Arbitrary-order nonlinear contribution to self-steepening

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On the basis of the recently published generalized Miller formulas, we derive the spectral dependence of the contribution of arbitrary-order nonlinear indices to the group-velocity index. We show that in the context of laser filamentation in gases, all experimentally accessible orders (up to the ninth-order nonlinear susceptibility $\chi^{(9)}$ in air and $\chi^{(11)}$ in argon) have contributions of alternative signs and similar magnitudes. Moreover, we show both analytically and numerically that the dispersion term of the nonlinear indices must be considered when computing the intensity-dependent group velocity. © 2010 Optical Society of America

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Nonlinear optics [1] relies on the nonlinear properties of the propagation medium, among which the successive orders of the nonlinear susceptibility are essential parameters. However, due to the difficulty in measuring them experimentally, their knowledge is generally limited to the first nonzero order ($\chi^{(2)}$ or $\chi^{(3)}$, depending on medium symmetry). Furthermore, the available laser sources drastically limit the wavelengths available for such measurements, so that reliable dispersion curves for higher-order susceptibilities cannot be deduced from the sparse experimental data available to date.

The lack of data led to the neglect of these higher order Kerr terms in most numerical simulations of, e.g., both self-guided filaments in ultrashort intense laser pulses [2–5] or the propagation of high-intensity pulses in hollow-core fibers [6]. Similarly, the Kerr contribution to the group velocity is most generally limited to the third order and treated as dispersionless in the lack of data about its dispersion [3,4]. Recently, however, the measurement of the higher-order refractive indices up to n_8 in N_2 and O_2, and up to n_{10} in argon [7,8], followed by the generalization of the Miller formulas [9] to any order of nonlinearity [10], provided a new insight into the spectral dependence of the nonlinear refractive index at high incident intensity.

This allowed us to show that these terms cannot be neglected and can even provide the dominant contribution stabilizing the self-guided filaments [11]. Furthermore, in argon-filled hollow-core fibers, these terms are necessary to obtain quantitative agreement of numerical simulations with experimental data [12,13]. But these works focused on the contribution of higher-order Kerr terms to phase velocity. The contribution of the spectral dispersion of higher-order indices to group velocity was not considered, although it can be expected to impact the propagation, and in particular the self-steepening term.

Here we derive an explicit expression for the contribution of any order of the nonlinear refractive indices to the group-velocity index. We show that in filamentation, all nonlinear orders of the group-velocity index can have similar orders of magnitude and must be considered. Furthermore, especially in the UV, the dispersion of the nonlinear indices cannot be neglected when calculating self-steepening.

At arbitrary intensity I and frequency ω, the refractive index can be expressed as [1]

$$n(\omega) = n_0(\omega) + n_2(\omega)I + n_4(\omega)I^2 + \ldots = \sum_{j=0}^{\infty} n_{2j}(\omega)I^j. \tag{1}$$

In gases, where $n - 1 \ll 1$, the refractive index $n(\omega) = \sqrt{1 + \sum_{j=0}^{\infty} \chi^{(2j+1)}(\omega)I^j}$ can be approximated by [1]

$$n_0(\omega) \approx 1 + \frac{1}{2}\chi^{(1)}(\omega), \tag{2}$$

$$n_{2j}(\omega) \approx Z^{(2j+1)}\chi^{(2j+1)}(\omega), \tag{3}$$

where $\chi^{(2j+1)}$ is the $(2j+1)$th-order nonlinear susceptibility and the $Z^{(2j+1)}$ are frequency-independent factors. If a pulse can be described as a carrier wave modulated by an envelope with a sufficiently narrow spectrum to allow neglection of the envelope deformations over short distances, then a group-velocity index can be defined as

$$n_g(\omega) = \frac{c}{v_g} = n(\omega) + \omega\frac{dn}{d\omega}, \tag{4}$$

where v_g is the group velocity and c is the speed of light in vacuum. Defining $Z^{(1)} = 1/2$ and considering Eqs. (1)–(3), n_g is rewritten:

$$n_g(\omega) = \sum_{j=0}^{\infty}\left(n_{2j}(\omega) + \omega\frac{dn_{2j}}{d\omega}\right)I^j \equiv \sum_{j=0}^{\infty} n_{g,2j}I^j, \tag{5}$$

$$\approx 1 + \sum_{j=0}^{\infty} Z^{(2j+1)}\left(\chi^{(2j+1)}(\omega) + \omega\frac{d\chi^{(2j+1)}}{d\omega}\right)I^j. \tag{6}$$

Identifying the terms for each power of the intensity, we obtain the contribution of each order of nonlinearity to the group-velocity index:

$$n_{g,0}(\omega) - 1 \approx \frac{1}{2}\left(\chi^{(1)} + \omega\frac{d\chi^{(1)}(\omega)}{d\omega}\right), \tag{7}$$

$$\forall\ j \geq 1, \qquad n_{g,2j}(\omega) \approx Z^{(2j+1)}\left(\chi^{(2j+1)} + \omega\frac{d\chi^{(2j+1)}(\omega)}{d\omega}\right). \tag{8}$$

The first expression corresponds to the usual linear contribution, while the first term in the expression of the group-velocity index $n_{2,g}$ corresponds to the classical self-steepening term [1,14,15]. Within the elastically bound electron model, the susceptibility of the arbitrary order is given up to any order by the generalized Miller formulas [10]:

$$\chi^{(1)}(\omega) = \frac{Ne^2}{m\epsilon_0({\omega_0}^2 - \omega^2 + i\omega_0\gamma)}, \tag{9}$$

$$\forall\ q \geq 2, \qquad \chi^{(q)}(\omega) = \frac{Ne}{\epsilon_0}\left(\frac{e}{m}\right)^q Q^{(q)}\frac{1}{\Omega(\omega)^{q+1}}, \tag{10}$$

where m and $-e$ are the electron mass and charge, ϵ_0 is the permittivity of vacuum, N is the density of dipoles in the propagation medium, γ is the width of the resonance at frequency ω_0, and $Q^{(q)}$ describes the potential well where the electron oscillates; $\Omega(\omega) = \omega_0^2 - \omega^2 + i\omega_0\gamma$. Inserting these expressions into Eqs. (7) and (8) yields

$$n_{g,0}(\omega) - 1 \approx (n_0(\omega) - 1)\frac{\omega_0^2 + \omega^2 + i\omega_0\gamma}{\omega_0^2 - \omega^2 + i\omega_0\gamma}, \tag{11}$$

$$\forall\ j \geq 1, \qquad n_{g,2j}(\omega) \approx n_{2j}(\omega)\frac{\omega_0^2 + (4j+3)\omega^2 + i\omega_0\gamma}{\Omega(\omega)}. \tag{12}$$

These equations provide a general expression of each nonlinear contribution to the group-velocity index n_g. As a consequence, it allows evaluation of the impact of higher-order Kerr terms on self-steepening in the context of laser filamentation in gases or the propagation of ultrashort pulses in hollow fibers. In the following, we consider the propagation of high-intensity pulses in transparent media, far from resonance. In this case, $|\omega_0 - \omega| \gg \gamma$, so that $\omega_0^2 + \omega^2 > |\omega_0^2 - \omega^2| \gg \omega_0\gamma$. The imaginary parts of Eqs. (11) and (12) become negligible:

$$n_{g,0}(\omega) - 1 \approx (n_0(\omega) - 1)\frac{\omega_0^2 + \omega^2}{\omega_0^2 - \omega^2}, \tag{13}$$

$$n_{g,2j}(\omega) \approx n_{2j}\frac{\omega_0^2 + (4j+3)\omega^2}{\omega_0^2 - \omega^2}. \tag{14}$$

Note that the negative values obtained for $\omega > \omega_0$ correspond to the well-known region of the negative group-velocity index [16]. Figure 1 displays the spectral dependence from Eq. (14), based on the recent experimental measurements of n_{2j} at 800 nm [7,8,11], extrapolated to the whole visible spectrum by applying generalized Miller formulas [10] and the dispersion data of Zhang *et al.* [17].

From Eqs. (13) and (14), we can estimate the ratio of the successive terms of the group-velocity index

$$\frac{n_{g,2}I}{n_{g,0} - 1} = \frac{n_2 I}{n_0 - 1}\frac{\omega_0^2 + 7\omega^2}{\omega_0^2 + \omega^2}, \tag{15}$$

$$\forall\ j \geq 1, \qquad \frac{n_{g,2j+2}I^{j+1}}{n_{g,2j}I^j} = \frac{n_{2j+2}I}{n_{2j}}\frac{\omega_0^2 + (4j+7)\omega^2}{\omega_0^2 + (4j+3)\omega^2}. \tag{16}$$

The second factor of Eqs. (15) and (16) is of the order of 1. Therefore, the orders of magnitude of the ratio of successive terms are driven by the ratio of the nonlinear indices n_{2j}, multiplied by I. The values displayed in Fig. 1 imply that, for $I < 10^{14}$ W/cm^2, $n_{2j}I^j \ll n_0 - 1$. Self-steepening is, therefore, as well known [14,15], a second-order term in the nonlinear Schrödinger equation (NLSE) describing the nonlinear propagation of light in a nonlinear transparent medium. Furthermore, all known terms in $n_{2j}I^j$ have alternate signs and comparable orders of magnitude [7,8,10]. The same, therefore, applies to the terms in $n_{g,2j}I^j$, which must all be taken into account when describing self-steepening e.g., in the context of filamentation, where the intensity is clamped around 5×10^{13} W/cm^2 [18,19], or of the propagation in hollow fibers.

Equation (14) provides an estimation of the error performed when neglecting the dispersion term in the contribution of the higher-order indices to the group-velocity index. Figure 2 displays the relative error $1 - n_{2j}/n_{g,2j}$ implied at atmospheric pressure when neglecting the dispersion terms of the Kerr contributions to the group velocity. The calculations are based on the same data as in Fig. 1. As is clear from Eq. (14), this error decreases for longer wavelengths, where dispersion is smoother. At 800 nm, it amounts to ~20% and may be considered acceptable, although not negligible. However, at blue or UV

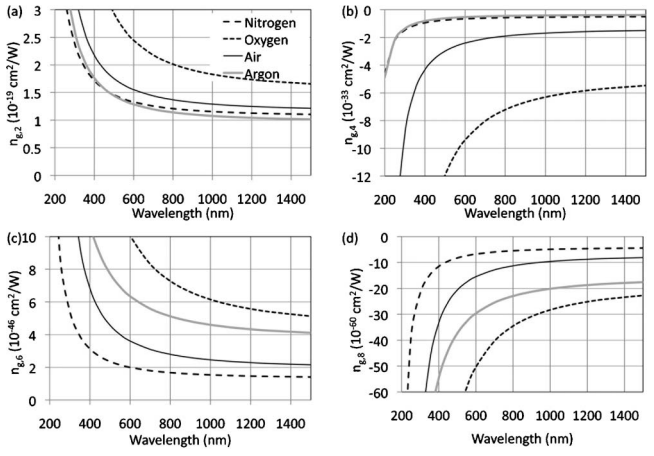

Fig. 1. Spectral dependence of the nonlinear group-velocity indices: (a) $n_{g,2}$, (b) $n_{g,4}$, (c) $n_{g,6}$, and (d) $n_{g,8}$ of O_2, N_2, air, and Ar at 1 atm.

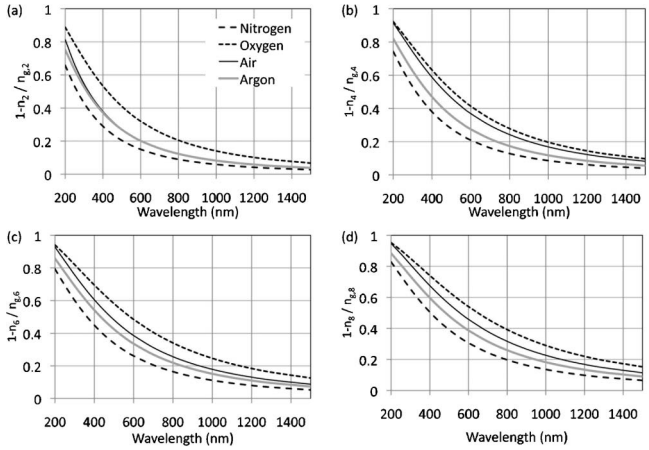

Fig. 2. Relative error induced when neglecting the dispersion of the Kerr terms in the group velocity: (a) $1 - n_2/n_{g,2}$, (b) $1 - n_4/n_{g,4}$, (c) $1 - n_6/n_{g,6}$, and (d) $1 - n_8/n_{g,8}$ of O_2, N_2, air, and Ar at 1 atm.

wavelengths, the dispersion term dominates and must be considered in the equations.

Numerical simulations of the propagation of a 35 fs pulse in a 1-m-long hollow-core fiber filled with 1.4 bars argon, confirm this finding. As described in detail earlier [13], the model implements the NLSE, including the higher-order Kerr terms. We compared the code output with and without the contribution of the higher-order indices to the group-velocity index up to the term in n_{10}, i.e., the terms of Eq. (6) for $1 \le j \le 5$. As can be seen in Fig. 3, the consideration of the full steepening term affects the spectrum by deforming the pulse

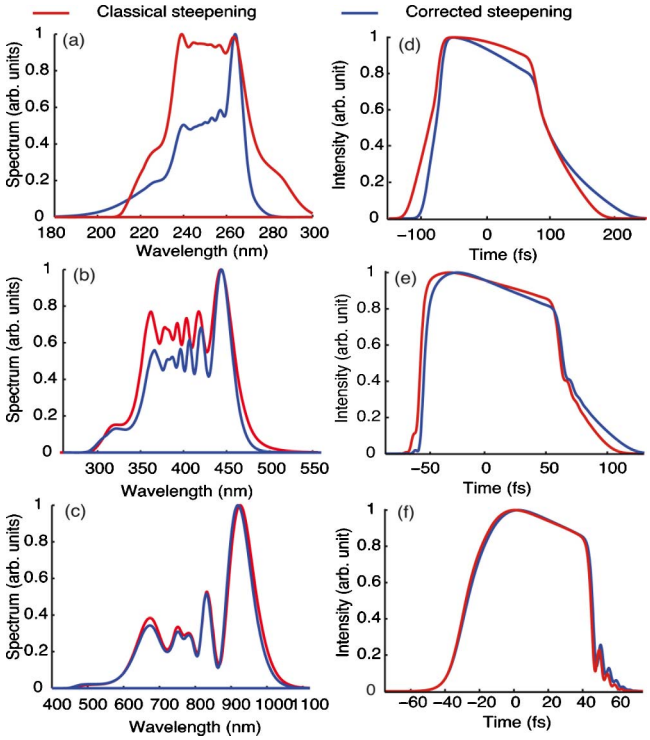

Fig. 3. (Color online) Influence of the higher-order indices contribution to the group-velocity index on the propagation of a 35 fs, pulse with fixed $n_2(\lambda)I$ in a 1-m-long hollow-core fiber filled with 1.4 bars argon: (a), (d) 250 nm, 286 μJ; (b), (e) 400 nm, 360 μJ; and (c), (f) 800 nm, 400 μJ.

envelope. It simultaneously redshifts the central part of the spectrum and blueshifts its edges. Furthermore, as predicted by the analytic calculations, the contribution of the dispersion of the higher-order Kerr terms is larger in the UV and negligible in the IR. These terms must, therefore, be considered in numerical simulations, especially while investigating spectral broadening.

In conclusion, based on the recent generalization of the Miller formulas, we have estimated the contribution of higher-order indices to the group-velocity index. These contributions define the self-steepening term of the NLSE. They have alternate signs and comparable absolute values in intensity regimes typical of filamentation. All nonlinear terms must, therefore, be considered in the evaluation of the self-steepening of ultrashort intense laser pulses propagating in transparent Kerr media. Furthermore, we demonstrate both analytically and numerically that their spectral dispersion cannot be neglected either, especially at shorter wavelengths.

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