Propagation of an intense ultrashort laser pulse under conditions of multiple nonlinear ionization

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A theory is presented of nonlinear propagation of an intense ultrashort laser pulse in a gas, which includes multiple ionization of the atoms by the optical field of the pulse itself, collisional ionization of heavy particles, and radiation absorption brought about by the ionization. A numerical simulation of the time-dependent, spatially two-dimensional problem for neon and lithium vapor is carried out. The physical conditions for the formation of an extended active medium of an x-ray laser (OFI x-ray laser) are investigated. The effect of small-scale ionization phases defocusing of laser radiation in a medium with an ionization-induced stepped profile of the electron number density is theoretically described. © *1995 American Institute of Physics.*

1. INTRODUCTION

In recent years, significant attention has been given to the study of the nonlinear propagation of intense ultra-short laser pulses in matter. Interest in questions of this type has been motivated both by the fundamental nature of the problem and by interesting applications which have arisen. For example, Refs. 1 and 2 examined self-channeling of ultrashort pulses in plasma. References 3 and 4 proposed the principle of a traveling-wave x-ray laser based on ionization and excitation of the medium by an ultrashort pulse of long-wavelength radiation [optical field ionization (OFI) x-ray laser]. There are also other interesting applications.[5]

Previously, the main attention was given to nonlinear propagation of ultrashort pulses in various media in the absence of breakdown[6] or in completely ionized plasma.[1,2,7–10] On the other hand, elementary processes of nonlinear ionization of atoms in strong optical fields were considered in detail in Ref. 11. References 12 and 14 considered an experiment and qualitative estimates of ionization defocusing of laser pulses in gases. However, the theoretical models describing the propagation of intense ultrashort laser pulses in matter with allowance for ionization of the medium in the strong optical field of the pulse itself are completely inadequate. We may mention here Ref. 15. It considered focusing of a short laser pulse with peak "vacuum" intensity $I \simeq 10^{16}$ W/cm^2 in argon under conditions in which the radiation was caused to converge on the focal spot directly through the gas. As a result of ionization defocusing, at the pulse transportation stage, the peak intensity of the radiation is decreased and the gas ionizes insignificnatly (the degree of ionization of the atoms is at most two or three). This case relates to the propagation of a pulse in a moderately ionized gas. For this reason, Ref. 15 did not consider the behavior of the process of nonlinear propagation of laser pulses when the medium's multiply ionized. The medium can become highly ionized when the radiation propagates to the focal spot in vacumm. In this way, intensities up to $I \simeq 10^{18}$ W/cm^2 (Ref. 2) were obtained in experiments on relativistic nonlinear self-channeling in gases.

In the present paper we numerically model the nonlinear propagation of an intense ultrashort laser pulse in a gas under when the atoms of the gas are multiply ionized by the optical field of the pulse itself. We consider the cases of neon and lithium vapor. We analyze some questions associated with the problem of an x-ray laser (OFI x-ray laser). We examine the physical situation arising with increase of the gas density.

2. THEORETICAL MODEL

We describe the propagation of a short intense laser pulse in an ionizing gas by a system of equations including the nonlinear Schrödinger (NLS) equation for the complex amplitude of the field and kinetic equations for the relative concentrations of the ions in the ionic component of the nascent plasma:

$$(v_g^{-1}\partial_t+\partial_z)a+\frac{i}{2k}\left[\Delta_\perp+k_p^2\left(1-\frac{\langle z\rangle}{Z}\right)\right]a+\frac{\mu^-}{2}a=0, \tag{1}$$

$$\partial_t\alpha=B\alpha, \quad \alpha=(\alpha_1,\alpha_2,\dots,\alpha_{Z+1})^T, \tag{2}$$

$$\langle z\rangle=\sum_{s=2}^{Z+1}(s-1)\alpha_s, \quad \sum_{s=1}^{Z+1}\alpha_s=1, \tag{3}$$

$$\mu^-=I^{-1}\partial_t(\rho\varepsilon_{\text{ion}}), \quad I=|a|^2, \tag{4}$$

$$\varepsilon_{\text{ion}}=\frac{k_6}{M}\sum_{s=2}^{Z+1}\sum_{n=1}^{s-1}J_n\alpha_s. \tag{5}$$

In Eq. (1) $\Delta_\perp=r^{-1}\partial_r+\partial_{rr}^2$ is the transverse Laplacian, $a(r,z,t)$ is the complex amplitude of the vector-potential of the electromagnetic field, v_g is the group velocity,

$k^2=k_0^2-k_p^2$, where $k_0=\omega/c$ is the wave number, $k_p=\omega_p/c$, $\omega_p=(4\pi e^2 N_0 Z/m_e)^{1/2}$ is the electron plasma frequency, N_0 is the number density of the heavy particles of the gas, Z is the nuclear charge of the heavy particles.

The mean charge $\langle z\rangle$ is given by (3), where the quantities α_s are the relative number densities of the ions of charge state s in the ionic composition of the plasma, which satisfy the system of kinetic equations (2).

The matrix of ionization rates has the form

$$B=\begin{pmatrix} -\gamma_1 & 0 & 0 & \cdots & 0 & 0 & 0 \\ \gamma_1 & -\gamma_2 & 0 & \cdots & 0 & 0 & 0 \\ \cdots & \cdots & \cdots & \cdots & \cdots & \cdots & \cdots \\ 0 & 0 & 0 & \cdots & \gamma_{Z-1} & -\gamma_Z & 0 \\ 0 & 0 & 0 & \cdots & 0 & \gamma_Z & 0 \end{pmatrix}. \tag{6}$$

The ionization rate $\gamma_s=\gamma_s^r+\gamma_s^e$, $s=1,\ldots,Z$ is the sum of the photoionization and collisional ionization rates. Since we are considering propagation of ultrashort laser pulses, recombination is not taken into consideration.

The coefficient of the absorption due to ionization, μ^-, is given by Eq. (4). The ionization energy per unit mass of the gas is given by Eq. (5), where k_B is the Boltzmann constant, $\rho=MN_0$ is the density of the gas, M is the mass of an ion, J_s is the ionization potential of the ions of multiplicity s, and I is the intensity of the radiation.

To complete the mathematical statement of the problem, system of equations (1)–(5) should be considered together with the initial and boundary conditions. For the case of propagation of a pulse for $t>0$ in an unbounded medium, we will use the natural initial

$$a(r,z,t=0)=a_0(r,z), \quad \alpha(r,z,t=0)=\alpha_0(r,z) \tag{7}$$

and boundary conditions

$$a(r=\infty,z,t)=0, \quad \partial_r a(r=0,z,t)=0. \tag{8}$$

The NLS equation (1) is valid when the envelope of the laser pulse varies slowly over distances on the order of a wavelength in the direction of propagation and over times on the order of a period of the high-frequency oscillations of the field: $\partial_z a,\ c^{-1}\partial_t a\ll ka$.

The problem as stated in Eqs. (1)–(8) allows for the following four effects: diffraction; refraction by inhomogeneities of the refractive index of the medium due to variations in the number density of the free electrons resulting from ionization of the atoms of the gas; field and collisional multiple ionization of the atoms; and absorption due to ionization.

The statement of the problem in Eqs. (1)–(8) agrees completely with Ref. 15. We will now specify the models of the different types of ionization.

Model of threshold ionization. There exist several models of nonlinear ionization of atoms by external radiation. One peculiarity of the problem under consideration is the necessity of describing the ionization over a wide range of values of the optical field for ions of different multiplicity. From this point of view, the model of threshold ionization in Ref. 16 is acceptable. It is distinguished by its simplicity and universality and provides a satisfactory description of the experimental data. The basis formulas of the model are

$$\gamma_s^r=\gamma_0\theta(I-I_{\text{thr}}^{(s)}), \quad I_{\text{thr}}^{(s)}=4\cdot 10^9\frac{J_s^4}{s^2}, \quad s=1,2,\ldots,Z, \tag{9}$$

where $\theta(x)$ is the unit step function, J_s are the ionization potentials, γ_0 is a general constant for all orders of ionization ($\gamma_0\ll t_0^{-1}$, where t_0 is the duration of the laser pulse. This model of threshold ionization assumes "instantaneous" ionization when the threshold intensity is exceeded.

Model of tunneling ionization. In Ref. 17 a quasiclassical formula is proposed to describe tunneling ionization of arbitrary atoms and ions. In our calculations we will make use of the following form of this formula:

$$\begin{aligned} \gamma_s^r&=\nu_0\left(\frac{3e}{\pi}\right)^{1/2}\frac{s^2}{n_s^{*4.5}}\left(\frac{4es^3}{\mathscr{E}n_s^{*4}}\right)^{2n_s^*-1.5} \\ &\quad\times\exp\left(-\frac{2s^3}{3\mathscr{E}n_s^{*3}}\right), \\ n_s^*&=s\left(\frac{Ry}{J_s}\right)^{1/2}, \quad \mathscr{E}[\text{a.u.}]=5.345 \\ &\quad\times 10^{-9}(I[W/\text{cm}^6])^{1/2}, \\ s&=1,2,\ldots,Z, \\ \nu_0&=m_e e_e^4/\hbar^3. \end{aligned} \tag{10}$$

Collisional ionization. In Ref. 18 an expression is proposed to describe collisional ionization specifically for atoms and ions of neon. In our calculations we will make use of the following formulation.

$$\begin{aligned} \gamma_s^e&=\begin{cases} N_e c\sigma_s, & W>J_s \\ 0, & W\leqslant J_s \end{cases}, \\ W&=m_e c^2[(1+I/I_r)^{1/2}-1], \\ \sigma_s&=\sigma_0\left(\frac{\Omega s}{2l_0+1}\right)\left(\frac{Ry}{J_s}\right)^2\Phi(u_s), \\ \sigma_0&=0.8794\times 10^{-16}\text{cm}^2, \quad \text{Ry}=13.606\ \text{eV}, \\ u_s&=(W-J_s)/J_s, \\ \Phi(u)&=\frac{C_s u}{(u+\phi_s)(u+1)}\begin{cases} \left(\dfrac{u}{u+1}\right)^2, & s=1 \\ 1, & s>1. \end{cases} \end{aligned}$$

These formulas take into account the relativistic factor, where I_r is the relativistic intensitym.[1,2] The values of the constants are given in Table 1.

Law of conservation of energy.

Expressing the energy in the pulse in the form

$$\varepsilon=2\pi v_g^{-1}\int_{-\infty}^{\infty}\int_0^{\infty}|a|^2 r\,dr\,dz \tag{11}$$

and the ionization energy of the medium as

TABLE I.

s	Configuration	Ω_s	l_{0s}	C_s	ϕ_s
1	$1s^2 2s^2 2p^6$	6	1	16.9	1.46
2	$1s^2 2s^2 2p^5$	5	1	16.9	1.46
3	$1s^2 2s^2 2p^4$	4	1	16.9	1.46
4	$1s^2 2s^2 2p^3$	3	1	16.9	1.46
5	$1s^2 2s^2 2p^2$	2	1	16.9	1.46
6	$1s^2 2s^2 2p$	1	1	16.9	1.46
7	$1s^2 2s^2$	2	0	5.28	1.65
8	$1s^2 2s$	1	0	5.28	1.65
9	$1s^2$	2	0	7.2	2.56
10	$1s$	1	0	7.2	2.56

$$U=2\pi\int_{-\infty}^{\infty}\int_{0}^{\infty}\rho\varepsilon_{\text{ion}}r\,dr\,dz, \tag{12}$$

we find in the usual way

$$\varepsilon+U=\text{const}, \tag{13}$$

i.e., the model under consideration energy conservation holds.

3. SPECIAL FEATURES OF THE NUMERICAL METHOD

To localize the laser pulse it is convenient to transform to new variables:

$$\xi=v_g t-z, \quad \tau=t. \tag{14}$$

In this case the basic relations (1), (3), and (5) take the form

$$v_g^{-1}\partial_\tau a+\frac{i}{2k}\left(\Delta_\perp+k_p^2\left[1-\frac{\langle z\rangle}{Z}\right]\right)a+\frac{\mu^-}{2}a=0, \tag{15}$$

$$(v_g^{-1}\partial_\tau+\partial_\xi)\alpha=B\alpha, \tag{16}$$

$$\mu^-=\frac{1}{I}(v_g^{-1}\partial_\tau+\partial_\xi)(\rho\varepsilon_{\text{ion}}). \tag{17}$$

For a focused Gaussian beam, the initial condition (8) can be written in the form

$$a_0(r,\xi,\tau=0)=a_0\exp\left[-\frac{1}{2}\left(\frac{r}{r_0}\right)^2-\frac{1}{2}\times\left(\frac{\xi-\xi_1}{\xi_0}\right)^2-ik\frac{r^2}{4R_f}\right], \tag{18}$$

where R_f is the focal length of the lens (in our calculations for an initially planar wavefront we set $R_f=\infty$), and r_0, ξ_0, and ξ_1 are the parameters of the beam. The boundary conditions (9) do not change.

4. SOLUTION OF THE THREE-DIMENSIONAL PROBLEM

The problem as stated by Eqs. (15)–(18) and (9) was solved using a spectral-difference numerical technique. The latter is a variant of the numerical methods considered in Refs. 19 and 20.

This formation takes account of the following effects: diffraction; refraction by inhomogeneities in the refractive index arising as a consequence of variations in the electron number density $N_e(r,z)$; generation of electrons as a consequence of ionization of atoms of the medium (taking into account both field and collisional ionization); and absorption arising from ionization and the medium. Our analytic estimates and numerical calculations show that for a laser pulse with fixed parameters propagating in a gaseous medium, the models describing the physical phenomena arising in the propagation of the pulse can be placed in a definite sequence depending on the density of the gas.

A) In a low-density gas, ionization of the medium does not act back on the propagating radiation. The corresponding criterion can be obtained from the properties of Gaussian beams. It is necessary that the variation of the optical path over the characteristic length of the Gaussian caustic be much less than the wavelength of the radiation:

$$\Delta nL\gg\lambda/10.$$

Noting the relations

$$\Delta n=1/2(\omega_p/\omega)^2, \quad L=\pi r_0^2/\lambda,$$

we obtain

$$N_e\ll 2m_e c^2/(e_e^2 r_0^2),$$

or in absolute units

$$N_e\ll 7\cdot 10^{19}(10^{-4}/r_0)^2\text{cm}^{-3}. \tag{19}$$

It is interesting that the wavelength does not enter into the lattice estimate.

B) As the gas density increases, i.e., when criterion (19), fails this ionization of the medium begins to react on the propagation radiation, thereby causing double refraction and a corresponding shortening of the "caustic."

C) With further increase of the gas density, absorption becomes pronounced, which leads to an "eating away" or "corrosion" of the leading edge of the pulse.

We will illustrate the above situations in specific cases below.

Let us consider the focusing of an ultrashort laser pulse ($\lambda=0.4$ μm) with aperture in the focal plane $r_0=10$ μm, peak intensity $I_0=2.0\cdot 10^{17}$ W/cm^2, length $\xi_0=15$ μm in lithium vapor with density $N_0=5.0\cdot 10^{18}$ cm^{-3}.

The solution of this problem is depicted in Fig. 1. The figure illustrates the two-dimensional spatial distribution of the mean charge, deposited by the laser pulse. In Fig. 1.1 the pulse is 0.31 cm behind the focal spot, Fig. 1.2 shows the pulse as it passes through the focus, and Fig. 1.3 shows the laser pulse when it has passed completely through the Gaussian caustic. In this calculation, the density of the medium is low and the appearance of free electrons has no effect on the propagating radiation.

The second case illustrates the situation when a neodymium laser pulse with peak intensity $I_0=10^{17}$ W/cm^2 and transverse and longitudinal dimensions $r_0=10$ μm and $\xi_0=12.53$ μm, propagates in neon with density $N_0=3\cdot 10^{19}$ cm^{-3}. Figure 2 depicts the evolution of the two-dimensional spatial distribution of the intensity of the laser pulse (in comoving coordinates) after the pulse has propagated a distance $\delta z=5.2+10.5j$ μm ($j=1,\dots,6$) from its starting

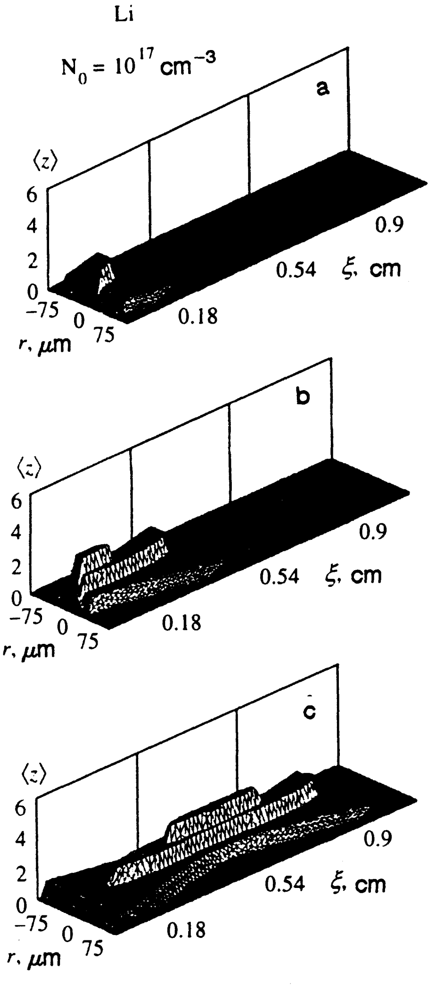

FIG. 1. Propagation of an ultrashort laser pulse in lithium vapor. Surface plots of the two-dimensional spatial distribution of the mean charge of the ionized plasma, deposited by the laser pulse. Threshold ionization model from Ref. 16.

point. Figure 3 depicts the mean charge distribution (in the co-moving coordinate system) along the propagation axis at times τ=4, 35, 44, 61 fs, respectively.

An important feature of this problem is the effect of small-scale ionization phase defocusing. The small-scale aspect of the effect is associated with the ionization state and was not observed in Ref. 15. The electron number density profile arising as a consequence of ionization has a stepped shape in z and r since both photoionization and collisional ionization have a threshold character. This is graphically demonstrated by Fig. 1.3. The radiation behaves as if it has passed through a phase palate. A complicated interference pattern arises, characterized by a large number of maxima and minima. The radiation starts to be scattered very strongly by the periphery: Note that direct numerical solution of a system of the form (2) with coefficients of the form (9) is hampered as a consequence of the discontinuous nature of the solutions. Therefore, the correct result is achieved by smoothing the steps of the ionization coefficients and using a nonuniform grid in the calculations, with bunching of the grid points in the region of intense ionization.

We will show that the Rayleigh length L_0 for radiation in

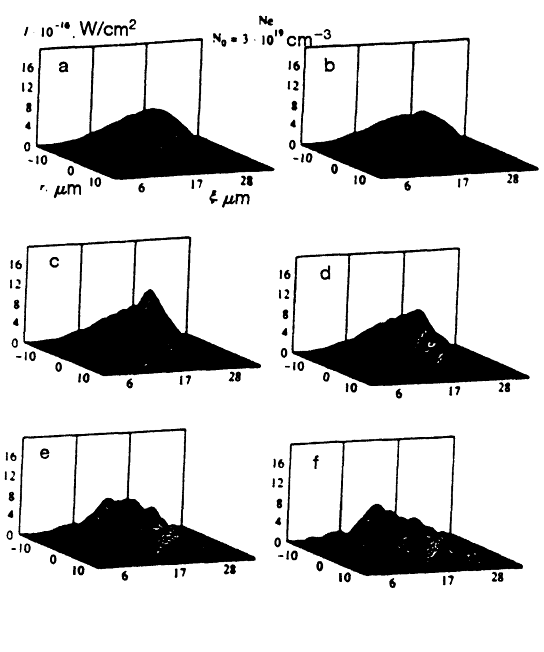

FIG. 2. Propagation of an ultrashort laser pulse in neon. Two-dimensional spatial distribution of the intensity. Tunneling ionization model from Ref. 17.

a medium with Z-fold ionization is shortened by a factor of Z^2. Indeed, the aperture of the pulse subdivides into Z transverse zones with average dimension r_0/Z. The Rayleigh length for an individual zone is equal to

$$L \simeq \frac{\pi(r_0/Z)^2}{\lambda} = \frac{1}{Z^2}\left(\frac{\pi r_0^2}{\lambda}\right) = \frac{1}{Z^2} L_0. \tag{20}$$

This means that a laser pulse which has been focused in a focal spot with transverse dimension r_0 and caustic length, L_0, having ionized the medium, Z timer will, if the density criterion (19) is exceeded, be scattered by a transverse distance r_0/Z and the propagation length of the pulse will be decreased by a factor Z^2. The scattering angle is increased by

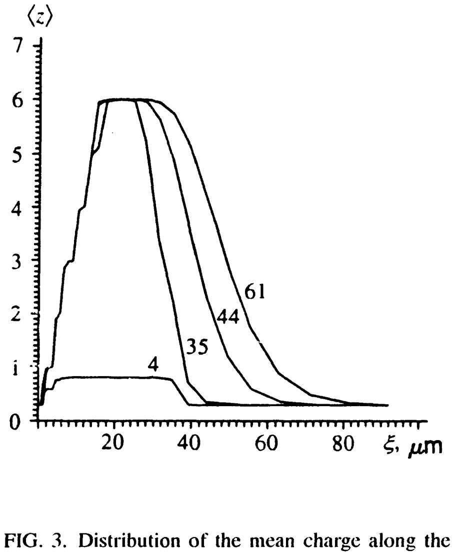

FIG. 3. Distribution of the mean charge along the propagation axis of the laser pulse. Threshold ionization model from Ref. 16.

a factor of Z: $\theta \simeq \lambda Z/r_0$. Note that the effect of small-scale ionization phase of defocusing can inhibit self-focusing of the laser pulse.

In this work we have performed calculations using two models of photoionization: the threshold ionization model and the tunneling ionization model. Both models are quite approximate. The first provides only a poor description of ionization far from the threshold; the second, on the contrary, provides a good description of ionization far from the threshold, but only a poor description of the behavior of the cross section near the threshold. Besides, the tunneling ionization model loses accuracy for radiation intensities above 10^{15} W/cm^2. Both models predict the effect of small-scale ionization defocusing. The tunneling ionization model smooths the cross section near the threshold, which leads to smoother profiles of $N_e(r,z)$. As a result, the second model gives small-scale phase distortions at distances roughly two times greater than the first.

Collisional ionization of the medium at the densities calculated is insignificant. Its role grows with the density of the gas. Note that in this work we did not take collisional ionization from the excited states of the atoms and ions into account.

In some situations the role of absorption due to ionization becomes noticeable. Absorption causes erosion of the leading edge of the pulse. This effect is noticeable in Fig. 2. At higher intensities, the effect of absorption due to ionization is insignificant since the pulse is scattered faster than it is absorbed.

5. CONCLUSION

The main conclusion of the present paper is the following: to realize x-ray lasers based on the principle of field ionization of the medium by a short powerful pulse from an exciting laser, it is necessary to satisfy the criterion (19), which ensures that the medium will not be strongly affected by the propagating pulse.

If (19) is violated, then if the laser pulse propagates to the focal region in vacuum small-scale ionization phase defocusing of the pulse can take place, due to the generation of a stepped profile of the electron number density. For Z-fold ionization of the medium, the pulse is scattered by a mean transverse distance r_0/Z, where r_0 is the aperture of the laser pulse. The scattering angle grows by a factor Z, and the Rayleigh length decreases by a factor Z^2. This regime of laser pulse propagation is unsuitable for the successful implementation of an OFI x-ray laser.

In some situations "corrosion" of the leading edge of the pulse is substantial, due to losses to ionization of the medium.

The field-threshold model[16] and the tunneling-ionization model[17] disagree by roughly a factor of two in the scattering lengths of the laser pulse. This being the case, the question arises of refining these models.

Note that the calculations in the present work were carried out for $P_0 < P_{cr}$, where P_{cr} is the critical power for relativistic nonlinear self-channeling of the laser pulse. In the regime of vacuum delivery of the pulse the question of the Kerr nonlinearity of the gas, due to deformation of the ion shells in a strong optical field, remains open. A deeper consideration of the ionization kinetics of the atoms and ions in a strong field is also possible.

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