NONLINEAR OPTICAL PHENOMENA

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Quasi-soliton and multifocal propagation of high-intensity laser pulses in silica glass

N.Yu. Vislobokov

***Abstract*. The results of numerical simulation of propagation of high-intensity ultrashort pulses in silica glass are presented. It is shown that in the case of a dynamic balance between self-focusing and defocusing induced by ionisation, radiation propagating in the glass over the distance of the order of a diffraction length can be captured to form a quasi-soliton. For certain relations between the parameters of radiation and glass over lengths exceeding the diffraction length, the multifocal propagation regime is possible.**

Keywords: *femtosecond pulse, ultrashort laser pulses, quasi-soliton, induced ionisation, multiphoton ionisation, multifocal propagation.*

1. Introduction

Many recent studies in nonlinear optics are devoted to the propagation and evolution of high-power ultrashort pulses (USPs) in air and solids. First the propagation of such USPs was investigated in different gases, in particular, air. It was shown, for example, that the propagation of an infrared laser USP in the atmosphere can result in its splitting into several narrow filaments with the peak intensity of the order of 10^{13} W cm^{-2}, which can propagate over long distances [1]. This effect is the result of dynamic competition between two nonlinearities affecting the propagation of USPs: defocusing and Kerr self-focusing. In this case, defocusing mainly occurs due to the negative contribution to the nonlinear part of the refractive index n_2 of the medium caused by the formation of a free electron plasma (FEP).

It is most difficult to study the propagation dynamics of such pulses in solids, in particular, in dielectrics (silica glass, sapphire). The results of these studies can be used, for example, for the development of control elements in laser systems and lasers themselves, as well as for the elaboration and manufacturing of microelectronic devices. The femtosecond laser modification of dielectrics such as silica glass can be used to vary the refractive index of materials to fabricate waveguides [2, 3]; in three-dimensional optical devices [4], for example, three-dimensional data storage devices [5]; and in chemistry for the selective etching of microstructures and microliquid channels [6]. Femtosecond laser microsurgery is applied in biological studies and is promising for applications directly in medicine [7]. Practical applications of femtosecond laser pulses require the knowledge of the pulse evolution during its propagation. Of special interest is the study of propagation of an USP in the regime when the spatiotemporal pulse profile does not change in essence (quasi-soliton) and/or changes quasi-periodically.

It was assumed first that it is impossible to obtain a quasi-soliton in a solid that would propagate over the distance exceeding 1 mm [8]. This is explained by the fact that the propagation of high-intense radiation pulses of duration $\tau_p > 10^{-12}$ s over a distance of only 1 mm is accompanied by avalanche ionisation resulting in the damage of a sample under study. The avalanche formation of a FEP took place even when the pulse duration was a few picoseconds. However, it was shown later that no avalanche formation of a FEP occurred in silica glass during the propagation of pulses of duration 200 fs and shorter with a peak intensity of no more than 2×10^{13} W cm^{-2} [9] or its influence on the femtosecond pulse was negligible.

In addition, to capture laser radiation to form a quasi-soliton propagating even over only a few millimetres in solids is much more difficult than in air because here the contribution of the Kerr nonlinearity is a few orders of magnitude greater.

In this paper, the study of the capture of an intense femtosecond laser radiation into a long-lived quasi-soliton in silica glass is presented.

2. Basic equations

In a dielectric with the positive Kerr nonlinearity, an USP whose power exceeds the critical self-focusing power tends to focus under the action of this effect. However, a number of defocusing effects such as group velocity dispersion (GVD) and multiphoton ionisation also act on the propagating pulse. The GVD can delay or stop self-focusing; however, it was found that pulses propagating in media with a high enough dispersion split into subpulses. Therefore, the use of media with high dispersion to obtain the quasi-soliton propagation regime is not promising. More promising are low-dispersion dielectric media (sapphire, silica glass) where self-focusing can compete with defocusing due to self-induced ionisation effects.

The influence of ionisation effects on a propagating pulse depends not only on the parameters of a medium in

N.Yu. Vislobokov Department of Physics, M.V. Lomonosov Moscow State University, Vorob'evy gory, 119992 Moscow, Russia; e-mail: nkt_2004@mail.ru

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which the pulse propagates but also on the parameters of the pulse itself, and in the case of a high-intense USP this influence is determined to a great extent by the peak intensity of the pulse. Due to a short duration of the pulse, free electrons can capture a great part of energy lost by the pulse due to multiphoton absorption before its transfer to the crystal lattice of a dielectric. This leads to the formation of a FEP which makes a negative contribution to the nonlinear part of the refractive index of the medium, thereby defocusing the propagating beam and delaying a collapse predicted by the paraxial model [4, 10].

Under certain conditions, self-focusing and defocusing can balance each other. Due to the dynamic equilibrium between these two effects, the laser pulse is captured into a quasi-soliton propagating in the medium until this dynamic equilibrium is preserved.

To describe this process correctly, it is necessary to modify the conventional Schrödinger equation by taking into account dispersion effects and ionisation processes [11–14]. According to studies [11, 12, 15], we assume that multiphoton absorption is the dominating mechanism of plasma generation at physical parameters used in computer experiments.

The equation for the field of a beam propagating in a sample has the form [15]

$$\frac{\partial E}{\partial z} = \frac{\mathrm{i}}{2k}\left(1 + \frac{\mathrm{i}}{\omega\tau_{\mathrm{p}}}\frac{\partial}{\partial\tau}\right)^{-1}\nabla_{\perp}^{2}E - \mathrm{i}\frac{\beta_2}{2}\frac{\partial^2 E}{\partial\tau^2} + P_{\mathrm{nl}}, \qquad (1)$$

$$P_{\mathrm{nl}} = \mathrm{i}k_0 n_2\left(1 + \frac{\mathrm{i}}{\omega\tau_{\mathrm{p}}}\frac{\partial}{\partial\tau}\right)|E|^2E - \frac{\sigma_{\mathrm{IBS}}}{2}(1 + \mathrm{i}\omega\tau_{\mathrm{c}})\rho E$$

$$- \frac{1}{2}\frac{W_{\mathrm{MPI}}(|E|)U}{|E|^2}E, \qquad (2)$$

where E is the electric field strength; z is the longitudinal coordinate; r is the transverse coordinate; τ is the propagation time; $\nabla_{\perp}^2$ is the Laplace operator over the transverse coordinate; k is the wave number; k_0 is the wave number at the entrance to the medium; β_2 is the GVD coefficient; n_2 is the nonlinear part of the refractive index; σ_{IBS} is the inverse bremsstrahlung cross section; ω is the laser frequency; τ_{p} is the pulse duration; τ_{c} is the momentum transfer collision time; ρ is the free electron density in the medium; $W_{\mathrm{MPI}}(|E|) = \sigma_{\mathrm{m}} I^m \rho_{\mathrm{at}}$ is the multiphoton ionisation rate; I is the beam intensity; σ_{m} is the multiphoton absorption coefficient; ρ_{at} is the background density of atoms; U is the gap potential of the dielectric; and m is the multiphoton transition order.

The first term in the right-hand side of (1) is caused by diffraction, the second one by the GVD whose coefficient for silica glass is $\beta_2 = 380$ fs^2 cm^{-1}, the third term describes the contribution of nonlinearity and induced ionisation transitions.

The model should take into account a change in the free electron density ρ due to multiphoton ionisation caused by the propagating pulse. The equation for the rate of a change in ρ (neglecting the avalanche and recombination [11, 12, 15]) has the form

$$\frac{\partial\rho}{\partial\tau} = W_{\mathrm{MPI}}(|E|). \qquad (3)$$

We assume that the incident beam has the Gaussian shape

$$E(r, z = 0, \tau) = E_0 \exp\left(-\frac{t^2}{2w_0^2} - \frac{\tau^2}{2\tau_{\mathrm{p}}^2}\right),$$

$$\rho(r, z, \tau = 0) = \rho_0. \qquad (4)$$

Here, w_0 is the initial width of the beam and E_0 is the initial amplitude of the electric field strength.

3. Computer experiment

The self-consistent system of equations (1)–(4) was solved by using a special program (developed by the author) based on the fast Fourier transform together with difference schemes, which provided the solution of the modified Schrödinger equation taking into account ionisation effects and variations in the free electron density. It was assumed that the reflected beam intensity is small compared to the incident beam intensity and does not noticeably affect the formation of a plasma and evolution of the pulse during its propagation.

The results of numerical experiments obtained for sapphire, silica glass, and BK7 glass showed that a high-intense USP propagating in these media is defocused due to the influence of a FEP. It was found experimentally earlier that induced ionisation effects can delay self-focusing in sapphire for $P/P_{\mathrm{cr}} = 1.8$ [13] (where P is the pump power and P_{cr} is the critical self-focusing power) or cause the filamentation of the pulse at distances of only several millimetres in silica glass for $P/P_{\mathrm{cr}} = 3$ and $\tau_{\mathrm{p}} = 160$ fs [8]. The results obtained with the help of our numerical model well agree with these experimental data.

Our numerical studies showed that the increase in the peak intensity of an USP up to 1 TW cm^{-2} and more and the simultaneous decrease in its duration down to a few tens of femtoseconds for the initial beam width not exceeding 70 μm can facilitate the formation of a soliton-like waveguide [10] propagating in a medium over the distance of the order of one diffraction length.

Parameters at which the quasi-soliton propagation of a femtosecond pulse is observed were determined by studying the evolution of the UPS in silica glass at a wavelength of 790 nm. This wavelength was selected taking into account the possibilities of real laser systems and the dependence of the absorption coefficient of silica glass on the incident radiation wavelength.

The pulse duration τ_{p} was varied between 50 and 90 fs and the beam width – between 30 and 70 μm. The order of the multiphoton transition was $m = 6$. The parameters of a dielectric favourable for the formation of a quasi-soliton were determined by studying numerically the behaviour of USPs propagating in media with different GVD coefficients β_2 and different refractive indices n_2. The ranges of variations in β_2 and n_2 were limited by the limits of application of the model and the properties of dielectrics under study: β_2 was varied from 361 to 1280 fs^2 cm^{-1} and n_2 from 2×10^{-16} to 5×10^{-16} cm^2 W^{-1}. Below, all the calculations were performed for silica glass as the most widespread and low-cost dielectric.

We also performed numerical simulations of the spatial dynamics of the beam intensity $I(r, z)$ and the dynamics of the temporal $I(t, z)$ and spatiotemporal $I(r, t)$ profiles of the propagating pulse. In addition, we studied the dynamics of the phase and spectrum of the femtosecond pulse.

Consider now how changes in the group-velocity coefficient and nonlinear part of the refractive index affect the formation and propagation (within one diffraction length) of a quasi-soliton. Our computer experiments showed that the increase of β_2 from 361 to 1280 fs^2 cm^{-1} has an adverse effect on the quasi-soliton propagation of the pulse, which is manifested in greater deviations of its shape from the initial one and violates the equilibrium between self-focusing and defocusing. However, for $\beta_2 = 0$ the deviation of the beam shape from its initial shape was greater than that for $\beta_2 = 361$ fs^2 cm^{-1}. A small dispersion ($\beta_2 \neq 0$) stabilises somewhat a quasi-soliton propagating in dielectrics such as sapphire or silica glass. At the same time, variations in the GVD coefficient in the range under study did not affect noticeably the conditions of quasi-soliton formation, which is consistent with assumptions [8] about the waveguide propagation of USPs. As for the nonlinear part of the refractive index n_2, it was found that its increase makes it possible to capture radiation into a quasi-soliton at lower intensities. Changes in the nonlinear part of the refractive index did not affect substantially the propagation of a quasi-soliton.

The numerical study performed in the paper gave the initial intensities at which radiation is captured already at the input to a quasi-soliton which propagates at least over one diffraction length. It was found that the deviation of the envelope of the beam intensity from its initial shape decreased somewhat with decreasing the beam width w_0 from 70 to 50 μm and remained unchanged as w_0 was further decreased. In the case of formation of quasi-solitons from a narrow beam, a higher initial intensity is naturally required, which, of course, limits the field of their applications. The increase in the pulse duration had a reverse effect on beams with $w_0 > 50$ μm.

The intensity range in which a quasi-soliton can be produced in a dielectric is important for the formation of quasi-solitons in real experiments. The broadest intensity range in silica glass was observed for $\tau_p = 70$ fs and $w = 50$ μm. For the specified beam duration and width, the most favourable initial intensity for the formation of a quasi-soliton was $I_0 = 5.71$ TW cm^{-1}. Figure 1a shows variations in the envelope of the quasi-soliton intensity in silica glass for these values of I_0, w_0, and τ_p taking ionisation into account.

For comparison, Fig. 1b shows a similar dependence neglecting ionisation effects. One can see that the quasi-soliton propagation of a high-intense femtosecond pulse in silica glass is possible mainly because the induced FEP stops self-focusing of the beam over some distance, i.e. ionisation is in fact the main effect making the beam self-defocusing sufficient to compete with self-focusing.

The dynamics of the phase and spectrum of a pulse propagating in silica glass shows that the phase of a quasi-soliton does not virtually change. The pulse spectrum also does not change significantly, although it deviates somewhat from the initial spectrum, as shown in Fig. 1c where the initial spectrum and spectra formed at lengths $z = 0.5L_{\rm df}$ and $z = L_{\rm df}$ are presented (where $L_{\rm df}$ is the diffraction length).

The behaviour of a beam propagating as a quasi-soliton within one diffraction length was also studied upon its further propagation in the medium. Figure 2 shows the propagation of the beam with the same input parameters as in Fig. 1a up to $z = 4L_{\rm df}$. One can see that the beam

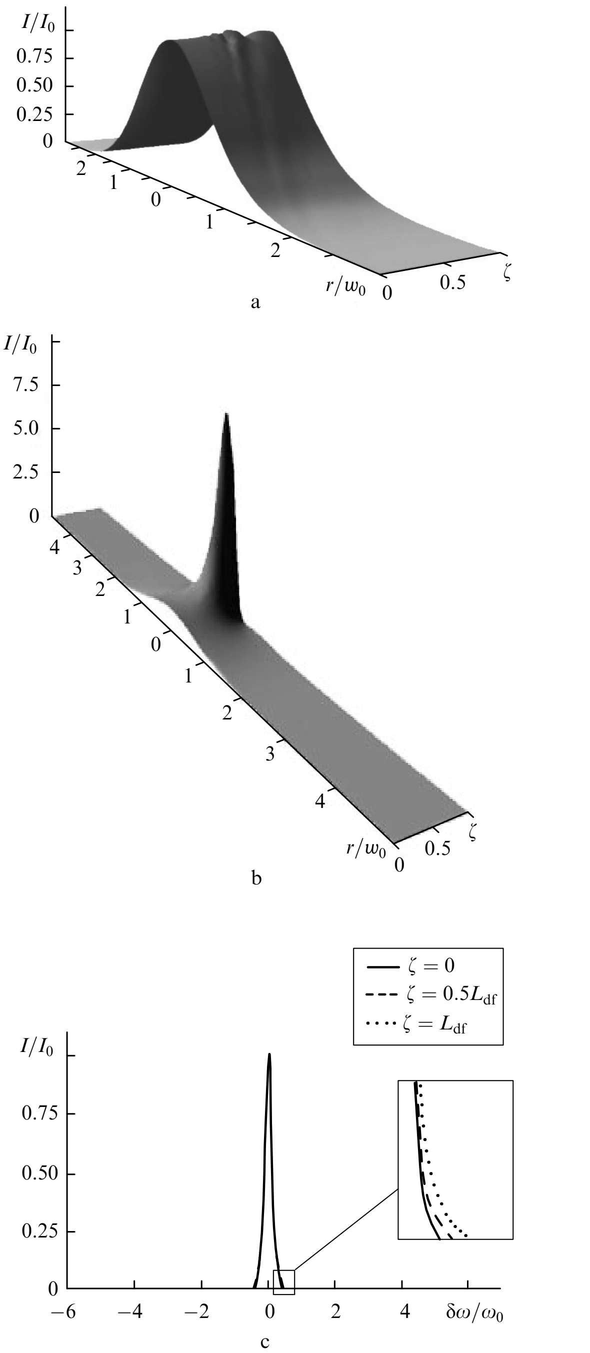

Figure 1. Change in the beam envelope at the centre of a pulse propagating along the longitudinal coordinate $\zeta = z/L_{\rm df}$ in silica glass taking into account (a) and neglecting (b) ionisation, and the change in the spectrum of a quasi-soliton propagating in silica glass at different points ζ (c) for $I_0 = 5.71$ TW cm^{-2}, $w_0 = 50$ μm, $\tau_p = 70$ fs, and $L_{\rm df} = 14.631$ mm ($\delta\omega = \omega - \omega_0$, ω_0 is the initial frequency of the pulse).

intensity gradually decreases with distance. It may seem that the problem can be solved simply by increasing the initial peak intensity of a femtosecond pulse; however, its drastic increase can violate the balance between self-focusing and defocusing so that the first effect will dominate, resulting in the self-focusing of the beam followed by its collapse.

If the pulse intensity at the input is lower than a threshold value at which defocusing caused by the FEP can compete with self-focusing, the beam propagating in a dielectric is gradually focused. Numerical simulations

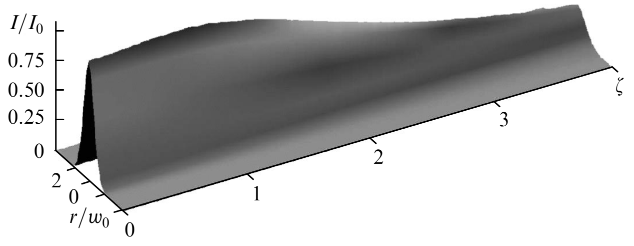

Figure 2. Change in the beam envelope (at distances up to $4L_{df}$) at the centre of a pulse propagating along the longitudinal coordinate $\zeta = z/L_{df}$ in silica glass for $I_0 = 5.71$ TW cm^{-2}, $w_0 = 50$ μm, and $L_{df} = 14.631$ mm.

showed that the beam self-focusing can be stopped by increasing the peak intensity up to a certain value when the pulse duration and beam width are small enough. Defocusing can compensate for self-focusing and exceed it. In this case, defocusing is mainly caused by the negative contribution of the FEP to the nonlinear part of the refractive index. The peak intensity gradually decreases during defocusing and the defocusing contribution of ionisation to the pulse propagation dynamics also correspondingly decreases. The contribution of focusing effects increases simultaneously and self-focusing of the pulse again begins at some distance. The so-called multifocal propagation regime takes place. Figure 3 presents the results of numerical study of the propagation of a high-intensity femtosecond pulse in silica glass over the distance up to 10 diffraction lengths (by 52.67 mm). Variation in the beam envelope is observed at the centre of the pulse propagating along the longitudinal coordinate taking ionisation effects into account.

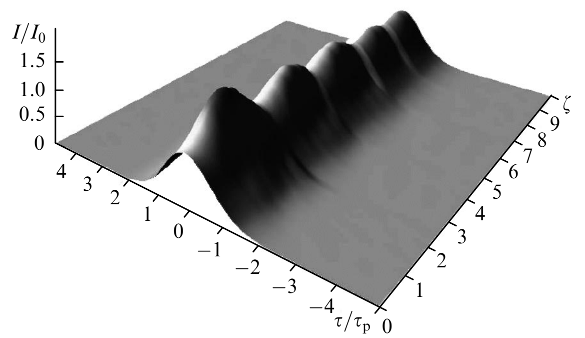

Figure 3. Spatiotemporal distribution of the intensity envelope for a pulse propagating in the multifocal regime over the distance $10L_{df}$ in silica glass taking into account ionisation effects ($\zeta = z/L_{df}$, $L_{df} = 5.267$ mm).

One can see that the beam propagates in the multifocal regime. The maximum intensity in self-focusing phases gradually decreases during pulse propagation. The temporal profile of the pulse does not change significantly. Although the width of the spatial profile gradually changes, it remains qualitatively Gaussian. The distance between the first and second peaks is smaller than that between the second and third peaks, and then it gradually increases with increasing ζ. This is explained by the fact that the pulse energy is gradually absorbed during pulse propagation, so that the distance between neighbouring intensity peaks increases with distance of these peaks from the input to a dielectric.

The obtained numerical results are in qualitative agreement with the experiments on multifocal propagation of 120-fs pulses at 800 nm in silica glass over small distances (up to 5 mm [5]).

4. Conclusions

It has been shown in this paper how plasma effects and self-focusing interact during the propagation of a high-intense laser pulse in silica glass. The interaction of ionisation and focusing effects and their influence on the propagation of this pulse in silica glass has been demonstrated. A number of parameters that are most convenient for capturing such a pulse into a quasi-soliton have been found.

The influence of parameters of radiation (I_0, w_0, τ_p) and medium (β_2 and n) on the propagating pulse has been analysed. It was pointed out that the propagation of radiation close to quasi-solitons was observed for broader beams at lower intensities, other parameters being the same. It has been shown that the quasi-soliton propagation of the beam in silica glass is facilitated by a low dispersion coefficient, while the increase in the refractive index n_2 makes it possible to capture radiation into a quasi-soliton at the lower peak intensity. In addition, numerical simulations have shown that the phase and spectrum of the propagating quasi-soliton do not change in fact. The radiation parameters that are most favourable for the formation of a quasi-soliton in silica glass have been proposed.

The evolution of a femtosecond pulse propagating in silica glass over the distance exceeding the diffraction length L_{df} has been considered. It has been shown that at certain parameters the beam can propagate over the distance at least up to $10L_{df}$ in the multifocal regime and the shape of its envelope does not change significantly, i.e. the beam intensity and width change during its propagation, but the beam shape remains Gaussian.

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