Possibility of sharp increase in the frequency of the radiation of ionizing laser pulse in gas

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It is shown that the mechanism of tunneling ionization of the atoms of a gas by the field of an electromagnetic wave can be used for the effective control of the spectrum of high-intensity laser radiation.

Success in the generation and amplification of subpicosecond laser pulses[1] has opened wide possibilities in the experimental investigation of the interaction of light with matter at field intensities reaching those inside the atom.[2] In such fields the probability of subbarrier tunneling of the electrons from the outer atomic subshells increases dramatically, and field ionization, which is accompanied by the interaction of the wave with the forming plasma, becomes the dominant nonlinearity mechanism in the propagation of the laser pulse in the gas.

As is well known,[3–7] the frequency of the ionizing radiation shifts as a result of variation of the refractive index of the forming plasma. Here the final picture of transformation of the spectrum depends not only on the time scale of the ionization of the gas at the given amplitude of the electromagnetic field, but also on the magnitude of the energy loss of the laser pulse due to the formation of the plasma. This loss in the range of the so-called superstrong fields (with energy of oscillation of the free electrons $\omega_{\sim} \gg U$, where U is the ionization potential of the atom) are determined in the first place not by the collisions of the electrons and not by loss of energy to ionization of the atoms, but by the loss due to the transfer of kinetic energy to the electrons which are created. In addition to the variable component of the velocity, which adiabatically follows the amplitude of the wave, the electrons also acquire a constant component which depends on the phase of the field at the instant of their generation and undergoes no further variation after the passage of the pulse. Tunneling ionization is the only mechanism which allows a minimization of losses (in contrast, e.g., with ionization by electron impact[5]) and thereby provides the fundamental possibility of covering entire frequency ranges as a result of the continuous transformation of the laser radiation spectrum.

To analyze this effect, we will make use of a one-dimensional interaction model based on the equation for the electric field $E(x,t)$ of the wave

$$E_{tt} - c^2 E_{xx} + \frac{4\pi e^2}{m} NE = 0 , \tag{1}$$

and the following expression for the rate of increase of the electron density $N(x,t)$ (Ref. 3)

$$N_t = W = \gamma N_m \exp(-E_a/|E|) \quad (\omega_{\sim} \gg U \gg \hbar\omega), \tag{2}$$

in which the density of the neutral gas N_m, the characteristic intraatomic field $E_a = 4(\sqrt{2m}\cdot U^{3/2})/3\hbar e$, and the maximum ionization probability $\gamma \approx U/\hbar$ are assumed to be given.[1)]

As can be seen from the structure of the nonlinearity in Eqs. (1) and (2), upon ionization of the medium in a locally quasiharmonic field with linear polarization $E = A\cos\varphi$ ($\varphi_t = \omega \gg \tau^{-1}$, $-\varphi_x = k \gg l^{-1}$, τ and l are the temporal and spatial scales of the envelope), the electron density, along with the smoothly growing component, contains oscillating even harmonics $N + N_0 + \frac{1}{2}(N_2 e^{2i\varphi} + \text{c.c}) + ...$ Closure of the equations through the second harmonic of the electron density N_2 (in the quasiharmonic approximation under consideration $N_2/N_0 \sim 1/\omega\tau \ll 1$) leads to the following system:

$$(A^2/8\pi)_t + (VA^2/8\pi)_x = -F, \tag{3}$$

$$(\omega^2)_t + V(\omega^2)_x = \frac{4\pi e^2}{m}(N_0)_t, \tag{4}$$

$$(N_0)_t = \langle W \rangle = \frac{2}{\pi}\gamma N_m Ki_1(E_a/A), \tag{5}$$

$$F = \langle W \sin^2\varphi \rangle \frac{e^2A^2}{2m\omega^2} = \gamma\omega_{pm}^2 A^2[Ki_1(E_a/A) - Ki_3(E_a/A)]/4\pi^2\omega^2 . \tag{6}$$

Here $V = c(1 - 4\pi e^2 N_0/m\omega^2)^{1/2}$ is the group velocity of the wave, $\omega_{pm}^2 = 4\pi e^2 N_m/m$, the angular brackets denote averaging over one wave period, and $Ki_{1,3}$ are multiple integrals of the modified Bessel function K_0.[9]

The intensity-transport Equation (3), with right-hand side given by Eq. (6), differs substantially from the equation which is obtained in the geometric-optics approximation[5] since it takes into account the existence of fast oscillations of the electron density (N_2) which are caused by the inhomogeneity in the generation of the secondary electrons over one wave period. The right-hand side $-F$, which determines the rate of dissipation of wave energy, is greatly decreased in comparison with the geometric optics value $-\langle W \rangle e^2 A^2/4m\omega^2$ if the ionization takes place preferentially in the phase of maximum of the field ($\cos\varphi = \pm 1$), where the constant component of the velocity of the newly created electrons is equal to zero.[2)] Precisely such a situation is realized at $A/E_a \ll 1$ for the dependence $W(|E|)$ which we considered. The right sides of Eqs. (5) and (3) transform in this case respectively to the forms $\gamma B N_m$ and $-\gamma B\omega_{pm}^2 A^2/8\pi\omega^2 E_a$, where $B = \sqrt{2A/\pi E_a}\exp(-E_a/A)$. Let us consider two instances of the solution of system (3)–(6) in this case.

If the ionization is "turned on" simultaneously during the entire length of a long, homogeneous pulse, the frequency of the wave will increase in accordance with the law

$$\omega = \omega_0[1 + 3\gamma t(\omega_{pm}^2/\omega_0^2)\sqrt{A_0/2\pi E_a}\exp(-E_a/A_0)]^{1/3}, \tag{7}$$

and the amplitude will vary only slightly:

$$A = A_0/[1 + (A_0/E_a)\ln(\omega/\omega_0)]$$

(ω_0 and A_0 are the values of the frequency and the amplitude at the initial time $(t=0)$). For the characteristic frequency doubling time we can write the expression

$$\tau_1 \approx 6\gamma^{-1}\sqrt{E_a/A_0}(\omega_0^2/\omega_{pm}^2)\exp(E_a/A_0).$$

If a short rectangular pulse (with a duration $\tau_u \ll \tau_1$) enters the medium and its parameters vary slowly (over the time scale τ_u) as a consequence of ionization, we obtain a second solution

$$\omega = \omega_0(A_0^2/E_a^2)\ln^2(\xi \ln^{-7/2}\xi), \tag{8}$$

where $\xi = \gamma t\omega_{pm}^2 E_a^4/\sqrt{8\pi}\omega_0^2 A_0^4 \gg 1$. The corresponding frequency doubling time is

$$\tau_2 \approx 17\gamma^{-1}\sqrt{A_0/E_a}(\omega_0^2/\omega_{pm}^2)\exp(\sqrt{2}E_a/A_0).$$

The coupling of amplitude with frequency (8) is determined here not by energy transfer from the wave to the electrons (as in the previous case), but by pulse broadening in the forming plasma as a result of the difference in the group velocities of the wave between the leading edge and the trailing edge of the pulse: the spatial extent of the pulse is $L = L_0\omega/\omega_0$. The total pulse energy $\varepsilon = LA^2/8\pi$ is nearly constant. Its relative decrease over any path $(\Delta\varepsilon/\varepsilon)_{\max}$ is approximately $2A_0/E_a$. Another important property of the transformation regime under consideration is the narrowing of the spectral line of the radiation as the frequency of the field in the rectangular pulse increases: the linewidth is $\Delta\omega \sim \omega^{-1}$.

In conclusion let us estimate the effect we are considering here for two cases which may be of considerable practical interest at present and which conform to the idealized approach used above: 1) a CO_2 laser pulse with duration $\tau_u < 1$ psec, energy flux density $S = 4\times10^{14}$ W/cm^2 in an atomic gas with $U \approx 15$ eV at a pressure $p = 300$ Torr; 2) a KrF laser pulse with $\tau_u < 0.1$ psec, $S = 3\times10^{15}$ W/cm^2 with $U \approx 25$ eV (He), $p = 30$ atm. In the first case we have $\tau_1 = 2.5$ psec, $\tau_2 \approx 25$ psec; in the second, $\tau_1 \approx 5$ psec, $\tau_2 \approx 25$ psec. This means that in both cases the pulses double their frequency after transversing a distance $c\tau_2 \approx 0.7$ cm. The possibility of a significant increase of frequency over a short path with simultaneous compression of the spectrum of the radiation with a minimal energy loss to plasma formation gives one good reason to consider the proposed transformation mechanism as very promising.

[1)] Note that Eq. (1) is valid for arbitrary rate of growth of the charged particle density; however, it is important that the velocity distribution of the free electrons at the time of formation be isotropic.

[2)] In the limiting case of a delta-function dependence, $W \sim \delta(|E| - A)$, the considered dissipation mechanism is absent ($F = 0$) and the calculation should take into account the small losses due to ionization of the atoms, the collisions of the electrons with each other, and their acceleration due to the average ponderomotive force, all of which were ignored here.

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