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Higher-order Kerr effect and harmonic cascading in gases

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The higher-order Kerr effect (HOKE) has been recently advocated to explain measurements of the saturation of the nonlinear refractive index in gases. Here we show that cascaded third-harmonic generation results in an effective fifth order nonlinearity that is negative and significant. Higher-order harmonic cascading will also occur from the HOKE, and the cascading contributions may significantly modify the observed nonlinear index change. At lower wavelengths cascading increases the HOKE saturation intensity, while for longer wavelengths cascading will decrease the HOKE saturation intensity. © 2012 Optical Society of America

OCIS codes: (190.3270);(190.2620);(320.7110);(190.5530);(190.4400);

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: $\Delta n = n_2 I + n_4 I^2 + ... + n_{2m} I^m$. 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] has sparked a vivid debate on the actual role of these higher-order terms in nonlinear beam-propagation dynamics. Indeed, it was predicted that the effective saturation of the HOKE nonlinearity could lead to filamentation of femtosecond pulses in gases without plasma generation [2]. The mechanism advocated in this case to arrest collapse of the wave packet was the saturation of the nonlinearity rather than the formation of plasma, as commonly accepted [3]. Experiments have either confirmed [4,5] or ruled out [6–8] a preponderant role of HOKE in filamentation dynamics.

In the strong cascading limit cascaded harmonic generation is known to give effective higher-order nonlinearities that compete with the inherent material ones. In particular, 2nd order $\chi^{(2)} : \chi^{(2)}$ cascading generates an effective $\chi^{(3)}$ [9,10], which can be used to generate a strong, tunable and ultrafast negative Kerr-like nonlinearity despite the material Kerr nonlinearity being positive [11], or 3rd order $\chi^{(3)} : \chi^{(3)}$ cascading generating an effective $\chi^{(5)}$ [12]. Here we study cascading contributions to the nonlinear HOKE coefficients in gases. This is unlike previous investigations that focussed on harmonic yield from cascading [5–7]. Cascading relies on the mth harmonic being phase mismatched $\Delta k_{1m} \neq 0$, so after two coherence lengths $2\pi/|\Delta k_{1m}|$ back-conversion to the pump is complete. As $\Delta k_{1m} \neq 0$ the back-converted pump photons are phase-shifted compared to the unconverted pump photons, and as this process is repeated the pump feels a nonlinear phase shift characterized by an *effective* higher-order nonlinear index $\propto -[\chi^{(m)}]^2/\Delta k_{1m}$. We show that in gases cascading can affect the nonlinear index change and in particular the saturation intensity. Specifically, to lowest order cascading comes from

a phase-mismatched third-harmonic generation (THG) process, which we show generates an n_4 term that turns out to be negative and of significant strength. Similarly, cascading of HOKE terms gives contributions to subsequent orders of nonlinearity (n_6, n_8, . . .) with values comparable with recent experimental [1] and theoretical HOKE coefficients [13]. This implies that in the strong cascading limit, which we discuss 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 $\omega_1 + \omega_1 + \omega_1 \rightarrow \omega_3$ in the plane-wave limit in an isotropic lossless medium are

$$\left(i\partial_z - \tfrac{1}{2}k_2(\omega_1)\partial_{\tau\tau}\right)E_1$$
$$+\Gamma^{(3)}_{\omega_1}\left[E_1|E_1|^2 + 2E_1|E_3|^2 + (E_1^*)^2 E_3 e^{i\Delta k_{13}z}\right] = 0 \quad (1)$$
$$\left(i\partial_z - id_{13}\partial_\tau - \tfrac{1}{2}k_2(\omega_3)\partial_{\tau\tau}\right)E_3$$
$$+\Gamma^{(3)}_{\omega_3}\left[E_3|E_3|^2 + 2E_3|E_1|^2 + \tfrac{1}{3}E_1^3 e^{-i\Delta k_{13}z}\right] = 0 \quad (2)$$

The harmonic phase mismatch is $\Delta k_{1m} = k(\omega_m) - mk(\omega_1)$, $d_{13} = k_1(\omega_1) - k_1(\omega_3)$ the group-velocity mismatch (GVM), $k_m(\omega_j) \equiv d^m k/d\omega^m|_{\omega=\omega_j}$ the dispersion coefficients, $k(\omega) = n(\omega)\omega/c$ the wave number, and $n(\omega)$ the linear refractive index. For simplicity higher-order dispersion is neglected. Finally $\Gamma^{(3)}_{\omega_j} = 3\omega_j \chi^{(3)}/8cn(\omega_j)$, making $n_2 = \Gamma^{(3)}_{\omega_1} 2/[\varepsilon_0 \omega_1 n(\omega_1)] = 3\chi^{(3)}/[4\varepsilon_0 c n^2(\omega_1)]$ the Kerr nonlinear index of the fundamental wave (FW).

Following [16], in the strong cascading limit, $\Delta k_{13} L \gg 1$ (L is the interaction length) and no pump depletion are assumed, so we can neglect third harmonic (TH) self- and cross-phase modulation in Eq. (2) and consider only the $E_1^3/3$ term. The ansatz $E_{3,\text{casc}}(z,\tau) = e^{-i\Delta k_{13}z}\psi_3(\tau)$ makes Eq. (2) a temporal ODE. Neglecting dispersion for the moment gives $E_{3,\text{casc}}(z,\tau) = -e^{-i\Delta k_{13}z}E_1^3(z,\tau)\Gamma^{(3)}_{\omega_3}/(3\Delta k_{13})$: the TH is effectively slaved to the FW. When inserting this in Eq. (1) yields the nonlinear term $\Gamma^{(3)}_{\omega_1}E_1|E_1|^2\left[1 - |E_1|^2\Gamma^{(3)}_{\omega_1}/\Delta k_{13} + 2|E_1|^4(\Gamma^{(3)}_{\omega_1}/\Delta k_{13})^2\right]$,

Table 1. Material [1] and cascading nonlinearities at $\lambda_1 = 0.8$ μm, $p = 1$ atm and $T = 293$ K. Δk_{13} was calculated using Sellmeier equations from [14], except for O_2 [15]. All units are SI (i.e. $[\Delta k_{13}] = $ m^{-1}, $[n_2] = $ m^2/W etc.).

	Δk_{13}	n_2	n_4	$n_{4,\mathrm{casc}}$	n_6	$n_{6,\mathrm{casc}}$	n_8	$n_{8,\mathrm{casc}}$	n_{10}	$n_{10,\mathrm{casc}}$
Ar	442	1.0E-23	-3.6E-42	-1.8E-42	40E-59	0.2E-59	-17.2E-76	-3.0E-76	0.9E-93	2.0E-93
Air	482	1.2E-23	-15.0E-42	-2.3E-42	21E-59	0.8E-59	-8.0E-76	-1.8E-76	-	1.0E-93
O_2	772	1.6E-23	-51.6E-42	-2.6E-42	48E-59	2.2E-59	-21.0E-76	-3.3E-76	-	2.2E-93
N_2	501	1.1E-23	-5.3E-42	-1.9E-42	14E-59	0.3E-59	-4.4E-76	-1.0E-76	-	0.5E-93

where we used $\Gamma_{\omega_3}^{(3)}/3 \simeq \Gamma_{\omega_1}^{(3)}$ and $\frac{n(\omega_1)}{n(\omega_3)} \simeq 1$. Converting to intensity $E_1 \to \sqrt{2/\varepsilon_0 n(\omega_1)c}A_1$ we get $(n_2 A_1|A_1|^2 + n_{4,\mathrm{casc}}A_1|A_1|^4 + n_{6,\mathrm{casc}}^{(3)}A_1|A_1|^6)\omega_1/c$. Thus, the cascaded TH gives effective 5$^{\mathrm{th}}$ and 7$^{\mathrm{th}}$ order *self-action* nonlinearities, where the 5$^{\mathrm{th}}$ order term is

$$n_{4,\mathrm{casc}} \equiv -\frac{n_2^2}{\Delta k_{13}}\frac{\omega_1}{c} \qquad (3)$$

This adds to the material quintic nonlinearity n_4 so $n_{4,\mathrm{tot}} = n_4 + n_{4,\mathrm{casc}}$. In other words, when THG is included in the description, $\chi^{(3)} : \chi^{(3)}$ cascading leads to a 5$^{\mathrm{th}}$ order HOKE contribution, and as Eq. (3) shows this contribution is negative when $\Delta k_{13} > 0$ (which is always true in bulk materials pumped far from resonances).

As shown above, the cascaded TH gave also a higher-order term $n_{6,\mathrm{casc}}^{(3)}$, and another term, $n_{6,\mathrm{casc}}^{(5)}$, appears from the cascaded TH in the next order nonlinear FW term (governed by n_4), i.e. $\chi^{(3)} : \chi^{(5)}$ cascading. Thus

$$n_{6,\mathrm{casc}}^{(3)} \equiv \frac{n_2^3}{n(\omega_3)\Delta k_{13}^2}\frac{\omega_1^2}{c^2}, \quad n_{6,\mathrm{casc}}^{(5)} \equiv -\frac{5n_2 n_4}{2\Delta k_{13}}\frac{\omega_1}{c} \qquad (4)$$

These add to the material septic nonlinearity n_6 so $n_{6,\mathrm{tot}} = n_6 + n_{6,\mathrm{casc}}^{(3)} + n_{6,\mathrm{casc}}^{(5)}$. The expressions for the next orders are lengthy, but briefly in the strong cascading limit the harmonic is reduced to $E_{m,\mathrm{casc}} \propto -(\Delta k_{1m})^{-1}E_1^m e^{-i\Delta k_{1m}z}$. Inserting these cascading terms in the FW equation, which contains all harmonic self- and cross-phase modulation terms, gives the cascaded higher-order self-action nonlinear coefficients.

In this way we calculated HOKE contributions from n_2 to n_{10}, and Table 1 shows the cascading terms up to $n_{10,\mathrm{casc}}$ for various gases at $\lambda_1 = 0.8$ μm. We observe that the cascading terms have similar strengths as the material nonlinearities at that level. Secondly, the Δk_{13} values for THG imply that the strong cascading limit can be achieved with propagation lengths fractions of a meter (the higher-order mismatches Δk_{1m} are always larger than Δk_{13}, so they are less critical).

Figure 1(a) shows the impact of cascading at $\lambda_1 = 0.8$ μm, where we plot the variation of the nonlinear refractive index as a function of the light intensity for the gases in Table 1. It is clear that the inclusion of the cascading effect shifts the inversion intensity, 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 is observable already at low intensities. In Fig. 1(b) the inversion intensities are
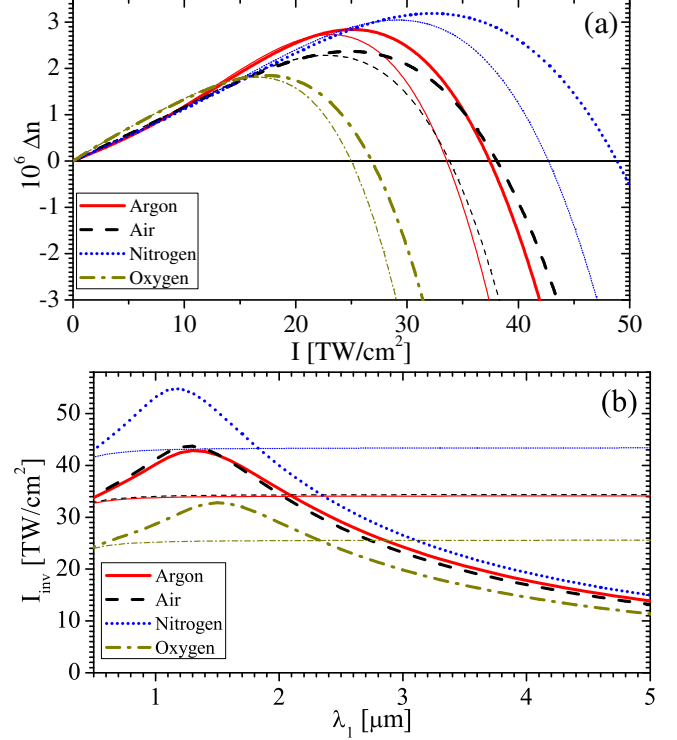

Fig. 1. (Color online) (a) The nonlinear index change Δn vs. pump intensity for $\lambda_1 = 0.8$ μm, based on data from Table 1. Thin lines show the HOKE nonlinear saturation [1], while the bold lines include cascading corrections. Note that the calculations naturally give orders beyond $n_{10,\mathrm{casc}}$ shown in Table 1, and these are included in the curves. (b) The inversion intensity of Δn vs. pump wavelength. The thin lines are the data from [1], while the bold lines include cascading corrections. The nonlinear coefficients are corrected for other wavelengths than $\lambda_1 = 0.8$ μm using the generalized Miller's formula [17].

plotted vs. pump wavelength to summarize these trends. Without cascading, only the slight wavelength scaling of the nonlinearities is seen. When including cascading the inversion intensity peaks in the near-IR, and is pushed to become lower than without cascading at the beginning of the mid-IR. The calculations are based on the HOKE material nonlinearities from [1], but we found similar results using the coefficients from [13] (seemingly, the HOKE coefficients of [1] and [13] 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,

O_2 and N_2) 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 $l_{\rm coh} = \pi/|\Delta k_{1m}|$ is much smaller than the interaction length L, expressed as $\Delta k_{1m}L \gg 1$, and also much smaller that the characteristic nonlinear length, expressed as $\Delta k_{1m}/[\Gamma_{\omega_m}^{(m)} E_{1,\rm in}^{m-1}] \gg 1$. To lowest order no pump depletion is found: $E_{3,\rm casc}$ above Eq. (3) is just phase modulated in z. Only $\Delta k_{1m}L \gg 1$ is needed to give a Kerr-like nonlinearity (Δn independent on z). For $\Delta k_{1m}L \simeq 1$ this no longer holds because to this next order the cascaded harmonic becomes amplitude modulated by $z\,{\rm sinc}(\Delta k_{1m}z/2)$. This gives a modulation factor $1 - {\rm sinc}(\Delta k_{1m}L)$ on the cascading coefficient, resolving the divergences at $\Delta k_{1m} = 0$ [see e.g. Eq. (3)] and giving as expected zero cascading for $\Delta k_{1m} = 0$. Despite this, even small $\Delta k_{1m}L$ values can give large cascading nonlinearities if the weak pump depletion requirement is relaxed, but since the cascading then grows in a step-like manner [10] we cannot assign the cascading nonlinear coefficients as parameters.

The increased cascading contributions predicted at longer wavelengths in Fig. 1(b) rely on reduced phase-mismatch values. Thus, fulfilling $\Delta k_{13}L \gg 1$ 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 sinc-oscillations stabilize. 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 Δn are enhanced.

The experiment determining the HOKE coefficients [1] used $L \simeq 1$ mm [18] (the pump-probe interaction length). Since the leading phase-mismatch values (see Table 1) are around 1/mm or less, the cascading contribution is reduced because the $1 - {\rm sinc}(\Delta k_{13}L)$ 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 measured values in [1] relate to the intrinsic material nonlinearity.

Even though cascading relies on a chain of up- and down-conversion steps, it can be considered instantaneous, much like an electronic Kerr nonlinearity: when dispersion is included, the cascading term in frequency domain reads $\tilde{E}_{3,\rm casc}(z,\Omega) \propto -\Delta k_{13}^{-1}\mathcal{F}[E_1^3(z,\tau)]R_{\rm casc}(\Omega)$ where $\mathcal{F}[\cdot]$ denotes the Fourier transform and $R_{\rm casc}(\Omega) = \Delta k_{13}/[k(\Omega + \omega_3) - k_1(\omega_1)\Omega - 3k(\omega_1)]$ is the cascading "response" function [16]. Neglecting higher-order dispersion the criterion $d_{13}^2 < 2k_2(\omega_3)\Delta k_{13}$ 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 [11].

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 third-harmonic contributes to the n_4 term, is always negative and significant compared to the material nonlinearities. For higher-order contributions similar conclusions were drawn; such terms rely on including higher-order material nonlinearities. At 800 nm the cascading from n_2 and higher-order coefficient (n_4 up to n_{10}) [1] 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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