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From higher-order Kerr nonlinearities to quantitative modeling of 3rd and 5th harmonic generation in argon

P. Béjot,1 E. Hertz,1 B. Lavoie1, J. Kasprian2, J.-P. Wolf2, and O. Faucher1,*

1Laboratoire Interdisciplinaire CARNOT de Bourgogne (ICB), UMR 5209 CNRS-Université de Bourgogne
BP 47870, 21078 Dijon Cedex, France
2Université de Genève, GAP-Biophotonics, 20 rue de l’Ecole de Médecine, 1211 Geneva 4, Switzerland

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The recent measurement of negative higher-order Kerr effect (HOKE) terms in gases has given rise to a controversial debate, fed by its impact on short laser pulse propagation. By comparing the experimentally measured yield of the third and fifth harmonics, with both an analytical and a full comprehensive numerical propagation model, we confirm the absolute and relative values of the reported HOKE indices. © 2011 Optical Society of America

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strications in the media, and a comprehensive model including linear and non-
linear propagation effects such as dispersion, self-phase modulation, ionization, and Kerr effect, is presented.

For a focused laser beam propagating linearly, the harmonic power of the qth harmonic in the perturbative regime is given by

\[ P_q = A_q N^2 |J_q(b\Delta k)|^2, \]

where \( N \) is the atomic density of the medium and

\[ A_q = \frac{q \omega_1^2}{4n_q^2(n_j^4)^q(\epsilon_0\pi)^{q-1}c^2w_0^2}\left(\frac{\chi^{(q)}}{2}\right)^2 P_1^q, \]

with \( P_1, \omega_1, \) and \( w_0 \) the power, the angular frequency, and the beam waist of the incident beam, respectively [13, 14]. \( \chi^{(q)} \) is the qth-order microscopic nonlinear susceptibility \((q = 3, 5)\) given in SI units, \( n_j^q \) are the linear refractive indices at the fundamental \((j = 1)\) and harmonic frequencies \((j = 3, 5)\), \( \epsilon_0 \) is the permittivity of vacuum, and \( c \) is the speed of light. \( J_q \) is a dimensionless function that accounts for the phase matching

\[ J_q = \int_{-2f/b}^{2(L-f)/b} d\xi \frac{\exp(-ib\Delta k\xi/2)}{(1 + i\xi)^q}, \]

with \( \Delta k = k_j - qk_1 = \frac{2\pi n_j^q}{\lambda_0} (n_j^q - n_j^1) \) the phase mismatch, with \( n_j^q - n_j^1 \) proportional to the pressure, and \( k_j (j = 1, q) \) the wave vectors, \( b \) the confocal parameter, \( L \) the length of the static cell, and \( f \) the position of the focus with respect to the entrance of the static cell [15].

According to Eqs. (1) and (2), the ratio of the FH to the TH power is

\[ \frac{P_3}{P_5} \approx \frac{5}{3\epsilon_0^2\pi^2c^2w_0^2} \left(\frac{\chi^{(5)}}{\chi^{(3)}}\right)^2 \left(\frac{N_3|J_3|}{N_5|J_3|}\right)^2 P_1^2, \]

where \( n_j^q \) have been approximated to unity in Eq. 2. \( N_3 \) and \( N_5 \) refer to the different atomic densities at the pressures maximizing the harmonic conversion for the 3rd and 5th orders, respectively. This equation provides a direct relationship between the power ratio of the harmonics and the ratio of the corresponding non-linear susceptibilities. The latter are related to the nonlinear refractive indices through the relation [7]

\[ n_{2j} \approx \frac{(2j + 1)!!}{2^{j+1}j!(j + 1)!!} \left(\frac{1}{(n_j^1)^2\epsilon_0c}\right)^2 \chi_{\text{Kerr}}^{(2j+1)}. \]
From the input energy $P$ both harmonics [12, 16]. This leads to a power ratio
so that

$$\frac{P_5}{P_3} \approx \frac{3}{5\pi^2 w_0^4} \left( \frac{n_4}{n_2} \right)^2 \left( \frac{N_5}{N_3} \right)^2 P_1^2,$$  \hspace{1cm} (6)

In the experiment by Kosma et al., $b=7.8$ cm, $n_0=100 \mu m$, $L=1.8$ cm, $f=L/2$, and the wavelength $\lambda_1=810$ nm [12]. The fundamental power, calculated from the input energy $E_1=710 \mu J$ and the pulse duration $t_1=12$ fs, is $P_1=59$ GW. They observed that the pressure maximizing the TH power ranged between 160 mbar [12] and 250 mbar [16], for similar experimental conditions. One single maximum, around 50 mbar, is observed for the FH. The maximum energies of the argon pressure relying on Eq. (3) (Fig. 1). This value confirms, especially considering the simplicity of the analytical model used.

Further comparison with the experiment was performed by computing the value of $N^2 |J_q|^2$ as a function of the argon pressure relying on Eq. (3) (Fig. 1). This function should reflect the pressure dependence of the harmonic powers. The analytical model predicts a maximum at about 300 mbar for TH, in line with the experimental results. It yields three maxima between 0 and 400 mbar for FH, the first of them close to the observed optimum pressure for the FH. This oscillatory structure, which is due to the periodic phase matching, was not observed in the experiments [18] probably due to nonlinear propagation effects which are not considered in the analytical model.

To overcome these limitations and take into account the perturbations of the fundamental pulse during its propagation through the gas sample, as well as the effect of the HOKE indices on the phase matching, we have solved the unidirectional pulse propagation equation for the experimental conditions of Kosma et al. More precisely, assuming a cylindrical symmetry around the propagation axis $z$, the angularly resolved spectrum $\tilde{E}(k_\perp, \omega)$ of the real electric field $E(r, t)$ follows the equation [19]

$$\partial_z \tilde{E} = i k_z \tilde{E} + \frac{1}{2k_\perp} \left( \frac{i\omega^2}{c^2} \tilde{P}_\text{NL} - \frac{\omega}{\epsilon_0 c} \tilde{J} \right),$$  \hspace{1cm} (7)

where $k_z = \sqrt{k^2(\omega) - k_\perp^2}$, $\tilde{P}_\text{NL}$ (resp., $\tilde{J}$) is the angularly resolved nonlinear polarization (resp., free charge induced current) spectrum, and $k(\omega) = \frac{n(\omega)}{19} c$.

The nonlinear polarization $P_\text{NL}$ is evaluated in the time domain as $P_\text{NL} = \chi^{(3)} E^3 + \chi^{(5)} E^5 + \chi^{(7)} E^7 + \chi^{(9)} E^9 + \chi^{(11)} E^{11}$. Since the nonlinear polarization is defined from the real electric field, Eq. 7 captures without any modifications all frequency-mixing processes induced by the total field. For numerical stability concerns, we considered only the part responsible for the refractive index change around $\omega_0$, neglecting harmonics generation induced by the terms proportional to $E^7$, $E^9$, and $E^{11}$. The current induced by the free charges is calculated in the frequency domain as $\tilde{J} = \vec{e} \cdot \vec{v}_i - \vec{e} \cdot \vec{v}_e - 55c$, where $c$ (resp., $m_e$) is the electron charge (resp., mass), $\nu_e$ is the effective collisional frequency, and $\rho$ is the electron density which is evaluated as

$$\partial_t \rho = W(I) (\rho_{at} - \rho) + \frac{\sigma}{U_i} I - \beta \rho^2,$$  \hspace{1cm} (8)

where $W(I)$ is the ionization probability evaluated with the Keldysh-PPT (Perelomov, Popov, Terent’ev) model [3], $\rho_{at}$ is the atomic number density, $\sigma$ is the inverse Bremsstrahlung cross-section, $\beta$ is the recombination constant (negligible on the time scale investigated in the present work), and $I$ is proportional to the time-averaged $\langle E^2 \rangle$.

Figure 2 displays the harmonics intensity as a function of argon pressure for an input pulse and a detection geometry matching the experimental parameters: 12 fs pulse duration (FWHM), 700 $\mu J$ input energy, and a beam radius of 4 mm before focusing. In order to mimic the experiment, the pulse first propagates in vacuum up to the position of the cell (99.1 cm after the $f=1$ m lens). After this focusing step, the pulse propagates over 1.8 cm in the argon cell. The optimal pressure for the FH is 50 mbar, in full agreement with the experiment [12]. The reduction of the second and third maxima of the FH, as compared to Fig. 1., results from the phase mismatch introduced by the HOKE at large pressure. The FH yield is maximal at 260 mbar, similar to the value reported in [16]. In full agreement with the experiment by Kosma al. [12], the ratio at 50 mbar is about 0.1 and becomes even larger at reduced pressures. Furthermore, the total FH and TH energies at their respective optimum pres-
Fig. 2. (Color online) Numerical calculation of the pressure dependence of the 3rd (dotted blue line) and 5th (open red circles) harmonics in argon integrated over the full radial distribution. To be compared with the Fig. 3 of [12]. The spectrum calculated at 50 mbar is shown in the inset.

Kerr effects quantitatively reproduces the ratio of the harmonic yields observed in the experiment, as well as the pressure dependence of both the 3rd and 5th harmonics. It even reproduces the absolute harmonics intensity within a fairly good accuracy.

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