From higher-order Kerr nonlinearities to quantitative modeling of third and fifth harmonic generation in argon

P. Béjot,[1] E. Hertz,[1] B. Lavorel,[1] J. Kasparian,[2] J.-P. Wolf,[2] and O. Faucher[1,*]

[1]*Laboratoire Interdisciplinaire CARNOT de Bourgogne (ICB), UMR 5209 CNRS-Université de Bourgogne BP 47870, 21078 Dijon Cedex, France*

[2]*Université de Genève, GAP-Biophotonics, 20 rue de l'Ecole de Médecine, 1211 Geneva 4, Switzerland*

**Corresponding author: Olivier.Faucher@u-bourgogne.fr*

Received December 9, 2010; revised January 21, 2011; accepted January 23, 2011; posted February 9, 2011 (Doc. ID 139397); published March 8, 2011

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.

OCIS codes: 320.2250, 190.2620, 320.7110.

In a recent experiment, we have shown that the electronic optical Kerr effect in Ar, N_2, O_2, and air exhibits a highly nonlinear behavior versus the applied intensity [1,2], resulting in a saturation of the nonlinear refractive index observed at moderate intensity, followed by a sign inversion at higher laser intensity. This observation has a substantial impact on the propagation of ultrashort and ultraintense laser pulses, especially in the context of laser filamentation [3–6], where the higher-order Kerr effect (HOKE), rather than the defocusing contribution of the free electrons, can play a key role in the self-guiding process [7], especially at long wavelengths [8] and for short pulses [9]. However, this issue is still controversial [10–12]. Therefore, an independent confirmation of our measurement of the HOKE is still needed. Recently, Kolesik *et al.* [10] have proposed such a test, based on the comparison of the yields of the third harmonic (TH) and the fifth harmonic (FH) radiations generated by the nonlinear frequency upconversion of a short and intense laser pulse in air. Based on numerical simulations, they suggested that, considering the HOKE indices, "the relative strength of the FH to the TH should reach values of the order of 10^{-1}" while, if omitting them, "this ratio should be about 4–5 orders smaller" [10].

So far, no measurement of the yield of the FH versus the TH have been achieved in air. However, Kosma *et al.* [13] measured the yields of TH and FH produced by a short and intense laser pulse in argon. The present Letter aims at confronting the results of this experiment with predictions based on the HOKE in argon [1,2].

In the first part, we confirm the ratio of the recently measured nonlinear indices [1,2] based on the analytical description of the harmonic generation. In the second part, a comprehensive model, including linear and nonlinear 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

$$P_q = A_q N^2 |J_q(b\Delta k)|^2, \tag{1}$$

where N is the atomic density of the medium and

$$A_q = \frac{q\omega_1^2}{4n_q^\ell (n_1^\ell)^q (\epsilon_0\pi)^{q-1} c^q w_0^{2q-2}} (\chi^{(q)})^2 P_1^q, \tag{2}$$

with P_1, ω_1, and w_0 the power, the angular frequency, and the beam waist of the incident beam, respectively [14,15]. $\chi^{(q)}$ is the qth-order microscopic nonlinear susceptibility ($q = 3$, 5) given in SI units, n_j^ℓ are the linear refractive indices at the fundamental ($j = 1$) and harmonic frequencies ($j = 3$, 5), ϵ_0 is the permittivity of the vacuum, and c is the speed of light. J_q is a dimensionless function that accounts for the phase matching

$$J_q = \int_{-2f/b}^{2(L-f)/b} \mathrm{d}\xi \frac{\exp(-ib\Delta k\xi/2)}{(1+i\xi)^{q-1}}, \tag{3}$$

with $\Delta k = k_q - qk_1 = \frac{2\pi q}{\lambda_1}(n_q^\ell - n_1^\ell)$ the phase mismatch (with $n_q^\ell - n_1^\ell$ 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 [16]. According to Eqs. (1) and (2), the ratio of the FH to the TH power is

$$\frac{P_5}{P_3} \approx \frac{5}{3\epsilon_0^2\pi^2c^2w_0^4}\left(\frac{\chi^{(5)}}{\chi^{(3)}}\right)^2\left(\frac{N_5|J_5|}{N_3|J_3|}\right)^2 P_1^2, \tag{4}$$

where n_j^ℓ 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 third and fifth orders, respectively. This equation provides a direct relationship between the power ratio of the harmonics and the ratio of the corresponding nonlinear susceptibilities. The latter are related to the nonlinear refractive indices through the relation [8]

$$n_{2j} = \frac{(2j+1)!}{2^{j+1}j!(j+1)!}\frac{1}{((n_1^\ell)^2\epsilon_0 c)^j}\chi_{\mathrm{Kerr}}^{(2j+1)}, \tag{5}$$

so that

$$\frac{P_5}{P_3} \approx \frac{3}{5\pi^2 w_0^4}\left(\frac{n_4}{n_2}\right)^2\left(\frac{N_5|J_5|}{N_3|J_3|}\right)^2 P_1^2. \tag{6}$$

In the experiment by Kosma *et al.*, $b = 7.8\,\mathrm{cm}$, $w_0 = 100\,\mu\mathrm{m}$, $L = 1.8\,\mathrm{cm}$, $f = L/2$, and the wavelength $\lambda_1 = 810\,\mathrm{nm}$ [13]. The fundamental power, calculated from the input energy $E_1 = 710\,\mu\mathrm{J}$ and the pulse duration $\tau_1 = 12\,\mathrm{fs}$, is $P_1 = 59\,\mathrm{GW}$. They observed that the pressure maximizing the TH power ranged between 160 [13] and 250 mbar [17] for similar experimental conditions. One single maximum, around 50 mbar, is observed for the FH. The maximum energies of the TH and FH measured at the respective optimal pressures reported in [13] are 140 and 4 nJ, respectively, while the pulse duration was estimated to be 11 fs for both harmonics [13,17]. This leads to the power ratio $P_5/P_3 = 0.028$. According to Eq. (6), where J_q of Eq. (3) has been calculated using $n_1^\ell = 1.00028$, $n_3^\ell = 1.00030$, and $n_5^\ell = 1.00035$ for the values of the refractive index of argon at 1 bar at 810, 270, and 162 nm, respectively [18], the corresponding ratio of the HOKE indices is $|n_4/n_2| = 6.8 \times 10^{-19}\,\mathrm{m}^2/\mathrm{W}$. This value confirms, within a factor of 2 compatible with the experimental error, the ratio of the experimental HOKE indices $n_2 = 10^{-23}\,\mathrm{m}^2/\mathrm{W}^1$ and $n_4 = -3.6 \times 10^{-42}\,\mathrm{m}^4/\mathrm{W}^2$ [1,2], resulting in $|n_4/n_2| = 3.6 \times 10^{-19}\,\mathrm{m}^2/\mathrm{W}$. The agreement is remarkable, 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 the TH, in line with the experimental results. It yields three maxima between 0 and 400 mbar for the 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 [19] 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 [20]

$$\partial_z \tilde{E} = ik_z \tilde{E} + \frac{1}{2k_z}\left(\frac{i\omega^2}{c^2}\tilde{\mathcal{P}}_{\mathrm{NL}} - \frac{\omega}{\epsilon_0 c^2}\tilde{\mathcal{J}}\right), \tag{7}$$

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

The nonlinear polarization P_{NL} is evaluated in the time domain as $P_{\mathrm{NL}} = \chi^{(3)}E^3 + \chi^{(5)}E^5 + \chi^{(7)}E^7 + \chi^{(9)}E^9 + \chi^{(11)}E^{11}$. Because 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 ω_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 $\tilde{\mathcal{J}} = \frac{e^2}{m_e}\frac{\nu_e + i\omega}{\nu_e^2 + \omega^2}\tilde{\rho\varepsilon}$, where e and m_e are the electron charge and the mass, respectively, ν_e is the effective collisional frequency, and ρ is the electron density, which is evaluated as

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

where $W(I)$ is the ionization probability evaluated with the Keldysh–PPT (Perelomov, Popov, Terent'ev) model [4], ρ_{at} is the atomic number density, σ is the inverse Bremsstrahlung cross section, β 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 μJ input energy, and a beam radius of 4 mm before focusing. In order to mimic the experiment, the pulse first propagates in a vacuum up to the position of the cell (99.1 cm after the $f = 1$ m lens). After

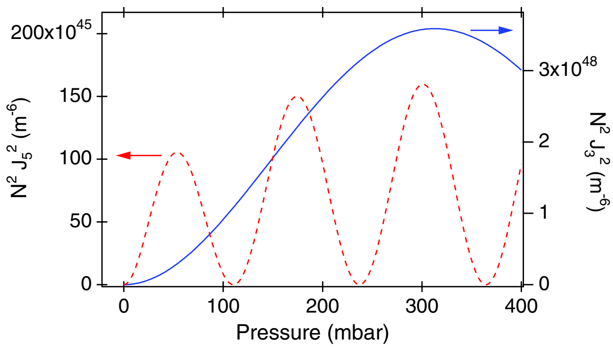

Fig. 1. (Color online) Analytical calculation of the pressure dependence of the third (solid blue line) and fifth (dashed red line) harmonics in argon.

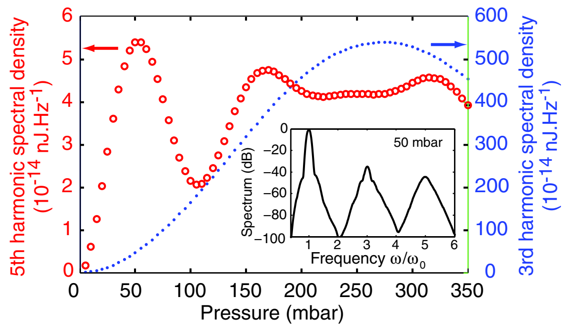

Fig. 2. (Color online) Numerical calculation of the pressure dependence of the third (dotted blue line) and fifth (open red circles) harmonics in argon integrated over the full radial distribution. To be compared with Fig. 3 of [13]. The spectrum calculated at 50 mbar is shown in the inset.

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 [13]. 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 high pressure. The TH yield is maximal at 260 mbar, similar to the value reported in [17]. In full agreement with the experiment by Kosma *et al.* [13], 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 pressures are 6 and 218 nJ, in good agreement with the experimental values of 4 and 140 nJ, where losses due to the setup lead to a slight underestimation of the output energies [13].

If the HOKE is not considered in the model, the ratio of the FH to the TH at a pressure of 50 mbar drops to 0.017, and the FH and TH energies are respectively 1.7 and 584 nJ: These values are inconsistent with the experimental results of Kosma *et al.* Furthermore, contrary to the experimental observations [19], the FH would exhibit strong maxima at 160 and 250 mbar. These discrepancies show that the HOKE is necessary to reproduce the experimental results [13,17], further validating their measured values [1,2]. Note that the ratio of 0.017 strongly depends on the propagation distance, so that it cannot be directly compared to that of 10,000 predicted by Kolesik *et al.* for the "classical" model over an unspecified propagation distance. For a propagation length of 220 μm, 80 times shorter than in our work but consistent with neglecting the phase matching, our calculation indeed predicts a ratio of 10,000.

In conclusion, as recently suggested in [10], we have compared the recent experimental measurements of the TH and FH yields in argon [13] with both analytical and numerical simulations. These results agree quantitatively with the measured high-order Kerr indices [1,2]. This conclusion is supported by the following findings. First, the harmonic yield reported in argon by Kosma *et al.* at the pressure that optimized the fifth harmonic leads to a ratio of about 0.1 between the fifth and the third harmonics. This ratio implies a ratio of the Kerr indices consistent with our measurement of the HOKE indices within their uncertainty range [1,2]. Second, the analytical model based on our HOKE indices reproduces the pressure maximizing the TH, as well as the first pressure maximum of the fifth harmonic yield. Third, a full numerical propagation model accounting for the dispersion and nonlinear effects such as ionization and higher-order Kerr effects quantitatively reproduces the ratio of the harmonic yields observed in the experiment, as well as the pressure dependence of both the third and fifth harmonics. It even reproduces the absolute harmonics intensity within a fairly good accuracy.

The authors are grateful to Werner Fuss and Kyriaki Kosma for fruitful discussions and critical reading of the manuscript. This work was supported by the Conseil Régional de Bourgogne, the Agence Nationale pour la Recherche (ANR) *COMOC*, the *FASTQUAST* Marie Curie Initial Training Networks (ITN) Program of the Seventh Framework Program and the Swiss National Science Foundation (SNSF), contract 200021-125315.

References

1. V. Loriot, E. Hertz, O. Faucher, and B. Lavorel, Opt. Express **17**, 13429 (2009).
2. V. Loriot, E. Hertz, O. Faucher, and B. Lavorel, Opt. Express **18**, 3011 (2010).
3. S. L. Chin, S. A. Hosseini, W. Liu, Q. Luo, F. Théberge, N. Aközbek, A. Becker, V. P. Kandidov, O. G. Kosareva, and H. Schröder, Can. J. Phys. **83**, 863 (2005).
4. L. Bergé, S. Skupin, R. Nuter, J. Kasparian, and J.-P. Wolf, Rep. Prog. Phys. **70**, 1633 (2007).
5. A. Couairon and A. Mysyrowicz, Phys. Rep. **441**, 47 (2007).
6. J. Kasparian and J.-P. Wolf, Opt. Express **16**, 466 (2008).
7. P. Béjot, J. Kasparian, S. Henin, V. Loriot, T. Vieillard, E. Hertz, O. Faucher, B. Lavorel, and J.-P. Wolf, Phys. Rev. Lett. **104**, 103903 (2010).
8. W. Ettoumi, P. Béjot, Y. Petit, V. Loriot, E. Hertz, O. Faucher, B. Lavorel, J. Kasparian, and J.-P. Wolf, Phys. Rev. A **82**, 033826 (2010).
9. V. Loriot, P. Béjot, W. Ettoumi, Y. Petit, J. Kasparian, S. Henin, E. Hertz, B. Lavorel, O. Faucher, and J.-P. Wolf, "On negative higher-order Kerr effect and filamentation," Laser Phys. (to be published).
10. M. Kolesik, E. M. Wright, and J. V. Moloney, Opt. Lett. **35**, 2550 (2010).
11. P. Polynkin, M. Kolesik, E. M. Wright, and J. V. Moloney, "Global fits of the minimal universal extra dimensions scenario," arxiv.org, http://arxiv.org/abs/1010.2023v2.
12. M. Kolesik, D. Mirell, J.-C. Diels, and J. V. Moloney, Opt. Lett. **35**, 3685 (2010).
13. K. Kosma, S. A. Trushin, W. E. Schmid, and W. Fuss, Opt. Lett. **33**, 723 (2008).
14. R. W. Boyd, *Nonlinear Optics*, 3rd ed. (Academic, 2008).
15. J. Reintjes and C. Y. She, Opt. Commun. **27**, 469 (1978).
16. G. C. Bjorklund, IEEE J. Quantum Electron. **11**, 287 (1975).
17. K. Kosma, S. A. Trushin, W. Fuss, and W. E. Schmid, Phys. Chem. Chem. Phys. **11**, 172 (2009).
18. A. Bideau, Y. Guern, R. Abjean, and A. Johannin-Gilles, J. Quant. Spectrosc. Radiat. Transfer **25**, 395 (1981).
19. W. Fuss, Max-Planck-Institut für Quantenoptik, D-85741 Garching, Germany (personal communication, 2010).
20. M. Kolesik and J. V. Moloney, Phys. Rev. E **70**, 036604 (2004).