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Ionization and absorption effects in high-order harmonic generation in gas-filled hollow fibers

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Abstract

The influence of absorption and ionization on high-order harmonic generation in a gas-filled hollow fiber is analyzed within the framework of slowly varying envelope approximation. Harmonic spectra and pressure dependencies are calculated for high-order harmonic generation in hollow fibers filled with different rare gases. Ionization of the gas filling the fiber gives rise to self-phase modulation of the pump pulse, changing the phase mismatch within the pump pulse and shifting the maxima in pressure dependencies of high-order harmonics.

1. INTRODUCTION

Nonlinear optical processes in hollow fibers are now extensively used for the generation of few-cycle pulses (Nisoli *et al.*, 1997) and frequency conversion through high-order-harmonic generation (Durfee *et al.*, 1997, 1999*a*, 1999*b*; Rundquist *et al.*, 1998; Tamaki *et al.*, 1998; Constant *et al.*, 1999). In particular, it was demonstrated that, due to the compensation of phase mismatch in hollow fibers, the efficiency of frequency conversion in high-order (up to the 45th order) harmonic generation can be increased by a factor of 100–1000 as compared with the efficiencies of frequency conversion attainable in experiments with gas jets (see, e.g., Ferray *et al.*, 1988; Li *et al.*, 1989; Balcou *et al.*, 1992; Macklin *et al.*, 1993). The conversion efficiency of $4 \cdot 10^{-5}$ has been recently achieved by Constant *et al.* (1999) for the 15th harmonic generated in a xenon-filled hollow fiber with the use of 40-fs 1.5-mJ 800-nm pulses.

To enhance the efficiency of nonlinear optical interactions, it is often desirable to increase the length of the fiber and to use laser pulses with higher intensities. Absorption and ionization effects may become significant under these conditions, especially in the case of high-order harmonic generation, as was shown recently both experimentally (Durfee *et al.*, 1999*b*) and theoretically (Naumov *et al.*, 2000).

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In this article, we apply the slowly varying envelope approximation to examine the influence of absorption and ionization effects on high-order harmonics in gas-filled fibers. We will calculate harmonic spectra and pressure dependencies of high-order harmonics generated in hollow fibers filled with different rare gases to demonstrate that the enhancement of harmonic-generation efficiency due to phase matching in hollow fibers may reach three orders of magnitude with an appropriate choice of the sort and the pressure of the gas and parameters of the fiber. We will also show that the ionization of the gas filling the fiber gives rise to a self-phase modulation of the pump pulse, thus changing the phase mismatch for the harmonic-generation process within the pump pulse, decreasing the overall efficiency of harmonic generation and making the harmonic-generation efficiency less sensitive to the gas pressure in the hollow fiber.

2. BASIC RELATIONS

Suppose that a fundamental (pump) wave and the wave of the qth harmonic propagating in a gas-filled hollow fiber along the z axis can be represented as

$$E_1 = A^n(\theta, z) f^n(\vec{\rho}) \exp[-i(\omega t - K^n z)] + c.c. \tag{1}$$

$$E_q = B_q(\theta, z, \vec{\rho}) \exp[-i(q\omega t - K_q z)] + c.c., \tag{2}$$

where ω is the frequency of fundamental radiation, q is the harmonic order, $f^n(\vec{\rho})$ is the transverse distribution of the

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light field, and K^n is the propagation constant of fundamental radiation corresponding to the EH_{1n} mode of the hollow fiber, K_q is the propagation constant of the qth harmonic, $A_n(\theta, z)$ is the slowly varying envelope of the fundamental pulse (by separating the dependencies of this envelope on the transverse and longitudinal coordinates, we assume that some definite waveguide mode was excited at the fundamental frequency), $B_q(\theta, z, \vec{\rho})$ is the slowly varying envelope of the harmonic pulse, and $\theta = t - z/v$, with v being the group velocity of the fundamental and harmonic pulses (i.e., we assume that the fiber length is chosen in such a way that the temporal walk-off of the fundamental and harmonic pulses due to the difference in their group velocities can be neglected; see the estimates in Zheltikov *et al.* (1999)). Expressions for the propagation constants of hollow-fiber waveguide modes and the transverse field distributions characteristic of these modes can be found in Marcatili and Schmeltzer (1964).

We will consider the case when the transverse distribution of the pump wave corresponds to the EH_{1n} waveguide mode. To include ionization effects, we assume that a small part of particles of the gas filling the fiber may be ionized during the pump pulse, giving rise to an electronic contribution to the refractive index of the gas and, consequently, to the phase modulation of the pump pulse. Since the number of atoms undergoing ionization is small, we neglect the decrease in the concentration of neutral atoms due to ionization and assume that this concentration remains independent of time. Then, in the range where the slowly varying envelope approximation is applicable, the equation for the envelope of the pump wave is written as

$$\frac{\partial A^n(\theta, z)}{\partial z} = \frac{2i\pi\omega^2}{K^n c^2} \chi_n^{(1)}(\theta) A^n(\theta, z). \tag{3}$$

Here, we introduced the effective linear susceptibility of plasma at the frequency of the pump pulse,

$$\chi_n^{(1)}(\theta) = \frac{-e^2}{m\omega^2} n_{\rm eff}^n(\theta), \tag{4}$$

where e and m are the electron charge and mass, respectively, and

$$n_{\rm eff}^n(\theta) = \frac{\displaystyle\int f^n(\vec{\rho}) n_e(\theta, \vec{\rho})\, d\vec{\rho}}{\displaystyle\int |f^n(\vec{\rho})|^2\, d\vec{\rho}} \tag{5}$$

is the effective electron concentration. The electron concentration $n_e(\theta, \vec{\rho})$ in Eq. (5) is given by

$$n_e(\theta, \vec{\rho}) = n_0 \int_{-\infty}^{\theta} w(E_1)\, d\theta, \tag{6}$$

where $w(E_1)$ is the ionization rate, which depends on the instantaneous pump field E_1, and n_0 is the concentration of neutral atoms.

The inclusion of ionization effects in the analysis of pulse propagation and harmonic generation in hollow fibers requires the use of some model of ionization allowing the calculation of the ionization rate $w(E_1)$. We shall examine below how tunneling ionization influences high-order harmonic generation in hollow fibers using the expression for the ionization rate derived by Ammosov *et al.* (1986):

$$w(E_1) = \left(\frac{3E_1}{\pi E_0}\right)^{1/2} \frac{2^{2n}}{n\Gamma(n+1)\Gamma(n)} \left(\frac{2E_0}{E_1}\right)^{2n-1} I_p \exp\left[-\frac{2E_0}{3E_1}\right], \tag{7}$$

where $E_0 = (2I_p)^{3/2}$, I_p is the ionization energy, $n = Z(2I_p)^{-1/2}$, Z is the residual ion charge, and atomic units are used in Eq. (7). In our simulations, the ionization rate $w(E_1)$ was averaged over the pump field cycle.

In the case when pump depletion is negligible, the equation for the amplitude $B_q = B_q(\theta, \vec{\rho}, z)$ of the qth harmonic is written as

$$\frac{dB_q}{dz} + \kappa_q B_q = \frac{2i\pi\omega^2 q^2}{K_q c^2} P_q^{NL}(\theta, \vec{\rho}) \times \exp[-i(K_q - qK^n - q\delta K^n(\theta))z], \tag{8}$$

where $2\kappa_q$ is the absorption coefficient of the qth harmonic, $P_q^{NL}(\theta, \vec{\rho})$ is the amplitude of the nonlinear polarization induced in the gas, and

$$\delta K^n(\theta) = \frac{-2\pi e^2}{K^n c^2 m} n_{\rm eff}^n(\theta) \tag{9}$$

is the addition to the propagation constant of the pump wave due to the ionization of the gas filling the fiber.

Solving Eq. (8), we derive the following expression for the intensity of the qth harmonic:

$$I_q(\theta, \vec{\rho}) = 2\pi K_q q\omega |P_q^{NL}(\theta, \vec{\rho})|^2 \left| \frac{1 - \exp(-i\Delta k_q^n(\theta) z - \kappa_q z)}{-i\Delta k_q^n(\theta) - \kappa_q} \right|^2, \tag{10}$$

where we set $K_q c \approx q\omega$, and

$$\Delta k_q^n(\theta) = K_q - qK^n - q\delta K^n(\theta) \approx \Delta k_0 + \Delta k_w^n + \delta k_q^n(\theta) \tag{11}$$

is the phase mismatch including the waveguide dispersion, with $\Delta k_0 = (q\omega/c)[n_1(q\omega) - n_1(\omega)]$, $\Delta k_w^n = (cq/\omega_1)(u^n/a)^2$, and $\delta k_q^n(\theta) = -q\delta K^n(\theta)$ being the phase-mismatch components due to the dispersion of the gas, waveguide, and electrons, respectively. Note that the first two components contribute to the phase mismatch in an additive way when the inequality $n_1(\omega) - 1 \ll 1$ is met.

The phase mismatch (11) involved in Eq. (10) depends not only on gas dispersion, but also on the dispersion of waveguide modes and time θ. As highlighted by Durfee *et al.* (1997), the phase mismatch dependence on the dispersion of waveguide modes provides an opportunity to improve phase matching by choosing an appropriate pair of transverse modes for the pump and third harmonic. As can be seen from Eqs. (10) and (11), a similar approach can be used to phase match higher order harmonics. However, in an ionizing gas, the phase mismatch is a function of time due to the generation of electrons, and phase matching in qth-harmonic generation can be achieved only for a certain moment of time θ.

In the case when the length of a hollow fiber is much larger than the absorption length of the qth harmonic, that is, when

$$\kappa_q z \gg 1, \tag{12}$$

Eq. (10) can be reduced to

$$I_q(\theta,\vec{\rho}) = 2\pi K_q q\omega |P_q^{NL}(\theta,\vec{\rho})|^2 [(\Delta k_q^n(\theta))^2 + (\kappa_q)^2]^{-1}. \tag{13}$$

Expression (13) gives the following estimate for the enhancement in the efficiency of the qth harmonic due to waveguide phase matching:

$$\eta = 1 + \left(\frac{\Delta k_0}{\kappa_q}\right)^2. \tag{14}$$

Formula (14) is very instructive from the physical point of view, as it shows that the maximum enhancement of harmonic-generation efficiency due to waveguide phase matching is determined by the dispersion and absorption of a gas medium rather than by the parameters of the fiber itself. In other words, if a hollow fiber allows the phase mismatch to be completely compensated, then the usefulness of a hollow fiber for harmonic generation increases with the growth in the ratio of the absorption length to the coherence lengths in a free gas. Therefore, a hollow fiber is especially efficient whenever a free gas is characterized by a considerable dispersion, becoming virtually useless in the case of strongly absorbing gases.

3. RESULTS AND DISCUSSION

To assess the influence of absorption effects on high-order harmonic generation, we employed Eqs. (11) and (14) to calculate the enhancement η of high-order harmonics of 790-nm fundamental radiation in a 150-μm-inner-diameter hollow fiber filled with different rare gases (helium, neon, and argon) in the absence of ionization (Fig. 1). The transverse distribution of the pump intensity was assumed to correspond to the EH_{11} waveguide mode. Keeping in mind a considerable absorption of rare gases in the short-wavelength spectral range, we assumed that condition (12) is satisfied

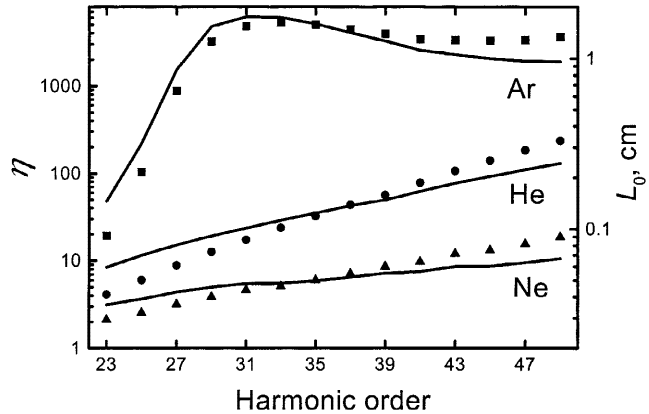

Fig. 1. The enhancement η of high-order harmonic generation due to waveguide phase matching (points) and the absorption length $L_0 = \frac{1}{2}\kappa_q$ corresponding to the optimal gas pressure p_0 allowing the harmonic-generation process to be phase matched (solid lines) in a 150-μm-inner-diameter hollow fiber filled with different rare gases (helium, neon, and argon) in the absence of ionization. The wavelength of pump radiation is 790 nm. The transverse distribution of the pump intensity corresponds to the EH_{11} waveguide mode.

(the absorption length $L_0 = \frac{1}{2}\kappa_q$ corresponding to the optimal gas pressure p_0 allowing the harmonic-generation process to be phase matched is also shown for comparison by solid lines in Fig. 1). The data on absorption coefficients and refractive indices of short-wavelength radiation in rare gases were taken from the data sheets by the Center for X-Ray Optics, Lawrence Berkeley National Laboratory, at http://cind.lbl.gov. The data on the refractive index at the pump frequency for rare gases can be found in Roth and Scheel (1931, 1935).

As can be seen from the data presented in Figure 1, the use of a hollow fiber allows the efficiency of high-order harmonic generation to be enhanced by two to three orders of magnitude, which is consistent with the results of experiments (Durfee *et al.*, 1997; Rundquist *et al.*, 1998; Tamaki *et al.*, 1998; Constant *et al.*, 1999). Especially high efficiencies of harmonic generation can be achieved with gases where the phase mismatch due to gas dispersion, Δk_0, is large as compared with the absorption coefficient $2\kappa_q$ at the harmonic frequency. This result agrees well with predictions of Eq. (14).

Figure 2 shows the energy $E_q = \iiint I_q(\theta,\vec{\rho})\,d\vec{\rho}\,d\theta\,(q = 41)$ of the 41st harmonic of 790-nm fundamental radiation generated in a 150-μm-inner-diameter 2-cm-long hollow fiber filled with argon (curves 1, 2), neon (curves 3, 4), and helium (curves 5, 6) calculated with the use of Eqs. (10) and (11) as a function of the gas pressure in the absence of ionization (solid lines 1, 3, 5) and in the regime of ionization governed by the ADK formula (7) (dashed lines 2, 4, 6). The energy of a 70-fs rectangular pump pulse was set equal to 0.65 mJ (2), 1.75 mJ (4), and 2.3 mJ (6), which corresponded to the ionization of 0.18% (2), 0.5% (4), and 0.7% (6) atoms of the gas, respectively.

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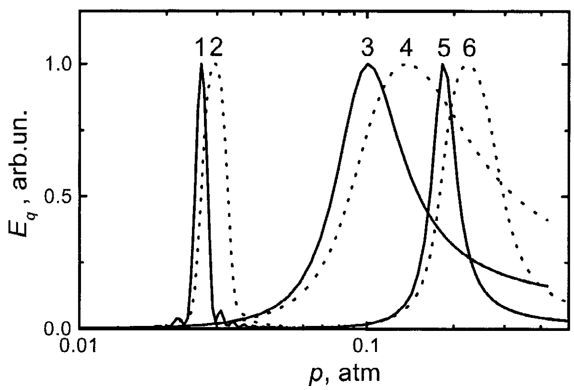

Fig. 2. The energy of the 41st harmonic of 790-nm fundamental radiation generated in a 150-μm-inner-diameter 2-cm-long hollow fiber filled with argon (curves 1, 2), neon (curves 3, 4), and helium (curves 5, 6) calculated with the use of Eqs. (10) and (11) as a function of the gas pressure in the absence of ionization (solid lines 1, 3, 5) and in the regime of ionization governed by the ADK formula (7) (dashed lines 2, 4, 6). The transverse distribution of the pump intensity corresponds to the EH_{11} waveguide mode. The energy of a 70-fs rectangular pump pulse was set equal to 0.65 mJ (2), 1.75 mJ (4), and 2.3 mJ (6).

As can be seen from Figures 2 and 3, the maximum energy of the 41st harmonic in the absence of ionization is achieved at the pressure corresponding to the phase-matching condition $\Delta k_q^1 = 0$. Due to the change in the phase of the pump pulse from its leading edge to the trailing edge, phase mismatch for harmonic generation changes within the pump pulse.

The shape of a laser pulse is assumed to be rectangular in our calculations, which is a gross oversimplification, of course. However, this approach allows us to reveal some important features of the influence of ionization on phase matching in harmonic generation in hollow fibers without specifying the harmonic generation model (as the nonlinear polarization amplitude at the harmonic frequency remains constant in the case of a rectangular pulse). In particular, a rectangular pulse gives rise to a linear growth in the effective electron concentration as a function of time θ. The phase mismatch in this regime also depends on θ. Therefore, different pressures would be required to phase match harmonic generation at different θ (see Fig. 3). The maxima in pressure dependencies of the harmonic signal are shifted toward higher pressures under these conditions (see Fig. 2). The net effect of ionization occurring in the gas filling the fiber is that it decreases the overall efficiency of harmonic generation and makes the harmonic-generation efficiency less sensitive to the gas pressure in the hollow fiber.

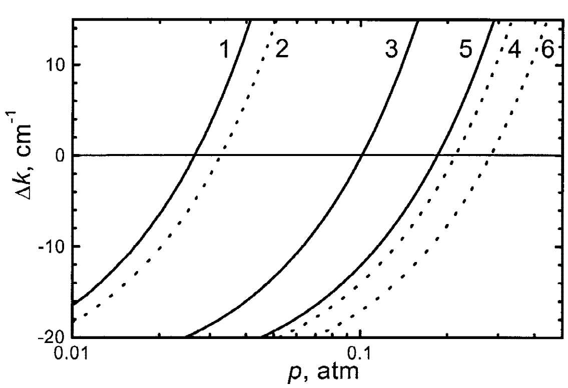

Fig. 3. The phase mismatch calculated with the use of Eq. (11) for the 41st harmonics of 790-nm fundamental radiation generated in a hollow fiber with an inner diameter of 150 μm and the length $L = 2$ cm filled with argon (1, 2), neon (3, 4), and helium (5, 6) as a function of the gas pressure on the leading edge of the pulse ($\theta/\tau = 0$), where ionization does not play an important role (solid lines 1, 3, 5), and on the trailing edge of the pulse ($\theta/\tau = 1$), where the number of ionized atoms is equal to n_e/n_0 (dashed lines 2, 4, 6). The transverse distribution of the pump intensity corresponds to the EH_{11} waveguide mode. The energy of a 70-fs rectangular pump pulse is 0.65 mJ (2), 1.75 mJ (4), and 2.3 mJ (6).

4. CONCLUSION

Theoretical investigation of high-order harmonic generation in gas-filled hollow fibers performed in this article within the framework of slowly varying envelope approximation allowed us to understand the influence of absorption and ionization effects on high-order harmonic generation in hollow fibers. We have shown that the enhancement of harmonic-generation efficiency due to phase matching in hollow fibers may reach three orders of magnitude with an appropriate choice of the sort and the pressure of the gas and parameters of the fiber. Especially high efficiencies of harmonic generation in hollow fibers can be achieved with gases where the phase mismatch due to gas dispersion is large as compared with the absorption coefficient at the harmonic frequency. Ionization of the gas filling the fiber during the pump pulse gives rise to an electronic contribution to the refractive index of the gas and, consequently, to the phase modulation of the pump pulse. Due to this change in the phase of the pump pulse from its leading edge to the trailing edge, the phase mismatch for harmonic generation changes within the pump pulse, decreasing the overall efficiency of harmonic generation, but making the harmonic-generation efficiency less sensitive to the gas pressure in the hollow fiber.

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