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Higher-order Kerr effect and harmonic cascading in gases

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In homogeneous, isotropic media, the nonlinear variation of the refractive index can be expressed in terms of a power series of the light intensity $I$: $\Delta n = n_2 I + n_4 I^2 + \ldots + n_{2n} I^n$. However, higher-order nonlinearities have usually been regarded as negligible, as compared to the leading third-order nonlinearity $n_2$ (Kerr effect). A recent experiment aimed at characterizing the higher-order Kerr effect (HOKE) [1,2] has sparked a vivid debate on the actual role of these higher-order terms in nonlinear beam-propagation dynamics. Indeed, the effective saturation of the HOKE nonlinearity could lead to filamentation of femtosecond pulses in gases without the plasma playing an active role [3]. The mechanism advocated in this case to arrest the collapse of the wave packet was the saturation of the nonlinearity rather than the formation of plasma, as commonly accepted [4]. Experiments have either confirmed [5–7] or ruled out [8–10] a preponderant role of HOKE in filamentation dynamics.

In the strong cascading limit, cascading of harmonics gives effective higher-order nonlinearities that compete with the inherent material ones. Second-order $\chi^{(2)}$ : $\chi^{(2)}$ cascading gives an effective $\chi^{(3)}$ nonlinearity [1,12], which can be made ultrafast and negative (self-defocusing) and even strong enough to overcome the self-focusing material Kerr nonlinearity [13]. Similarly, third-order $\chi^{(3)}$ : $\chi^{(3)}$ cascading gives an effective $\chi^{(3)}$ [14]. Here we study cascading contributions to the nonlinear HOKE coefficients in gases. This is unlike previous investigations that focused on harmonic yield from cascading [7–9]. Cascading relies on a phase mismatch $\Delta k_{1\text{in}} = k_1(o_m) - mk(o_1) \neq 0$ between the fundamental wave (FW, $o_1$) and the mth harmonic ($o_m$). After two coherence lengths $2\pi/|\Delta k_{1\text{in}}|$ backconversion to the FW is complete, and as $\Delta k_{1\text{in}} \neq 0$ the harmonic has a different phase velocity than the FW. Thus, the backconverted FW photons are phase shifted with respect to the unconverted FW photons. For a strong phase mismatch $\Delta k_{1\text{in}} L \gg 2\pi$ ($L$ is the interaction length), this process repeats many times and the FW feels a nonlinear phase shift parameterized by an effective higher-order nonlinear index $n_{2\text{casc}} \propto \Delta k_{1\text{in}}$. We show that in gases cascading can affect the nonlinear index change and in particular the saturation intensity. Specifically, lowest-order cascading comes from a phase-mismatched third-harmonic generation (THG) process, which we show generates an $n_4$ term that is negative and of significant strength. Similarly, cascading of HOKE terms gives contributions to subsequent orders of nonlinearity ($n_6, n_8, \ldots$) with values comparable with recent experimental [1,2] and theoretical HOKE coefficients [15]. This implies that in the strong cascading limit, defined later, cascading may contribute significantly to the nonlinear dynamics.

We now discuss cascaded THG, and will generalize to higher harmonics later. In the slowly varying envelope approximation the mks electric-field coupled-wave propagation equations for THG $o_1 + o_1 + o_1 \rightarrow o_3$ in the plane-wave limit in an isotropic lossless medium are

\begin{align} 
\left(i\frac{\partial}{\partial z} - \frac{1}{2}k_2(o_1)\partial_{\tau}\right) E_1 + \Gamma_{o_1}^3 |E_1|^2 E_1^3 + 2|E_1|^2 |E_3|^2 = 0, \\
\left(i\frac{\partial}{\partial z} - i\Delta k_3/2 - \frac{1}{2}k_2(o_3)\partial_{\tau}\right) E_3 + \Gamma_{o_3}^3 |E_3|^2 E_3^3 + 2|E_3|^2 |E_1|^2 + \frac{1}{3} |E_3|^4 e^{i\Delta k_3 z} = 0. 
\end{align}

$k(o) = n(o) o/c$ is the wave number, $n(o)$ is the linear refractive index, $k_m(o) \equiv d\omega/k(o)/d\omega^m$ are the dispersion coefficients, $d_1 = k_1(o_1) - k_3(o_3)$ is the group-velocity mismatch, and $\Gamma_{o_j}^3 = 3\omega_j \chi^{(3)}(\omega_j)/8cn(o_j)$, $j = 1, 3$. In the strong cascading limit, $\Delta k_{3\text{in}} L \gg 2\pi$ and no FW depletion are assumed [16], so third-harmonic (TH) self- and cross-phase modulation in Eq. (2) are negligible and only the $E_3/3$ term is relevant. The ansatz $E_{3\text{casc}}(z, \tau) = e^{-i\Delta k_{3\text{in}} z} E_3^3(z, \tau) \Gamma_{o_3}^3/(\Delta k_{3\text{in}})$: the TH is effectively slaved to the FW. Inserting this result in Eq. (1) yields the nonlinear term $\Gamma_{o_1}^3 |E_1|^2 [1 - |E_1|^2 \Gamma_{o_3}^3/\Delta k_{3\text{in}} + 2|E_1|^4 \Gamma_{o_3}^3/\Delta k_{3\text{in}}^3]$, where we used $\Gamma_{o_j}^3/3 \simeq \Gamma_{o_j}^3$ and $n(o_1)/n(o_3) \approx 1$. Converting to intensity $I \rightarrow |E|^2/|E_0|^2$, we get $(n_2 A_1|A_1|^2 + n_4 A_2 A_1|A_1|^4 + n_6 A_3 A_2 A_1|A_1|^6)/c$, where $n_2 = 3\chi^{(3)}/4e\epsilon_0 c^2 n_0(o_1)$ is the Kerr nonlinearity. Thus, the cascaded
When including cascading the inversion intensity peaks in the near-IR, and is pushed to become lower without cascading at the beginning of the mid-IR. The calculations are based on the HOKE material nonlinearities from [1,2], but we found similar results using the coefficients from [15] (seemingly, the HOKE coefficients of [1,2] and [15] do not match, but this is in part due to the large difference in the number of HOKE orders). Note that the diatomic gases (air, O₂, and N₂) also have fractional contributions from rotational and vibrational Raman effects that contribute to the overall observed nonlinearity as well, and this might make it more difficult to observe effects of cascading.

The strong cascading limit [16] assumes that the coherence length \( l_{\text{coh}} = \pi / |\Delta k_{13}| \) is much smaller than (a) the interaction length (\( |\Delta k_{13}|L \gg 2\pi \)) and (b) the characteristic nonlinear length \( |\Delta k_{13}|/|\Gamma_{\text{non}(m)}E_{\text{in}}^{(m)-1}| \gg 1 \). To lowest order, no pump depletion is found: \( E_{\text{in}} \) is just phase modulated in \( z \). When \( |\Delta k_{13}|L \gg 2\pi \), the expressions for the following orders are lengthy, but briefly in the strong cascading limit the harmonic is reduced to \( E_{\text{in}} \Delta n \propto -E_{\text{in}}^{(p)} e^{-|\Delta k_{13}|L/2} \). Inserting these cascading terms in the FW equation, which contains all possible harmonic self- and cross-phase modulation terms, gives the cascaded higher-order self-action nonlinear coefficients. In this way we calculated HOKE contributions from third- to 11th-harmonic generation (nonlinearities governed by \( n_2 \) to \( n_{10} \)), and Table 1 shows the cascading terms up to \( n_{10} \) for various gases at \( \lambda_1 = 0.8 \mu m \). Note first that the cascading terms have similar strengths as the material nonlinearities at that level. Second, the \( |\Delta k_{13}| \) values for THG imply that the strong cascading limit can be achieved with propagation lengths fractions of a meter (the higher-order \( |\Delta k_{13}| \) are always larger than \( |\Delta k_{13}| \), so they are less critical).

Figure 1(a) shows the impact of cascading at \( \lambda_1 = 0.8 \mu m \), where we plot the variation of the nonlinear refractive index as a function of the pump intensity for the gases in Table 1. It is clear that the inclusion of the cascading effect shifts the inversion intensity \( I_{\text{inv}} \), where \( \Delta n = 0 \), to higher values. However, at longer pump wavelengths the flattened gas dispersion enhances cascading. As a result the inversion intensity becomes smaller and departures from the ideal Kerr effect are observable already at low intensities. In Fig. 1(b) the inversion intensities are plotted versus pump wavelength to summarize these trends. Without cascading, only a slight wavelength scaling [19] of the nonlinearities is seen.

Table 1. Material [1,2] and Cascading Nonlinearities at \( \lambda_1 = 0.8 \mu m \), \( p = 1 \) atm, and \( T = 293 \) K

<table>
<thead>
<tr>
<th>( \Delta k_{13} ) *</th>
<th>( n_2 )</th>
<th>( n_4 )</th>
<th>( n_{4,\text{casc}} )</th>
<th>( n_6 )</th>
<th>( n_{6,\text{casc}} )</th>
<th>( n_8 )</th>
<th>( n_{8,\text{casc}} )</th>
<th>( n_{10} )</th>
<th>( n_{10,\text{casc}} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ar</td>
<td>442</td>
<td>1.0E-23</td>
<td>-3.6E-42</td>
<td>-1.8E-42</td>
<td>40E-59</td>
<td>0.2E-59</td>
<td>-17.2E-76</td>
<td>-3.0E-76</td>
<td>0.9E-93</td>
</tr>
<tr>
<td>Air</td>
<td>482</td>
<td>1.2E-23</td>
<td>-15.0E-42</td>
<td>-2.3E-42</td>
<td>21E-59</td>
<td>0.8E-59</td>
<td>-0.8E-76</td>
<td>-1.8E-76</td>
<td>—</td>
</tr>
<tr>
<td>O₂</td>
<td>772</td>
<td>1.6E-23</td>
<td>-51.6E-42</td>
<td>-2.6E-42</td>
<td>48E-59</td>
<td>2.2E-59</td>
<td>-21.0E-76</td>
<td>-3.3E-76</td>
<td>—</td>
</tr>
<tr>
<td>N₂</td>
<td>501</td>
<td>1.1E-23</td>
<td>-8.5E-42</td>
<td>-1.9E-42</td>
<td>14E-59</td>
<td>0.3E-59</td>
<td>-4.4E-76</td>
<td>-1.0E-76</td>
<td>—</td>
</tr>
</tbody>
</table>

\*Calculated using Sellmeier equations [17,18]. All units are SI (i.e., \( [\Delta k_{13}] = m^{-1} \), \( [n_2] = m^2/W \) etc.). Note: the cascading calculations gave orders beyond \( n_{10,\text{casc}} \), which are included in Fig. 1. The propagated error in the leading cascading term \( n_{4,\text{casc}} \) is 12%-35%.
cascading gives an effective Kerr-like nonlinearity \( (\Delta n \text{ independent on } z) \), and when \( \Delta k_{1m}/[\Gamma_{\omega n}^{(m)}E_{1m}^{m-1}] > 1 \), the approximation \( \Delta n = \sum \gamma_j \beta_j / \Delta k_{1m} \) holds. For \( \Delta k_{1m}L \approx 2\pi \), this breaks down as the cascaded harmonic becomes amplitude modulated by \( z \sin(c/\Delta k_{1m}z/2) \). The cascading terms must then be corrected by \( 1 - \sin(c/\Delta k_{1m}L) \) (resolving the unphysical divergences at \( \Delta k_{1m} = 0 \)). Nonetheless, even for \( \Delta k_{1m}L \ll 2\pi \), strong cascading is possible if \( \Delta k_{1m}/[\Gamma_{\omega n}^{(m)}E_{1m}^{m-1}] \sim 1 \) (strong FW depletion), but the cascading will then grow step-wise in \( z \) [12], so the cascading nonlinear coefficients cannot be parameterized.

The increased cascading contributions predicted at longer wavelengths in Fig. 1(b) rely on reduced phase-mismatch values. Thus, fulfilling \( \Delta k_{13}L > 2\pi \) can become problematic except for very long cells. Filamentation experiments will therefore make an ideal test bed for cascading as they inherently use very long interaction lengths, where the \( \sin(c/\Delta k_{13}L) \)-oscillations vanish. Alternatively, a solution could be to increase the pressure: the inversion intensity remains the same, but the phase mismatch values increase and overall we find that the cascading contributions to \( \Delta n \) are enhanced.

The experiment behind the HOKE coefficients [1,2] used a pump-probe interaction length \( L \approx 1 \text{mm} \) [20]. Since the leading phase-mismatch values (see Table 1) are around \( 1/\text{mm} \) or less, the cascading contribution is reduced because the \( 1 - \sin(c/\Delta k_{13}L) \) term must be applied. Specifically, we found that cascading corrections to all HOKE terms for the various gases are within the error bars of the measurements. Thus, the HOKE values in [1,2] correspond to the intrinsic material nonlinearities.

Even if cascading relies on a chain of upconversion and downconversion steps, it can be considered instantaneous, much like an electronic Kerr nonlinearity: with full dispersion, the cascading term in frequency domain becomes \( E_{\text{3-casc}}(\omega, \Omega) \propto -\Delta k_{13}/[\mathcal{F}[E] \mathcal{R}_{\text{casc}}(\Omega)] \), where \( \mathcal{F} \) denotes the Fourier transform and \( \mathcal{R}_{\text{casc}}(\Omega) = \Delta k_{13}/[k(\Omega + \omega_2) - k(\omega_1)\Omega - 3k(\omega_1)] \) is the cascading “response” function [16]. Neglecting higher-order dispersion the criterion \( |\omega_1^2| \ll 2k_2(\omega_2)^2 \Delta k_{13} \) approximately determines when the cascading response is nonresonant and thus broadband [16]. We checked that this inequality is always fulfilled, and moreover since octave-spanning bandwidths are supported the corresponding temporal response of the cascading is extremely fast [13].

In conclusion, cascading in Kerr media from phase-mismatched harmonic generation may significantly contribute to the observed nonlinear index change in gases. The calculations rely on the strong phase-mismatch limit, and the lowest-order contribution from \( n_2 \), where \( \chi^{(3)}:\chi^{(3)} \) cascading from a phase-mismatched TH gives an effective \( n_3 \) term, is always negative and significant compared to the material \( n_4 \). A unidirectional pulse propagation equation can model such harmonic dynamics with a proper nonlinear expansion [21]. For higher-order harmonics the conclusions were similar; such terms rely on including higher-order material nonlinearities. At 800 nm the cascading from \( n_2 \) and higher-order coefficient \( n_4 \) [1,2] delayed the saturation point to higher intensities than what the material nonlinearities dictate. This might explain why indications of saturation for near-IR filaments have been elusive. Instead for longer (mid-IR) wavelengths, cascading pushed the saturation point to lower intensities, which will enhance saturation and possibly affect filamentation.

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References and Notes

20. B. Lavorel, CNRS-Université de Bourgogne (personal communication, 2012).