel field [13]. This field is produced spontaneously by the plasma instability due to inhomogeneous velocity distribution of free electrons ejected as a result of field ionization. In the case of linear polarization, the Weibel field has the same polarization as the exciting laser field (the electric Weibel field is much less than the magnetic Weibel field). Electron current of even harmonics is rotated by this field with the Larmor frequency in the plane produced by the direction of laser polarization and direction of propagation of the laser pulse. In the ultrarelativistic case, the Larmor frequency is given by

$$\omega_L=\frac{B_{\mathrm{Weibel}}}{F}\,\omega\ll\frac{B_{\mathrm{Weibel}}}{c}.$$

The maximum Weibel magnetic field is [13] (though this estimate is valid for circularly polarized field; in the case of linearly polarized field, the Weibel field is weaker)

$$B_{\mathrm{Weibel}}^{\max}\sim\omega_p c.$$

Thus, the Larmor frequency ω_L is small compared to the plasma frequency ω_p. Rotation over the right angle makes even harmonics to be observable.

Of course, the Weibel field can be produced experimentally also by the spatial nonuniformity of the laser plasma at the irradiation of solid targets by intense laser pulses. Such a nonuniformity can produce also harmonics at the collisions of free electrons with diffuse surface of exploding target, instead of collisions with multicharged atomic ions.

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