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

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#### 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 [1] or cascading quadratic nonlinearities [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):  $\partial_z A = -ik_2 A_{tt}/2 +$  $i\gamma |\dot{A}|^2 \dot{A}$ . Here,  $\dot{k}_2 = \partial^2 \dot{k}(\omega) / \partial \omega^2 |_{\omega = \omega_0}$  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

#### ABSTRACT

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.

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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. [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 [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.

#### 2. Derivation of the governing equation

#### 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



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the absence of free charges  $\rho_f = 0$  and free currents  $\vec{J_f} = 0$ , a nonmagnetic medium  $\vec{B} = \mu_0 \vec{H}$  and the material to be nonlinear in the sense that  $\vec{D} = \epsilon_0 \vec{E} + \vec{P}$ . It is common to express the polarization  $\vec{P}$  in a power series in  $\vec{E} : \vec{P} = \vec{P}^{(1)} + \vec{P}^{(3)} + \vec{P}^{(5)} + \cdots, \vec{P}^{(j)} \sim \vec{E^j}$ , where we already accounted for the vanishing of contributions of even power in  $\vec{E}$  because we are dealing with centrosymmetric media. In this work, we keep only terms up to third order and refer to  $\vec{P}^{(1)} = \vec{P}_L$  as the linear polarization and to  $\vec{P}^{(3)} = \vec{P}_{NL}$  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

$$\nabla^2 \vec{E} = \mu_0 \epsilon_0 \frac{\partial^2 \vec{E}}{\partial t^2} + \mu_0 \frac{\partial^2 \vec{P}_L}{\partial t^2} + \mu_0 \frac{\partial^2 \vec{P}_{NL}}{\partial t^2} + \nabla (\nabla \cdot \vec{E}).$$
(1)

For small polarization  $|\vec{P}| \ll |\epsilon_0 \vec{E}|$ ,  $\vec{\nabla} \cdot \vec{E}$  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  $\tilde{P}_L(\omega) = \epsilon_0 \chi^{(1)}(\omega)\tilde{E}(\omega)$  and after introducing the wave number  $k(\omega) = \sqrt{1 + \chi^{(1)}(\omega)\omega/c}$  we obtain the wave equation in Fourier space

$$\nabla^2 \tilde{E} + k^2(\omega)\tilde{E} = -\omega^2 \mu_0 \tilde{P}_{NL}.$$
(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 mainly along the  $z \ge 0$  direction and therefore decompose it into forward propagating plane waves with wave numbers  $k(\omega)$  and the envelope  $\tilde{A}$ 

$$E(x, y, z, t) = \int d\omega \tilde{A}(x, y, z, \omega) e^{i[k(\omega)z - \omega t]}.$$
(3)

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

$$\partial_{z}\tilde{A} = \frac{i}{2k(\omega)}\nabla_{\perp}^{2}\tilde{A} + ik(\omega)\tilde{A} + \frac{i\mu_{0}\omega^{2}}{2k(\omega)}\tilde{P}_{NL}.$$
(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

$$k(\omega) = k_0 + \frac{k_1}{1!}\bar{\omega} + \frac{k_2}{2!}\bar{\omega}^2 + \cdots,$$
 (5)

with  $k_j = \partial^j k(\omega)/\partial \omega^j|_{\omega = \omega_0}$  and  $\bar{\omega} = \omega - \omega_0$ . Moreover, for convenience in the numerics, we split fast oscillations in *z* and go over to a co-moving time frame, defined by  $t \rightarrow t + k_1 z$ . In Fourier space, this leads to the introduction of the (normalized) slowly varying complex envelope

$$\tilde{\varepsilon}(\bar{\omega}) = \Theta(\bar{\omega} + \omega_0) \frac{1}{\sqrt{s}} \tilde{E}(\bar{\omega} + \omega_0) e^{-i(k_0 + k_1\bar{\omega})z}, \quad s = \frac{1}{2n_0\epsilon_0 c}, \quad (6)$$

with  $|\mathcal{E}|^2 = I$  being the laser intensity,  $\Theta(x)$  the usual Heaviside function, and  $n_0 = k_0 c / \omega_0$ .

#### 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

$$\tilde{P}_{NL}(\omega) = \epsilon_0 s^{3/2} \iint d\omega_1 d\omega_2 \chi^{(3)}(-\omega; \omega - \omega_1 - \omega_2, \omega_2, \omega_1) \times 3\tilde{\varepsilon}(\bar{\omega}_1) \tilde{\varepsilon}(\bar{\omega}_2) \tilde{\varepsilon}^*(\bar{\omega}_1 + \bar{\omega}_2 - \bar{\omega}) e^{i(k_0 + k_1 \bar{\omega})z},$$
(7)

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

$$\partial_{z}\tilde{\mathcal{E}} = \frac{1}{2k(\omega)}\nabla_{\perp}^{2}\tilde{\mathcal{E}} + i[k(\omega) - k_{0} - k_{1}\bar{\omega}]\tilde{\mathcal{E}} + \frac{3i\omega^{2}s}{2k(\omega)c^{2}}\iint d\omega_{1}d\omega_{2}\chi^{(3)}(-\omega;\omega-\omega_{1}-\omega_{2},\omega_{2},\omega_{1}) \times \tilde{\mathcal{E}}(\bar{\omega}_{1})\tilde{\mathcal{E}}(\bar{\omega}_{2})\tilde{\mathcal{E}}^{*}(\bar{\omega}_{1}+\bar{\omega}_{2}-\bar{\omega}).$$
(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 ( $\nabla_{\perp}^2$ ) 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  $\omega_0$ :

$$\chi^{(3)}(-\omega; \omega - \omega_1 - \omega_2, \omega_2, \omega_1) = \chi_0^{(3)} + \chi_1^{(3)} \bar{\omega}_1 + \chi_2^{(3)} \bar{\omega}_2 + \chi_3^{(3)} \bar{\omega} + \cdots,$$
(9)

where  $\bar{\omega}_j = \omega_j - \omega_0$  and

$$\begin{split} \chi_{0}^{(3)} &= \chi^{(3)}(-\omega_{0}; -\omega_{0}, \omega_{0}, \omega_{0}), \\ \chi_{1,2}^{(3)} &= \partial_{\omega_{1,2}} \chi^{(3)}(-\omega; \omega - \omega_{1} - \omega_{2}, \omega_{2}, \omega_{1})|_{\omega_{1} = \omega_{2} = \omega = \omega_{0}}, \\ \chi_{3}^{(3)} &= \partial_{\omega} \chi^{(3)}(-\omega; \omega - \omega_{1} - \omega_{2}, \omega_{2}, \omega_{1})|_{\omega_{1} = \omega_{2} = \omega = \omega_{0}}. \end{split}$$

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 ( $\chi_{1,2,3}^{(3)} = 0$ ) 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  $\omega_{--} = 7.73 \times 10^{15} \text{ s}^{-1}$  for the characteristic central frequency ensuring a negative  $n_2$  and additionally  $\omega_{00} = 7.91 \times 10^{-15} \text{ s}^{-1}$  for investigations with vanishing  $n_2$ . That is, a laser pulse with central frequency  $\omega_0 = \omega_{--}$  experiences defocusing as well as effects originating from nonlinear dispersion, whereas a pulse with central frequency  $\omega_0 = \omega_{00}$  lacks the usual Kerr effect  $\sim n_2 = 0$  and only undergoes nonlinear dispersive modulations.

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