Excitation of ultrashort bursts of harmonics of the radiation during ionization of a gas by an intense light pulse

E. V. Vanin, A. V. Kim, and A. M. Sergeev
Institute of Applied Physics, Russian Academy of Sciences, 603600 Nizhniĭ Novgorod, Russia

M. C. Downer
The University of Texas at Austin, Austin, Texas 78712, USA

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A self-consistent model for a nonlinear interaction of an intense femtosecond-range light pulse with a gas of quantum particles (atoms) is discussed. During fast multiphoton ionization of the gas, the spectral lines of harmonics of the radiation undergo a substantial broadening and a blue shift. A pulse of the third harmonic with a duration equal to less than three periods of the exciting optical field can be generated at an efficiency $\sim 10\%$ in terms of the field amplitude.

Excitation of harmonics of optical radiation at atoms is an interesting aspect of the interaction of intense laser pulses with matter and the subject of active research.[1-3] The spectra of the radiated harmonics contain information on the nonlinear response of an individual atom and also on collective features of the excitation of harmonics along the interaction path. At ultrahigh intensities of the laser fields, $I > 10^{15}$ W/cm^2, the generation of harmonics occurs in the course of fast bound–free transitions of optical electrons of the atoms, which substantially alter the dielectric properties of the gaseous medium. On the one hand, the strong dispersion of the plasma which is produced degrades the conditions for phase matching and group matching in the wave packets at the fundamental frequency and the higher harmonics, with the result that the excitation of these harmonics reaches saturation.[1,4] On the other hand, the ionization of the medium affects the properties of the light at the fundamental frequency, causing (in addition to an absorption of energy) an adiabatic frequency shift of this light in the blue direction.[5,6] This shift introduces an additional mismatch in the space–time dynamics of the harmonic generation. These collective wave effects will obviously be an obstacle in efforts to raise the radiation power at high frequencies by raising the gas density (or by increasing the length of the interaction path). One is led to ask whether it is possible, in general, to achieve anything approaching a significant conversion of the fundamental frequency into harmonics in the case of a fast multiphoton ionization of atoms. In the example discussed below, which is a self-consistent model of a nonlinear interaction of an ultraintense laser pulse with a gas of quantum particles, we show that it is possible to generate a burst of harmonics with a relative power $\sim 1\%$ and a duration amounting to only a few periods of the exciting optical field.

The theoretical model of the interaction includes a 1D wave equation (along the z axis) for the linearly polarized electric field of the pulse, $\mathbf{E} = E\mathbf{x}^0$, and a 1D Schrödinger equation (in this case along the x axis) for an electron which is subjected to the "intraatomic" potential $V(x)$ and the optical radiation in the dipole approximation:

$$\frac{\partial^2 E}{\partial t^2} - c^2 \frac{\partial^2 E}{\partial z^2} = -4\pi \frac{\partial^2 P}{\partial t^2}, \tag{1}$$

$$i\hbar \frac{\partial \Psi}{\partial t} = -\frac{\hbar^2}{2m} \frac{\partial^2 \Psi}{\partial x^2} + V(x)\Psi + exE\Psi. \tag{2}$$

The polarization P of the medium, which consists of model one-electron atoms with a number density N, is

$$P(z,t) = -eN \int_{-\infty}^{+\infty} x|\Psi|^2 \, dx. \tag{3}$$

The nonlinearity of the medium stems from the interaction with the external field, $\sim E\Psi$; the specific manifestations of this nonlinearity are governed by the potential $V(x)$.

It is convenient to write the source on the right side of the wave equation in the form

$$\frac{\partial^2 P}{\partial t^2} = \frac{e^2 NE}{m} + \frac{eN}{m} \int |\Psi|^2 \frac{\partial V}{\partial x} \, dx. \tag{4}$$

The first term describes the response of the free electrons in the absence of the intraatomic confining potential. The second term is proportional to the number of atoms which are not ionized. The decrease in this term in the course of the interaction tells us the rate of gas ionization.

If the gas is not too dense ($\omega_p^2 = 4\pi Ne^2/m \ll \omega^2$, where ω is the frequency of the laser light), and if there are no strong resonant effects in the interaction of the optical field with the atom, we can ignore the backscattering of a short pulse and restrict the discussion to the copropagating waves. Mathematically, we do this by transforming to a frame of reference of the local time, $\tau = t - z/c$, $z = z$, and by ignoring the second derivative with respect to what is now the "slow" coordinate z in the second equation: $\partial^2/\partial z^2 \ll (1/c)\partial^2/\partial z \partial \tau$. Imposing the standard normalization of variables, in terms of characteristic atomic values,

$$\tau_0 = \frac{\hbar^3}{me^4}, \quad x_0 = \frac{\hbar^2}{me^2}, \quad E_0 = \frac{m^2 e^5}{\hbar^4}, \quad V_0 = \frac{me^4}{\hbar^2}, \quad \omega_0 = \frac{me^4}{\hbar^3}, \quad z_0 = \frac{cme^4}{\omega_p^2 \hbar^3}, \tag{5}$$

we find our basic system of dimensionless equations:

$$\frac{\partial^2 E}{\partial z \partial \tau} = -\frac{1}{2}(E + R), \tag{6}$$

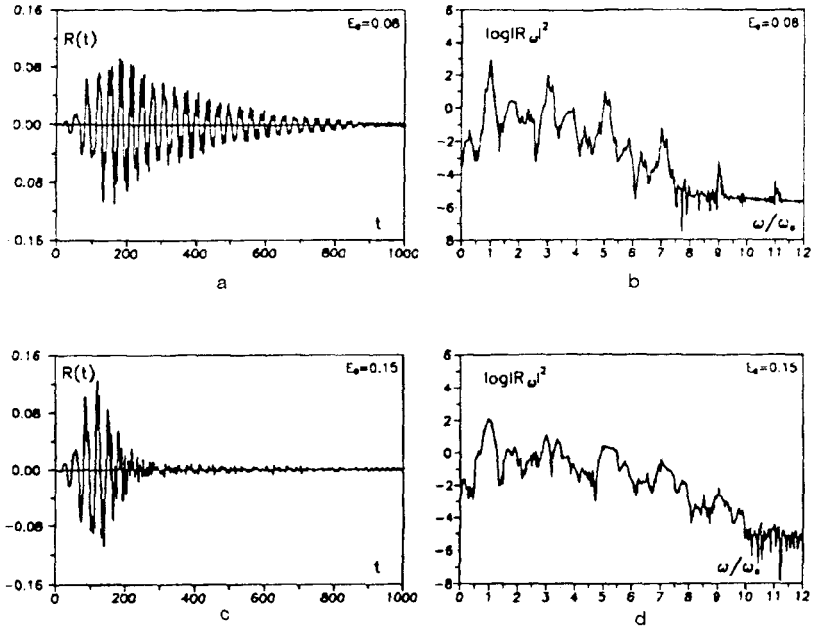

FIG. 1. a, c—Responses of a model atom to the field of a laser pulse with the profile shown by the dashed curve in Fig. 2a; b, d—spectra of these responses.

$$i\frac{\partial\Psi}{\partial\tau}=-\frac{1}{2}\frac{\partial^2\Psi}{\partial x^2}+V\Psi+xE\Psi, \tag{7}$$

$$R=\int_{-\infty}^{+\infty}|\Psi|^2\frac{\partial V}{\partial x}\,dx. \tag{8}$$

As the intraatomic potential we use $V=-(1+x^2)^{-1/2}$, which has recently been discussed as a standard potential for modeling the dynamics of an atom in strong fields.[7,8] The energy of the ground state in this potential is $u_0=-0.67$ ($\simeq 18$ eV in dimensional units). The frequency of the laser pulse for the results presented below is $\omega=0.2$, so direct detachment of an electron from the atom occurs upon the absorption of at least four photons from the optical field. We are interested in high intensities of the laser pulse ($E\geqslant 0.1$), at which the atom is ionized over a few oscillation periods of the field. In this case, the particular shape of the pulse over times greater than the lifetime of the atom is unimportant from the standpoint of harmonic excitation. This circumstance simplifies the modeling substantially, allowing us to work with short field pulses, a few tens of periods long, and to focus the computation effort on the detailed time resolution of the process.

Figure 1, a and c, shows the responses (R) of an individual atom in the fields of laser pulses with amplitudes $E=0.08$ and $E=0.15$ and with the profile shown by the dashed curve in Fig. 2a. Figure 1, b and d, shows corresponding polarization spectra.

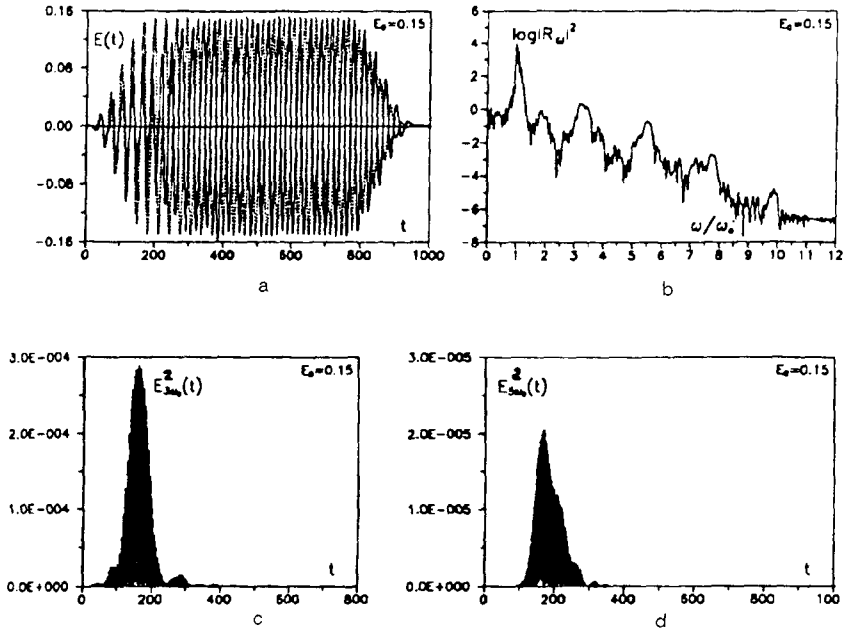

FIG. 2. a (solid curve)—Field of the laser pulse at the exit from the layer; b—the spectrum of this pulse; c, d—corresponding pulses of the intensities of the third and fifth harmonics, respectively.

The intermediate critical range of values of E ($\sim$0.11–0.13) is characterized by a rapid enrichment of the harmonic spectrum and a broadening of the lines. These effects are clearly due to the decrease in the time over which they are excited if the atom decays rapidly. As the leading edge of the radiation becomes steeper, the time interval corresponding to passage through the field interval, which is critical for the atom, obviously becomes shorter. We run into an unexpected conclusion: The higher the amplitude of a laser pulse of a given duration, the smaller the number of harmonics distinguishable in the emission spectrum of an individual atom. The higher harmonics, which are effectively excited over a time interval $\leqslant 2\pi/\omega$, overlap, and the continuum boundary shifts toward lower indices with increasing radiation intensity.

To reach the maximum intensity of harmonics of the electric field, we must optimize the length of the interaction region [or optimize the total number of atoms along the path, as follows from the last scaling relation in (5)]. For simplicity we assume that the interaction region is filled with the gas in a uniform manner. Let us look at various factors which would limit the growth of field harmonics: a phase mismatch, a group mismatch, an ionization-related frequency shift, and a depletion of the pump at the fundamental frequency due to ionization. If we assume that the plasma which is produced is the primary source of wave dispersion, we can write the following dimensional estimates for the limiting distances (z^*) over which the amplitude of the nth harmonic will grow:

1) In the case of a phase mismatch,

$$z_{\text{ph}}^{*} \simeq \frac{\lambda}{n} \frac{\omega^2}{\omega_p^2} \frac{N}{N_{\text{int}}},$$

where λ is the fundamental wavelength, and $N_{\text{int}} \sim (0.2\text{–}0.5)N$ is the number density of the plasma in the region in which harmonic generation is most efficient.

2) In the case of a group mismatch,

$$z_g^{*} \simeq z_{\text{ph}}^{*} \frac{\tau_{\text{ion}}}{\tau_n} > z_{\text{ph}}^{*},$$

where τ_{ion} is the time scale for ionization of the atom, and τ_n is the period of the harmonic.

3) In the case of an ionization-induced blue shift of the radiation frequency,[6]

$$z_{\text{sh}}^{*} \simeq \frac{\lambda}{n} \frac{\omega^2}{\omega_p^2}.$$

4) In the case of a depletion of the fundamental harmonic due to ionization,

$$z_{\text{abs}}^{*} = 2c\tau_{\text{ion}} \frac{U_{\sim}}{U_i} \frac{\omega^2}{\omega_p^2},$$

where $U_{\sim}/U_i$ is the ratio of the average oscillatory energy of an electron to the ionization potential of the ground state.

Since the ionization of the atom occurs over a small number of periods of the optical field in the intensity range of interest here, and since the oscillatory energy is not very large ($U_{\sim} \sim 0.1 U_i$), the depletion and blue shift of the pump are the factors of primary importance in limiting the growth of the most intense harmonics ($n=3$ and 5). At larger values of n the blue shift becomes predominant. We should stress that this shift is always more important than the phase mismatch.

Figure 2, a and b, shows a pulse of the optical field (the solid curve) and its spectrum after passage through a layer of gas of thickness $\lambda\omega^2/\omega_p^2$ ($L=0.4\pi$ in dimensionless units). In this case the amplitude of the third harmonic has reached saturation. The source depletion effect and the effect of the frequency increase are obviously predominant in the deformation of the pulse. Since the blue shift is multiplied with increasing n, it becomes possible to achieve a substantial restructuring of the harmonic frequencies. For example, the center of the fifth-harmonic line is shifted 0.5 ω_0, while the ninth harmonic has essentially taken the place of the tenth. Figure 2, c and d, shows corresponding pulses of excited harmonics of the electric field with $n=3$ and $n=5$. The maximum amplitude of the third-harmonic packet is $E_3 \simeq 1.75 \times 10^{-2}$, which corresponds to $> 10\%$ of the exciting field. The length of the pulses of harmonics at the exit from the layer is no more than two or three periods of the carrier frequency; i.e., these pulses are much shorter than the fundamental pulse. A further increase in the length of the interaction region results in an increase in the duration of the harmonic pulses without any significant change in their intensity.

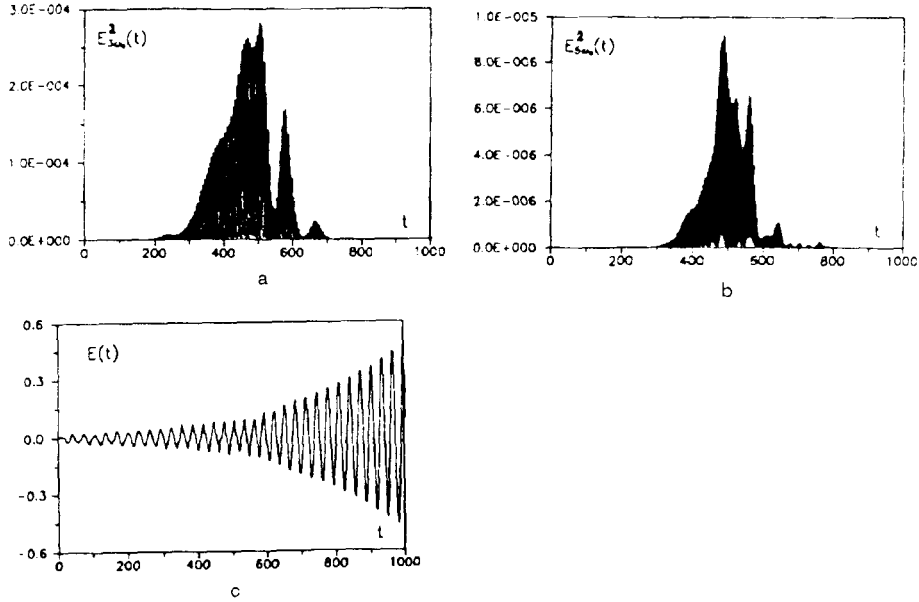

FIG. 3. Bursts of generation of the third (a) and fifth (b) harmonics of the laser field with a gently rising leading edge (c) at the exit from the layer.

In the femtosecond pulses which would actually be used in a laser experiment, the profile of the applied optical field would be smoother than in the case described above. Nevertheless, the excitation of ultrashort bursts of harmonics, with a length equal to a few periods of the carrier frequency, would still occur. It results from a rapid increase in the probability for the ionization of an atom when the field amplitude reaches critical values $E \simeq 0.1E_a$. This assertion is supported by the pulses of harmonics shown in Fig. 3, which are achieved during passage through a gas layer of thickness $\lambda\omega^2/\omega_p^2$ of a laser pulse whose amplitude at the leading edge rises from $E=0.05$ to $E=0.5$ over 25 field oscillation periods.

In summary, and in contrast with the prevailing opinion that ionization of the gas is a hindrance in the excitation of harmonics, we can conclude from the results presented here that it is precisely because of the rapid decay of the atom in an intense optical field that a mechanism operates to generate intense bursts of harmonics of the radiation. This mechanism might be utilized to develop tunable sources of ultrashort pulses in the UV frequency range.

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