Spectrum Evolution of Accelerating or Slowing down Soliton at its Propagation in a Medium with Gold Nanorods

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Abstract. We investigate both numerically and analytically the spectrum evolution of a novel type soliton - nonlinear chirped accelerating or decelerating soliton - at a femtosecond pulse propagation in a medium containing noble nanoparticles. In our consideration, we take into account one- or two-photon absorption 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. Accelerating or slowing down chirped soliton formation is accompanied by the soliton spectrum blue or red shift. To prove our numerical results, we derived the approximate analytical law for the spectrum maximum intensity evolution along the propagation coordinate, based on earlier developed approximate analytical solutions for accelerating and decelerating solitons.

1. Introduction
Noble metal nanoparticles reveal the unique optical properties, allowing their use in devices of ultrahigh density information storage [1-6]. The optical properties of metal nanoparticles are based on their high sensitivity to the wavelength and polarization of laser radiation. That is why the influence of the noble metal nanoparticle aspect ratio and orientation, and local environment on the nanoparticle optical response, is widely investigating nowadays [7-14].

Nanoparticles aspect ratio changing because of photo-thermal reshaping [15-19] provides the ability for five-dimensional recording [1-6]. Further development of five-dimension memory can be related to the information density increase, for example, due to the decrease of the space between the recording layers. In our opinion, the solution of this problem may be related to the self-similar mode of laser radiation propagation in the medium containing noble metal nanorods. Indeed, nanoparticles reshaping due to the laser radiation absorption leads to the laser pulse spectrum changing caused by the pulse chirping. These spectrum distortions can result in violation of optimal conditions for information recording, as well as (that is more essential) in the false information recording and reading.

In our previous papers [20-25] we investigated a femtosecond pulse propagation in a medium containing nanorods under conditions of non-instantaneous absorption due to the nanorods reshaping and dependence of laser radiation absorption on nanoparticle aspect ratio. In particular, we investigated accelerating and slowing down soliton mode of laser radiation propagation in the medium containing nanorods under one- and multi-photon absorption [21-24]. In papers [22] and
[23], we derived the approximate analytical solutions for these modes of laser radiation propagation, and compared these solutions with the computer simulation results. In numerical simulations, red or blue shift of the pulse spectrum was observed for accelerating or slowing down soliton propagation, respectively.

In present paper, we use the approximate analytical solutions, derived in [23] and [24] for accelerating and decelerating soliton modes of laser radiation propagation in the medium containing nanorods, to develop the analytical law for the soliton spectrum evolution observed in computer simulations.

2. Problem statement

We consider a femtosecond laser pulse propagation in a medium with nanorods, taking into account the nanorods aspect ratio changing due to their reshaping because of laser energy multi-photon absorption (MPA). In the framework of slowly varying envelope of wave packet, this process can be described by the following dimensionless nonlinear equations [21]:

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

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

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

\[ A(z,t)|_{z \to \pm \infty} = 0, \quad \frac{\partial A(z,t)}{\partial t}|_{z \to \pm \infty} = 0, \quad A(0,t) = A_0(t), \quad \varepsilon(z,t=0) = \varepsilon_0, \quad 0 \leq z \leq L. \]

Above A is a 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. \( k \) is the number of photons involved in the absorption: \( k = 1 \) corresponds to one-photon absorption, \( k = 2 \) - to two-photon absorption. 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; \( L_\varepsilon \) is a dimensionless time interval, during which the laser pulse interaction with nanorods is analyzed, \( L_\varepsilon \) 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 \( \xi \) 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 \( \xi = 0 \) 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 (\( \xi \neq 0 \)), 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 \( \varepsilon \). The Boyd’s and Shen’s shape factor expression [26] for the absorption coefficient of gold nanorods, calculated in the dipole approximation [27] for physical parameters \( \varepsilon_1 = -22.4, \varepsilon_2 = 1.8 \) for Au at \( \lambda = 800 \) and \( \varepsilon_m = 3 \), gives the following dependence
\[
f(\varepsilon) = \left(\frac{\varepsilon_2/A^2}{\varepsilon_1 + \frac{1-A}{A}\varepsilon_m}\right)^2 + \varepsilon_2^2.
\] (3)

\[
A = \left[1 - \frac{\xi Q_1(\xi)}{Q_1(\xi)}\right]^{-1}, \quad \xi = \left[1 - \left(\frac{1}{\varepsilon}\right)^2\right]^{-1/2}, \quad Q_1(\xi) = \left(\frac{\xi}{2}\right) \ln \left[\frac{\xi + 1}{\xi - 1}\right] - 1, \quad Q_1(\xi) = \frac{dQ_1(\xi)}{d\xi},
\] (4)

In equation (3), \(\varepsilon_m\) is the dielectric permittivity of the ambient medium, \(\varepsilon_1 + i\varepsilon_2\) is the complex dielectric permittivity of the nanorods. Below we consider the linear approximation of this dependence:

\[
f(\varepsilon) = \varepsilon - 1
\] (5)

The linear dependence (3) adequately approximates dependence (3)-(4) (figure 1, red line) if the aspect ratio varies from 1 to 2. Below we choose initial value of aspect ratio as \(\varepsilon_0 = 2\).

Figure 1. Dependence \(f(\varepsilon)\) defined by formulas (3)-(4) for gold nanorods at the falling radiation with \(\lambda = 800\) nm (black line), or by formula (5) (red 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 (5) (figure 1, red line) adequately approximates dependence (3)-(4) (figure 1, black line). Below we choose the following initial value of aspect ratios \(\varepsilon_0 = 2\).

We specify the incident Gaussian pulse

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

\(\tau\) is a dimensionless pulse duration.

We follow the laser pulse centre position along the z-coordinate

\[
\tau_z(z) = \int_0^{L_z} \left|A(z,t)\right|^2 dt / \int_0^{L_z} \left|A(z,t)\right|^2 dt
\]

and the maximum position for the laser pulse spectra along the z-coordinate.

\[
\omega_{\text{max}}(z) = \arg \max_{\omega} \left|A(z, \omega)\right|^2,
\]

which means that, for a fixed value of the variable \(z\), the function \(\left|A(z, \omega)\right|^2\) takes the maximum value for \(\omega = \omega_{\text{max}}(z)\).
3. Analytical consideration

In our previous papers [21-23], we investigated numerically and analytically the accelerating and slowing down solitons for the problem under consideration. In particular, we derived the approximate analytical solution, which describes the soliton propagation, and computed the soliton center shift along the z-coordinate \( \tau_c(z) \). In our investigations, the obvious shift of the soliton spectrum into the area of lower or higher frequencies was clearly seen. Present research is devoted to the derivation of the analytical expression for the maximum spectrum shift along the z-coordinate \( \omega_{\text{max}}(z) \).

In [22,23], we considered the following approximation of the accelerating or slowing down soliton

\[
A(z,t) = B(z) \exp(-is(z,\zeta)) \cdot \varepsilon(z,\zeta), \quad \varepsilon(z,\zeta) = e^{i(\tau_c(z) - t_0)},
\]

\[
s(z,\zeta) = a(z) + b(z)\zeta + h(z)\ln(\cosh(\zeta))
\]

\[
B(0) = B_0, \quad \tau_c(0) = \tau_{i0}.
\]

Here \( A(z,t) \) is the soliton complex amplitude, functions \( B(z), s(z,\zeta), \tau_c(z) \) and \( \tau_{i0} \) describe the soliton amplitude, phase distribution, and pulse center and its duration, respectively, with their initial values \( B_0, t_0 \) and \( \tau_{i0} \) for the time moment \( t = 0 \). Parameters \( a_0, b_0 \) and \( h_0 \) define the incident pulse phase distribution. Function \( a(z) \) describes a soliton phase shift along the z-coordinate, and is unchangeable in time. Functions \( b(z), h(z) \) describe the soliton frequency evolution along the propagation coordinate and a pulse chirp evolution, respectively.

Note that, as it follows from our approximate analytical solution for accelerating and decelerating soliton, the function \( h(z) \) does not depend on the z-coordinate, so that \( h(z) \equiv h_0 \) [22, 23]. In this case, one can write the following sequence of equalities for the Fourier transform \( A(z,\omega) \) of the soliton complex amplitude \( A(z,t) \):

\[
A(z,\omega) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} B(z) \exp(-is(z,\zeta)) \cdot \exp(i\omega t) dt = \frac{B(z)}{\sqrt{2\pi}} \int_{-\infty}^{\infty} \exp(-ia(z)) \cdot \exp(-i(b(z)\zeta + h_0\ln(\cosh(\zeta))) \cdot \exp(i\omega t) dt = \frac{B(z)}{\sqrt{2\pi}} \tau_c(z) \exp\left(-i\left(a(z) - \omega t_c(z)\right)\right) \times \int_{-\infty}^{\infty} \exp(-ib(z)\zeta + h_0\ln(\cosh(\zeta))) \cdot \exp(-i\omega\tau_c(z)) d\zeta = \frac{B(z)}{\sqrt{2\pi}} \tau_c(z) \exp\left(-i\left(a(z) - \omega t_c(z)\right)\right) \int_{-\infty}^{\infty} \exp(-ib(z)\zeta + h_0\ln(\cosh(\zeta))) \cdot \exp(i\omega(\tau_c(z) - b(z))\zeta) d\zeta
\]

Using (9), the spectrum intensity can be computed as

\[
|A_{\omega}(z,\omega)|^2 = B^2(z)\tau_c^2(z)F(\omega\tau_c(z) + b(z)), \quad F(x) = \left| \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} \exp(-ib\zeta + h_0\ln(\cosh(\zeta))) \cdot \exp(-ix\zeta) d\zeta \right|^2
\]

The maximum spectrum intensity position for the given medium section \( z = \text{const} \) is determined from (10) by the position of the function \( F(x) \) maximum.
\( \omega_{\text{max}}(z) \tau_s(z) - b(z) = \arg \max_x F(x). \)  \hspace{1cm} (11)

Eq. (11) means that the function \( F(x) \) takes the maximum value for \( x = \omega_{\text{max}}(z) \tau_s(z) - b(z) \).

Taking into account that the position of the maximum of the function \( F(x) \), determined by (10), does not depend on the \( z \)-coordinate, the following equality is valid

\[ \omega_{\text{max}}(z) \tau_s(z) - b(z) = \arg \max_x F(x) = \omega_{\text{max}}(0) \tau_s(0) - b(0). \]  \hspace{1cm} (12)

It is possible to obtain the dependence of the maximum spectra intensity position \( \omega_{\text{max}}(z) \) on the \( z \)-coordinate from equality (12) if the position \( \omega_{\text{max}}(0) \) of the maximum of the incident pulse spectrum intensity is known, as well as the dependences \( \tau_s(z) \) and \( b(z) \).

Obviously, the maximum spectra position of the incident Gaussian pulse spectrum in the problem under consideration is

\[ \omega_{\text{max}}(0) = 0 \]

In our approximation, the pulse duration is a constant value: \( \tau_s(z) = \tau_{s0} \) [22, 23]. So, equation (12) determines the sought-of dependence for the maximum intensity position of the soliton spectrum:

\[ \omega_{\text{max}}(z) = \frac{b(z) - b(0)}{\tau_{s0}}. \]  \hspace{1cm} (13)

Equation (13) shows that the soliton maximum spectrum intensity position does not depend on the soliton amplitude \( B(z) \) or the soliton center \( \tau_z(z) \), but it is determined by the pulse duration \( \tau_{s0} \) and the evolution \( b(z) \) of the soliton frequency along the propagation coordinate.

4. Computer simulation results

This section is devoted to the comparison of the soliton maximum spectrum intensity position evolution given by analytical prediction (13) and obtained in computer simulation. In section 4.1, this comparison is provided for decelerating soliton in the medium containing nanorods under one-photon absorption. In section 4.1, we compare the analytical and computer simulation results for accelerating soliton in the medium containing nanorods under two-photon absorption.

4.1. Deceleration soliton

In paper [22], we have shown that the decelerating soliton is formed in the medium containing nanorods under one-photon absorption and negative phase grating for parameters \( \delta_0 = 0.2, \xi = -5, \bar{\delta} = 5, D = 0.1 \) (figure 2). The soliton shape of the deceleration soliton is confirmed by its spectrum evolution: during the decelerating soliton propagation, the soliton spectrum preserves its shape [22] and shifts into the area of higher (lower) frequencies.

In our approximation (6)-(8), the soliton centre evolution (figure 2b) is described by the quadratic dependence

\[ t_c(z) = t_0 + \left( \frac{2D}{\tau_{s0}} b_0 + \delta_0 (\epsilon_0 - 1) \bar{\delta} \tau_{s0}^2 B_0^2 \exp \left( -\bar{\delta} B_0^2 \tau_{s0} \right) \right) z - \]
\[ - D \left( \xi - h \cdot \delta_0 \right) (\epsilon_0 - 1) \bar{\delta} B_0^2 \exp \left( -\bar{\delta} B_0^2 \tau_{s0} \right) z^2 \]  \hspace{1cm} (14)

In our approximation (6)-(8), the soliton centre evolution (figure 2b) is described by the quadratic dependence
Dependence (14) was obtained in paper [22] under assumption that the soliton amplitude remains the same for a large trace of propagation (it takes place, for example, if laser energy depletion is weak). This assumption is valid for the considered case. Under this assumption, the evolution of the soliton frequency along the z-coordinate is a linear function [22]

\[ b(z) = b_0 - \left( \xi - h \cdot \delta_0 \right) (\epsilon_0 - 1) \delta \tau_\alpha B^2 \exp \left( -\delta B^2 \tau_\alpha \right) z \]  

(15)

Substituting dependence (15) into equation (13), we obtain that the evolution of the position of the soliton maximum spectrum intensity is also a linear function

\[ \omega_{max}(z) = - \left( \xi - h \cdot \delta_0 \right) (\epsilon_0 - 1) \delta \delta B^2 \exp \left( -\delta B^2 \tau_\alpha \right) z \]  

(16)

As it follows from equations (14) and (16), the coefficient at \( z \) in equation (16) differ from the coefficient of the second degree of \( z \) in equation (16) by the multiplier \( D \).

![Figure 2](image1.png)  
**Figure 2.** Decelerating soliton shape (black solid line), incident Gaussian pulse shape (black dashed line) and aspect ratio distribution (red line) at the section \( z = 10 \) (a), maximal intensity position (b) at the incident Gaussian pulse propagation under the one-photon absorption in the medium with negative phase grating. Dashed line in the figure b is given by (17).

![Figure 3](image2.png)  
**Figure 3.** Decelerating soliton spectrum evolution (a), maximum spectrum intensity position (b) at the incident Gaussian pulse propagation under the one-photon absorption in the medium with negative phase grating. Dashed line in the figure b is given by (18).
For the decelerating soliton under consideration, the following values of approximation soliton (6) parameters follow from computer simulation results [22]

\[ b_0 = 0, \quad \tau_{x0} = 0.5, \quad B_0 \approx 0.668 \]

So, the dependences (14) and (16) take the following form

\[ t_c(z) = 50 + 0.3z^2 \quad (17) \]

\[ \omega_{\text{max}}(z) \approx 3z. \quad (18) \]

Dependence (17) was obtained in [22] and is shown in figure 2b by the red dashed line. Dependence (18) is depicted in figure 3b by the dashed line. Good coincidence with the computer simulation results is well seen in figure 3b.

Figure 3 shows the spectrum evolution for the decelerating soliton. As it is well seen, the soliton spectrum undergoes blue shifting – during the soliton propagation, its spectrum preserves its shape and shifts into the area of higher (slower) frequencies (figure 3a).

### 4.2. Acceleration soliton

In paper [21,23], we have shown that the accelerating soliton is formed in the medium containing nanorods under two-photon absorption and positive phase grating for the following parameters \( \delta_0 = 0.005, \quad \xi = 5, \quad \tilde{\delta} = 5, \quad D = 0.1 \) (figure 4).

The soliton shape of the left sub-pulse is confirmed by its spectrum evolution (figure 5): the left part of the spectrum gradually shifts into the area of lower frequencies (red shifting).

![Figure 4](image-url)

**Figure 4.** Accelerating sub-pulses shapes (black solid line) and aspect ratio distribution (red line) at the section \( z = 40 \), incident Gaussian pulse shape (black dashed line) (a), maximal intensity position (b) at the incident Gaussian pulse propagation under the two-photon absorption in the medium with positive phase grating. Dashed line in the figure b is given by (19).

The left sub-pulse acceleration can be described by the following quadratic dependence obtained in [23] (red dashed line in figure 4):

\[ t_c(z) = 215 - 3z - 0.05z^2 \quad (19) \]

The absolute value of coefficient at the second degree of \( z \) in dependence (19), is about five times smaller than the corresponding coefficient in our analytical dependence for the soliton centre evolution [23]
\[ t_s(z) = t_0 + 2 \frac{D}{\tau_{s,0}} \xi - D \xi (e_0 - 1) B_0^6 \exp \left( -2/3) \tilde{\delta} B_0^4 \tau_{s,0} \right) z^2, \quad (20) \]

calculated for approximation soliton parameters obtained in computer experiment for the left sub-pulse at section \( z=40 \):

\[ \tau_{s,0} = 0.3, \quad B_0 \approx 0.748. \quad (21) \]

Dependence (20) takes place if the soliton amplitude remains the same for a large trace of propagation which is valid for the parameters under consideration. Under this assumption, the evolution of the soliton frequency along the z-coordinate is a linear function [23]

\[ b(z) = b_0 - \xi (e_0 - 1) \tilde{\delta} \tau_{s,0} B_0^6 \exp \left( -2/3) \tilde{\delta} B_0^4 \tau_{s,0} \right) z \quad (22) \]

Substituting (22) into (13), one obtains the linear evolution of the maximum intensity position of the soliton spectrum along the z-coordinate

\[ \omega_{\text{max}}(z) = -\xi (e_0 - 1) \tilde{\delta} B_0^6 \exp \left( -2/3) \tilde{\delta} B_0^4 \tau_{s,0} \right) z \quad (23) \]

Taking into account parameters (21) of soliton approximation (6), it is possible to calculate the coefficient at \( z \) in (23), which absolute value occurs about five times bigger than the corresponding coefficient in the linear approximation

\[ \omega_{\text{max}}(z) = -0.66z \quad (24) \]

shown in figure 9b by the red dashed line.

As it follows from equations (20) and (22), the coefficient at \( z \) in formula (20) differ from the coefficient of the second degree of \( z \) in equation (22) by the multiplier \( D \). So, it is possible to use equation (19) to calculate the coefficient at \( z \) in dependence (23):

\[ \omega_{\text{max}}(z) = -0.5z \]

which is close to approximation (24).

Figure 5. Accelerating soliton spectrum formation (a), maximal spectrum intensity position (b) at the incident Gaussian pulse propagation under the two-photon absorption in the medium with positive phase grating. Dashed line in the figure b is given by (24).
5. Conclusions

We investigated numerically and analytically the spectrum evolution of the nonlinear chipped solitons - accelerating and slowing down solitons, which are formed at the laser radiation propagation in the medium containing nanorods. We considered decelerating soliton which is formed under one-photon absorption and accelerating soliton which is formed under two-photon absorption. Earlier we have shown that the spectrum of the accelerating soliton undergoes red shifting, while the spectrum of the slowing down soliton undergoes blue shifting.

Using the approximate analytical solution for accelerating and slowing down soliton, we derive the approximate analytical law for the spectrum maximum intensity evolution. We confirmed this law by the computer simulation results. It is essential that the spectrum maximum intensity evolution obeys a linear law with the negative or positive slope depending on the soliton type.

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