On harmonic generation in a photoionized gas

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The laws characterizing the radiation of high harmonics due to the coherent bremsstrahlung effect are indicated in the limit of high intensity of the laser pump photoionizing a gas in regime of suppression of the ionization barrier. It is shown that the intensity of the harmonics is determined by the quantum properties of the electron distribution in an atom before it is ionized. © 1999 American Institute of Physics.

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The generation of high harmonics of optical radiation has been attracting a great deal of attention in the last few years, both as a general problem of nonlinear optics and as a problem whose solution opens up prospects for many applications.1 The study of the generation of very high harmonics started with plasma as the nonlinear medium.2,3 In the last few years harmonic generation has been studied in neutral gases, see, for example, Refs. 4 and 5. However, as the lasers employed are improved and the energy flux density of the laser radiation increases, neutral gases become ionized.6–8 Thus attention once again turns to plasma as a nonlinear medium for generating high harmonics of laser radiation. There is a tendency in experiments to increase the intensity of the laser radiation when photoionization is the process used to ionize the gas.

In this letter we present the basic laws characterizing the efficiency of harmonic generation in a photoionized plasma, which are determined by the quantum distribution of electrons in an atom. The exposition is mainly of a qualitative nature and is based on an associative generalization of the existing results of the theory of harmonic generation in a prepared classical plasma. In what follows we shall concentrate on the limit of a pump field which is strong enough that photoionization occurs under conditions of suppression of the ionization barrier (SIB).9,10 In so doing, first, we shall assume the ionization potential $I_i$ of the atoms to be small compared with the energy $(1/2)mV_E^2$ of the electron oscillations in the pump electric field $E(t)=E \cos \omega t$, where $V_E = |eE/m\omega|$ is the amplitude of the oscillatory electron velocity and $e$ and $m$ are the electron charge and mass, respectively. This means that the Keldysh parameter of the theory of tunneling ionization is small: $\gamma = (2I_i/mV_E^2)^{1/2} \ll 1$ (Ref. 11). Second, we shall assume the parameter $\beta = (1/16Z)(I_i/I_H)^2(E_{at}/E)$ to be small. Here $Z$ is the nuclear charge of the atom, $I_H$ is the ionization potential of the hydrogen atom, and $E_{at} = 5.13 \times 10^9$ V/cm is the atomic unit of electric field intensity. In the case of interest here, where $\beta < 1$, according to Refs. 9 and 10 an electron is ionized from an atom without tunneling. In other words, photo-
ionization in the SIB model is determined by the free, virtually unimpeded, escape of an electron from an atom. For a spatially uniform distribution of atoms in a plasma this makes it possible to express the electron momentum distribution function in the form

\[ f(p, t) = F(p^2/m_u(t)). \]

Here \( u(t) \) is the time-dependent electron velocity in the electric field of the laser pump ionizing the atoms. This form of the electron distribution is similar to that ordinarily arising in the theory of harmonic generation in a plasma in the first collisionless approximation. However, in the present case, in a coordinate system oscillating together with the electron it is the electron momentum distribution inside an atom before the radiation acts on the plasma. For a pure quantum state, to within a normalization factor,

\[ F \sim p^4 \]

where \( a_q(p) \) is the electron wave function in the momentum representation and the summation extends over all quantum numbers corresponding to the electron distribution inside the atom. Such a distribution is physically obvious. It can also be obtained directly by using the equation for the density matrix in the Wigner representation.

The similarity so arising and, at the same time, the obvious difference between the cases of a preprepared plasma and a photoionized plasma in the SIB regime makes it possible to establish the following general law characterizing the generation efficiency

\[ h(l) = \frac{\eta^{(2l+1)}}{q} \frac{\eta^{(2l+1)}}{q} \left( \frac{V}{V_E} \right)^2, \]

where \( q^{(2l+1)} \) and \( q \) are, respectively, the energy flux density of the \((2l+1)\)-st harmonic and the pump. In Eq. (1) the function \( \nu(V) \) is the electron-velocity-dependent electron–ion collision frequency, which corresponds to the cause responsible for the harmonic generation — bremsstrahlung due to the oscillatory motion of the electrons in the Coulomb field of the ions, and \( h(l) \) is a function of the number of the harmonic and in the case of a plane-wave geometry for both the pump and generated harmonic fields is given by:

\[ h(l) = \left[ l(l+1/2)^2 (l+1) \right]^{-1}. \]

Finally, the function \( S \), which is determined by the electron distribution inside the atom, depends in the high-field regime \( V_E \gg V \) on the argument \( \left( [2l+1]V/V_E \right) \), where \( V \) is the characteristic electron velocity for some electron velocity distribution that does not take braking effects into account. This is similar to the results obtained in Refs. 13–15, and it can also be established directly for the photoelectron distribution in the SIB regime (see below).

In order to use our general expression (1) to examine the quantum properties of the efficiency of harmonic generation in a photoionized plasma we shall first use the expression for the electron–ion collision frequency corresponding to the Fokker–Planck–Landau collision integral \( \nu(V) = 4 \pi e^2 \epsilon^2 n_i \Lambda / m^2 V^3 \), where \( e = Z_i |e| \) is the ion charge, \( n_i \) is the ion number density, and \( \Lambda \) is the Coulomb logarithm. For ionization in the SIB