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# Interaction between optical fields and their conjugates in nonlinear media

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**Abstract:** Motivated by recent experimental results, we demonstrate that the ubiquitous pulse propagation equation based on a single generalized nonlinear Schrödinger equation is incomplete and inadequate to explain the formation of the so called negative-frequency resonant radiation emitted by optical solitons. The origin of this deficiency is due to the absence of a peculiar nonlinear coupling between the positive and negative frequency components of the pulse spectrum during propagation, a feature that the slowly-varying envelope approximation is unable to capture. We therefore introduce a conceptually new model, based on the envelope of the analytic signal, that takes into account the full spectral dynamics of all frequency components, is prone to analytical treatment and retains the simulation efficiency of the nonlinear Schrödinger equation. We use our new equation to derive from first principles the phase-matching condition of the negative-frequency resonant radiation observed in previously reported experiments.

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## 1. Introduction and motivations

The study of supercontinuum generation (SCG), i.e. the explosive broadening of the spectrum of an intense and short input pulse due to nonlinear effects in a medium, typically an optical fiber or a bulk crystal, has been an active area of research since its first discovery in 1970 [1], due to the many applications in metrology and device characterization [2, 3].

Constructing a theory of SCG has proved to be crucial in order to understand and control the dynamics of pulses in optical fibers [4]. Such theory is based on the so-called generalized nonlinear Schrödinger equation (GNLSE), an enhanced version of the integrable nonlinear Schrödinger equation [2]. The GNLSE, based on the concept of slowly varying envelope approximation (SVEA) of the electric field, is paradigmatic in nonlinear optics, and has been extremely successful in explaining most of the features of SCG [2, 3]. One of the most successful predictions was the emission of dispersive waves from optical solitons, which are phase-matched at specific wavelengths, usually referred to as resonant radiation (RR) or Cherenkov

radiation [5–8]. RR contributes significantly to the formation of SCG spectra and can have many applications especially when using photonic crystal fibers [4, 9, 10].

It is known that the conventional GNLSE fails when considering few- or single-cycle pulses. Many approaches for deriving accurate propagation equations with minimal approximations have been successfully developed for this situation during the years [11, 12], which are capable of dealing with broadband spectral evolution, Raman effect and the inclusion of backward waves. However, recent experiments have revealed that new resonant frequencies [referred to as negative-frequency resonant radiation (NRR)] can be emitted by solitons, which are not predicted by any GNLSE formulation [13–15]. Such frequencies can be numerically predicted by using the full Maxwell equations [solved with the finite difference time domain (FDTD, [16]) or the pseudo-spectral spatial domain (PSSD, [17]) techniques], or alternatively by the so-called unidirectional pulse propagation equation (UPPE, [18]), which includes only forward propagating waves but uses the full oscillating electric field, while the phase-matching condition for NRR formation has been derived heuristically [13, 15]. NRR has been attributed to the presence of negative frequency components in the UPPE, which are absent in the GNLSE due to SVEA. However, this claim sparked some controversy in the community [19], due to a lack of a solid theoretical support that could confirm or disprove the given interpretation. For example, this radiation could be confused with that generated by backward waves or by the conventional four-wave mixing (FWM) between the soliton and co-propagating radiation as in [20]. It is also interesting to notice that, despite the fact that negative frequencies are routinely used in quantum optics [21] (where they are associated to the photon creation operator), quantum field theory [22] and water waves [23], in nonlinear optics there is still some resistance in accepting this concept, which we aim to solve in this manuscript.

In the present work we introduce a new equation for a properly defined pulse envelope that is able to capture the surprising and peculiar interaction between positive and negative frequency components during the propagation of an ultrashort pulse. Such an interaction is able to generate phase-matched dispersive waves that would not exist in any model based on the conventional envelope defined when deriving the NLSE, currently referred to as NRR in the literature. We demonstrate that our new equation can be efficiently solve numerically and gives an analytical insight into the very nature of ultrashort pulse propagation in any dielectric medium. Moreover, in this paper we also show that there are some serious deficiencies in the universally adopted equation based on the GNLSE, since the latter neglects the contribution of the cross-phase modulation between the positive and negative frequency parts of the spectrum, which gives rise to new and unexpected nonlinear phenomena that have been previously overlooked.

The structure of the paper is the following. In section 2 we discuss the physical relevance and the existence of positive and negative frequency states, first in linear optics, and then in nonlinear optics. We support our claims with simple yet robust mathematical arguments, and we discuss the necessity for including negative energy states in the dispersion relation. In section 3 we introduce our notation for the analytic signal, and we derive our new equation based on the envelope of the analytic signal, starting from the UPPE. The transparency of this equation when compared to the UPPE allows us to identify the term responsible for the NRR. In section 4 we derive the phase-matching conditions of the radiated frequencies, and we discover new phase-matched frequencies that were previously unknown. In section 5 we present detailed numerical simulations supporting our theory, and in particular we focus our attention to the dynamics of the NRR. Discussion and conclusions are given in section 6, while appendix A and B treat the delicate issue of the energy conservation of our new propagation equation, and the possibility to control and observe the NRR in bulk crystals, respectively.

## 2. The existence and reality of negative frequencies in optics

In this section we discuss the somewhat disputed physical reality and the interpretation of negative frequencies in optics. The logical conclusion is that both positive and negative frequencies must be treated on equal footing and must be included in any meaningful formulation of a fully consistent pulse propagation equation.

Let us discuss light propagation in the vacuum case for simplicity. The linear curl Maxwell equations can be written (in the Heaviside-Lorentz unit system) as

$$\nabla \times \mathbf{E} = -\frac{1}{c} \partial_t \mathbf{B}, \quad (1)$$

$$\nabla \times \mathbf{B} = \frac{1}{c} \partial_t \mathbf{E}, \quad (2)$$

where  $\mathbf{E}$  and  $\mathbf{B}$  are the electric and magnetic field vectors, and  $c$  is the speed of light in vacuum. In order to analyze the frequency content of these equations, we note that Eqs. (1)-(2) are exactly equivalent to a linear relativistic Dirac equation. In order to see this we follow [24], and we define a 'Dirac spinor' as  $\psi = (E_y + iE_x, -iE_z, -B_x + iB_y, B_z)^T$ . This is only one possible choice out of a total of eight equivalent vectors [24]. With this definition, we can write Eqs. (1)-(2) in the following form:

$$\gamma^\mu \partial_\mu \psi = 0, \quad (3)$$

where  $\gamma^\mu$  are the  $4 \times 4$  Dirac matrices and  $\partial_\mu \equiv (\frac{1}{c} \partial_t, \nabla)$  is the derivative four-vector, with  $\mu = \{0, 1, 2, 3\}$ . Equation (3) is exactly the massless Dirac equation that is encountered in the theory of relativistic quantum mechanics of spin-1/2 particles. This equation is fully Lorentz-invariant, as it should be since Maxwell equations have also this property. The two degrees of freedom that correspond to the two states of spin in quantum mechanics, here represent the two possible circular polarizations of light. Alternative (but equivalent) ways to write a Dirac-like equation for Maxwell's equations make use, for instance, of the Riemann-Silberstein vector [25], and are treated in some textbooks as a mere curiosity [26].

From Eq. (3) one derives the wave equation,  $i\partial_t \psi = -ic\alpha \cdot \nabla \psi$ , where  $\alpha^j \equiv \gamma^0 \gamma^j = \begin{pmatrix} 0 & \sigma_j \\ \sigma_j & 0 \end{pmatrix}$ , and  $j = \{1, 2, 3\}$ . The Hamiltonian associated with this wave equation is thus  $\hat{H} = -ic\alpha \cdot \nabla$ . Eigenvalues of the Hamiltonian are found by solving the secular Eq.  $\det(c\alpha \cdot \mathbf{k} - \lambda) = 0$ . By using the well-known relation  $(\alpha \cdot \mathbf{k})^2 = |\mathbf{k}|^2$ , we finally obtain  $\lambda_\pm = \pm |\mathbf{k}|c$ . This brings us to the core conclusion of this section: Eq. (3) exhibits two frequency eigenvalues with opposite sign, irrespective of the direction of propagation of the wave. This is an unavoidable consequence of the fact that Maxwell's equations are relativistic and can be cast in a form identical to the massless Dirac equation, at least in the vacuum case. These two eigenvalues correspond to positive and negative energy states: in full analogy with the classical solutions of the Dirac equation, negative energy states are not unphysical solutions that should be discarded. On the contrary, one must include them *on equal footing* with the positive energy states, in order to preserve the internal consistency of Maxwell's equations.

In Fig. 1 we show a full plot of the dispersion for plane waves for bulk silica. There are four quadrants, containing all the combinations of forward/backward propagation and positive/negative energy states. It is customary in nonlinear optics, but certainly not complete, to consider only the upper-rightmost quadrant with positive frequencies and forward propagation when neglecting the backward waves, for example in the UPPE formulation. This is incorrect and incomplete, since forward waves are described also by the bottom-leftmost quadrant of Fig. 1, which contains negative frequency states. Note that the forward propagation constant must satisfy (for lossless media) the relation  $\beta(-\omega) = -\beta(\omega)$ , and thus it must be an odd

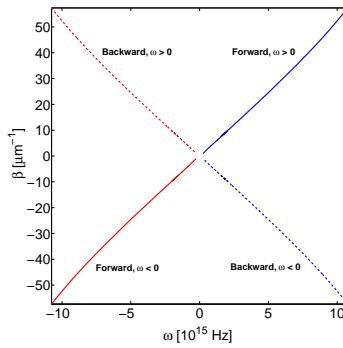


Fig. 1. Dispersion curves for bulk silica. All possible combinations of forward/backward waves and positive/negative energy states appear in this plot.

function with respect to the axis  $\omega = 0$ . As a consequence, the real refractive index must satisfy  $n(\omega) = n(-\omega)$ , and it is thus an even function of the frequency.

In a medium different from vacuum, Maxwell's equations cannot be written in the Dirac form of Eq. (3), since the presence of the medium introduces a preferred reference frame, breaking the Lorentz invariance. However, in this case, Maxwell's equations can be cast in a form similar to the Schrödinger equation by using  $6 \times 6$  matrices, see [27]. The associated Hamiltonian operator has positive and negative eigenvalues, exactly as in the vacuum case [27]. This result can be directly generalized to the nonlinear case by using perturbation theory, assuming that the nonlinear polarization does not perturb considerably the linear modes.

### 3. Derivation of the envelope equation for the analytic signal from the unidirectional pulse propagation equation

We now introduce some important definitions that will be crucial for the following discussion, mainly following [28–31].

The real electric field propagating in the fiber is denoted by  $E(z, t)$ , where  $z$  is the propagation direction and  $t$  is the time variable. The Fourier transform of the electric field is denoted by  $E_\omega(z) \equiv \mathcal{F}[E(z, t)] = \int_{-\infty}^{+\infty} E(z, t) e^{i\omega t} dt$ . The *analytic signal* of the electric field, i.e. the positive frequency part of the field, which is a complex function, is defined as  $\mathcal{E}(z, t) \equiv \pi^{-1} \int_0^\infty E_\omega(z) e^{-i\omega t} d\omega$ . The analytic signal can also be defined alternatively by using the Hilbert transform:  $\mathcal{E}(z, t) = E(z, t) - i\mathcal{H}[E(z, t)]$ , where  $\mathcal{H}[E(z, t)] \equiv \pi^{-1} \mathbb{P} \int_{-\infty}^{+\infty} dt' E(z, t') / (t - t')$ , and the symbol  $\mathbb{P} \int_{-\infty}^{+\infty}$  indicates that the integral must be taken in the sense of the Cauchy principal value. With these definitions, the Fourier transform of the electric field can be written as the sum  $E_\omega = [\mathcal{E}_\omega + (\mathcal{E}_{-\omega})^*] / 2$  since only the positive (or negative) frequency part of the spectrum carries information, while for the same reason the electric field itself is real and is given by  $E(z, t) = [\mathcal{E}(z, t) + \mathcal{E}^*(z, t)] / 2$ . The analytic signal satisfies the following requirements:  $\mathcal{E}_{\omega > 0} = 2E_\omega$ ,  $\mathcal{E}_{\omega < 0} = 0$  and  $\mathcal{E}_{\omega=0} = E_{\omega=0}$ . Note that  $(\mathcal{E}^*)_\omega$  and  $(\mathcal{E}_\omega)^*$  are different in general and must be distinguished.

It can be proved that the fields  $\mathcal{E}$  and  $\mathcal{E}^*$  are the classical analogues of the annihilation and creation operators  $a$  and  $a^\dagger$  used after quantization of the electromagnetic field, see e.g. [28, 29]. This fact is quite understandable since  $a$  and  $a^\dagger$  are related to the positive and negative energy parts of the electric field, which in quantum optics correspond to absorption and emission of a photon [22]. Since the concept of discrete absorption or emission is extraneous to classical electromagnetism, in a pre-second quantization context one is forced to talk about 'conjugated'

fields or 'negative frequency' fields, as we shall do in the following.

Operators  $a$  and  $a^\dagger$  are not linked to observables in quantum optics, since they are not Hermitian. Only the real electric field, which is a Hermitian combination of  $a$  and  $a^\dagger$ , is an observable quantity [22]. Due to the requirement that the electric field be real at all moments of time and at all locations in space, the positive and negative energy content of any pulse in optics and in quantum optics must be absolutely identical. This means that, since photons and antiphotons are equivalent, i.e. they carry the same information, there are no quantum numbers that can distinguish between them, and this results in a perfectly real-valued electric field [25]. An interesting, albeit somewhat trivial, corollary is that (contrary to common belief) a measurement can only detect the positive and negative frequency content of a pulse *simultaneously*, and never one or the other individually. For instance, a real-valued cosine wave is the combination of exponentials with both positive and negative energies. The exponential form is the correct basis that has to be used, since only exponentials are eigenfunctions of the Hamiltonian  $\hat{H}$  written in the previous section, which is a first order operator.

The starting point of our discussion is the so-called unidirectional pulse propagation equation (UPPE) [18], which is a reduction of Maxwell's equations that accounts only for the forward propagating part of the electric field:

$$i \frac{\partial E_\omega}{\partial z} + \beta(\omega) E_\omega + \frac{\omega}{2cn(\omega)} P_{\text{NL},\omega} = 0, \quad (4)$$

where  $\beta(\omega)$  is the full propagation constant of the medium,  $c$  is the speed of light in vacuum,  $n(\omega)$  is the linear refractive index, and  $P_{\text{NL},\omega}(z) \equiv \chi^{(3)} \mathcal{F}[E(z,t)^3]_\omega$  is the nonlinear Kerr polarization. Particular care must be devoted to the definition of the complex envelope, since we do not want to put any limitation to the frequency extent of the signals. This aspect is overlooked in the literature, and it is taken for granted that the frequency bandwidth of the envelope is narrow with respect to the carrier wave.

The key element that we introduce here is that only a proper definition of the envelope is able to capture the correct coupling between the positive and the negative frequency parts of the spectrum. The 'envelope' we introduce here is based on the analytic signal and is defined as:

$$A(z,t) \equiv \mathcal{E}(z,t) e^{-i\beta_0 z + i\omega_0 t}, \quad (5)$$

i.e. the frequency components of the analytic signal are 'shifted' by an amount  $-\omega_0$ . By doing this, we shift the carrier frequency of the analytic signal to zero, so that we deal with frequency detuning  $\Delta\omega$  from  $\omega_0$ , and not with absolute frequencies, in analogy with the conventional definition of envelope done in many textbooks [3]. However, *there is a key difference* between the conventional definition of envelope (see e.g. [3]) and Eq. (5): the former is adequate only if the spectral extension of the pulse evolution is much smaller than the pulse central frequency,  $|\Delta\omega| \equiv |\omega - \omega_0| \ll \omega_0$ , i.e. only under SVEA conditions, while the envelope of the analytic signal  $A(z,t)$  considered here does not suffer from this limitation, and so  $\text{supp}\{A_{\Delta\omega}(z)\} = [-\omega_0, +\infty)$ . By clearly dividing the envelope associated to the positive frequency components from that associated to the negative frequency components, we will be able to write the envelope equation that correctly describes the dynamics of pulses *of arbitrary duration and spectral extension*, taking into account the peculiar and non-trivial interaction between positive and negative frequencies that arises due to the nonlinear polarization.

With the above definitions, the nonlinear polarization is now written as:

$$P_{\text{NL}}(z,t) = \frac{\chi^{(3)}}{8} \left[ A^3 e^{-3i\omega_0 t + 3i\beta_0 z} + A^*{}^3 e^{3i\omega_0 t - 3i\beta_0 z} + 3|A|^2 A e^{-i\omega_0 t + i\beta_0 z} + 3|A|^2 A^* e^{i\omega_0 t - i\beta_0 z} \right]. \quad (6)$$

Due to our definition of  $A$ , the first (second) term in the square brackets contains only positive (negative) frequencies, and they are responsible for third harmonic generation (THG). The third

and fourth terms *contain both positive and negative frequencies*, because the Fourier transform of  $|A|^2$  has a frequency support (i.e. a domain of existence) that extends from  $-\infty$  to  $+\infty$ . In fact  $\mathcal{F}[|A|^2]_{\Delta\omega}$  is the convolution between  $A_{\Delta\omega}$ , whose support is  $[-\omega_0, +\infty)$ , and  $A_{\Delta\omega}^*$ , whose support is  $(-\infty, +\omega_0]$ . By applying the Titchmarsh convolution theorem (i.e. the support of the convolution is contained in the sum of the supports of its individual terms [32]), it immediately follows that  $\text{supp}\{\mathcal{F}[|A|^2]_{\Delta\omega}\} \subseteq (-\infty, +\infty)$ . This means that although in absence of nonlinearities positive and negative frequencies live a completely separate existence, *in presence of nonlinear terms they can interact nonlinearly*. Such an interaction is also present in the traditional Kerr term  $|A|^2A$ . However, new nonlinear effects will only be visible numerically and experimentally in the presence of resonant processes, such as the emission of RR and NRR from solitons. If we denote with  $\mathcal{P}_{\text{NL}}(z, t)$  the analytic signal for the nonlinear polarization, then its envelope  $A_p(z, t) = \mathcal{P}_{\text{NL}}e^{-i\beta_0z+i\omega_0t}$  can be expressed as:

$$A_p(z, t) = \frac{3\chi^{(3)}}{4} \left[ |A|^2A + |A|^2A^*e^{2i\omega_0t-2i\beta_0z} + \frac{1}{3}A^3e^{-2i\omega_0t+2i\beta_0z} \right]_+ \quad (7)$$

The subscript '+' prescribes that only positive frequencies must be taken (i.e.  $\Delta\omega > -\omega_0$ ) and is a shorthand notation to indicate the positive frequency *spectral filtering* involved in the analytic signal, and operated in the time domain by the Hilbert transform, which is crucial in our formulation. The first and third terms in Eq. (7) are the conventional Kerr term and the THG term, respectively. The second term, which we call a *conjugated Kerr term*, is the new feature of our formulation and it emerges as a consequence of the analytic signal envelope.

Finally, with all the above ingredients, one can write an equation for the analytic signal envelope  $A$  which contains only positive frequencies:

$$i\partial_\xi A + \hat{D}(i\partial_\tau)A + \gamma\hat{S}(i\partial_\tau) \left[ |A|^2A + |A|^2A^*e^{2i\omega_0\tau+2i\Delta k\xi} + \frac{1}{3}A^3e^{-2i\omega_0\tau-2i\Delta k\xi} \right]_+ = 0, \quad (8)$$

where  $\Delta k \equiv (\beta_1\omega_0 - \beta_0)$  (this is a central quantity in this work),  $\xi \equiv z$  and  $\tau \equiv t - \beta_1z$  are the new space-time variables in the co-moving frame, the dispersive operator  $\hat{D}(i\partial_\tau) \equiv \sum_{m=2}^{\infty} \beta_m(i\partial_\tau)^m/m!$ ,  $\gamma$  is the nonlinear coefficient of the medium, and  $\hat{S}(i\partial_\tau)$  is the operator accounting for the dispersion of the nonlinearity [provided in the Fourier space by the factor  $\omega/n(\omega)$  in Eq. (4)], which is necessary to include since the equations are broadband and SVEA is not used. For our purposes, and without loss of generality (our results are valid for any form of this operator, provided it includes at least the first order expansion term), it will be sufficient to perform the traditionally adopted approximation  $\hat{S}(i\partial_\tau) \simeq 1 + i\partial_\tau/\omega_0$ . Note that the field  $A$  feels a dispersion given by  $D(\Delta\omega) = \sum_{m=2}^{\infty} \beta_m\Delta\omega^m/m!$  (where  $\Delta\omega$  is the detuning from  $\omega_0$ ) and a positive nonlinearity, while the field  $A^*$  feels a different, 'conjugate' dispersion  $-D(-\Delta\omega) \neq D(\Delta\omega)$  and a negative nonlinearity, and both fields are forward-propagating.

Equation (8) is the central result of this paper. Since  $A$  and  $A^*$  carry the same amount of information, it is sufficient to consider a single equation only: indeed the dynamics around the positive carrier frequency ( $\omega_0$ ) must be the mirror image of the dynamics around the negative carrier frequency ( $-\omega_0$ ), due to the requirement that the electric field  $E$  must be real. The two modes  $A$  and  $A^*$  do not see each other in the absence of nonlinearity, but they mutually exchange energy when the nonlinear terms are included, thus generating new frequencies. Since the interaction modifies the phase, new resonant nonlinear effects occur. It is possible to prove (we give the non-trivial derivation in appendix A) that in Eq. (8) the energy is perfectly conserved, i.e.  $\partial_\xi \int_{-\infty}^{+\infty} |A(\xi, \tau)|^2 d\tau = 0$ , due to the detailed balance of the energy flow from  $A$  to  $A^*$  and back. It is interesting to note that the presence of the shock operator and the THG term are *essential* for energy conservation, which establishes a *deep and previously un-noticed connection* between the shock operator, THG and negative frequencies. In the absence of THG terms, Eq.



(8) would exhibit a small non-conservation of energy proportional to the missing THG energy. Another crucial point to notice is that, although Eq. (8) looks at first sight as a kind of GNLSE written for an envelope, this equation is in its physical content *completely equivalent to the UPPE* [Eq. (4)]: the analytic signal and the filtering procedure is used to completely separate positive from negative frequency parts in the UPPE, and the introduction of the envelope of the analytic signal is used only to give Eq. (8) a formal resemblance with the GNLSE, but our equation is not restricted by any of the limitations of by the latter.

#### 4. Phase-matching conditions between soliton and radiation

In order to derive phase-matching conditions between a soliton and its resonant radiations, we follow a standard procedure described in [20]. We first pose  $A(\xi, \tau) = F(\tau)e^{iq\xi} + g(\xi, \tau)$ , where  $F(\tau)$  is the (purely real) envelope of the optical soliton,  $q$  is the nonlinear mismatch and  $g$  is a small amplitude dispersive wave. After substitution into Eq. (8), and by taking only the fundamental and first order terms, one obtains (neglecting the shock term for simplicity, without loss of generality):

$$(i\partial_\xi + \hat{D})g + \gamma F^2 g^* e^{2iq\xi} + 2\gamma F^2 g = - \left( \hat{D} + \frac{1}{2}\beta_2 \partial_\tau^2 \right) F e^{iq\xi} + \gamma F^3 e^{2i\omega_0\tau + 2i\Delta k\xi - iq\xi} - \frac{1}{3}\gamma F^3 e^{-2i\omega_0\tau - 2i\Delta k\xi + 3iq\xi}. \quad (9)$$

The phase-matching conditions derived from Eq. (9) are then easily found:

$$D(\Delta\omega) = q, \quad (10)$$

$$D(\Delta\omega) = 2\Delta k - q, \quad (11)$$

$$D(\Delta\omega) = -2\Delta k + 3q. \quad (12)$$

Solving Eqs. (10)-(12) for  $\Delta\omega$  will provide all the phase-matched frequencies. In particular, Eq. (10) is very well known [5, 7] and corresponds to the positive-frequency RR, while Eq. (11), found experimentally in [13] and heuristically in [13, 15], corresponds to the negative-frequency RR. Equation (12) represents the phase-matching condition of the non-solitonic radiation due to THG (which we call *third harmonic resonant radiation*, or TH-RR), and is also a new unexpected feature of our model, which is vindicated by our numerical simulations. A curious and unexpected feature of the TH-RR radiation is that, even though this is due to the interaction between the soliton and its third harmonic waves, it is strongly detuned to long wavelengths, and is thus very feeble and unobservable in bulk crystals, but it is possible that in small-core waveguides such radiation could become experimentally accessible.

Equations (11) and (12) are impossible to find by using a single GNLSE based on SVEA and thus correspond to new features of our envelope model Eq. (8). In all the transparent bulk crystals we have examined,  $\Delta k > 0$  and  $2\Delta k \gg q$ , thus the NRR is usually strongly blueshifted with respect to the RR. This fact is ultimately due to the structure of the Lorentz oscillator theory, and we give the proof in appendix B. However, this restriction seems not to be fundamental for transparent waveguides and there might well be waveguide structures with a range of frequencies for which  $\Delta k < 0$ , so that they would be able to exhibit redshifted NRR. This interesting and potentially important question is left for future investigations.

The last important thing to mention regarding the NRR generation, is that there is an intimate relation between the NRR and the FWM between the soliton and THG, which can contribute to its amplitude at the second order of perturbation theory. In fact, one can prove by using the theory reported in [20], that the FWM between soliton and THG gives *exactly* the phase-matching condition Eq. (11). Thus, the formation of the NRR is due to two different contributions, one

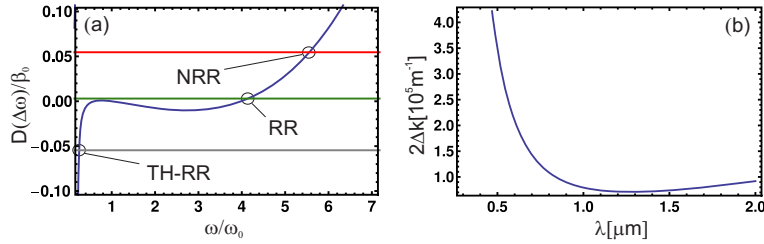


Fig. 2. (a) Phase-matching curve (normalized to  $\beta_0$ ) derived by using Eqs. (10-11) in bulk silica.  $q/\beta_0$ ,  $(2\Delta k - q)/\beta_0$  and  $(-2\Delta k + 3q)/\beta_0$  are indicated by the green, red and gray horizontal lines, respectively. RR, NRR and TH-RR frequencies are indicated by circles. (b)  $\Delta k$  for bulk silica vs. pump wavelength.

(of first order) coming directly from the conjugated Kerr term, and the other one (of second order) coming indirectly from the process of FWM of soliton and CW waves, but the amplitudes of these two contributions could be quite different since they are of different order, as we will see below. This interesting 'coincidence' has deep roots in the structure itself of Eq. (8), and in particular in the energy conservation law (see appendix A).

Figure 2(a) shows the phase-matching curve  $D(\Delta\omega)$  versus pump frequency (normalized to  $\beta_0$  and  $\omega_0$  respectively), together with its intersections with  $q$ ,  $2\Delta k - q$  and  $-2\Delta k + 3q$ , which give respectively the RR, NRR and TH-RR frequencies. Figure 2(b) shows the value of  $2\Delta k$  versus pump wavelength, showing that in bulk silica there is an optimal pump wavelength (in the normal dispersion regime) for which the NRR would be closer to the pump frequency, and thus would have an unusually large amplitude.

The above procedure is able to give only the exact frequency position of each resonant radiation that is emitted by the soliton. However, the procedure is not able to provide a correct value for the amplitude of the NRR, for the following reason. When we wrote Eq. (9), we have assumed implicitly that an analytical solution for the soliton field  $F$  is known. Although it is reasonable to assume, at least for pulses that are not sub-cycle, that the conventional Schrödinger soliton is a good approximation, this cannot be completely correct since a true soliton solution of Eq. (8) must include the conjugated Kerr term and the THG term as well. Such solution is not currently available (we leave this problem to future investigations, with few indications in the literature on how to do that [33, 34]), and thus we are temporarily forced to use the conjugated Kerr term as a source term in Eq. (9), which gives the correct phase-matching condition but cannot give the correct amplitude, since the amplitude of the NRR must be proportional to the third order dispersion  $\beta_3$ , as it happens for the normal RR. We can however always find the emitted NRR amplitude numerically, as it was done in Fig. 4.

## 5. Numerical simulations

In this section we support the above theory with numerical simulations performed by integrating Eq. (8). In Fig. 3(a) we show the spectral evolution of a 15 fs sech pulse, with peak intensity 1.4 TW/cm<sup>2</sup> propagating in bulk silica, for a pump wavelength  $\lambda_0 = 2 \mu\text{m}$ , obtained by solving Eq. (8) when the THG term is neglected. Both RR and NRR emissions are visible. Vertical black dashed lines indicate the predictions given by Eqs. (10) and (11), see also Fig. 2(a). Figure 3(b) shows the same as Fig. 3(a), when omitting also the second nonlinear term inside the square brackets in Eq. (8). No NRR radiation is generated in this case, showing that such radiation is indeed coming from the interaction between the positive and the negative frequency spectral components. Figure 3(c) shows the same simulation as in Fig. 3(a) but when switching off the

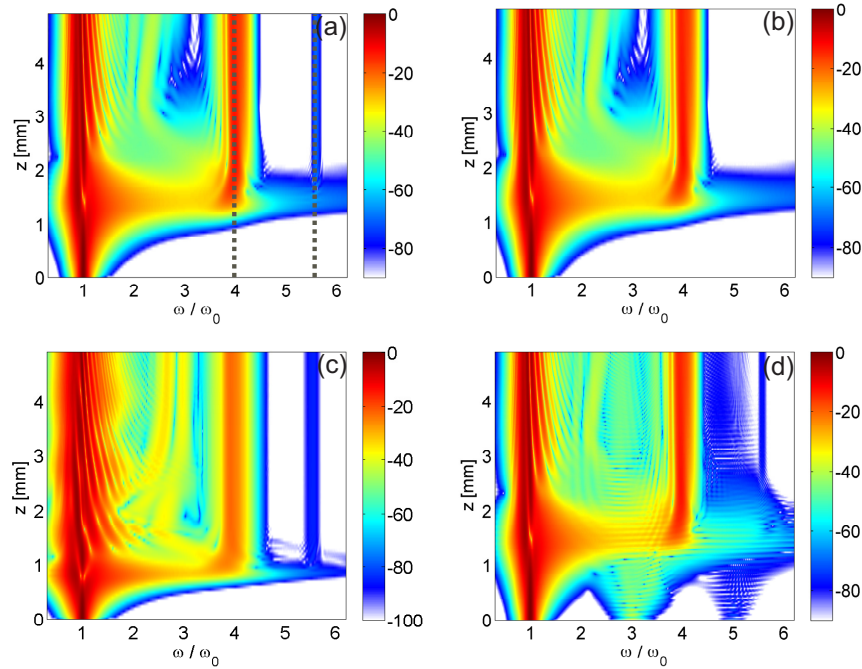


Fig. 3. (a) Contour plot of the spectral evolution of a short sech pulse in bulk silica, obtained by direct simulation of Eq. (8), when THG is neglected. The pulse is pumped at  $\lambda_0 = 2 \mu\text{m}$ , with a peak intensity of  $1.4 \text{ TW/cm}^2$  and a duration  $t_0 = 15 \text{ fsec}$ . The formation of RR and NRR is clearly visible. Vertical black dashed lines indicate the position of the radiations as predicted by Eqs. (10-11), compare with Fig. 2(a). (b) Same as (a) when also switching off the second nonlinear term inside the square brackets of Eq. (8), i.e. the conjugated Kerr term. The NRR line has completely disappeared. (c) Same as (a) but when switching off the shock operator, and for a peak intensity  $2.6 \text{ TW/cm}^2$ . (d) Results obtained with the UPPE of Eq. (4), using the same parameters as in (a). All plots are in logarithmic scale.

shock term, i.e.  $\hat{S}(i\partial_\tau) = 1$ , and for a peak intensity of  $2.6 \text{ TW/cm}^2$ . One can see that both RR and NRR are visible, conclusively proving that NRR is not due to the shock effect, even though the shock helps to further broaden the spectrum and thus to feed the soliton tail that excites the NRR. Finally, Fig. 3(d) shows the evolution of the pulse by solving the full-field UPPE, Eq. (4), which also shows evidence of small THG. Exactly the same figure is obtained by solving Eq. (4), showing that our envelope model based on the analytic signal is indeed correct.

Figure 4 shows the comparison between the amplitudes of the generated NRR when switching on and off the various terms of Eq. (8). Parameters are the same as in Fig. 3, and the spectra are recorded after  $z = 5 \text{ mm}$  of propagation. One can notice that, for the chosen parameters, the conjugated term alone overestimates the radiation amplitude, while the THG alone underestimates it with respect to the case when both terms are maintained. Figure 4 first of all proves that the FWM between soliton and THG on one hand, and the contribution of the conjugated term on the other hand give exactly the same phase-matching point for the NRR, as we have predicted in the previous section. Moreover, since when omitting the THG or the conjugated Kerr term energy is not conserved, the radiation amplitude is not correctly predicted in these cases, and the only consistent way to simulate correctly the problem is to consider all the terms in Eq. (8), which strictly conserves energy.

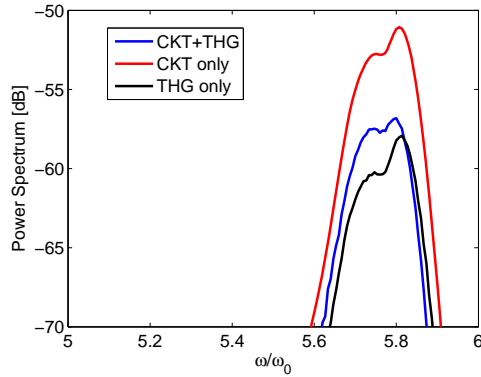


Fig. 4. Comparison between the amplitudes of the generated NRR when the conjugated Kerr term (CKT) is present but THG is absent (red line), when THG is present but conjugated Kerr term is absent (black line), and in the case when both terms are present (blue line). Equation (8) has been used in the simulation, and parameters are the same as in Fig. 3. Spectra are recorded after  $z = 5$  mm of propagation, and the vertical scale is logarithmic.

## 6. Discussion and conclusions

In conclusion, we have derived an equation that correctly describes the nonlinear interaction between the positive and the negative frequency parts of the spectrum of optical pulses. The key concept is that the envelope function is now defined in terms of the analytic signal of the electric field, therefore clearly dividing the dynamics of the negative and positive frequency parts of the spectrum, and avoiding SVEA altogether, while still retaining an envelope formulation. The interaction between positive and negative frequencies is due to the presence of a cross-phase-modulation-like term in the nonlinear polarization, the role of which we have elucidated here for the first time. By using the new equation we have analytically derived the phase-matching conditions between a soliton and the positive- and negative-frequency resonant radiation emitted by it. Our theory opens up a new realm in nonlinear optics and in other areas that are described by NLSE-like equations (for instance BEC, plasmas, etc.), since it proves that conventional treatments based on GNLSE are deficient, due to the lack of the negative frequency terms. These interactions are of course present in the UPPE and in the nonlinear Maxwell's equations, which are however less transparent and less suitable for analytical treatment than Eq. (8). Exciting future perspectives are represented by the inclusion of the Raman nonlinearity, which could provide additional unexplored non-linear effects that are not captured by conventional GNLSE based on SVEA.

### Appendix A: Proof of energy conservation of Eq. (8)

In this appendix we provide the nontrivial proof of the global energy conservation of Eq. (8), i.e. the relation

$$\frac{dN}{d\xi} = \frac{d}{d\xi} \int_{-\infty}^{+\infty} |A(\xi, \tau)|^2 d\tau = \int_{-\infty}^{+\infty} [A_\xi A^* + \text{c.c.}] d\tau = 0. \quad (13)$$

We start with the case in which the filtering procedure (i.e. the removal of the negative frequency part in the equation) is absent. Note that the usual  $|A|^2 A$  term, is conservative by itself, but the conjugated Kerr term  $|A|^2 A^*$  and THG term  $A^3/3$  must be both retained to conserve energy. Dispersive term is obviously conservative by itself.

Let us start from the usual SPM term, so let's demonstrate that  $A_\xi = i\gamma(1 + i/\omega_0\partial_\tau)|A|^2A$  conserve energy. We get

$$\begin{aligned}\frac{dN}{d\xi} &= \int d\tau \left\{ i\gamma|A|^4 - \frac{\gamma}{\omega_0}(|A|^2A)_\tau A^* - i\gamma|A|^4 - \frac{\gamma}{\omega_0}(|A|^2A^*)_\tau \right\} A = \\ &= -\frac{\gamma}{\omega_0} \int d\tau \{ 2(|A|^2)_\tau |A|^2 + A_\tau A^* |A|^2 + A_\tau^* A |A|^2 \} = \\ &= -\frac{\gamma}{\omega_0} \int d\tau 3(|A|^2)_\tau |A|^2 = 0.\end{aligned}\quad (14)$$

Last step can be verified by integrating by parts and assuming vanishing boundary conditions for the fields.

Now let us show that  $A_\xi = i\gamma(1 + i/\omega_0\partial_\tau)(|A|^2A^*e^{2i\phi} + A^3/3e^{-2i\phi})$  conserves energy ( $\phi \equiv \omega_0\tau + \Delta k\xi$ ). We get (all integrals are performed in  $d\tau$ ):

$$\begin{aligned}\frac{dN}{d\xi} &= \int i\gamma|A|^2A^{2*}e^{2i\phi} + i\frac{\gamma}{3}|A|^2A^2e^{-2i\phi} - \frac{\gamma}{\omega_0} \left[ (|A|^2A^*)_\tau + 2i\omega_0|A|^2A^* \right] A^*e^{2i\phi} + \\ &\quad - \frac{\gamma}{\omega_0} \left[ A^2A_\tau - 2i\frac{\omega_0}{3}A^3 \right] A^*e^{-2i\phi} + \text{c.c.} = \\ &= \int -i\gamma|A|^2A^{2*}e^{2i\phi} + i\gamma|A|^2A^2e^{-2i\phi} - \frac{\gamma}{\omega_0} \left[ A_\tau A^{3*} + 2A_\tau^* A^* |A|^2 \right] e^{2i\phi} + \\ &\quad - \frac{\gamma}{\omega_0} \left[ A_\tau A |A|^2 \right] e^{-2i\phi} + \text{c.c.} = \\ &= \frac{\gamma}{2\omega_0} \int (A_\tau A^{3*} + 3|A|^2A^*A_\tau^*)e^{2i\phi} + (A^3A_\tau^* + 3|A|^2AA_\tau)e^{-2i\phi} + \\ &\quad - 2(A_\tau A^{3*} + 2|A|^2A^*A_\tau^*)e^{2i\phi} - 2|A|^2AA_\tau e^{-2i\phi} + \text{c.c.} = \\ &= \frac{\gamma}{2\omega_0} \int (2A_\tau A^{3*} + 6|A|^2A^*A_\tau^* - 2A_\tau A^{3*} - 6|A|^2A^*A_\tau^*)e^{2i\phi} + \text{c.c.} = 0.\end{aligned}$$

We now turn to the full case in which the filtering is introduced. Useful relations:

$$\mathcal{H}[E(t)] = \frac{1}{\pi t} \otimes E(t) = \mathbb{P} \int_{-\infty}^{+\infty} \frac{E(t-t')}{\pi t'} dt' \quad (15)$$

$$\mathcal{H}[E(t)e^{\pm i\omega_0 t}] = \mathbb{P} \int_{-\infty}^{+\infty} \frac{E(t-t')e^{\pm i\omega_0(t-t')}}{\pi t'} dt' = e^{\pm i\omega_0 t} \left[ \frac{e^{\mp i\omega_0 t}}{\pi t} \otimes E(t) \right] \quad (16)$$

$$\mathcal{F} \left\{ -\frac{i}{\pi t} \right\} = \text{sgn}(\omega), \quad \mathcal{F} \left\{ -\frac{ie^{\pm i\omega_0 t}}{\pi t} \right\} = \text{sgn}(\omega \pm \omega_0) \quad (17)$$

Defining the operator  $\hat{Q}[\cdot] = \hat{I} - i\mathcal{H}[\cdot]$  as the operator that when applied to an arbitrary real function  $f(t)$  gives its analytic signal ( $\hat{I}$  is the identity operator), we have

$$\mathcal{F} \{ \mathcal{E}(t) \} \equiv F \{ \hat{Q}[E(t)] \} = \mathcal{F} \{ E(t) - i\mathcal{H}[E(t)] \} = E_\omega + \text{sgn}(\omega)E_\omega = 2E_{\omega>0} \quad (18)$$

and

$$\mathcal{F} \{ \hat{Q}[E(t)e^{-i\omega_0 t}] \} = E_{\omega-\omega_0} + \text{sgn}(\omega)E_{\omega-\omega_0} = 2E_{\Delta\omega>-\omega_0}. \quad (19)$$

The analytic signal of the nonlinear polarization can be written as

$$\begin{aligned}\hat{Q}[P_{NL}(z,t)] &= \frac{3\chi^{(3)}}{8} \left[ |A|^2Ae^{-i\omega_0 t + i\beta_0 z} - i \left[ \frac{e^{i\omega_0 t}}{\pi t} \otimes |A|^2A \right] e^{-i\omega_0 t + i\beta_0 z} + |A|^2A^*e^{i\omega_0 t - i\beta_0 z} + \right. \\ &\quad \left. - i \left[ \frac{e^{-i\omega_0 t}}{\pi t} \otimes |A|^2A^* \right] e^{i\omega_0 t - i\beta_0 z} + \frac{2}{3}A^3e^{-3i\omega_0 t + 3i\beta_0 z} \right] \quad (20)\end{aligned}$$

and the nonlinear polarization

$$A_p(z,t) = \frac{3\chi^{(3)}}{8} \left\{ |A|^2 A - i \left[ \frac{e^{i\omega_0 t}}{\pi t} \otimes |A|^2 A \right] + \right. \\ \left. + |A|^2 A^* e^{2i\omega_0 t - 2i\beta_0 z} - i \left[ \frac{e^{-i\omega_0 t}}{\pi t} \otimes |A|^2 A^* \right] e^{2i\omega_0 t - 2i\beta_0 z} + \frac{2}{3} A^3 e^{-2i\omega_0 t + 2i\beta_0 z} \right\}$$

Equation (8) in the time domain, written in the reference frame moving with group velocity  $\beta_1^{-1}$ , reads (again we define  $\phi \equiv \omega_0 \tau + \Delta k \xi$ ,  $\Delta k \equiv \beta_1 \omega_0 - \beta_0$ ):

$$i \frac{\partial A}{\partial \xi} + \hat{D}(i\partial_\tau)A + \frac{\gamma}{2} \left( 1 + \frac{i}{\omega_0} \frac{\partial}{\partial \tau} \right) \left\{ |A|^2 A - i \left[ \frac{e^{i\omega_0 \tau}}{\pi \tau} \otimes |A|^2 A \right] + \right. \\ \left. + |A|^2 A^* e^{2i\phi} - i \left[ \frac{e^{-i\omega_0 \tau}}{\pi \tau} \otimes |A|^2 A^* \right] e^{2i\phi} + \frac{2}{3} A^3 e^{-2i\phi} \right\} = 0 \quad (21)$$

Terms with same color in the Eq. (21) are expected to conserve energy independently. The term in green color (first term inside curly brackets) obviously conserves energy: it corresponds to the conventional unfiltered case of the derivative nonlinear Schrödinger equation, for which it is known that energy conservation holds, as showed above.

Let's show that the red term (second term inside curly brackets) is conservative, i.e.  $\partial_z A = i\gamma/2(1 + i/\omega_0 \partial_\tau) \left[ -i \frac{e^{i\omega_0 \tau}}{\pi \tau} \otimes |A|^2 A \right]$  conserves energy. In order to do this, we go from the time domain to the frequency domain by repeatedly using Parseval's theorem, and exploit the fact that  $\text{sgn}(\omega + \omega_0) \mathcal{F}[A^*] = \mathcal{F}[A^*]$  and  $\text{sgn}(\omega - \omega_0) \mathcal{F}[A^*]^* = -\mathcal{F}[A^*]^*$ :

$$\begin{aligned} \frac{dN}{d\xi} &= \frac{\gamma}{2} \int d\tau i \left[ \left( 1 + \frac{i}{\omega_0} \frac{\partial}{\partial \tau} \right) \left( -i \frac{e^{i\omega_0 \tau}}{\pi \tau} \otimes |A|^2 A \right) \right] A^* + \\ &\quad - i \left[ \left( 1 - \frac{i}{\omega_0} \frac{\partial}{\partial \tau} \right) \left( i \frac{e^{-i\omega_0 \tau}}{\pi \tau} \otimes |A|^2 A^* \right) \right] A = \\ &= \frac{\gamma}{2} \int d\omega i \left[ \left( 1 + \frac{\omega}{\omega_0} \right) \text{sgn}(\omega + \omega_0) \mathcal{F}\{|A|^2 A\} \right] \mathcal{F}[A]^* + \\ &\quad + i \left[ \left( 1 - \frac{\omega}{\omega_0} \right) \text{sgn}(\omega - \omega_0) \mathcal{F}\{|A|^2 A^*\} \right] \mathcal{F}[A^*]^* = \\ &= \frac{\gamma}{2} \int d\omega i \left[ \left( 1 + \frac{\omega}{\omega_0} \right) \mathcal{F}\{|A|^2 A\} \right] \mathcal{F}[A]^* - i \left[ \left( 1 - \frac{\omega}{\omega_0} \right) \mathcal{F}\{|A|^2 A^*\} \right] \mathcal{F}[A^*]^* = \\ &= \frac{\gamma}{2} \int d\tau i \left[ \left( 1 + \frac{i}{\omega_0} \frac{\partial}{\partial \tau} \right) |A|^2 A \right] A^* - i \left[ \left( 1 - \frac{i}{\omega_0} \frac{\partial}{\partial \tau} \right) |A|^2 A^* \right] A = \quad (22) \\ &= \frac{\gamma}{2} \int d\tau \left[ i|A|^4 - \frac{1}{\omega_0} (|A|^2 A)_\tau A^* - i|A|^4 - \frac{1}{\omega_0} (|A|^2 A^*)_\tau A \right] = \\ &= -\frac{\gamma}{2\omega_0} \int d\tau \left[ 2(|A|^2)_\tau |A|^2 + A_\tau A^* |A|^2 + A^* A |A|^2 \right] = -\frac{\gamma}{2\omega_0} \int d\tau 3(|A|^2)_\tau |A|^2 = 0, \end{aligned}$$

where in the last step we have assumed that all fields are localized in time. It is worth noting that Eq. (22) is equivalent to the energy conservation of the term  $(\gamma/2)(1 + i/\omega_0 \partial_\tau)|A|^2 A$  without filtering. In practice, when calculating  $dN/d\xi$  the Hilbert transform has disappeared.

Let us now show that the blue terms (last three terms inside curly brackets) are conservative.

We consider now the term  $A_\xi = i\gamma/2(1 + i/\omega_0 \partial_\tau) \left[ \left( -i \frac{e^{-i\omega_0 \tau}}{\pi \tau} \otimes |A|^2 A^* \right) e^{2i\phi} \right]$ . We have for

this term:

$$\begin{aligned}
\frac{dN}{d\xi} &= \frac{\gamma}{2} \int d\tau i \left\{ \left( 1 + \frac{i}{\omega_0} \frac{\partial}{\partial \tau} \right) \left[ \left( -i \frac{e^{-i\omega_0 t}}{\pi \tau} \otimes |A|^2 A^* \right) e^{2i\phi} \right] \right\} A^* + \\
&\quad - i \left\{ \left( 1 - \frac{i}{\omega_0} \frac{\partial}{\partial \tau} \right) \left[ \left( i \frac{e^{i\omega_0 t}}{\pi \tau} \otimes |A|^2 A \right) e^{-2i\phi} \right] \right\} A = \\
&= \frac{\gamma}{2} \int d\omega i \left\{ \left( 1 + \frac{\omega}{\omega_0} \right) \left[ \text{sgn}(\omega + \omega_0) \mathcal{F}\{|A|^2 A^*\}_{\omega+2\omega_0} \right] \right\} \mathcal{F}[A]^* e^{2i\Delta k \xi} + \\
&\quad + i \left\{ \left( 1 - \frac{\omega}{\omega_0} \right) \left[ \text{sgn}(\omega - \omega_0) \mathcal{F}\{|A|^2 A\}_{\omega-2\omega_0} \right] \right\} \mathcal{F}[A^*]^* e^{-2i\Delta k \xi} = \\
&= \frac{\gamma}{2} \int d\tau i \left\{ \left( 1 + \frac{i}{\omega_0} \frac{\partial}{\partial \tau} \right) \left[ |A|^2 A^* e^{2i\phi} \right] \right\} A^* - i \left\{ \left( 1 - \frac{i}{\omega_0} \frac{\partial}{\partial \tau} \right) \left[ |A|^2 A e^{-2i\phi} \right] \right\} A.
\end{aligned}$$

Again the Hilbert transform has disappeared. The considered term gives the same contributions to the energy as  $|A|^2 A^*$ , so the total energy variation corresponding to the blue terms can be calculated considering the equation  $A_\xi = i\gamma(1 + i/\omega_0 \partial_\tau)(|A|^2 A^* e^{2i\phi} + A^3/3e^{-2i\phi})$ , that we showed before (unfiltered case) to conserve energy.

## Appendix B: Positiveness of $\Delta k$ for transparent bulk media

Transparent bulk media, i.e. media without a waveguide dispersion contribution, can be described by the Sellmeier equation

$$n(\omega) = \sqrt{1 + \sum_{j=1}^m \frac{B_j \omega_j^2}{\omega_j^2 - \omega^2}}, \quad (23)$$

where  $B_j$  are appropriate dimensionless coefficients, and  $\omega_j$  are material resonances. Near one particular resonance  $j = R$ ,  $\omega = \omega_R + \Delta\omega$ , with  $\Delta\omega \ll \omega_R$ , one can consider a single term in the sum:

$$n(\omega) \simeq \sqrt{1 + \frac{B_R \omega_R^2}{\omega_R^2 - \omega^2}} \simeq \sqrt{1 - \frac{\mu}{2(\omega - \omega_R)}}, \quad (24)$$

with  $\mu \equiv B_R \omega_R$ . The physical requirements that this refractive index must satisfy in order to have a value of  $\Delta k < 0$  in the anomalous dispersion, where the soliton can propagate, are  $n(\omega) > 0$ ,  $\partial_\omega n < 0$  and  $\partial_\omega^2 n < (2/\omega)|\partial_\omega n|$ . These three conditions are impossible to fulfill simultaneously for a positive  $\mu$ , and therefore in bulk media NRR emitted by solitons can only be observed in the blue part of the spectrum.

Things can change drastically in a lossy bulk medium. Again, near a resonance one has

$$n(\omega) \simeq \sqrt{1 - \frac{\mu}{2(\omega - \omega_R + i\gamma)}}, \quad (25)$$

where  $\gamma$  is the resonance damping parameter. In this case, one can easily show that in the approximate range  $\omega_R - \gamma < \omega < \omega_R$  one can fulfill the above conditions for  $\Delta k < 0$ , but this range is located around the maximum of absorption. In practical experiments, this limits strongly the observability of NRR in the red part of the spectrum in bulk media - for instance in metals the region where the soliton should be pumped is typically located in close proximity of the plasma frequency. This reasoning does not hold in general for waveguides, for which the waveguide dispersion can compete with the material dispersion, however it remains to be seen whether the above three conditions can be satisfied simultaneously in realistic optical fibers or other confining microstructures.