Soliton self-formation in the medium with nanorods under conditions of the absorption spectrum finite width

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Abstract. We investigate numerically an influence of the absorption spectrum width and the absorption spectrum central frequency shift on novel type of solitons (chirped soliton) appearance at a femtosecond pulse propagation in a medium with noble nanoparticles. We take into account the TPA of laser radiation by nanorods, and time-dependent nanorod aspect ratio changing due to their melting or reshaping because of laser energy absorption. The chirped solitons are formed due to the trapping of laser radiation by the nanorods reshaping fronts, if a positive or negative phase-amplitude grating is induced by laser radiation. We consider both linear and strong nonlinear dependences of absorption coefficient on the nanorod aspect ratio. We discuss a physical mechanism of the absorption spectrum width influence on the accelerating soliton formation.

1. Introduction
The influence of the noble metal nanoparticle aspect ratio and orientation, and local environment on the nanoparticle optical response has been widely investigated in recent years [1-12] because of nanorods using in many novel technologies. For example, nanoparticles aspect ratio changing because of photo-thermal reshaping provides the ability for five-dimensional recording [1-6]. Other very important question is the physical mechanism for photo-thermal reshaping of nanorods or wires at the temperatures less than the bulk melting [13-16]. Some models are proposed up to now to explain this phenomenon [15,16].

Among a number of problems of laser radiation interaction with a medium containing nanorods, the self-similar mode of laser pulse propagation is of great importance. This is due to the fact that the laser radiation spectrum distortions taking place due to nonlinear refraction can cause false information recording in all-optical data storage devices. Indeed, nanoparticles reshaping due to the laser radiation absorption leads to the pulse spectrum changing caused by the pulse chirping. This results in violation of optimal conditions for information recording, as well as (that is more essential) in the false information recording and reading. That is why the investigation of the laser pulse interaction with the medium, containing nanoparticles, is an actual problem.

In our previous papers [17-20] we investigated a femtosecond pulse propagation in a medium with nanorods under the conditions of nanorod aspect ratio changing and dependence of laser radiation absorption on nanoparticle aspect ratio. In particular, using the density matrix formalism, we derived the equation set which describes the femtosecond pulse propagation in the medium with nanorods reshaping, and obtained some analytical formulas for such propagation [17]. We also demonstrated the possibility for the superluminality effect implementation [21-22] in such media and discussed a
physical mechanism of chirped soliton formation and light acceleration. In particular, we showed that 
the light acceleration takes place for a chirped incident pulse and demonstrated that a nonlinear 
frequency chirp is crucial for a self-similar mode realization in a medium with nanorods [18]. We 
analyzed the influence of the relation between the nanorods absorption spectrum bandwidth and laser 
pulse spectrum bandwidth on the laser pulse spectrum transform [19] and the laser pulse acceleration 
[20] under its propagation in a medium containing nanorods.

Our attention in this paper is attracted by an influence of the absorption spectrum central frequency 
on the effects of superliminality and soliton formation at laser pulse propagation in a medium with 
nanorods under the condition of a narrow bandwidth of the pulse spectrum.

2. Problem statement

We consider a femtosecond laser pulse propagation in a medium with nanorods by taking into account 
the nanorod aspect ratio changing due to nanorod reshaping because of TPA of optical energy. In the 
framework of slowly varying envelope of wave packet, this process can be described by the following 
dimensionless nonlinear equations:

$$\frac{\partial A}{\partial z} + iD \frac{\partial^2 A}{\partial t^2} + f(\varepsilon)(\delta_0 + i\tilde{\xi})|A|^2 \cdot A \cdot \int_{-\infty}^{\infty} \delta(\omega)|A_{\omega}|^2 d\omega = 0 ,$$  

$$\frac{\partial^{\circ} E}{\partial^t} = \tilde{\delta} f(\varepsilon)|A|^4 \int_{-\infty}^{\infty} \tilde{\delta}(\omega)|A_{\omega}|^2 d\omega .$$  

with initial and boundary conditions for complex amplitude and nanorod aspect ratio

$$A(z,0) = A(z,L_z) = 0, \quad 0 \leq z \leq L_z , \quad A(0,t) = A_0(t), \quad 0 \leq t \leq L_t , \quad \varepsilon(z,t = 0) = \varepsilon_0, \quad 0 \leq z \leq L_z .$$

Above $A$ is dimensionless slowly varying envelope of a wave packet normalized on a square root 
from the maximum incident pulse intensity ($z = 0$). $\varepsilon = a/b$ is a nanorod aspect ratio, where $a$ and $b$ 
are major and minor axes of nanorods, $\varepsilon_0$ is its initial value. Variable $z$ is a dimensionless longitudinal 
coordinate, along which the optical radiation propagates; $t$ is a dimensionless time in the system of 
coordinates moving with a pulse, time is measured in the units of $\tau_{pulse}$ - duration of the incident 
pulse; $L_z$ is a dimensionless time interval, during which the laser pulse interaction with nanorods is 
analyzed, $L_t$ is a dimensionless length of nonlinear medium. Parameter $D$ characterizes the group 
velocity dispersion (GVD). Parameters $\delta_0$ and $\tilde{\delta}$ characterize the absorption of laser light and a part 
of absorbed energy spent on nanorods reshaping, correspondingly. Coefficient $\tilde{\xi}$ characterizes the 
laser pulse self-action due to the wave packet carrier frequency detuning from the central frequency of 
the nanorod absorption spectrum. The case of $\tilde{\xi} = 0$ corresponds to an optical pulse propagation in a 
medium with pure amplitude grating. It means an influence only of a laser energy absorption on the 
laser pulse propagation. In the opposite case ($\tilde{\xi} \neq 0$), the phase grating is also induced by the laser 
radiation. It should be mentioned, that the positive sign of the parameter $\xi$ (this case is named by us 
as positive grating) corresponds to pulse compression and the laser pulse decompression occurs at 
negative sign of this parameter (this case is named by us as negative grating).
The function $f(\varepsilon)$ describes the dependence of TPA process on the nanorod aspect ratio $\varepsilon$. Below we consider two possible types of this dependence:

\begin{align}
  f(\varepsilon) &= \varepsilon - 1 \\
  f(\varepsilon) &= \exp(5(\varepsilon - 2)), \quad \varepsilon \leq 2.7,
\end{align}

These dependences approximate the dependence

\begin{equation}
  f(\varepsilon) = \left( \frac{\varepsilon_2 / A^2}{\varepsilon_1 + \frac{1 - A}{A} \varepsilon_m^2} \right) + \varepsilon_2^2,
\end{equation}

\begin{equation}
  A = \left[ 1 - \frac{\xi Q_1(\xi)}{Q_1(\xi)} \right]^{-1}, \quad \xi = \left[ 1 - \left( \frac{1}{\varepsilon} \right)^2 \right]^{-1/2}, \quad Q_1(\xi) = \left( \frac{\xi}{2} \right) \ln \left[ \frac{\xi + 1}{\xi - 1} \right] - 1, \quad \frac{dQ_1(\xi)}{d\xi},
\end{equation}

which follows from Boyd and Shen shape factor expression [23] for the absorption coefficient of gold nanorods, calculated in the dipole approximation [24] for physical parameters $\varepsilon_1 = -22.4, \varepsilon_2 = 1.8$ for Au at $\lambda = 800$ and $\varepsilon_m = 3$. In Eq.(5)-(6), $\varepsilon_m$ is the dielectric permittivity of the ambient medium, $\varepsilon_1 + i \varepsilon_2$ is the complex dielectric permittivity of the nanorods.

Figure 1. Dependence $f(\varepsilon)$ defined by formulas (5)-(6) for gold nanorods at the falling radiation with $\lambda = 800$ nm (solid line), or by formula (3) (dashed line), or by formula (4) (dotted line).

If the aspect ratio varies from 1 to 2, then a laser radiation interaction with nanorods occurs far from the nonlinear absorption resonance. In this case, a linear dependence (3) adequately approximates dependence (5)-(6) (figure1, dashed line). Strongly non-linear dependence (4) can be used to approximate the dependence (5)-(6) if the aspect ratio changes from 1 to 2.6 (2, dotted line). The expression (4) is more preferable for a laser pulse interaction with nanorods near the nonlinear absorption resonance. Below we consider both dependences and choose initial value of aspect ratio as $\varepsilon_0 = 2$ for the dependence (4) and $\varepsilon_0 = 2.6$ for the dependence (5).

In Eqs. (1)-(2), we also take into account the absorption spectrum bandwidth influence on a laser energy absorption. At writing the spectral absorption coefficient $\delta(\omega)$, we consider the Lorentz law for the absorbance cross-section on light frequency. Hence, it is written as

\begin{equation}
  \delta(\omega) = \frac{1}{1 + 4 \left( \frac{\omega_0 - \omega}{\Delta \omega} \right)^2},
\end{equation}

with the absorption spectrum width and the central absorption frequency denoted as $\Delta \omega$ and $\omega_0$, respectively. Obviously, the absorption spectrum central frequency $\omega_0$ does not influence TPA, if the absorption spectrum width is many times larger than the laser pulse spectrum width. Indeed, in this case.
case the integral dependence in (1) and (2) can be omitted because the second item in the denominator of the expression (6) becomes negligible. So, it is possible to choose \( \omega_0 = 0 \) for the wide absorption spectrum. Below we show that the central absorption frequency \( \omega_0 \) greatly influences the process of soliton formation if the absorption spectrum is comparable with the pulse spectrum or narrower.

We specify the following complex amplitude of incident pulse

\[
A(z=0,t) = A_0(t) = \exp\left(-((t-L_z/2)/\tau)^{m}\right), 0 \leq t \leq L_z,
\]

\( \tau \) is a dimensionless pulse duration, \( m = 2 \) describes the Gaussian pulse shape.

We also follow the laser pulse centre position

\[
\tau_c(z) = \int_0^{L_z} (t-L_z/2) |A(z,t)|^2 \, dt \int_0^{L_z} |A(z,t)|^2 \, dt
\]

3. Computer simulation results

Below we present computer simulation results for the incident Gaussian pulse \( (m=2, \tau = 1) \) propagation at \( D = 0.1 \) and positive phase-amplitude grating \( \xi = 5 \), and sufficiently small strength of reshaping \( \delta = 5 \) for both types (3) and (4) of the absorption coefficient dependence on aspect ratio. Let us mention that the considered value of GVD corresponds to the pulse duration about 500 fs. We consider narrow absorption spectrum with bandwidth \( \Delta \omega = 1 \), which is less than the pulse spectrum width and three values of the central absorption frequency \( \omega_0 = [0, 1, -1] \) with respect to the carrier frequency of a wave packet.

Before we discuss the computer simulation results, let us note that the absorption spectrum bandwidth \( \Delta \omega \) affects the pulse spectrum changing and the pulse propagation only if it becomes comparable with the initial spectrum width. This is valid for all signs of the phase grating as well as for the pure amplitude grating [19]. Moreover, as it was demonstrated in [20], if the absorption spectrum bandwidth is comparable with the incident pulse spectrum bandwidth or narrower than it, a light acceleration is not so pronounced as in the case of the absorption spectrum that is much wider than the incident pulse spectrum. The pulse splitting also depends on the absorption spectrum bandwidth: it occurs only for a wide spectrum or for the spectrum comparable with the incident pulse spectrum bandwidth. If the absorption spectrum is narrower than the incident pulse spectrum, no pulse splitting occurs, and the pulse propagates as a whole at the velocity corresponding to the pulse front propagation in a linear medium.

![Figure 2](image-url)

Figure 2. Pulse centre shifting along the z-coordinate (a), maximal intensity evolution (b) for the three values of the central absorption frequency \( \omega_0 \) (shown in Figure), positive phase amplitude grating \( \xi = 5 \) and \( \delta_0 = 0.005, \delta = 5, \Delta \omega = 1 \).
3.1. Soliton formation far from nonlinear absorption resonance
We provide computer simulations for the linear dependence (3) of absorption coefficient on nanorod aspect ratio which is valid far from the nonlinear absorption resonance. We considered a small depletion ($\delta_0 = 0.005$) of laser energy due to TPA at a short distance ($z \approx 1$). It should be mentioned that the considered values of phase-amplitude grating and energy depletion correspond to the dimensionless frequency detuning parameter $\theta = (2\omega_p - \omega_{21}) T_\perp = \frac{\xi}{\delta_0} = 10^3$, which is much bigger than the dimensionless value of $\omega_p T_\perp \approx 236$, corresponding to $\lambda = 800$ nm (we consider $T_\perp \approx 10^{-13}$ s [25]). Nevertheless, this parameter values are very important for theoretical consideration.

As it is well seen in figures 2-4, if the central absorption frequency belongs to low frequency area ($\omega_0 = -1$) then the pulse acceleration enhancing occurs in comparison with its zero-value ($\omega_0 = 0$). Shift of the central absorption frequency to the high frequency area ($\omega_0 = 1$) results in the pulse acceleration decreasing.

![Figure 3](image3.png)

**Figure 3.** Soliton propagation in the medium with positive phase grating $\xi = 5$, $\delta_0 = 0.005$, $\tilde{\delta} = 5$ for $\Delta \omega = 1$, $\omega_0 = 0$ (a), -1(b).

![Figure 4](image4.png)

**Figure 4.** Maximal intensity position at the incident Gaussian pulse propagation in the medium with positive phase grating $\xi = 5$ and $\delta_0 = 0.005$, $\tilde{\delta} = 5$ for $\Delta \omega = 1$, $\omega_0 = 0$ (a),1(b), -1 (c). The solid lines at the top panels show soliton shapes at the exit section. Dashed line shows pulse shape at its propagation in linear medium (a) or aspect ratio distribution (b, c) at the exit section.
Note, that in the considered case, the soliton position coincides with the pulse front position at its propagation in a linear medium without nanorods (dashed line in figure 4a), if the central absorption frequency is equal to zero ($\omega_0 = 0$). The pulse compression observed at the initial stage of propagation is due to the positive phase grating action and small laser energy depletion (figure 2b). Moreover, a negative value of the central absorption frequency ($\omega_0 = -1$) results in a larger average maximal intensity of the soliton compared to its positive value (compare lines 1 and -1 in figure 2b).

These laser pulse propagation peculiarities can be explained using the physical mechanism of the soliton acceleration due to the laser radiation trapping by the nanorods reshaping front [17]. The trapping of laser radiation is well-seen in figures 4b, 4c, in which the dashed lines show the nanorod aspect ratio distribution. Indeed, as is well seen in figure 5b, up to the section $z=30$, the most part of the pulse leaves the absorption area in the frequency range if $\omega_0 = 1$. So, after this section, the pulse acceleration stops because the slow waves, which are at the pulse front due to the positive phase grating action, are no longer absorbed. If the central absorption frequency is negative ($\omega_0 = -1$), the pulse remains in the frequency area corresponding to a laser energy absorption for a larger distance of
propagation, and therefore its acceleration is greater (figure 5c). As a result, the pulse possesses a higher velocity.

3.2. Soliton formation for strong nonlinear dependence of laser energy absorption

We explore the strong nonlinear dependence (4) of the laser energy absorption on the nanorod aspect ratio for its description near the nonlinear absorption resonance. For example, we choose a maximal value of the absorption coefficient as $\delta_0 = 0.1$. In this case, the dimensionless frequency detuning parameter $\theta = (2\omega_p - \omega_2)T_\perp = \xi / \delta_0 = 50$ corresponds to the frequency detuning of $\approx 0.2\omega_p$ for $\omega_p \approx 2360$ THz ($\lambda = 800$ nm). This frequency detuning is available in physical experiments.

Figure 6. Soliton propagation in the medium with positive phase grating $\xi = 5$ and $\delta_0 = 0.1$, $\bar{\delta} = 5$ for $\Delta \omega = 1$, $\omega_0 = 0$ (a), -1(b).

Figure 7. Maximal intensity position at the incident Gaussian pulse propagation in the medium with positive phase grating $\xi = 5$ and $\delta_0 = 0.1$, $\bar{\delta} = 5$ for $\omega_0 = 0$, $\Delta \omega = 1000$ (a), $\omega_0 = -1$, $\Delta \omega = 1$ (c) near the nonlinear absorption resonance. The solid lines at the top panels show soliton shapes at the exit section. Dashed line shows pulse shape at its propagation in a linear medium (a) or aspect ratio distribution (b, c) at the exit section.
Figure 8. Pulse centre shifting along z-coordinate (a), maximal intensity evolution (b) for positive phase grating \( \xi = 5 \) and \( \delta_0 = 0.1, \tilde{\delta} = 5 \). The values of \( \Delta \omega \) and \( \omega_0 \) are shown in the figure by the pairs of numbers (the first number is for \( \Delta \omega \) and the second is for \( \omega_0 \)).

Figure 9. Pulse spectrum at the incident Gaussian pulse propagation in the medium with positive phase grating \( \xi = 5 \) and \( \delta_0 = 0.1, \tilde{\delta} = 5 \) for \( \Delta \omega = 1000 \) (a), 1(b,c) and \( \omega_0 = 0 \) (a,b), -1(c) and near the nonlinear absorption resonance. Dashed lines show the absorption spectrum.
Let us notice that the function $f(\varepsilon)$ in the form (4) instead of (3), does not influence on the dimensionless frequency detuning $\theta$, but this function type strongly changes the optical energy absorption and the strength of induced phase grating, and, thus, decrease the propagation distance at which the effects under consideration can be observed.

In this case, the multiple solitons formation occurs (figures 6, 7). Nevertheless, similar to the previous case of the dependence (3), the absorption spectrum bandwidth influence on the sub-pulses acceleration becomes noticeable only for the absorption spectrum bandwidth which is comparable with the incident pulse spectrum. Also, the acceleration of the sub-pulses decreases and their number decreases with the absorption spectrum bandwidth decreasing (compare figures 7a and 7b; solid and dashed-dotted curves in figure 8a, marked 1000,0 and 1.0, respectively).

The action of the central absorption frequency position in a spectral domain on the sub-pulses accelerating is also similar to the previous case (section 3.2): its negative value ($\omega_0 = -1$) promotes a pulse acceleration in comparison with its zero-value ($\omega_0 = 0$) (figures 6, 7b,c). But this promotion is much less pronounced then in the case of the linear dependence (3) (compare figures 2a and 8a). Indeed, similar to the previous case, the laser energy absorption occurs for the larger propagation distance if $\omega_0 = -1$ (figures 9b,c). Therefore, the acceleration is more pronounced in this case. For the wide absorption spectrum, the sub-pulses do not leave the absorption area in spectral domain, so they achieve higher velocities (figure 9a).

Due to the stronger depletion of light energy and the nonlinear function $f(\varepsilon)$, there is no compression of the light pulse at the initial stage of propagation and the maximum intensities of the sub-pulses at the medium exit are less 0.2 dimensionless units (figure 8b).

4. Conclusions
We investigated the influence of the central absorption frequency on the process of soliton self-formation and light acceleration for the absorption far and close to the nonlinear absorption resonance. This influence occurs mainly if the absorption spectrum bandwidth is compared to the incident pulse spectrum bandwidth.

We showed that the number of self-formed sub-pulses depends on the absorption spectrum bandwidth: it decreases with the absorption spectrum bandwidth decreasing. In particular, if the absorption spectrum is narrower than the initial pulse spectrum, no pulse splitting occurs for the absorption which is far from the nonlinear absorption resonance.

Acceleration of the sub-pulses is the most pronounced if the absorption spectrum is much wider than the incident pulse spectrum. For the narrow absorption spectrum, the shift of its central absorption frequency into the area of lower frequencies promotes acceleration of the sub-pulses, while the shift into the area of higher frequencies results in a lower light velocity.

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