Dynamics of nonlinear absorption of light in solids

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The problem of the passage of a laser pulse in a medium in the presence of multiphoton ionization is solved. It is shown that the accumulation of nonequilibrium carriers in the interaction region leads to shortening of the laser pulse and deformation of the intensity distribution over the cross section of the light beam. The maximum intensity of the transmitted radiation depends to a large extent on the slope of the leading edge of the incident radiation pulse and on the exponent of the multiphoton process. The extremal values of the intensity, absorbed energy and concentration of the nonequilibrium carriers in the lens caustic are obtained for focused beams. The effect of recombination and carrier heating on the dynamics of the light absorption is discussed.

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1. INTRODUCTION

The passage of a powerful laser radiation through a solid is usually accompanied by a number of nonlinear processes, which change both the characteristics of the medium through which the light passes and the very dynamics of propagation of the electromagnetic wave. Among such processes, the foremost is the process of generation of free charge carriers on account of multiphoton, impact, and thermal ionization. Thermal ionization is a very simple and well-studied mechanism of such generation, causing thermal breakdown of the medium.[1] However, in the case of nanosecond pulses of radiation of high intensity ($I > 10^5$ W/cm^2), the greatest interest attaches to the mechanism of generation due to multiphoton or cascade absorption and impact ionization. The concentration of the nonequilibrium carriers in this case can be very large and reach values 10^{17} cm^{-3} and higher. In the case of such densities, the chief effect on the propagation of the radiation is provided by the nonlinear absorption and by processes of self-focusing by the free carriers.

There is interest in the study of the role of nonlinear absorption in the dynamics of light propagation. In a number of researches (see, for example, Refs. 2 and 3), analysis of the light propagation in the medium in the presence of multiphoton absorption was carried out by numerical methods. However, numerical calculation, as applied to a single specific case, does not allow us to explain all the features of the given process.

2. DYNAMICS OF NONLINEAR ABSORPTION IN A PLANE LAYER

The equations of propagation of light radiation in a medium in the presence of multiphoton absorption, with account of the absorption by the free carriers, which take place in the process of multiphoton ionization, have the form

$$\frac{\partial I}{\partial z}=-\alpha_0 I-\alpha_k I^k-\sigma NI, \qquad \frac{\partial N}{\partial t}=\frac{\alpha_k}{k\hbar\omega}I^k-\frac{N}{\tau}. \qquad (1)$$

Here I is the intensity of the light wave propagating in the z direction, α_0 is the coefficient of linear absorption, α_k is the coefficient of k-photon absorption, N is the density of nonequilibrium carriers, σ is the cross section for absorption by them, $\hbar\omega$ is the energy of the light quantum, and τ is the recombination time of the free carriers.

The solution of the problem of wave propagation in the medium without account of absorption at the charge carriers ($\sigma N = 0$) is well known (see, for example, Ref. 4). However, the real process of multiphoton absorption is accompanied by generation of charge carriers which begin to absorb effectively and which change the entire dynamics of propagation of the light wave. It should also be noted that for a correct determination of the probability of k-photon absorption, it is always necessary to solve the complete set of equations (1).

We estimate which of the last two terms in the first equation of the set (1) will play the principle role in the dynamics of light absorption. Recognizing that the carrier density is

$$N=\frac{\alpha_k}{k\hbar\omega}\int_0^t I^k\,dt' \approx \frac{\alpha_k}{k\hbar\omega} I^k\,\Delta t,$$

we find that $\alpha_k I^k = \sigma NI$ at $\Delta t = k\hbar\omega/I\sigma$. For light pulses $I \approx 10^6$ W/cm^2, $\sigma = 10^{-7} - 10^{-18}$ cm^{-2} and $\hbar\omega = 1$ eV, we have $\Delta t = 10^{-14} - 10^{-15}$ s. These simple estimates show that the principal role in the dynamics of nonlinear absorption of a powerful electromagnetic wave is played by absorption by the free carriers which are created in the process of multiphoton absorption.

Two extreme cases are considered in here: when the duration of the pulse τ_p is much shorter than the recombination time τ, and the case of fast recombination times $\tau \ll \tau_p$. The latter case will be considered below.

In the absence of recombination and with account of the estimates given above, we obtain a set of nonlinear equations that describe the dynamics of light propagation in the medium in the case of k-photon absorption:

$$\frac{\partial I}{\partial z}=-\sigma NI, \qquad \frac{\partial N}{\partial t}=\beta I^k, \qquad \beta=\frac{\alpha_k}{k\hbar\omega}. \qquad (2)$$

For simplicity here, we do not take linear absorption into account.

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The solution of the problem of the passage of light through an absorbing medium will be carried out in the thin-layer approximation, i.e., it is assumed that the length of the sample z_0 is much less than the diffraction length of the light beam l_d, and that the initially plane wave front of the beam is not distorted by the processes of defocusing. This latter assumption is valid if the nonlinear phase shift $\Delta\varphi$ in a layer, due to defocusing, is $\Delta\varphi \ll l_d/z_0$. In this case, we can assume that the light beams pass through the investigated material rectilinearly.

For a specified form $I = I_0\varphi(t)$ of the pulse of initial radiation, the boundary and initial conditions for (2) have the form

$$I(0,t)=I_0\varphi(t), \quad N(0,t)=\beta I_0^k \int_0^t \varphi^k(t')\,dt', \qquad (3)$$
$$I(z,0)=I_0\varphi(0), \quad N(z,0)=0.$$

Here I_0 is the peak value of the intensity of the incident wave.

Using the substitution $u = k\ln[I/I_0\varphi(t)]$, we obtain from (2) the following equation for the function u:

$$\frac{\partial^2 u}{\partial\xi\,\partial z_{\text{н}}} = -e^u, \qquad (4)$$
$$u(z_{\text{н}},0)=u(0,\xi)=0, \quad \left.\frac{\partial u}{\partial z_{\text{н}}}\right|_{z_{\text{н}}=0} = -\xi,$$
$$\xi=\int_0^t \varphi^k(t')\,dt', \quad z_{\text{н}}=k\sigma\beta z.$$

The solution for an equation of the form (4) was first obtained by Liouville. In our case, since Eq. (4) and the boundary conditions corresponding to it are symmetric relative to the variables z and ξ, the introduction of a new variable ξz allows us to obtain the desired solution, which has the form

$$I(z,t)=\frac{I_0\varphi(t)}{[1+z\xi(t)k\sigma\beta I_0^k/2]^{2/k}}, \qquad (5)$$
$$N(z,t)=\frac{\beta I_0^k\xi(t)}{1+z\xi(t)k\sigma\beta I_0^k/2}. \qquad (6)$$

The expressions that have been obtained described the space-time dynamics of light propagation, since (5) is the solution along the trajectory of light ray, while I_0 is the distribution of the intensity of the beam on the entry surface. Attention should be called to the fact that, in contrast to the absorption of instantaneous and nonintegrating type,[4] the absorption of the form (5) does not undergo saturation, and at high intensities of incident radiation $k\sigma\beta z\xi I_0^k/2 \gg 1$, the intensity of the transmitted light begins to fall off according to the law $I \propto I_0^{-1}$ for all forms of the nonlinearity. This leads to the result that a dip in the intensity is formed at the center of the beam at its exit from the plate. The total power of the beam can change very weakly or remain constant upon increase in the power of the incident radiation.

It should be noted that in contrast to absorption of the non-integrating type, which leads to a flattening of the transmitted radiation pulse and its effective broadening, absorption of the integrating type produces a shortening of the transmitted pulse, its "cutoff." For a determination of the time of the cutoff and the peak value of the intensity of the transmitted light, we consider the dynamics of radiation exiting from the plate. For simplicity, we assume that at the leading edge of the pulse, the light intensity at the entry into the medium increases according to the law $I = I_0 t^m$. In this case, the dynamics of radiation at the exit from the plate is described by the expression

$$I(t,z)=I_0 t^m\left[1+t^{km+1}\frac{I_0^k z k\sigma\beta}{2(km+1)}\right]^{-2/k}. \qquad (7)$$

We note that if at small t (or small I_0) the radiation at the exit repeats the shape of the pulse of incident radiation $\varphi(t)$, at high I_0 the intensity of the light at the exit will have a maximum at the leading edge of the pulse $\varphi(t)$. It is not difficult to obtain the cutoff time t_0 from (7):

$$t_0^{km+1}=2m(km+1)/(km+2)z\sigma\beta I_0^k, \quad \text{i.e.,} \quad t_0\sim I_0^{-k/(km+1)}, \qquad (8)$$

and the maximum intensity at the exit:

$$I_{max}=I_0 t_0^m\left(1+\frac{km}{km+2}\right)^{-2/k}\sim I_0^{1/(km+1)}. \qquad (9)$$

Estimates for the exit energy density, obtained in this case, give

$$U\approx I_{max}t_0\sim I_0^{(1-k)/(km+1)}.$$

Thus, the exit energy at high amplitudes of the incident radiation I_0 begins to fall off and, it appears, a certain I_0 exists which guarantees a maximum energy density of the exiting radiation.

The solution (5) and (6) was obtained without account of the linear absorption. By the simple method of substitution $I = I\exp(-\alpha_0 z)$ and renormalization of N, we can obtain the general solution at $\alpha_0 \neq 0$:

$$I(z,t)=I_0\varphi(t)e^{-\alpha_0 z}\left[1+\frac{\sigma\beta\xi(t)I_0^k}{2\alpha_0}(1-e^{-k\alpha_0 z})\right]^{-2/k}, \qquad (10)$$
$$N(z,t)=\beta I_0^k\xi(t)e^{-k\alpha_0 z}\left[1+\frac{\sigma\beta\xi(t)I_0^k}{2\alpha_0}(1-e^{-k\alpha_0 z})\right]^{-1}.$$

In the analysis above it was assumed that the absorption cross section σ does not depend on the radiation intensity. However, this assumption can be violated, for example, because of heating of the free charge carriers in the field of an intense electromagnetic wave. We shall not discuss here the conditions under which such an effect is significant, since it depends on the type of the medium, on the character of the scattering of the free carriers, on the frequency of the incident radiation, and so on.

In a number of cases, $\sigma(I)$ can be approximated by a dependence of the form $\sigma(I) = \sigma\eta I^\gamma$, where $\gamma \leqslant 1$, as occurs in certain semiconductors (see, for example, Ref. 5). In this case, the solution for the intensity $I(z,t)$ can be obtained in the following way. We introduce the new variable

$$x=\gamma z\xi(t)\sigma_0\beta\varphi^\gamma(t)I_0^{\gamma+k}.$$

Then $I(z,t) = I_0\varphi(t)/w^{2/(\gamma+k)}$, where $w(x)$ satisfies the equation

$$w\frac{dw}{dx}-\frac{k-\gamma}{k+\gamma}x\left(\frac{dw}{dx}\right)^2+xw\frac{d^2w}{dx^2}=\frac{k+\gamma}{2\gamma}. \tag{11}$$

The solution of Eq. (11) can be obtained, for example, in the form of a power series in x:

$$w(x)=1+\sum_{n=1}^{\infty}c_nx^n, \tag{12}$$

where the coefficients c_n satisfy the recurrence relation

$$c_{n+1}=\frac{2k}{(1+n)^2(k+\gamma)}\sum_{i=1}^{n}i(n-i+1)c_ic_{n-i+1}-\frac{1}{2}\sum_{i=1}^{n}c_ic_{n-i+1},$$

$$c_1=\frac{k+\gamma}{2\gamma}. \tag{13}$$

Analysis of Eq. (13) shows that at $x<(1+k/2\gamma)^{-1}$ the series (12) is convergent.

It should be noted that the values of x corresponding to the instant of appearance of the cutoff are located in the region of convergence of the series (12) under the condition $\gamma m<2$, where m, as in (7), describes the curvature of the leading edge of the pulse of incident radiation.

The study of the dynamics of nonlinear absorption at $\gamma\lesssim 1$ shows that in general it is analogous to the case $\sigma=$ const. Thus, for example, the maximum value of the light intensity which can be obtained in passage through a nonlinear layer depends on the intensity of the incident radiation according to the law [compare with (9)]

$$I_{max}\propto I_0^{1/[m(k+\gamma)+1]}.$$

3. DYNAMICS OF THE NONLINEAR ABSORPTION OF A FOCUSED BEAM

We considered above the dynamics of the absorption of a non-focused beam. However, in studies of the processes of nonlinear absorption, and, in particular, processes of decay, high intensities are usually required. These intensities are achievable with the help of focusing of the incident light beam inside the investigated material. In such a formulation of the problem. the question arises so to what maximum intensities can be achieved in the sample as function of the conditions at the input surface, and what is the magnitude of the energy release in the interaction region.

The problem will be solved in the geometric-optics approximation, without account of processes of self-focusing of the light. Taking into account the initial focusing, the equation for the intensity I along the trajectory of the ray takes the form

$$\frac{\partial I}{\partial z}=\frac{2I}{R-z}-\sigma NI. \tag{14}$$

Here R is the radius of curvature of the wave front of the beam at the entrance to the medium. With the help of the substitution $\tilde{I}=I/(1-z/R)^2$ and the introduction of the new variable $x=1/(1-z/R)^{2k-1}-1$ the system of equations for $\tilde{I}$ and N can be solved by the method described above. The solution has the form

$$I(z,t)=I_0\varphi(t)\left(1-\frac{z}{R}\right)^{-2}\times\left\{1+\frac{k\sigma\beta RI_0^k\xi(t)}{2(2k-1)}\left[\left(1-\frac{z}{R}\right)^{1-2k}-1\right]\right\}^{-2/k} \tag{15}$$

$$N(z,t)=\beta I_0^k\xi(t)\left(1-\frac{z}{R}\right)^{-2k}\times\left\{1+\frac{k\sigma\beta RI_0^k\xi(t)}{2(2k-1)}\left[\left(1-\frac{z}{R}\right)^{1-2k}-1\right]\right\}^{-1}. \tag{16}$$

We note that although (15) is valid only in the region $(R-z)>l_c$ (l_c is the length of the region of the caustic of the focused beam), it can be used for the estimate of the maximum intensity also in the region of the caustic if we set $z=R-l_c$.

Analysis of Eq. (15) shows that the distribution of the intensity along the beam axis has a maximum (focus) whose position on the z axis is given by the expression

$$\left(1-\frac{z}{R}\right)^{2k-1}=\frac{(k-1)\sigma\beta RI_0^k\xi(t)}{2(2k-1)-k\sigma\beta RI_0^k\xi(t)}. \tag{17}$$

It is seen from this expression that at sufficiently high intensities of input radiation I_0 there exists an instant of time t_c such that at $t<t_c$ the maximum of the intensity (the focus) is located in the region of the caustic, while at $t>t_c$ it begins to be shifted toward the entry surface. Under the condition $\alpha\beta RI_0^k\xi(t)\gg 2$ the maximum intensity is always located at the entry surface. Thus the nonlinear absorptions significantly weakens the focusing of the light and can change appreciably the maximum values of the intensity in comparison with linear optics. It is obvious that if $t_c>\tau_0$ (τ_0 is the duration of the leading edge of the pulse), then the absolute maximum of the intensity is realized in the caustic at the instant of time $t\approx\tau_0$ and differs little from the value given by linear optics. In the opposite case, $t_c<\tau_0$, the effect of nonlinear absorption is large. We estimate the value of the maximum intensity in this case. For this purpose, we assume that the leading edge of the pulse has the form $\varphi(t)=t^m$. From the condition $\partial I/\partial t=0$, we obtain the peak value of the intensity I_a and the time of cutoff t_0:

$$I_a=\frac{I_0t_0^m}{(1-z/R)^2}\left(\frac{km+2}{2km+2}\right)^{2/k},$$
$$t_0^{km+1}=\frac{2(2k-1)m(km+1)}{(km+2)\sigma\beta RI_0^k[(1-z/R)^{1-2k}-1]}. \tag{18}$$

It is seen from this that the greatest shortening of the pulse occurs in the caustic of the lens and the absolute maximum intensity is reached there. Thus, within the time of the pulse τ_p the nonlinear focus (maximum intensity along z) shifts into the region of smaller z as time passes and leaves the caustic of the lens; its absolute value here falls off but the maximum value I is achieved in the caustic of the lens at the leading edge of the pulse.

Since greatest interest attaches to the near-focal region, assuming $1 - z/R \ll 1$, we obtain the following value of the absolute maximum of the intensity:

$$I_{max} = \left(\frac{km+2}{2km+2}\right)^{(km+2)/k(km+1)} \left[\frac{m(2k-1)}{\sigma\beta R}\right]^{m/(km+1)} \times \frac{I_0^{1/(km+1)}}{(1-z/R)^{(m+2)/(km+1)}} \quad (19)$$

Thus, whereas linear optics yields $I_{max} = I_0(l_c^2/R^2)^{-1}$, in nonlinear absorption, I_{max} is determined by the slope of the leading edge of the pulse and by the nonlinearity of the absorption, and $I_{max} \propto [I_0(l_c/R)^{-(m+2)}]^{1/(km+1)}$.

Since the processes of nonlinear absorption can be accompanied by a significant energy release, it is of interest to estimate the value of the absorbed energy density P. In the region $1 - z/R \ll 1$, we have

$$P = \int_0^{\tau_p} \sigma N I \, dt = \frac{2(2k-1)\tau_p}{k(R-z)^{1+1/k}} \left(\frac{4k-2}{k\sigma\beta\tau_p}\right)^{1/k} \int_0^{\infty} \frac{A^{1+1/k}\xi(t)\varphi(t)\,dt}{[1+A\xi(t)]^{1+2/k}}, \quad (20)$$

$$A = k\sigma\beta R I_0^k \tau_p / 2(2k-1)(1-z/R)^{2k-1}.$$

Analysis of (20) shows that in the case of strong absorption $A\xi \gg 1$, the maximum of the quantity P is achieved in the caustic, for all real values of the parameters β and σ, and its value depends on the shape of the pulse and the value of k. We give data on the value of the integral in (20) for a bell-shaped pulse:

$$\varphi(t) = (t/\tau_p)^2 \exp\{1-(t/\tau_p)^2\}. \quad (21)$$

Upon change of k from 2 to 7, the maximum value of the integral changes in the limits 0.6–0.8 while the quantity A, which corresponds to its maximum, changes from 25 to 120 in the same case. Upon further increase in the intensity of the incident radiation, the quantity P_{max} falls off slightly.

It should be kept in mind that Eqs. (6) and (7) were obtained with neglect of processes of self-action, in particular, the effect of self-focusing by the free carriers. Estimate of the carriers density corresponding to the self-focusing threshold can be obtained from the simple relation[6] $\varepsilon'/\varepsilon_0 \approx \Theta^2$, where θ is the focusing angle of the incident light and ε' is the contribution of the free carriers to the permittivity of the material.

4. EFFECT OF RECOMBINATION ON THE DYNAMICS OF NONLINEAR ABSORPTION

The investigations set forth above were performed without account of the recombination of the free charge carriers, i.e., $\beta I^k \ll N/\tau$. In the case of a plane layer, this condition will have the form

$$\frac{\varphi^k(t)}{1+\xi(t) z k\sigma\beta I_0^k/2} \gg \frac{\xi(t)}{\tau}. \quad (22)$$

Analysis of this expression shows that upon increase in the quantity zI_0^k, the effect of recombination manifests itself mainly at the trailing edge of the pulse $\varphi(t)$. At the same time, the effect of recombination on the leading edge of the front $\varphi(t)$ is much weaker and up to the instant of the appearance of cutoff it is determined by the condition $t/\tau \ll 1 + km/2$. Here, as in (18), m is determined by the slope of the leading edge of the pulse.

For an explanation of the physical meaning of the condition (22), we carry out the solution for very short recombination times $\tau \ll \tau_p$ (quasistationary regime):[7]

$$I_{st} = \frac{I_0\varphi(t)}{[1+kz\tau\sigma\beta I_0^k\varphi^k(t)]^{1/k}}.$$

We note that the condition $I \gg I_{st}$ is similar to the condition (22) and its violation in some region (z,t) means that in this region the effect of the recombination becomes substantial.

At small recombination times, we can obtain a general solution for the system of equations (1) by the method of successive approximations. Taking the first terms of the expansion in powers of τ into account, we obtain

$$I = \frac{I_0\varphi(t)}{[1+A\varphi^k(t)]^{[1-\exp(-t/\tau)]/k}} \exp\frac{\tau\,\partial\varphi/\partial t}{\varphi(t)[1+A\varphi^k(t)]},$$

$$N = \frac{\beta\tau I_0^k\varphi^k(t)}{1+A\varphi^k(t)}\left\{1-e^{-t/\tau} - \frac{k\tau\,\partial\varphi/\partial t}{\varphi(t)[1+A\varphi^k(t)]}\right\},$$

where $A = kz\tau\sigma\beta I_0^k$. It is seen from these equations that a limitation on the intensity, and consequently on the density of free charge carriers, occurs for the transmitted radiation. The crest of the transmitted pulse is flattened.

A similar picture exists also in the case of focused beams.

5. TWO-PHOTON LIGHT ABSORPTION IN GERMANIUM.

As shown in Ref. 8, in the propagation of powerful infrared radiation with $\lambda = 2.76\,\mu$, two-photon generation of electron-hole pairs takes place in germanium ($\beta = 1.3\times 10^9$ $W^{-2}s^{-1}cm^{-3}$). Assuming that the absorption takes place by the free holes ($\sigma = 2\times 10^{-16}$ cm^2), for a bell-shaped laser pulse (21) with duration $\tau_p = 10^{-7}$ we obtain from (5) that a germanium layer of thickness 1 cm loses its transparency at $I_0 \geqslant 10^7$ W/cm^2. At the same intensities, the distribution of the density of free charge carriers over the thickness of the sample ceases to be uniform and at large distances from the entry surface of the sample the density does not exceed $N = 1/\sigma z \approx 10^{16}$ cm^{-3}.

We now consider the case in which the radiation is focused inside the smaple by a lens with focal length $R = 1$ cm. At a caustic length $l_c = 2\times 10^{-2}$ cm,[9] estimates of the maximum carrier density give $N_{max} = 3/\sigma l_c \approx 10^{18}$ cm^{-3}; the greatest energy-release density in the caustic does not exceed $P_{max} \approx 500$ J/cm. The values mentioned are reached at an intensity of the initial radiation $I_0 > 1.4\times 10^5$ W/cm^2. It should be noted that the foregoing analysis was carried out without account of the processes of self-focusing at the free carriers and their diffusion from the interaction region. The effect of these processes also decrease the value of the maximum energy release, which means, in turn, the absence of damage in the case of two-photon excitation in germanium.[1]

Thus, in the present work we have obtained analytic expressions for the dependence of the intensity of the caustic of the lens on the intensity of the incident radiation, and which also allow the determination of the maximum energy-release density and free-carriers density in multiphoton ionization in transparent dielectrics. Knowledge of these values is necessary, both in the investigation of multiphoton ionization and in the study of processes accompanying it, such as laser damage and self-action of light.

[1)] A similar absence of damage was observed earlier in CdS crystals and has also been attributed to the effect of self-focusing.[10]

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