

# Photon-conserving generalized nonlinear Schrödinger equation for frequency-dependent nonlinearities

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Pulse propagation in nonlinear waveguides is most frequently modeled by resorting to the generalized nonlinear Schrödinger equation (GNLSE). In recent times, exciting new materials with peculiar nonlinear properties, such as negative nonlinear coefficients and a zero-nonlinearity wavelength, have been demonstrated. Unfortunately, the GNLSE may lead to unphysical results in these cases since, in general, it does not preserve the number of photons and, in the presence of a negative nonlinearity, predicts a blue shift due to Raman scattering. In this paper, we put forth a modified GNLSE that can be used to model the propagation in media with an arbitrary, even negative, nonlinear coefficient. This novel photon-conserving GNLSE (pcGNLSE) ensures preservation of the photon number and can be solved by the same tried and trusted numerical algorithms used for the standard GNLSE. Finally, we compare results for soliton dynamics in fibers with different nonlinear coefficients obtained with the pcGNLSE and the GNLSE.

## 1. INTRODUCTION

For many years, the generalized nonlinear Schrödinger equation (GNLSE) [1] has been proved to accurately model propagation of light pulses in nonlinear Kerr media. Moreover, the existence of powerful numerical algorithms [1,2] that can efficiently solve the GNLSE has rendered its use widespread. However, the validity of the GNLSE is limited, and there have been many attempts to introduce modifications in order to extend the range of its applicability, for instance, to shorter pulses [3–10]. Moreover, it is well known that the approximations involved in the derivation of the GNLSE do not conserve some physical quantities, such as the photon number, unless the transmission medium nonlinearity satisfies some special and restrictive conditions [11–13]. These conditions pose a serious limitation to the application of the GNLSE to new kinds of materials, such as metamaterials [14–19] and nanoparticle-doped glasses [20–24]. The nonlinear refractive-index of these materials is strongly frequency dependent, giving rise to unusual phenomena, such as solitons and modulation instability in the normal-dispersion regime, the existence of a zero-nonlinearity

wavelength, or a controllable self-steepening parameter [15,23]. It can be shown that the straightforward addition of a complex wavelength dependence of the nonlinear coefficient on the GNLSE does not satisfy the strict requirements for photon number conservation and, hence, leads to unphysical predictions. Thus, a careful modification of the GNLSE, accounting for the frequency dependence of the nonlinear coefficient, is needed to adequately model propagation in these peculiar media [12,15,19,25–28]. In this work, we put forth one such modification based on a simple quantum theory of the GNLSE developed in Ref. [29]. In this sense, we extend the work in Ref. [30], which introduced a modified photon-conserving nonlinear Schrödinger equation, neglecting the effect of the Raman (delayed) response of the medium.

The rest of the paper is organized as follows. In Section 2, we briefly note the problems of the GNLSE to model frequency-dependent nonlinearities. In Section 3, we summarize our proposed modification to the propagation equation. Numerical results are presented in Section 4, and we close with some final remarks in Section 5.

## 2. GNLSE WITH A FREQUENCY-DEPENDENT NONLINEARITY

Let us recall that the GNLSE in the frequency domain reads [1]

$$\frac{\partial \tilde{A}_\omega}{\partial z} = i\beta(\omega)\tilde{A}_\omega + i\gamma(\omega)\mathcal{F}(A|A|^2) + if_R\gamma(\omega)\mathcal{F} \times \left( A \int_0^\infty h(\tau)|A(t-\tau)|^2 d\tau - A|A|^2 \right), \quad (1)$$

where  $z$  is the direction of propagation,  $A$  is the complex envelope of the electric field of a light pulse with central frequency  $\omega_0$  normalized such that  $|A|^2$  is the optical power,  $\tilde{A}_\omega = \mathcal{F}(A)$ , and  $\mathcal{F}$  stands for the Fourier transform. Since we focus on the lossless case, the mode-propagation profile  $\beta(\omega)$  is assumed to be real valued.  $f_R$  is the fractional Raman contribution [31], and  $h(t)$  is the delayed Raman response. As shown in Ref. [30], Eq. (1) does not conserve the number of photons in general, a fact inconsistent with the underlying photon-conserving physical processes, namely, dispersion, four-wave mixing, and Raman scattering. Moreover, although Raman scattering involves the annihilation of a photon at frequency  $\omega$  and the creation of a photon at frequency  $\omega - \mu$  and a phonon of frequency  $\mu$ , leading to a transfer of energy toward lower frequencies (red shift), an unphysical soliton blue shift is predicted when a negative nonlinearity is considered [23].

Limitations of Eq. (1) to model both Raman scattering and arbitrary nonlinearities can be best understood by resorting to some simple calculations. Let us consider the interaction between a continuous-wave intense pump and a small signal. We propose the solution  $A(z, t) = A_p(z)e^{-i(\omega_0+\omega_p)t} + A_s(z)e^{-i(\omega_0+\omega_s)t}$  for Eq. (1), where  $\omega_s = \omega_p - \mu$ ,  $\mu$  represents the phonon frequency, and  $A_p$  and  $A_s$  are the pump and signal amplitudes, respectively;  $\omega_0$  is an arbitrary central frequency such that all other frequencies in this paper represent a detuning from  $\omega_0$ .

For the sake of simplicity, we neglect the four-wave interaction with an anti-Stokes signal. The evolution of photon fluxes,  $\Phi_{p,s} = |A_{p,s}|^2 / \hbar(\omega_0 + \omega_{p,s})$ , is given by (see, e.g., Chapter 10 in Ref. [32])

$$\frac{\partial \Phi_p}{\partial z} = -\frac{2f_R\gamma(\omega_p)\tilde{h}_\mu^I}{\hbar(\omega_0 + \omega_p)} P_p P_s, \quad (2)$$

$$\frac{\partial \Phi_s}{\partial z} = \frac{2f_R\gamma(\omega_s)\tilde{h}_\mu^I}{\hbar(\omega_0 + \omega_s)} P_p P_s, \quad (3)$$

where  $P_{p,s} = |A_{p,s}|^2$  is the optical power and  $\tilde{h}^I = \text{Im}\{\mathcal{F}(h(t))\}$ . We focus on the case  $\omega_s < \omega_p$ , where a photon-flux transfer from pump to signal is expected. First, it must be noted that the photon-number conservation,  $\partial_z \Phi_p + \partial_z \Phi_s = 0$ , is satisfied only if  $\gamma(\omega_p)/\gamma(\omega_s) = (1 + \omega_p/\omega_0)/(1 + \omega_s/\omega_0)$ . Since both  $\omega_p$  and  $\mu$  are arbitrary, the conservation of the number of photons requires that  $\gamma(\omega) = \gamma_0(1 + \omega/\omega_0)$ , for some constant  $\gamma_0$  and all  $\omega$ . This requirement, already found by Blow and Wood [11], not only prevents the modeling of a more general linear frequency dependence of the nonlinear coefficient, but it also precludes the modeling of far more interesting phenomena such as the

existence of zero-nonlinearity wavelengths. Furthermore, these equations necessitate that  $\gamma_0 > 0$ ; otherwise, a negative nonlinearity would lead to a transfer of photons from the signal to the pump. All in all, these observations point to the fact that Eq. (1) is not suitable when applied to arbitrary  $\gamma(\omega)$  profiles.

## 3. PHOTON-CONSERVING GENERALIZED NONLINEAR SCHRÖDINGER EQUATION

In order to circumvent the difficulties of Eq. (1), we resort to a simple quantum theory of the GNLSE developed in Ref. [29]. Following an approach similar to that in Lai and Haus [33], it is shown that, for a standard nonlinearity  $\gamma(\omega) = \gamma_0(1 + \omega/\omega_0)$  with  $\gamma_0 > 0$ , Eq. (1) can be derived from the quantum master equation

$$\frac{d\rho}{dz} = i[\hat{H}_{\text{Kerr}} + \hat{H}_R, \rho] + \int_0^\infty \left[ \hat{L}_\mu \rho \hat{L}_\mu^\dagger - \frac{1}{2} \{ \rho, \hat{L}_\mu^\dagger \hat{L}_\mu \} \right] d\mu, \quad (4)$$

where  $\rho$  is the density matrix representing the quantum state of the electromagnetic field,  $\hat{H}_{\text{Kerr}}$  is the four-wave mixing operator associated with the Kerr effect,

$$\hat{H}_{\text{Kerr}} = \iiint \frac{\gamma_0}{4\pi\hbar\omega_0} \hat{A}_{\omega_1}^\dagger \hat{A}_{\omega_2}^\dagger \hat{A}_{\omega_1-\mu} \hat{A}_{\omega_2+\mu} d\omega_1 d\omega_2 d\mu, \quad (5)$$

$\hat{H}_R$  is the four-wave mixing operator associated with the real part of the Raman response [34–36],

$$\hat{H}_R = \iiint \frac{\gamma_0 f_R (\tilde{h}_\mu^R - 1)}{4\pi\hbar\omega_0} \hat{A}_{\omega_1}^\dagger \hat{A}_{\omega_2}^\dagger \hat{A}_{\omega_1-\mu} \hat{A}_{\omega_2+\mu} d\omega_1 d\omega_2 d\mu, \quad (6)$$

and  $\hat{L}_\mu$  is the Lindbladian operator associated with the creation of a phonon of frequency  $\mu$ ,

$$\hat{L}_\mu = \int \sqrt{\frac{\gamma_0 f_R \tilde{h}_\mu^I}{\pi\hbar\omega_0}} \hat{A}_{\omega'-\mu}^\dagger \hat{A}_{\omega'} d\omega'. \quad (7)$$

The field operators  $\hat{A}_\omega$ , related to the annihilation operators by  $\hat{A}_\omega = \sqrt{\hbar(\omega_0 + \omega)} \hat{a}_\omega$ , provide a clear quantum picture of the four-wave mixing and Raman scattering processes in terms of the creation and annihilation of photons. The standard GNLSE can be straightforwardly obtained as the mean-value evolution of these operators (for full details of this calculation, we refer the interested reader to Ref. [29]):

$$\frac{d\langle \hat{A}_\omega \rangle}{dz} = i\langle [\hat{A}_\omega, \hat{H}_{\text{Kerr}} + \hat{H}_R] \rangle + \frac{1}{2} \int_0^\infty \langle [\hat{L}_\mu^\dagger, \hat{A}_\omega] \hat{L}_\mu - \hat{L}_\mu^\dagger [\hat{L}_\mu, \hat{A}_\omega] \rangle d\mu. \quad (8)$$

Both Eqs. (5) and (6) must be modified in order to preserve the number of photons when an arbitrary frequency-dependent nonlinear coefficient  $\gamma(\omega)$  is considered. It can be shown that a modified photon- and energy-conserving Kerr operator is given by [30]

$$\hat{H}_{\text{Kerr}} = \frac{1}{8\pi\hbar} \iiint \left( \hat{B}_{\omega_1}^\dagger \hat{B}_{\omega_2}^\dagger \hat{C}_{\omega_1-\mu} \hat{C}_{\omega_2+\mu} + \hat{C}_{\omega_1}^\dagger \hat{C}_{\omega_2}^\dagger \hat{B}_{\omega_1-\mu} \hat{B}_{\omega_2+\mu} \right) d\omega_1 d\omega_2 d\mu, \quad (9)$$

where the operators are defined as  $\hat{B}_\omega = \sqrt[4]{\gamma(\omega)/(\omega_0 + \omega)} \hat{A}_\omega$  and  $\hat{C}_\omega = (\sqrt[4]{\gamma(\omega)/(\omega_0 + \omega)})^* \hat{A}_\omega$ .

In order to include the effect of Raman scattering in the presence of arbitrary nonlinearity profiles, we propose rewriting the Raman operators [Eqs. (6) and (7)] in terms of  $\hat{B}$ , obtaining

$$\hat{H}_R = \iiint \frac{f_R(\tilde{h}_\mu^R - 1)}{4\pi\hbar} \hat{B}_{\omega_1}^\dagger \hat{B}_{\omega_2}^\dagger \hat{B}_{\omega_1 - \mu} \hat{B}_{\omega_2 + \mu} d\omega_1 d\omega_2 d\mu, \quad (10)$$

$$\hat{L}_\mu = \int \sqrt{\frac{f_R \tilde{h}_\mu^I}{\pi\hbar}} \hat{B}_{\omega' - \mu}^\dagger \hat{B}_{\omega'} d\omega'. \quad (11)$$

It can be easily shown that Eqs. (9)–(11) are consistent with Eqs. (5)–(7) when setting  $\gamma(\omega) = \gamma_0(1 + \omega/\omega_0)$  and  $\gamma_0 > 0$  in the definition of  $\hat{B}_\omega$ . That is, these equations agree with the GNLSE when the frequency dependence of the nonlinear coefficient  $\gamma(\omega)$  is linear. However, note that our proposal involves a subtle difference with the usual approach of preserving the GNLSE and adding an arbitrary  $\gamma(\omega)$  in straightforward fashion; instead, we keep the quantum master equation and allow  $\gamma(\omega)$  to take arbitrary values. This way, we ensure that the

physical processes represented by the equation remain the same, but weighted by different coefficients.

By substituting Eqs. (9)–(11) in Eq. (8), we obtain a photon-conserving GNLSE (pcGNLSE):

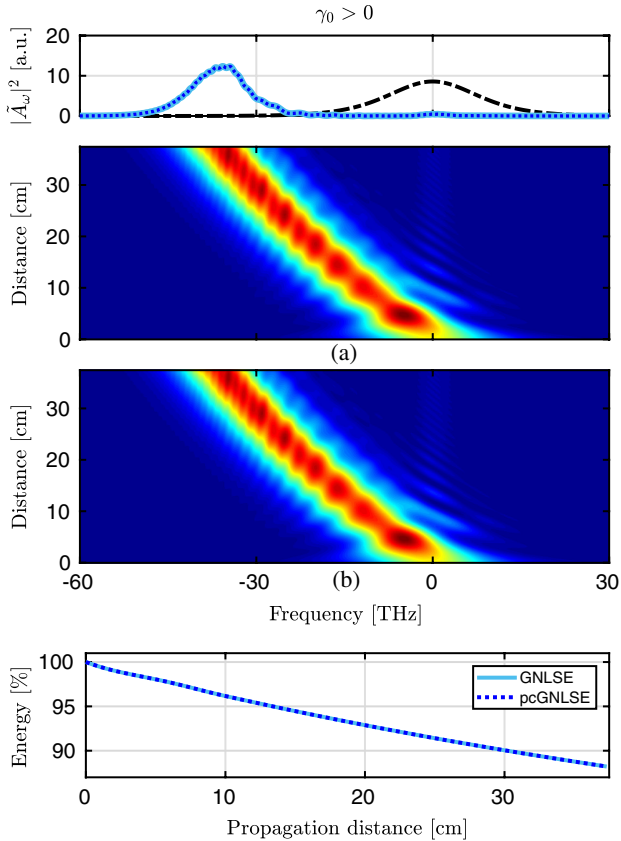
$$\begin{aligned} \frac{\partial \tilde{A}_\omega}{\partial z} = & i\beta(\omega)\tilde{A}_\omega + i\frac{\tilde{\gamma}(\omega)}{2}\mathcal{F}(C^*B^2) + i\frac{\tilde{\gamma}^*(\omega)}{2}\mathcal{F}(B^*C^2) \\ & + i f_R \tilde{\gamma}^*(\omega) \mathcal{F}\left(B \int_0^\infty h_R(\tau) |B(t-\tau)|^2 d\tau - B|B|^2\right), \end{aligned} \quad (12)$$

with

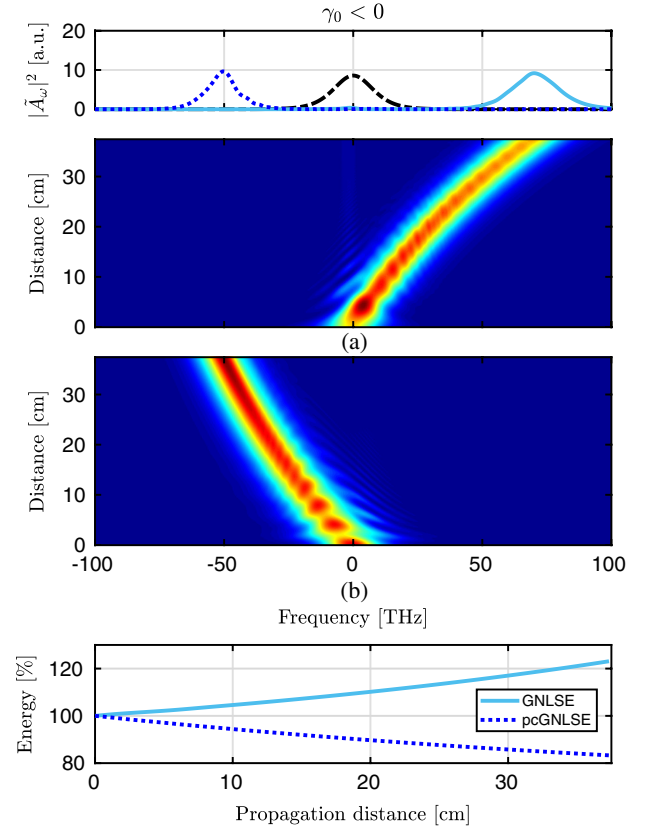
$$\tilde{B}_\omega = \sqrt[4]{\frac{\gamma(\omega)}{\omega_0 + \omega}} \tilde{A}_\omega, \quad \tilde{C}_\omega = \left(\sqrt[4]{\frac{\gamma(\omega)}{\omega_0 + \omega}}\right)^* \tilde{A}_\omega, \quad (13)$$

$$\tilde{\gamma}(\omega) = \sqrt[4]{\gamma(\omega) \times (\omega_0 + \omega)^3}. \quad (14)$$

It must be remarked that Eq. (12) can be efficiently solved using numerical methods similar to those used for the standard GNLSE, such as, e.g., split-step Fourier or Runge–Kutta interaction picture (Ref. [2]): aside from the extra calculation of the auxiliary fields  $B$  and  $C$ , which can be readily obtained from the



**Fig. 1.** Propagation of a fundamental soliton in the anomalous dispersion regime of a medium with a positive nonlinear coefficient and  $\gamma(\omega) = \gamma_0(1 + \omega/\omega_0)$ . Top panel: input spectrum (dashed-dotted line) and output spectra (GNLSE, solid line; pcGNLSE, dotted line). Spectral evolution: (a) GNLSE and (b) pcGNLSE. Bottom panel: energy evolution along propagation. In this scenario, the GNLSE and pcGNLSE equations yield the exact same spectral shifts and energy evolution.



**Fig. 2.** Propagation of a fundamental soliton in the normal dispersion regime of a medium with a negative nonlinear coefficient. Top panel: input spectrum (dashed-dotted line) and output spectra (GNLSE, solid line; pcGNLSE, dotted line). Spectral evolution: (a) GNLSE and (b) pcGNLSE. Bottom panel: energy evolution along propagation. In this scenario, the pcGNLSE predicts a physically sound soliton red shift, while the GNLSE produces an unphysical blue shift.

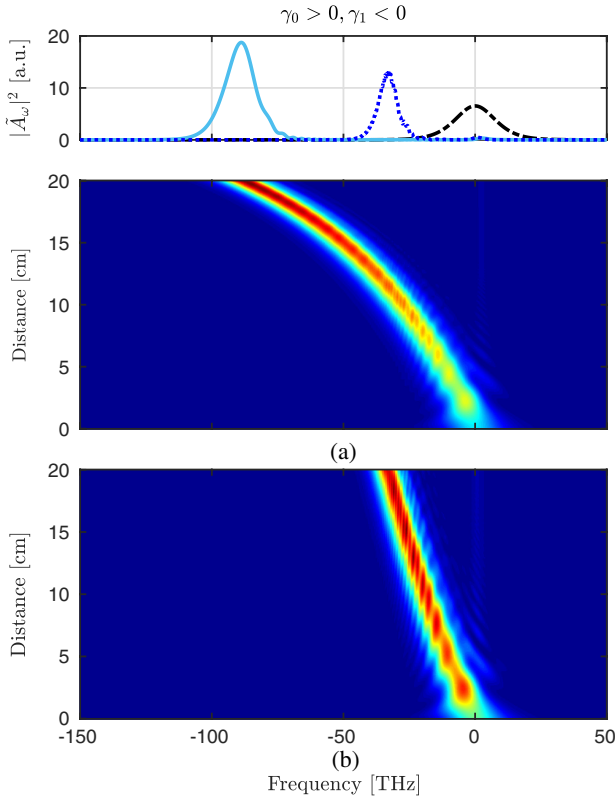
knowledge of  $\tilde{A}_\omega$  and  $\gamma(\omega)$  using Eq. (13), numerical methods can be applied in the exact same manner.

As we did in the case of the GNLSE [viz. Eqs. (2) and (3)], we can analyze the behavior of photon fluxes in the simple case of a pump and a signal. We can arrive at these flux equations governing the Raman scattering process by considering a pump plus signal field  $A(z, t) = A_p(z)e^{-i\omega_p t} + A_s(z)e^{-i\omega_s t}$  in Eq. (12), neglecting all four-wave mixing terms (i.e., keeping only terms in the original pump and signal frequencies) and then obtaining coupled differential equations for  $\partial_z A_{s,p}(z)$ . After some cumbersome but straightforward calculations, the fluxes  $\partial_z \Phi_{s,p}(z) = 2\text{Re}[\frac{A_{s,p}^*(z)\partial_z A_{s,p}(z)}{\hbar(\omega_0 + \omega_{s,p})}]$  are found to be

$$\frac{\partial \Phi_p}{\partial z} = -\frac{2f_R\sqrt{|\gamma(\omega_s)\gamma(\omega_p)|}\tilde{h}_\mu^I}{\hbar\sqrt{(\omega_0 + \omega_s)(\omega_0 + \omega_p)}}P_pP_s, \quad (15)$$

$$\frac{\partial \Phi_s}{\partial z} = \frac{2f_R\sqrt{|\gamma(\omega_s)\gamma(\omega_p)|}\tilde{h}_\mu^I}{\hbar\sqrt{(\omega_0 + \omega_s)(\omega_0 + \omega_p)}}P_pP_s. \quad (16)$$

It is easy to verify that the photon number is conserved ( $\partial_z \Phi_s + \partial_z \Phi_p = 0$ ). Moreover, since  $\partial_z \Phi_p \leq 0$  and  $\partial_z \Phi_s \geq 0$ , Raman scattering cannot produce a shift towards higher frequencies, i.e., only a red shift is possible, as expected.



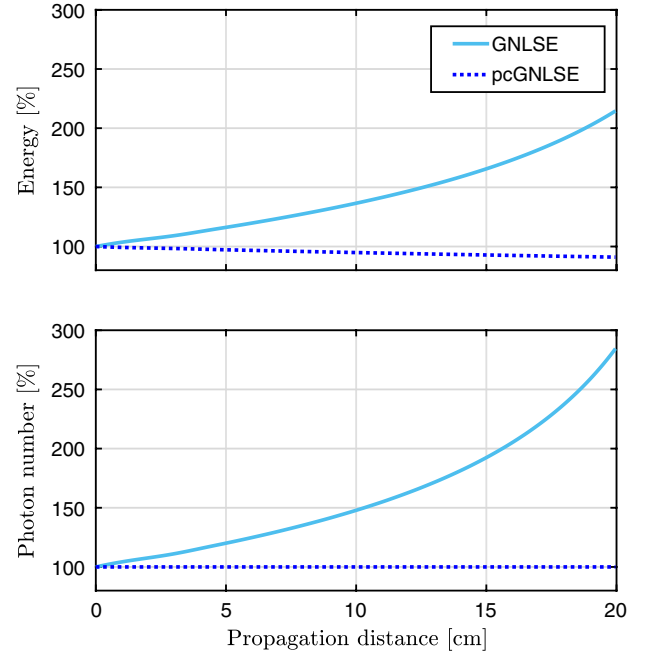
**Fig. 3.** Propagation of a fundamental soliton in the anomalous dispersion regime of a medium with a positive nonlinear coefficient and a negative self-steepening parameter. Top panel: input spectrum (dashed-dotted line) and output spectra (GNLSE, solid line; pcGNLSE, dotted line). Spectral evolution: (a) GNLSE and (b) pcGNLSE. In this scenario, the GNLSE and the pcGNLSE predict different soliton red shifts.

## 4. NUMERICAL RESULTS

In what follows, we show substantially different results obtained with the GNLSE and the proposed pcGNLSE. Also, we show a simulation of soliton dynamics with parameters taken from Ref. [24] where an alternative model, including the frequency-dependent  $\gamma(\omega)$  only in the instantaneous response of the medium, is introduced.

Figure 1 shows the evolution of a fundamental soliton of 10-fs half-width (at the  $1/e$ -intensity point), at a center wavelength of 1000 nm, in a fiber with a nonlinearity  $\gamma(\omega) = \gamma_0(1 + \omega/\omega_0)$  and  $\gamma_0 = +10^{-2}1/\text{W m}$ , and dispersion profile  $\beta(\omega) = \frac{1}{2}\beta_2(\omega)\omega^2$ , where the group velocity dispersion parameter is  $\beta_2(\omega) = \beta_2 = -21 \text{ ps}^2/\text{km}$ ; the Raman response function is  $h(\tau) = \frac{\tau_1^2 + \tau_2^2}{\tau_1\tau_2}e^{-t/\tau_2}\sin(t/\tau_1)$ , where  $\tau_1 = 0.0155 \text{ ps}$  and  $\tau_2 = 0.2305 \text{ ps}$ , and the Raman fractional contribution is  $f_R = 0.18$  [1]. As expected, in this case, both equations predict the exact same results. The decreasing of the pulse energy due to the Raman-scattering red shift is displayed in Fig. 1 (bottom). More interestingly, differences arise in the case shown in Fig. 2, where the nonlinear coefficient is negative ( $\gamma_0$  and  $\beta_2$  signs are alternated with respect to that in Fig. 1). The GNLSE predicts unphysical results, namely, a soliton blue shift together with an increase in the total energy (bottom of Fig. 2). On the other hand, the pcGNLSE predicts a red shift of the soliton and an energy decrease due to Raman scattering. Closer observation of the top panel of Fig. 2 reveals not only the difference in the soliton frequency shift, but also that the pcGNLSE predicts a soliton of narrower bandwidth.

In order to further assess the physical consistency of the proposed equation, we compare the evolution of a



**Fig. 4.** Energy and photon number evolution corresponding to Fig. 3: GNLSE, solid line; pcGNLSE, dotted line. Note that, while the pcGNLSE consistently conserves the photon count and reflects energy losses due to Raman scattering, the GNLSE predicts the increase in both photon number and energy.

fundamental soliton, as obtained with the GNLSE and the pcGNLSE, now including an arbitrary negative self-steepening parameter  $\gamma_1 \neq \gamma_0/\omega_0$ . The simulation parameters are 3.48 kW peak power,  $t_0 = 10$  fs,  $\lambda_0 = 835$  nm,  $\beta_2 = -38.3$  ps<sup>2</sup>/km,  $\beta_3 = 0.25$  ps<sup>3</sup>/km,  $\gamma_0 = 0.111$ /W m, and  $\gamma_1 = -2.25 \times 10^{-4}$  ps/W m. These parameters are taken from Ref. [24], where soliton dynamics in a photonic-crystal fiber with a frequency-dependent Kerr nonlinearity are analyzed with an alternative ad hoc model. Results obtained with the pcGNLSE agree well with those in this reference.

In Fig. 3, we observe that, while the GNLSE and the pcGNLSE predict a physically sound soliton red shift, the predicted soliton frequency-shift rate and output spectra differ substantially. Figure 4 shows the energy and photon-number evolution for both cases; while the pcGNLSE predicts an energy decrease due to the soliton red shift and conserves the number of photons, the GNLSE produces unphysical results for both quantities.

## 5. CONCLUSION

We showed that the GNLSE does not preserve the photon number, and thus predicts unphysical results when dealing with arbitrary frequency-dependent nonlinearities. By resorting to the quantum-mechanical theory of four-wave mixing and Raman scattering, we derived a new equation, named pcGNLSE, circumventing this problem. We showed examples comparing results from the pcGNLSE with the standard GNLSE. Although the ultimate accuracy of the proposed pcGNLSE needs to be put to the experimental test, the equation is shown to reduce to the standard GNLSE for a standard nonlinearity profile  $\gamma(\omega) = \gamma_0(1 + \omega/\omega_0)$ , and to yield physically sound results such as a soliton red shift even in the presence of a negative nonlinear coefficient, while strictly preserving the number of photons. Furthermore, the pcGNLSE can be solved using the same efficient numerical algorithms used for the GNLSE, and since it can be successfully applied to study arbitrary  $\gamma(\omega)$ , even the ones associated with negative-index materials, it puts forth a powerful tool to assess nonlinear propagation in new and interesting media.

**Disclosures.** The authors declare no conflicts of interest.

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