Soliton Formation and Superluminality Effect due to Nonlinear Absorption of Femtosecond Laser Pulse Energy by the Medium Containing Nanorods

Vyacheslav A. Trofimov, Tatiana M. Lysak
Lomonosov Moscow State University, Moscow 119992, Russia

E-mail vatro@cs.msu.ru

Abstract. We investigate a femtosecond pulse propagation in a medium, containing nanorods, with taking into account the dependence of multi-photon absorption from the aspect ratio of nanorods. Nanorods melting due to the laser energy absorption leads to the non-stationary interaction of laser pulse with the medium and time-dependent nanorod aspect ratio changing. Under certain conditions, we found out the soliton-like mode of a laser pulse propagation and the superluminality effect: acceleration of light (fast light) in comparison with light propagation in a linear medium. We discuss a physical mechanism of superluminality effect for considering laser pulse propagation. Using spatio-temporal analogy, one can see the similarity between the pulse centre evolution along longitudinal coordinate and the beam centre evolution under the infrared optical radiation propagation in a cloud, or fog, which moves across the beam, with taking into account its thermal blooming.

1. Introduction
In recent years, thin films, doped with noble (gold or silver) metal nanoparticles, have attracted attention as recording media due to their strong nonlinear response resonance to the frequency of incident optical radiation [1-4]. Detailed understanding of laser pulse interaction with metal nanoparticles is of great interest for various fields of application. For example, nanoparticles aspect ratio changing because of photo-thermal melting provides the ability for five-dimensional recording [1,5]. A number of investigations are devoted to the measurements on a single-particle level, focusing on the dependence of nanoparticle optical response on the nanoparticle aspect ratio, orientation and local environment [6-11].

Among a number of problems of laser radiation interaction with the medium containing nanorods, self-similar mode of propagation is of great importance. This is due to the fact that the laser radiation spectrum distortions, because of nonlinear refraction, can cause false recording of information in all-optical data storage devices. Indeed, nanoparticles melting due to the laser radiation absorption leads to the laser pulse spectrum changing caused by the pulse chirping. This results in the condition violation for optimal 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 dynamic with the medium, containing nanoparticles, is an actual problem.

In our previous papers [12-14] we have investigated a femtosecond pulse propagation in a medium with nanorods under the conditions of both changing the ellipticity (aspect ratio) of nanorods and dependence of TPA from nanoparticle aspect ratio. We analyzed the influence of the relation between
the nanorods absorption spectrum bandwidth and laser pulse spectrum bandwidth on laser pulse
spectrum transformation under the pulse propagation in a medium containing nanorods, and found
that, under certain conditions, the laser pulse spectrum distortion is really absent despite the action of
various nonlinearities.

Our attention in this paper (see also [14]) is attracted by the effects of superliminality [15-19], as
well as a soliton-like formation of a laser pulse at its propagation in a medium with nanorods. Such
kind of the light propagation occurs if weak laser energy absorption takes place on the laser pulse
dispersion length. We also take into account one- or multi-photon of laser radiation by nanorods and
time-dependent nanorod aspect ratio changing due to their melting.

Due to the superluminality effect, the total time interval of light pulse interaction with nanoparticles decreases thus decreasing the
nonlinear response. In this paper, we compare the features of the superluminality effect in the media
with one-photon and multi-photon (three- and four-photon) absorption with the similar effect in the
media with two-photon absorption, discussed in [14].

The light acceleration effect takes place for a chirped incident pulse, and we confirmed this fact by
analytical consideration in [14]. Using spatio-temporal analogy, one can see the similarity between the
pulse centre evolution along longitudinal coordinate and the beam centre evolution at the infrared
optical radiation propagation in a cloud, or fog, which moves across the beam, under the condition of
thermal blooming [20-23]. As it is well known, the beam centre changing appears due to the refraction
of the beam on the thermal blooming grating and amplitude grating induced by nonlinear absorption of
laser energy by the cloud particles.

2. Problem statement
We consider a femtosecond laser pulse propagation in a medium with nanorods, taking into account
the aspect ratio (ellipticity) of nanorods changing due to their melting because of multi-photon
absorption (MPA) of optical energy. In the framework of slowly varying envelope of wave packet, this
process can be described by the following dimensionless nonlinear:

\[
\frac{\partial A}{\partial z} + iD \frac{\partial^2 A}{\partial t^2} + (\varepsilon - 1)(\delta_0 + i\xi)|A|^{2(k-1)} A = 0 ,
\]

\[
\frac{\partial \varepsilon}{\partial t} = -\tilde{\delta}(\varepsilon - 1)|A|^{2k}
\]

with initial and boundary conditions for complex amplitude and aspect ratio of nanorods

\[A(z,0) = A(z,L_z) = 0, \quad 0 \leq z \leq L_z, \quad A(0,t) = A_0(t), \quad 0 \leq t \leq t_L, \quad \varepsilon(z,t = 0) = \varepsilon_0, \quad 0 \leq z \leq L_z.\]

Above \(A\) is dimensionless slowly varying envelope of electric field pulse, normalized on the
maximum value of square root from the incident pulse intensity \((z = 0)\). The nanorods are melting under the action of laser light. As a consequence, their aspect ratio \((\varepsilon = a/b)\) is varied, where \(a\) and \(b\)
are major and minor axes of nanorods, \(\varepsilon_0\) is its initial value. Parameter \(k\) is the number of photons,
involved in the absorption. E.g., \(k=1\) is one-photon absorption, \(k=2\) is two-photon absorption
(TPA) and so on. 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 \(\tau_{\text{pulse}}\) - duration of the incident pulse; the dimensionless duration of laser
pulse is denoted as \(\tau_p\); \(t_L\) is a dimensionless time interval, during which the laser pulse interaction
with nanorods is analyzed, \(L_z\) 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. Coefficient \(\xi\) characterizes self-action of laser pulse due to detuning of the carrier
frequency of wave packet from the central frequency of the nanorod absorption spectrum. The case of \( \xi = 0 \) corresponds to an optical pulse propagation in a medium with pure amplitude grating. It means an influence only of absorption on the laser pulse propagation. In the opposite case (\( \xi \neq 0 \)), 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).

We specify the following complex amplitude of incident pulse

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

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

We also follow the laser pulse centre position

\[
\tau_c(z) = \int_0^{L/2} \left(\int_0^L [A(z, t)]^2 \, dt\right) \, dz,
\]

3. Analytical analysis

Below we develop the solution of equations (1)-(2) in the framework of nonlinear geometrical optics approximation [24] for one-photon absorption medium and a chirped incident pulse. This helps us to understand and explain the features of laser pulse propagation in a medium, containing nanorods, with a phase grating induced by laser radiation as well.

Let us represent the complex amplitude as \( A = \sqrt{I} \exp(-iS) \), \( S \) is a real function describing a pulse phase. In the new variables, these equations are transformed to

\[
\frac{\partial}{\partial z} I + 2D \frac{\partial}{\partial t} I \frac{\partial}{\partial t} S + 2D \frac{\partial^2}{\partial t^2} S + 2\delta_0 f I = 0,
\]

\[
\frac{\partial}{\partial z} S + D\left(\frac{\partial}{\partial t} S\right)^2 - D \frac{1}{\sqrt{I}} \frac{\partial^2 \sqrt{I}}{\partial t^2} - \xi f = 0,
\]

\[
\frac{\partial f}{\partial t} = -\delta I.
\]

Introducing the instantaneous frequency \( \Omega(z, t) = \frac{\partial S(z, t)}{\partial t} \) of the optical pulse and omitting the third term in equation (3) we get the following set of equations

\[
\frac{\partial}{\partial z} I + 2D \frac{\partial}{\partial t} I \left(\frac{\partial}{\partial t} \frac{\partial}{\partial t} \Omega\right) - 2\delta_0 \frac{\partial}{\partial \delta} f = 0,
\]

\[
\frac{\partial}{\partial z} \Omega + 2D \frac{\partial}{\partial t} \Omega - \xi \frac{\partial}{\partial t} f = 0,
\]

\[
\frac{\partial f}{\partial t} = -\delta I.
\]

Integrating equation (4) with respect to \( t \) and taking into account the zero-value pulse intensity till \( t \leq 0 \), we get

\[
\frac{\partial}{\partial z} P + 2D\Omega \frac{\partial}{\partial t} P - 2\delta_0 \frac{\partial}{\partial \delta} f_0 \left(\exp(-\delta P) - 1\right) = 0,
\]

\[
\frac{\partial}{\partial z} \Omega + 2D\Omega \frac{\partial}{\partial t} \Omega - \xi f_0 \left(\exp(-\delta P) - 1\right) = 0
\]

(5)
where $P(z,t) = \int_0 I(z,\tau)d\tau$, $I(z,t) = |A(z,t)|^2$, is the laser pulse energy, passed through a medium section $z$ till time moment $t$. We can solve these equations for a pure amplitude grating ($\xi = 0$). In this case, the solution of the Cauchy problem for these equations can be constructed [14] by the characteristics method

$$\Omega(z,t) = \hat{\Omega}_0(t - 2D\Omega z), \quad w(z,t) = \hat{w}_0(t - 2D\Omega z)$$

where $\hat{\Omega}_0(t) = \Omega(0,t) = \frac{\partial S(z = 0,t)}{\partial t}$ is an instantaneous frequency of the incident wave packet, and

$$w = \ln\left(2\delta_0 f_0 \left(\exp\left(\delta P\right) - 1\right) + 2\delta_0 f_0 z\right).$$

If the incident pulse is a chirped one

$$S(z = 0,t) \equiv \Omega_0(t + b)^2,$$

an instantaneous frequency of incident laser pulse is $\hat{\Omega}_0(t) \equiv 2\Omega_0(t + b)$ and the solution of equation (5) is

$$\Omega(z,t) \equiv \frac{2\Omega_0(t + b)}{1 + 4D\Omega_0 z}.$$

Therefore, the solution of equation (4) with respect to the beam intensity is

$$I(z,t) = \frac{I_0(\zeta)}{1 + \exp\left(-\delta P(\zeta)\right)} \left|\frac{1}{1 + 4D\Omega_0 z}\right|, \quad \zeta = \frac{t - 4D\Omega_0 z b}{1 + 4D\Omega_0 z b}. \tag{7}$$

Let us note, that formula (7) is invalid for $z$-sections satisfying inequality: $1 + 4D\Omega_0 z << 1$, because we derived (7) using the geometrical optics approximation.

A laser pulse shift in time coordinate depends on the sign of parameter $\Omega_0$. Let us remember that the initial pulse is up-chirped (has positive chirp) for positive value of $\Omega_0$ and down-chirped (has negative chirp) for negative value of $\Omega_0$. In figure 1 we compare the computer simulation results (solid lines) with the results obtained on the basis of analytical formula (7) derived for incident Gaussian pulse shape and quadratic law of the frequency distribution (6) (dashed lines). As we can clearly see, the solid and dashed lines are aligned at a distance of no more than 0.05 dimensionless units despite the negligible GVD under the analytical consideration.

It is well known that an initially up-chirped pulse, propagating in a linear medium with normal GVD, is broaden more rapidly, while a down-chirped pulse first is compressed and then it is broaden [25], and, of course, for both signs of $\Omega_0$ no pulse center shift is observed. In the medium, containing nanorods, the pulse can shift towards the area of time increasing or time decreasing. The nanorods
melting leads to the slowing down of the laser pulse propagation. On the other hand, the affect of nanorods melting on the incident down-chirped pulse can reverse the pulse center shifting direction and the laser pulse starts accelerating (figure 1). As it will be shown in the next Section 4, this effect is similar to a light propagation in a medium with a positive phase-amplitude grating (compare curves -1 in figure 1 and solid line 1 in figure 2a).

4. Computer simulation results
Below we present computer simulation results for the propagation of an incident Gaussian pulse \( m_r = 2, \tau = 1 \) at \( D = 0.1 \) and positive phase-amplitude grating \( h = 5 \), and sufficiently small strength of melting \( \delta = 5 \), and small enough depletion \( \delta_0 = 0.005 \) of laser energy under propagation on a short distance \( z \approx 1 \). We compare the process of soliton formation and light acceleration discussed in [14] for TPA of light energy with the similar superluminality effects for one-photon \( k = 1 \) and multi-photon \( k = 3, 4 \) absorption of light energy.

For all considered number of photons, involved in the process of absorption, the effect of pulse centre shifting in the direction of time decreasing takes place (figure 2a). Nevertheless, soliton formation occurs only for two- and three photon absorption (compare figures 3, 4, 8, 9).

Figure 2. Pulse centre shifting along z-coordinate (a), maximal intensity evolution (b) for the positive phase amplitude grating \( h = 5, \delta_0 = 0.005, \delta = 5 \). Dashed lines correspond to the pulse propagation in a linear medium. Number of photons, involved in the absorption process, is shown by figures.

Figure 3. Soliton formation (a), maximal intensity position for both sub-pulses (b) at the incident Gaussian pulse propagation for one-photon absorption \( k = 1 \) of light energy. The top plane in Figure b shows sub-pulse shapes at section \( z = 40 \).
In the case of one-photon absorption, the broadening of the initial pulse in the direction of time decreasing occurs with a well pronounced peak at the pulse front (figure 3). This broadening is accompanied by significant decreasing of maximal pulse intensity, which is much stronger than even the intensity decreasing for the pulse propagation in a linear medium (compare curve 1 and dashed line in Figure 2b).

If two photons are involved in the process of absorption, two solitons are formed as a result of the incident pulse splitting at the initial stage of propagation (figures 4,5). Maximal intensity growth occurs at the initial stage of propagation as a result of a nonlinear absorption. After splitting, the left sub-pulse transforms into a soliton and propagates with high velocity without its shape changing for at least 30 dimensionless units (figure 4b). The right sub-pulse also gradually transforms into a soliton. However, its velocity corresponds to the dispersion spreading of the pulse in a linear medium, while the left sub-pulse velocity exceeds it many times. Figure 5 clearly shows that the velocity of the right sub-pulse is close to the velocity of the pulse front motion, propagating in a linear medium.

As it was already mentioned, the positive phase grating action is similar to the incident chirped pulse propagation (see (6)) with $\Omega_0 < 0$. Moreover, in this case, the processes of higher frequency waves absorption due to nanorods melting and lower frequency waves generation due to the positive phase grating are not limited by rapid energy absorption. Therefore, the higher velocity sub-pulse (left sub-pulse in Figures 4,5) transforms into a soliton at its further propagation.

To clarify the soliton formation mechanism and the pulse acceleration we combined in figure 6 the pulse shape and frequency chirp $\frac{ds}{dt}$ ($s(t)$ is the pulse phase) with the aspect ratio distribution. Mention that the negative slope of a frequency chirp distribution in figure 6 corresponds to the positive chirp (up-chirp) in generally accepted terminology and is the result of our notations. At the initial stage of propagation, the pulse is splitting into two sub-pulses. A positive phase grating, which is formed at the pulse propagation, produces a negative chirp (see Section 3 above). This negative chirp compensates the positive chirp caused by the normal dispersion action. As a result, waves with lower frequencies go to the trailing edge of the pulse, and waves with higher frequencies – to its leading edge. This leads to the pulse compression, the wave packet power grows and the nanorods melting increases due to the high intensity waves absorption. So, the pulse splitting into two sub-pulses occurs. The waves with lower frequencies are concentrated in the left sub-pulse, while the waves with higher frequencies – in the right sub-pulse. So, the left sub-pulse moves with higher velocity than the right sub-pulse does. The further compression of the sub-pulses due to the action of positive phase-amplitude grating is restricted by the normal dispersion and energy absorption. So, after a certain distance of propagation, each sub-pulse is transformed into a soliton and becomes trapped at the boundary between domains with high and low nanorod aspect ratio. The soliton form of the self sub-pulse is also proved by figure 7, where the pulse spectrum is also split into two parts, the one with lower frequencies corresponds to the higher velocity soliton. It should be mentioned that, in figure 7, there are the frequencies with zero value spectral amplitude between the two parts of the spectrum. Up to the section $z = 30$, the right sub-pulse takes a soliton form as well (figures 4,5), being trapped by the melting front. Constant values of frequency chirp for both sub-pulses are well seen in figure 6b.

The influence of the positive phase grating results in a permanent shifting of higher frequency waves to the sub-pulse front, where they are absorbed by nanorods. So, the spectrum of both sub-pulses is permanently shifting into the range of lower frequencies, which is well seen in figure 6, especially for the high velocity left sub-pulse. As a result, the velocity of each sub-pulse is growing. In figure 4 the acceleration of the higher velocity left sub-pulse is also well seen. As for the right sub-pulse, its velocity is close to the velocity of the pulse front in a linear medium. Indeed, lower nanorod aspect ratio decreases the self-action in the area of the right sub-pulse. As a result, the sub-pulse compression decreases and the right sub-pulse trapping occurs at a lower peak intensity compared to the left sub-pulse. As it was already mentioned, the right sub-pulse consists of higher frequency waves compared to the left sub-pulse. That’s why its velocity is much smaller than the velocity of the left
sub-pulse, though it is bigger than the velocity of the pulse, propagating in a linear medium. As a result of both sub-pulses accelerating, the pulse center is shifting in the direction of time decreasing (fast light, line 2 in figure 2a).

![Figure 4. Soliton formation (a), maximal intensity position for both sub-pulses (b) at the incident Gaussian pulse propagation for TPA \((k = 2)\) of light energy. The top plane in Figure b shows sub-pulse shapes at section \(z=40\).](image4.png)

The similar process of soliton formation and light acceleration also takes place for laser pulse propagation in a medium with three-photon absorption (figure 8). Nevertheless, in comparison with TPA, pulse centre shifting is smaller, but the left sub-pulse maximal intensity is greater (figure 2). And the left sub-pulse does not transform into a soliton up to section \(z=40\). This is obviously due to the smaller effective strength of phase grating and laser energy depletion because of the fourth and the sixth power of the amplitude modulus in Equations (1) and (2), correspondingly.

Further photon number increase results in decreasing of the phase grating effective strength and laser energy depletion. So, the optimum ratio of GVD and strength of phase grating, necessary for soliton formation, is violated. As a result, after the pulse splitting, both sub-pulses start spreading and the decreasing of maximal intensity is similar to the one for the pulse propagation in a linear medium (figure 2b, compare curve 4 and dashed line). Nevertheless, even for \(k=4\), the pulse centre shift in the direction of time increasing occurs because of slow waves absorption for nanorods melting (figure 2a).

![Figure 5. Shapes of both sub-pulses combined at various sections for TPA \((k = 2)\) of light energy. Dashed lines correspond to section \(z = 40\) for the pulse, propagating in a linear medium.](image5.png)
Figure 6. Pulse shape (curve 1) and aspect ratio distribution (curve 2) (a); frequency chirp (curve 3) and aspect ratio distribution (curve 2) (b) at z=40 for TPA ($k = 2$) of light energy. Dashed lines show corresponding distributions for the pulse, propagating in a linear medium.

Figure 7. Spectrum of laser pulse propagating in a medium with TPA ($k = 2$) of light energy. Dashed lines correspond to the incident light spectrum.

Figure 8. Soliton formation (a), maximal intensity position for both sub-pulses (b) at the incident Gaussian pulse propagation for three-photon absorption ($k = 3$) of light energy. The top plane in Figure b shows sub-pulse shapes at section z=40.
5. Conclusions
Under the weak absorption on the dispersion length, we found out acceleration of light (fast light) in comparison with light propagation in a linear medium with one-photon and multi-photon (three- or four-photon) absorption.

The physical reason for laser pulse acceleration is the pulse chirping due to the induced positive phase-amplitude grating and absorption of light energy for nanorods melting which leads to non-stationary interaction of laser radiation with the medium and time-dependent changing of aspect ratio of nanorods. Acceleration of light is accompanied by the soliton formation due to the trapping of laser radiation by nanorods melting fronts for TPA and three-photon absorption. As a result of soliton formation, very slow decrease of maximal intensity occurs at the pulse propagation in the medium with TPA or three-photon absorption, especially in comparison with the pulse propagation in a linear medium.

Figure 9. Soliton formation (a), maximal intensity position for both sub-pulses (b) at the incident Gaussian pulse propagation for four-photon absorption (\(k = 4\)) of light energy. The top plane in Figure b shows sub-pulse shapes at section z=40.

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