Effect of nonlinear dispersion on pulse self-compression in a defocusing noble gas

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\textbf{A B S T R A C T}

Media with a negative Kerr index ($n_2$) offer an intriguing possibility to self-compress ultrashort laser pulses without the risk of spatial wave collapse. However, in the relevant frequency regions, the negative nonlinearity turns out to be highly dispersive as well. Here, we study the influence of nonlinear dispersion on the pulse self-compression in a defocusing xenon gas. Purely temporal (1 + 1)-dimensional investigations reveal and fully spatio-temporal simulations confirm that a temporal shift of high intensity zones of the compressed pulse due to the nonlinear dispersion is the main effect on the modulational instability (MI) mediated compression mechanism. In the special case of vanishing $n_2$ for the center frequency, pulse compression leading to the ejection of a soliton is examined, which cannot be explained by MI.

\begin{figure}
\centering
\includegraphics[width=\textwidth]{figure1.png}
\caption{Schematic diagram of the experimental setup.}
\end{figure}

\section{1. Introduction}

The common property of laser pulse compression schemes is the utilization of nonlinear effects the pulse undergoes upon propagation. The omnipresent idea hereby is to broaden the pulse spectrum and ensure a flat spectral phase at the same time. Among others, such as frequency conversion in filaments \cite{1} or cascading quadratic nonlinearities \cite{2-4}, the spectral broadening ability of self-phase modulation (SPM) is often exploited in the guided configuration (waveguides, fibers). The resulting phase can be accounted for by applying some post-compression mechanisms such as Bragg gratings and chirped mirrors or, even more convenient, by a counteracting term on the phase contributions, that is in our case the group velocity dispersion (GVD). The simplest model that captures both processes is expressed by the famous nonlinear Schrödinger (NLS) equation for the slowly varying envelope of an optical pulse $A(z, t)$:

\begin{equation}
\frac{\partial A}{\partial z} = -i k_2 A + 2 + i \gamma |A|^2 A.
\end{equation}

Here, $k_2 = \frac{\partial^2 k(\omega)}{\partial \omega^2}|_{\omega=\omega_0}$ is the GVD coefficient at center frequency $\omega_0$ and $\gamma = n_2 \omega_0 / c$ is the Kerr coefficient with index $n_2 \propto \chi^{(3)}$ defined by the third order susceptibility tensor $\chi^{(3)}$. The standard case described above would be anomalous GVD ($k_2 < 0$) counteracting on positive Kerr response ($\gamma > 0$), since a negative second derivative of the wave number $k_2$ is more common than a negative $\gamma$. Trying to transfer this “solitary compression mechanism” to the unguided bulk setup gives rise to problems due to self-focusing and subsequent collapse of the beam in the transverse spatial directions. Resolving this problem and still sticking to the “solitary compression” (phase cancelation due to different signs of GVD $k_2$ and SPM $\gamma$) means to ensure a negative $\gamma$. Indeed, negative values for the Kerr response can be found, e.g., in the UV near two photon resonances in Xe (Ref. \cite{5}). However, the strong dispersion of the nonlinearity near these resonances questions the compression mechanism which is based on a constant nonlinearity. Nevertheless, it was shown recently \cite{6} and will be elucidated in more detail in this work, that the basic compression mechanism survives these obstacles. Moreover, we will present an alternative scenario where pulse compression is actually caused by the nonlinear dispersion, while the Kerr index $n_2$ approaches zero at center wavelength.

The paper is organized as follows. In Section 2, we give a short derivation of our wave equation, followed by a discussion of the temporal dynamics in a (1 + 1)-dimensional configuration in Section 3. Finally, in Section 4, results for the (3 + 1)-dimensional setup with radial symmetry are presented and discussed. Conclusions are drawn in the last section.

\section{2. Derivation of the governing equation}

\subsection{2.1. From Maxwell's to wave equations}

The propagation of light through a (nonlinear) optical medium is described by the macroscopic Maxwell’s equations. We assume

\begin{equation}
\frac{\partial E}{\partial t} = \nabla \times H - \frac{n^2}{c} \frac{\partial B}{\partial t}.
\end{equation}

Where $E$ is the electric field, $H$ is the magnetic field, $B$ is the magnetic induction, $n$ is the index of refraction, and $c$ is the speed of light. The term $\nabla \times H$ represents the magnetic field divergence and $\frac{\partial B}{\partial t}$ represents the magnetic field time derivative. The term $- \frac{n^2}{c} \frac{\partial B}{\partial t}$ represents the effect of the nonlinear medium on the magnetic field.

\begin{equation}
\frac{\partial B}{\partial t} = \nabla \times E - \frac{n^2}{c} \frac{\partial E}{\partial t}.
\end{equation}

Finally, the term $\nabla \times E$ represents the electric field divergence and $\frac{\partial E}{\partial t}$ represents the electric field time derivative. The term $\frac{n^2}{c} \frac{\partial B}{\partial t}$ represents the effect of the nonlinear medium on the electric field.

\begin{equation}
\frac{\partial H}{\partial t} = \nabla \times E.
\end{equation}

Where $\nabla \times E$ represents the electric field curl and $\frac{\partial H}{\partial t}$ represents the magnetic field time derivative. The term $\nabla \times E$ represents the effect of the nonlinear medium on the magnetic field.

\begin{equation}
\frac{\partial E}{\partial t} = \nabla \times H.
\end{equation}

Where $\nabla \times H$ represents the magnetic field curl and $\frac{\partial E}{\partial t}$ represents the electric field time derivative. The term $\nabla \times H$ represents the effect of the nonlinear medium on the electric field.

Therefore, we can write the wave equation as:

\begin{equation}
\nabla^2 E + \frac{n^2}{c^2} \frac{\partial^2 E}{\partial t^2} = 0.
\end{equation}

And the wave equation for $H$ is:

\begin{equation}
\nabla^2 H - \frac{n^2}{c^2} \frac{\partial^2 H}{\partial t^2} = 0.
\end{equation}

These wave equations are the basis for the derivation of the nonlinear Schrödinger equation.
the absence of free charges \( \rho \neq 0 \) and free currents \( \mathbf{j} \neq 0 \), a
nonmagnetic medium \( \mathbf{B} = \mu_0 \mathbf{H} \) and the material to be nonlinear in
the sense that \( D = \varepsilon_0 \mathbf{E} + \mathbf{P} \). It is common to express the polarization
\( \mathbf{P} \) in a power series in \( \mathbf{E} \); \( \mathbf{P} = \mathbf{P}^{(1)} + \mathbf{P}^{(3)} + \mathbf{P}^{(5)} + \ldots \), \( \mathbf{P}^{(3)} \sim \mathbf{E} \),
where we already accounted for the vanishing of contributions of
even power in \( \mathbf{E} \) because we are dealing with centrosymmetric media. In this work, we keep only terms up to third order and refer to \( \mathbf{P}^{(1)} = \mathbf{P}_L \) as the linear polarization and to \( \mathbf{P}^{(3)} = \mathbf{P}_N \) as the nonlinear part. Putting all together, we get a wave equation in a
form that is often used as starting point for further calculations

\[
\nabla^2 \vec{E} = \mu_0 \varepsilon_0 \frac{\partial^2 \vec{E}}{\partial t^2} + \mu_0 \frac{\partial^2 \mathbf{P}_L}{\partial t^2} + \mu_0 \frac{\partial^2 \mathbf{P}_N}{\partial t^2} + \nabla (\nabla \cdot \vec{E}).
\]

(1)

For small polarization \( |\mathbf{P}| \ll |\varepsilon_0 \mathbf{E}| \), \( \mathbf{E} \cdot \mathbf{E} \) is negligible. Furthermore, we restrict our analysis to linearly polarized light and disregard the
tensor nature of the material response. Together with the
paraxiality assumption, we can omit vector arrows and treat a scalar equation for the dominating field component. In Fourier
space, the linear polarization can be expressed as \( \mathbf{P}_L(\omega) = \varepsilon_0 \chi^{(1)}(\omega) \mathbf{E}(\omega) \) and after introducing the wave number \( k(\omega) = \sqrt{1 + \chi^{(1)}(\omega) / \varepsilon} \) we obtain the wave equation in Fourier space

\[
\nabla^2 \vec{k} + k^2(\omega) \vec{E} = -\omega^2 \mu_0 \mathbf{P}_N.
\]

(2)

2.2. Forward propagating equation for the complex field

In this work, we focus on the description of forward propagating
waves only, since backscattered waves as well as the coupling between
both directions are usually weak for our beam configurations [7]. We assume the field to propagate main along the
\( z \geq 0 \) direction and therefore decompose it into forward propagating
plane waves with wave numbers \( k(\omega) \) and the envelope \( \mathbf{A} \)

\[
\mathbf{E}(x, y, z, t) = \int \omega \mathbf{A}(x, y, z, \omega) e^{i[k(\omega)z - \omega t]} d\omega.
\]

(3)

Distinction of the directions by \( \nabla^2 \vec{k} + \omega^2 \vec{A} \) allows us to neglect the fast varying backward traveling part of \( \mathbf{A} \) by skipping terms \( \sim \omega^2 \) as long as the slowly varying envelope condition \( |\omega^2| \ll |k(\omega)| \) holds for the forward propagating part. Then, in Fourier space the so-called forward Maxwell equation [8] emerges naturally

\[
\hat{\omega} \mathbf{A} = \frac{1}{2\omega(\omega^2 - 1)} \mathbf{E} + ik(\omega) \mathbf{A} + \frac{1}{\varepsilon_0 \varepsilon_0} \mathbf{P}_N.
\]

(4)

Eq. (4) is in principle valid in the whole spectral domain. However, in this work, our radiation is limited to a finite frequency window around the laser operating frequency \( \omega_0 \). Hence, it is useful to Taylorize the dispersion relation

\[
k(\omega) = k_0 + \frac{k_1}{1!} \hat{\omega} + \frac{k_2}{2!} \hat{\omega}^2 + \ldots,
\]

(5)

with \( k_i = \partial \omega(k(\omega)) / \partial \omega \big|_{\omega=\omega_0} \) and \( \hat{\omega} = \omega - \omega_0 \). Moreover, for convenience in the numerics, we split fast oscillations in \( z \) and go over to a co-moving time frame, defined by \( t \rightarrow t + k L \). In Fourier space, this leads to the introduction of the (normalized) slowly varying complex envelope

\[
\tilde{E}(\hat{\omega}) = \Theta(\hat{\omega} + \omega_0) \frac{1}{s} \mathbf{E} + \omega_0 e^{i(k_0 \omega_0 + k L)}
\]

(6)

with \( |\omega|^2 = 1 \) being the laser intensity, \( \Theta(x) \) the usual Heaviside function, and \( n_0 = \varepsilon_0 \varepsilon_0 / \omega_0 \).

2.3. Including the nonlinearity

As for the linear polarization, the tensor nature of the nonlinear response function \( \chi^{(3)} \) is neglected. Furthermore, we are only interested in contributions having approximately the same frequency \( \omega_0 \) as our initial pulse and therefore omit terms responsible for higher order harmonics generation. Hence, the resulting expression for a dispersive third order nonlinear polarization in Fourier domain is

\[
n\mathbf{P}_N(\omega) = \varepsilon_0 S^{3/2} \int \omega_1 \omega_2 \chi^{(3)}(-\omega; \omega - \omega_1 - \omega_2, \omega_2, \omega_1) \]

\[
\times \mathbf{E}(\omega_1) \mathbf{E}(\omega_2) \mathbf{E}^*(\omega_1 + \omega_2 - \omega) e^{i(k_0 + k_1 \omega_0)}.
\]

(7)

where \( \sqimath \) means complex conjugate. With this expression, we can finally write down the propagation equation for the slowly varying complex envelope

\[
\hat{\omega}_1 \tilde{E} = \frac{1}{2\omega(\omega^2 - 1)} \mathbf{E} + ik(\omega) \mathbf{A} + \mathbf{P}_N(\omega) + \frac{3i\omega^2 s}{2k(\omega)c^2} \int \omega_1 \omega_2 \chi^{(3)}(-\omega; \omega - \omega_1 - \omega_2, \omega_2, \omega_1) \]

\[
\times \mathbf{E}(\omega_1) \mathbf{E}(\omega_2) \mathbf{E}^*(\omega_1 + \omega_2 - \omega).
\]

(8)

In the following analysis, we take the linear dispersion for Xe from Ref. [9] and the nonlinear dispersion \( \chi^{(3)} \) is given in Ref. [10].

3. (1 + 1)-dimensional setup

In a first approach we want to investigate purely temporal influence of the dispersive nonlinearity on pulse dynamics, thus skipping the transverse spatial derivatives (\( \nabla^2 \)) in Eq. (8). Even if the numerical solution of this equation is straightforward, it does not elucidate the underlying mechanisms for compression. In order to get some deeper insight, we include subsequently increasing orders of nonlinear dispersion, originating from a Taylor expansion around the central frequency \( \omega_0 \):

\[
\chi^{(3)}(-\omega; \omega - \omega_1 - \omega_2, \omega_2, \omega_1)
\]

\[
= \chi^{(3)}(\omega_0) + \chi_{1,3}^{(3)} \omega_1 + \chi_{2}^{(3)} \omega_2 + \chi_{3}^{(3)} \omega + \ldots,
\]

(9)

where \( \omega_0 = \omega_0 - \omega_0 \) and

\[
\chi_{0}^{(3)} = \chi^{(3)}(-\omega_0; -\omega_0, \omega_0, \omega_0),
\]

\[
\chi_{1,3}^{(3)} = \partial_{\omega_0} \chi^{(3)}(-\omega_0; -\omega_0 - \omega_1 - \omega_2, \omega_2, \omega_1)|_{\omega_0=\omega_0=\omega_0=\omega_0},
\]

\[
\chi_{3}^{(3)} = \partial_{\omega_0} \chi^{(3)}(-\omega_0; -\omega_0 - \omega_1 - \omega_2, \omega_2, \omega_1)|_{\omega_0=\omega_0=\omega_0=\omega_0}.
\]

By doing so, we are able to attribute specific effects to the corresponding orders of nonlinear dispersion. In a first step, we neglect nonlinear dispersion (\( \chi^{(3)} = 0 \)) and explain the basic mechanisms of compression occurring in this case. In a second step, first order terms of nonlinear dispersion are included and their influence upon compression is investigated. Finally, the results are compared with the ones obtained from the fully dispersive (1 + 1)-dimensional model.

As mentioned above, we are interested in setups with negative
\( n_2 \sim \chi^{(3)} \), which can be found near resonances in Xe [see Fig. 1]. So for the coming simulations, we introduce the abbreviation \( \omega_\infty = 7.73 \times 10^{15} \text{s}^{-1} \) for the characteristic central frequency ensuring a negative \( n_2 \) and additionally \( \omega_0 = 7.91 \times 10^{15} \text{s}^{-1} \) for investigations with vanishing \( n_2 \). That is, a laser pulse with central frequency \( \omega_0 = \omega_\infty \) experiences defocusing as well as effects originating from nonlinear dispersion, whereas a pulse with central frequency \( \omega_0 = \omega_0 \) lacks the usual Kerr effect \( n_2 = 0 \) and only undergoes nonlinear dispersive modulations.
previously made approximations. It turns out (see Section 3.1.2. The NLSN Dequation since \( \omega \) pulse compression. Note that on this strong approximation of the effects, which can be used to estimate suitable input conditions for propagation. SincetheNLSequation offers a rich variety of propagation effects, where we introduced the nonlinearsusceptibility \( \chi^{(3)} \) for Xe Ref. [10].

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>( k_2 (s^2/m) )</td>
<td>( 7.6 \times 10^{-28} )</td>
</tr>
<tr>
<td>( \text{Re}[\chi^{(3)}] (m^2/V^2) )</td>
<td>( -1.7 \times 10^{-24} )</td>
</tr>
<tr>
<td>( \text{Im}[\chi^{(3)}] (m^2/V^2) )</td>
<td>( 1.0 \times 10^{-27} )</td>
</tr>
<tr>
<td>( n_2 (cm^2/W) )</td>
<td>( -0.47 \times 10^{-17} )</td>
</tr>
<tr>
<td>( p_0 (W) )</td>
<td>( 2.0 \times 10^3 )</td>
</tr>
<tr>
<td>( \text{Re}[\chi^{(3)}] (m^2/V^2) )</td>
<td>( 6.0 \times 10^{-29} )</td>
</tr>
<tr>
<td>( \text{Im}[\chi^{(3)}] (m^2/V^2) )</td>
<td>( -3.0 \times 10^{-42} )</td>
</tr>
<tr>
<td>( \text{Re}[\chi^{(3)}] (m^2/V^2) )</td>
<td>( 6.2 \times 10^{-39} )</td>
</tr>
<tr>
<td>( \text{Im}[\chi^{(3)}] (m^2/V^2) )</td>
<td>( -3.1 \times 10^{-42} )</td>
</tr>
<tr>
<td>( \text{Re}[\chi^{(3)}] (m^2/V^2) )</td>
<td>( 1.7 \times 10^{-40} )</td>
</tr>
<tr>
<td>( \text{Im}[\chi^{(3)}] (m^2/V^2) )</td>
<td>( 1.0 \times 10^{-43} )</td>
</tr>
</tbody>
</table>

3.1. Model equations

3.1.1. The NLS equation

We now summarize approximations to Eq. (8) without the transverse Laplacian and here deal with the one dimensional nonlinear Schrödinger (NLS) equation. First, only the leading order of linear dispersion is kept, that is \( k(\omega) - k_0 - \kappa \omega \approx k_2 \omega^2 / 2 \). Second, we neglect nonlinear dispersion and due to the smallness of the imaginary part (see Table 1) we have \( \chi^{(3)} (\omega - \omega_1 - \omega_2, \omega_1, \omega_2) \approx \text{Re}[\chi^{(3)}] \). Finally, we neglect the frequency dependency of the prefactor in front of the nonlinearity. Transforming back to time domain gives

\[
\partial_t \xi = -i \frac{k_2}{2} \partial_z^2 \xi + i \frac{\omega_0}{c} n_2 |\xi|^2 \xi,
\]  

(10)

where we introduced the nonlinear refractive index \( n_2 = 3 \text{Re}[\chi^{(3)}] / 4n_0^2 e c \). Eq. (10) is the well known nonlinear Schrödinger equation. Since the NLS equation offers a rich variety of propagation effects, we have to identify our simulation parameters which lead to the desired compression. We therefore present two well studied effects, which can be used to estimate suitable input conditions for pulse compression. Note that on this strong approximation of the pure NLS equation we will treat the case \( \omega_0 = \omega - n_2 \omega_0 < 0 \) only, since \( n_2 \) vanishes for \( \omega_0 = \omega_0 \).

3.1.2. The NLSND equation

In order to get a qualitative idea of the action of nonlinear dispersion, we use the expansion of the nonlinear susceptibility \( \chi^{(3)} \) up to first order derived in Eq. (9) and keep all other previously made approximations. It turns out (see Table 1) that \( \text{Re}[\chi^{(3)}] \ll |\chi^{(3)}| \). Additionally, \( |\chi^{(3)}| \ll |\chi^{(3)}| \) and since \( \Delta \omega \sim \Delta \omega_1, \omega_2 \), the dispersive nonlinearity can be further simplified to

\[
\chi^{(3)} (-\omega, \omega - \omega_1 - \omega_2, \omega_1, \omega_2) \approx \text{Re}[\chi^{(3)}] + \text{Im}[\chi^{(3)}] \omega_1 + \text{Im}[\chi^{(3)}] \omega_2.
\]  

(11)

With this simplification, transforming back to time domain leads to

\[
\partial_t \xi = -i \frac{k_2}{2} \partial_z^2 \xi + i \gamma |\xi|^2 \xi - \delta |\xi|^2 \partial_z \xi,
\]  

(12)

where we introduced \( \delta = 3 \omega_0 (|\chi^{(3)}| + 3 |\chi^{(3)}|) / 4 |n_0^2 e c \). The new term \( \sim \delta \) (compared to the NLS equation) is a so-called wave-breaking term. It acts like an intensity-dependent group velocity and shifts zones with higher intensity stronger to the front/rear of the pulse, depending on the sign of \( \delta \). It is interesting to note that a similar term can be obtained for nondispersive nonlinearities beyond the slowly varying envelope approximation, but with much smaller prefactor.

3.2. Estimating compression parameters via modulational instability (MI)

The phenomenon of modulational instability [11,12] is responsible for an inherent instability of the propagation of a continuous wave background for certain values of \( k_2 \) and \( n_2 \), and originates from the interplay of nonlinear (SPM) and dispersive (GVD) effects. This instability (MI) leads to the splitting of the continuous wave into a train of pulses with a well defined period usually fixed by the maximum of the instability growth rate. That property will be exploited to approximate the simulation parameters needed for compression.

Applying a standard linearization approach [13] to Eqs. (10) and (12) for the perturbed steady state

\[
\xi = (\sqrt{\xi_0} + p(z, t)) e^{i\gamma \xi z},
\]  

(13)

where \( \xi_0 \) is the background intensity and \( p \) the perturbation expressed as

\[
p(z, t) = a_1 e^{i(\xi_2 - \Omega t)} + a_2 e^{-i(\xi_2 - \Omega t)} \quad (a_1, a_2 \ll \sqrt{\xi_0})
\]  

(14)

leads to the dispersion relation for the perturbation wavenumber \( K \) and perturbation frequency \( \Omega \)

\[
K = \delta \xi_0 \Omega \pm \sqrt{\frac{k_2 \Omega^4}{4} + k_2 \gamma |\xi_0| \Omega^2}.
\]  

(15)

Temporal modulations with \( \Omega \) grow, whenever the corresponding wavenumber \( K \) possesses imaginary contributions. These define the instability growth rate (or gain)

\[
g(\Omega) = \Im \left( \frac{k_2 \Omega^4}{4} + k_2 \gamma |\xi_0| \Omega^2 \right),
\]  

(16)

which reaches its maximum for

\[
\Omega = \Omega_{\text{max}} = \pm \sqrt{\frac{2|\gamma| |\xi_0|}{k_2}}.
\]  

(17)

Since our center frequency \( \omega_0 \) determines the values of \( k_2 \) and \( \gamma \), we are able to calculate the necessary intensity for a fixed, desired modulation frequency \( \Omega_{\text{max}} \) to occur. To achieve single peak pulse compression, this frequency \( \Omega_{\text{max}} \) should be of the order of the inverse initial pulse duration. Of course, this can only be a rough approximation, because our input pulse is far from a constant background. However, those estimates turn out to be useful and quite reliable when compared to simulation results (see Section 3.4).

It is worth noticing, that the growth rate \( g(\Omega) \) and therefore also the optimum frequency \( \Omega_{\text{max}} \) are equal for both cases of NLS and NLSND. This is approved numerically by plotting the initial (\( z = 0 \), red curves) and propagated (\( z = 0.04 \), blue...
curves) spectral intensity for the NLS and NLSND (see Fig. 2(a) and (b), respectively). The only difference becomes apparent in the dispersion relation Eq. (15), where the perturbation wavenumber \( K \) is purely real (propagating perturbations) or purely imaginary (growing perturbations) for the NLS (\( \delta = 0 \)), whereas \( K \) always has a real contribution \( \sim \delta k_0 \Omega \) in the NLSND case. These contributions correspond to additional transverse velocities of the growing perturbations oscillating at \( \Omega \), resulting in their temporal shift upon propagation, which is numerically demonstrated in Fig. 2(c) and (d).

Since we are able to tune the laser center frequency \( \omega_0 \) and can therefore adjust \( \gamma \sim \omega_0 k_0^2 \), we can reach propagation regimes, in which the effect of MI is not occurring, namely in the case where \( \gamma \approx 0 \) (\( \omega_0 \approx \omega_{0p0} \)). Then no statements concerning the pulses dynamics can be derived in the framework of MI theory. Nevertheless, as will be seen below, even in this case pulse self-compression is possible due to soliton formation.

### 3.3. Pulse compression via soliton dynamics

Another effect based on the interplay of SPM and GVD is the possibility of soliton formation in the NLS equation (see, e.g., Ref. [13]). The solitons with initial sech shape are characterized by a single integer, the soliton order

\[
N = \sqrt{\frac{|\gamma| \omega_0^2}{k_2}} \tag{18}
\]

where \( \tau_0 \) is the pulse duration and \( I_0 \) is the peak intensity. For \( N = 2, 3, \ldots \) the evolution patterns are known: the initial shape periodically undergoes several modulations, including compression at certain propagation distances. Moreover, these solitons are stable against perturbations on the soliton order \( N \) [14] as well as against perturbations of the initial pulse shape. That is, a perturbed input pulse relaxes to the evolution pattern of the corresponding soliton upon propagation. So we can make use of the stability by fixing the soliton order \( N \) (and therefore the approximate evolution pattern) and subsequently estimating our input intensity \( I_0 \) from Eq. (18) for given \( \tau_0 \). In order to achieve compression, we want to have single peaked wave forms preserving their shape over long distances of propagation, so we should choose neither a too high \( N \) (strong compression over short propagation distance) nor a too low one (weak compression for long distances). Of course, the obtained intensity \( I_0 \) is a lower boundary, since the energy dispersed away during the soliton formation process is no longer available for the soliton itself. The actual estimation is carried out below.

Concerning the NLSND equation, it is known [15] that Eq. (12) still allows soliton solutions. These solitons naturally coincide with the ones for NLS equation for \( \delta \to 0 \) and are therefore expected to be stable against perturbations as well. In the other limit case with \( \gamma = 0 \) (and \( \delta \neq 0 \)) the amplitude of the soliton solution is given by

\[
|E_{\text{sol}}| = \frac{2\lambda k_0/\delta}{\sinh^2(\lambda t) + \cosh^2(\lambda t)}, \tag{19}
\]

with \( \lambda \) the family parameter which determines the intensity peak value and duration. Interestingly, the soliton fluence is a constant and can be evaluated as \( E_{\text{sol}} = \int |E_{\text{sol}}|^2 dt = \pi k_0 / \delta \). An estimate for the minimal input fluence a Gaussian pulse has to carry for exciting a soliton follows from \( E_{\text{sol}} > E_{\text{Gauss}} \), giving the condition \( \sqrt{2\pi k_0 / \delta} < k_0 \tau_0 \).

### 3.4. Simulation results

#### 3.4.1. NLS equation

Let us now present some results for the simulation of the NLS equation. We assume Gaussian input pulses

\[
E(z = 0) = \sqrt{I_0} \exp(-t^2 / \tau_0^2), \tag{20}
\]

with intensity \( I_0 \) and pulse duration \( \tau_0 \) at center frequency \( \omega_0 \).

First, we follow the approach to estimate optimum compression parameters via MI, according to Eq. (17). Choosing \( \omega_0 = \omega_{0p0} \) and \( \tau_0 = 100 \) fs implies \( k_0 = 7.6 \) fs/cm and \( \gamma = 1.2 \times 10^{-14} \) m/W. Now we demand \( \Omega_{\text{min}} = 2\pi / \tau_0 \sqrt{2 \ln 2} = 5.3 \times 10^{13} \) s\(^{-1} \), which gives \( I_0 = 9.0 \times 10^{14} \) W/m\(^2 \) for the initial intensity. Our second estimation is based on Eq. (18). For \( N = 4 \), we find with the same parameters as above \( I_0 = 1.0 \times 10^{14} \) W/m\(^2 \) for the initial intensity.
Compared to numerical simulations, the above estimates give reasonable values for the optimum field intensity. For $t_\text{0} = 100$ fs and $\omega_0 = \omega_\infty$, we find effective compression at $l_\text{0} = 2.1 \times 10^{14}$ W/m$^2$. Simulation results are summarized in Fig. 3(a) and (d). Fig. 3(a) shows the intensity plotted against propagation distance $z$ and time $t$. The propagation pattern can be determined to correspond to a 4th order soliton, undergoing the typical periodic modulations. Thus the dynamics can be clearly attributed to soliton propagation dynamics, giving that compression scheme its name. Fig. 3(d) details the intensity of the initial pulse at $z = 0$ m and after $z = 1.4$ m of propagation. The full-width at half-maximum intensity (FWHM) is $\tau_\text{FWHM} \sim 120$ fs at $z = 0$ m and $\tau_\text{FWHM} \sim 10$ fs at $z = 1.4$ m, which corresponds to temporal compression by a factor of 12.

3.4.2. NLSND equation

Let us now confront the above predictions on NLSND solutions with numerical simulations. As before, we simulate Gaussian input pulses with intensity $I_\text{0}$, pulse duration $\tau_\text{0}$ and central frequency $\omega_0$. First we follow the former approach to estimate parameters by MI and as mentioned earlier, obtain the same parameters as in the previous Section 3.4.1. For comparability with results from NLS we use again $I_\text{0} = 2.1 \times 10^{14}$ W/m$^2$ for the intensity at $t_\text{0} = 100$ fs for the pulse duration and $\omega_0 = \omega_\infty$ as central frequency. The simulation results are presented in Fig. 3(b) and (d). The temporal dynamics in (b) show the evolution pattern for a fourth order soliton which shifts to later times upon propagation, which can be attributed to the action of the wave-breaking term $\sim \delta$. Fig. 3(d) shows the initial pulse and the temporal profile at $z = 1.4$ m. The initial pulse is compressed from $\tau_\text{FWHM} = 117$ fs down to $\tau_\text{FWHM} = 8$ fs at $z = 1.4$ m, that is compression by a factor of 15. Again, the action of the additional intensity-dependent group velocity is apparent, since the peak value is occurring at later times. Furthermore, the peak value is somewhat decreased compared to the pure NLS case.

3.4.3. (1 + 1)-dimensional full model equation

In this section, simulation results for the fully dispersive (linear and nonlinear) wave Eq. (8) without transverse spatial dimensions are presented. The same simulation parameters as in the corresponding setup for NLSND are used.

Fig. 3(c) and (d) show results for Gaussian input pulse with intensity $I_\text{0} = 2.1 \times 10^{14}$ W/m$^2$, pulse width $\tau_\text{0} = 100$ fs at central frequency $\omega_0 = \omega_\infty$. Temporal dynamics in Fig. 3(c) reveal the splitting of the initial pulse into a singly peaked waveform which is undergoing slight modulations in the peak value and shifted to later times upon propagation. According to the last sections, these modulations can be attributed to the soliton character of the evolution, whereas the shift to later times is due to corrections from the nonlinear dispersion. Fig. 3(d) shows, that the pulse is compressed by a factor of 9 from initially $\tau_\text{FWHM} = 117$ fs down to $\tau_\text{FWHM} = 12.5$ fs at $z = 1.4$ m.

It is important to underline that, as Fig. 4(d) and (e) reveal, the pulse has contributions in the spectral range where the nonlinearity exhibits resonances, so that the whole model becomes questionable. Moreover, due to numerical issues, these resonances cannot be resolved properly in the simulations, hence the results at propagation distances where the spectrum hits some resonances may not be reliable. Nevertheless, one can estimate the shortest pulse duration for which its spectrum stays in the nonresonant range. Starting from a center frequency $\omega_0 = \omega_0$ and assuming the allowed spectral width to be twice the distance to the nearest resonance at $7.54 \times 10^{13}$ s$^{-1}$, we get a minimal pulse duration of $\tau_\text{min} \approx 15$ fs. So what can be claimed is, that upon propagation the pulse shortens at least down to 15 fs.

However, the similarity of the temporal dynamics for NLSND and for the full (1 + 1)-dimensional equation indicates, that the obtained dynamics are qualitatively correct and the error originating from the resonances is small. Thus, the main action of nonlinear dispersion can be summarized in shifting the maximum intensity region of the pulse to later times without altering the solitary character of the compression scheme.

3.5. Pulse compression at vanishing $n_2$

The original situation we are interested in is the propagation regime where $\gamma \approx 0$, which corresponds to a central frequency $\omega_0 = \omega_0$ ($\lambda_0 = 238$ nm as central wavelength).

If we fix the pulse duration to $t_\text{0} = 100$ fs and employ the NLSND model, we get from the above considerations an estimate for the minimum intensity of $I_\text{0} = 3.7 \times 10^{14}$ W/m$^2$ necessary to trigger soliton formation. This is a lower boundary for the intensity and in order to see the ejection of a soliton from the input pulse...
we have to choose our initial intensity somewhat higher in the simulations, namely $I_0 = 1.9 \times 10^{15}$ W/m$^2$.

The temporal dynamics obtained in Fig. 4(a) show the splitting of the initial pulse into a mainly single peaked structure which is shifted to later times upon propagation. From Fig. 4(c) we estimated the peak intensity and the FWHM pulse duration and compared these with the values for the soliton and were able to validate the assumption of a soliton being emitted from the initial pulse.

Further simulations suggest that it should be possible to produce solitons with shorter widths by just increasing the input intensity. However, at some point Eq. (12) boarders its validity, since for shorter pulses higher order linear and nonlinear dispersion start to come into play.

Let us return to the full model equation. Fig. 4(b) and (c) show the results for Gaussian input with intensity $I_0 = 1.9 \times 10^{15}$ W/m$^2$, pulse duration $\tau_0 = 100$ fs at central frequency $\omega_0 = \omega_0(k)$. The results are in good qualitative agreement with the ones for the NLS equation. Temporal dynamics in Fig. 4(b) show the ejection of the soliton whose width is determined from the cut in Fig. 4(c) at $z = 2.4$ m to be $\sim 12.5$ fs. In this setup, the pulse spectral intensity (see Fig. 4(e), green dashed line) does not reach frequencies where $\chi^3$ is resonant, and the found dynamics can be trusted.

Another point concerns the higher order nonlinearities $P^{(5)} \sim \alpha^5$, ..., since the cubic term $P^{(3)}$ is considerably smaller in the vicinity of $\omega_0(k)$ at which $n_2$ almost vanishes. Nevertheless, it can be shown, that the length scales on which specific orders act can be estimated by $|P_{(p)}|/|P_{(3)}| \approx |P^{(5)}|/|P^{(3)}| \approx |E/E_0|^2$ where $E$ is the laser field and $E_0 = 5 \times 10^{10}$ V/m [16]. We used field strengths $E \approx 0.4 \times 10^{10}$ V/m, giving $|P_{(5)}|/|P_{(3)}| \approx 2 \times 10^{-4}$, so $P^{(5)}$ is negligible. Furthermore, the pulses launched at $\omega_0(k)$ do have a spectral width, thus experiencing, e.g., a $n_2 \approx 2.7 \times 10^{-23}$ m$^2$/W at the half-maximum frequency due to dispersion of the nonlinearity, which is already comparable to usual values of $n_2 \sim 1.0 \times 10^{-23}$ m$^2$/W. Therefore, neglect of higher order nonlinearities is still justified for center frequencies close to $\omega_0(k)$.

4. (3 + 1)-dimensional setup in radial symmetry

In this section, we want to investigate the full spatio-temporal propagation dynamics of Eq. (8) in radial symmetry. As before, we use a simplified equation, the (3 + 1)-dimensional NLS equation, to estimate simulation parameters for the full equation. Once obtained, we check whether temporal compression still occurs with dispersive nonlinearity. Finally, we try to transfer the new compression mechanism found for $\chi \approx 0$ in the (1 + 1)-dimensional case to bulk configuration.

4.1. NLS equation

Let us first estimate parameters suitable for pulse compression in Eq. (8). For this purpose, a simplified (3 + 1)-dimensional NLS equation is used, whose approximate dynamics can be described analytically by a two scale variational approach, followed by a standard MI analysis which finally gives the desired parameters.

The (3 + 1)-dimensional NLS equation is obtained by including transverse coordinates in Eq. (10) and accounting for radial symmetry ($r^2 = x^2 + y^2$), giving

$$\partial_t \mathcal{E} = \frac{i}{2k_0} \partial_r r \partial_r \mathcal{E} - \frac{k_2}{2} \partial_r^2 \mathcal{E} + i \frac{\omega_0}{c} n_2 |\mathcal{E}|^2 \mathcal{E}.$$ (21)

This equation is used to estimate the spatio-temporal dynamics for input Gaussian pulses

$$\mathcal{E} = \sqrt{I_0} \exp{-\frac{r^2}{\tau_0^2} - \frac{r^2}{u_0^2}}$$ (22)

by a variational approach [17]. This approach minimizes the generalized action, leading to the dynamical system

$$w^2 k_0^2 d^2 w = 1 + \frac{p r_0}{\sqrt{2r}} + \frac{r^3 k_2}{4k_0} d^2 r = k_2 - \frac{p r_0}{\sqrt{2k_0} w^2},$$ (23)

for the beam waist $w(z)$ and pulse duration $r(z)$. Here, $p = P_{in}/P_{cr}$ with the critical power $P_{cr} = 2 \pi c^2 / \omega_0^2 n_2^2 |n_2|$. The presence of a lens is included by the initial condition $d_w |_{z=0} = -w_0 / f$, where $f$ is the focal length. This is combined with an additional MI analysis for plane waves [18] in the sense, that the maximal intensity resulting from the variational approach $l_{max}$ serves as background plane wave intensity. Then, the growth rate for perturbations with frequency $\omega$ and transverse wave number $k_\perp$

$$g(\omega, k_\perp) = \Re(2\omega_0 |n_2| l_{max}/c - \Omega^2),$$ (24)
where $\Omega^2 = k_0 \omega_0^2 / 2 - k^2_\perp / 2k_0$, reaches its maximum for $\omega_{\text{max}}$ and $k^\text{max}_\perp$, being linked by

$$\omega_{\text{max}} \approx \sqrt{\frac{2\omega_0 n_2 I_{\text{max}}}{k_2 c}} + \frac{(k^\text{max}_\perp)^2}{k_0 k_2}. \quad (25)$$

A necessary condition [19] for MI to occur is $k^\text{max}_\perp > \sqrt{2\pi / w_0}$ and $\omega_{\text{max}} > \sqrt{2\pi / t_0}$ for any given intensity. Fixing $k^\text{max}_\perp = \sqrt{2\pi / w_{\text{min}}}$, we use the highest perturbation frequency $\omega_{\text{high}} = \sqrt{2\pi / t_{\text{min}}}$ to fulfill the condition for compression $\omega_{\text{high}} \approx \omega_{\text{max}}$ [6].

In our case where $\omega_0 = \omega_{\text{max}}$, an optimum set of parameters is $P_{in} = 5P_{cr}$ for input power ($I_0 = 7.1 \times 10^{14} \text{ W/m}^2$, $t_0 = 100 \text{ fs}$ for pulse duration, $w_0 = 0.3 \times 10^{-3} \text{ m}$ as beam radius and a focal length of $f = 1 \text{ m}$).

### 4.2. Full equation

In this section, results for the full model Eq. (8) will be presented. We assume Gaussian input pulses with simulation parameters given above. In Fig. 5 simulation results are summarized, where Fig. 5(b) details the evolution of the initial Gaussian pulse into a singly peaked structure with minimal FWHM duration of 17 fs [see Fig. 5(c)], which remains almost constant over propagation distances of $\sim 0.5 \text{ m}$. Fig. 5(d) validates that the compression is not constrained to the on-axis intensity profiles but homogeneous in radial direction. Again, the pulse spectrum reaches regions where the nonlinearity is resonant and the previous discussion of validity has to be considered.

Further on, results for the configuration with $\gamma \approx 0$ (central frequency $\omega_0 = \omega_{\text{high}}$) are presented. As learned before, a MI analysis is not suitable to estimate dynamics in that case. Therefore parameters from the $(1 + 1)$-dimensional configuration previously applied to the NLSND model are taken for intensity and pulse width ($I_0 = 1.9 \times 10^{15} \text{ W/m}^2$ and $t_0 = 100 \text{ fs}$, respectively). In order to keep transverse dynamics at bay, we choose a broad initial beam radius $w_0 = 3 \text{ mm}$, implying a long diffraction length of $L_{\text{diff}} \sim 120 \text{ m}$ compared to typical propagation distances $\sim 5 \text{ m}$.

As revealed in Fig. 6(a), the temporal dynamics agree well with the ones obtained in the $(1 + 1)$-dimensional setup for propagation distances up to $z \sim 3 \text{ m}$, for which the transverse profile remains constant [see Fig. 6(b)]. Upon further propagation, the blue part ($\omega > \omega_{\text{high}}$) of the pulse spectrum, experiencing a positive $n_2 \sim \chi^{(3)}(-\omega; -\omega, \omega, \omega)$, self-focuses and finally collapses at $z \sim 4 \text{ m}$. This slowly propagating, focusing part of the spectrum appears as a high intensity zone at later times in the (on-axis) temporal dynamics plot at $z \sim 3.5 \text{ m}$ [see Fig. 6(a)]. Increasing the input intensity would be an option to rise effectiveness of the soliton ejection (scaling wave-breaking length $\sim 1 / I_0$ against self-focusing length $\sim 1 / \sqrt{I_0}$). However, this option is limited by plasma generation setting in, which is not described within our model.

### 5. Conclusions

We demonstrated the effect of nonlinear dispersion on solitary compression for normal GVD and negative $n_2$. We showed that the main effects of nonlinear dispersion can be captured by using
first order Taylor expansion of the nonlinear susceptibility. The resulting shift of high intensity zones to later times does not change the qualitative mechanism of compression being mediated by MI. Because we are able to control MI in the unguided configuration by adjusting, e.g., the laser intensity, this mechanism provides an effective and simple way of compressing laser pulses by just focusing them into a gas cell. In the special case of vanishing $n_2$ at center frequency, we showed that nonlinear dispersion enables an alternative compression mechanism. However, while working perfectly in purely temporal ($1+1$)-dimensional configuration, the important influence of spatial dynamics on temporal compression effects is underlined by the pulse self-collapse after soliton ejection in bulk configuration.

References


