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, 1999a, 1999b; 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 · 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., 1999b) and theoretically (Naumov et al., 2000).

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

\[ E_1 = A^n(\theta, z)f^n(\hat{p})\exp[-i(\omega t - K_n z)] + c.c. \]  

\[ E_q = B_q(\theta, z, \hat{p})\exp[-i(q \omega t - K_q z)] + c.c., \]

where \( \omega \) is the frequency of fundamental radiation, \( q \) is the harmonic order, \( f^n(\hat{p}) \) is the transverse distribution of the
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 $q$th 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), $B_q(\theta, z, \hat{\rho})$ 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 Marcanti 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

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

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

$$\chi^{(1)}_n(\theta) = -\frac{e^2}{m\omega^2} n_{\text{eff}}^e(\theta), \tag{4}$$

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

$$n_{\text{eff}}^e(\theta) = \frac{\int f^n(\hat{\rho}) n_e(\theta, \hat{\rho}) d\hat{\rho}}{\int |f^n(\hat{\rho})|^2 d\hat{\rho}} \tag{5}$$

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

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

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):

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

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 $B_q = B_q(\theta, \hat{\rho}, z)$ of the $q$th harmonic is written as

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

where $2\kappa_q$ is the absorption coefficient of the $q$th harmonic, $P_{\text{NL}}(\theta, \hat{\rho})$ is the amplitude of the nonlinear polarization induced in the gas, and

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

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 $q$th harmonic:

$$I_q(\theta, \hat{\rho}) = 2\pi K_q q\omega |P_{\text{NL}}^q(\theta, \hat{\rho})|^2 \left[ 1 - \frac{\exp(-i\Delta k_q^0(\theta) z - \kappa_q z)}{-i\Delta k_q^0(\theta) - \kappa_q} \right]^2 \tag{10}$$

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

$$\Delta k_q^0(\theta) = K_q - qK^n - q\delta K^n(\theta) \approx \Delta k_0^q + \Delta k_0^\omega + \delta k_q^0(\theta) \tag{11}$$

is the phase mismatch including the waveguide dispersion, with $\Delta k_0^q = (q\omega/c)[n_1(q\omega) - n_1(\omega)]$, $\Delta k_0^\omega = (c\omega/\omega_1)(n^*/n)^2$, and $\delta k_q^0(\theta) = -q\delta K^n(\theta)$ 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.
Ionization and absorption effects in high-order harmonic generation

The phase mismatch (11) involved in Eq. (10) depends not only on gas dispersion, but also on the dispersion of waveguide modes and time \( \theta \). 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 \( q \)th-harmonic generation can be achieved only for a certain moment of time \( \theta \).

In the case when the length of a hollow fiber is much larger than the absorption length of the \( q \)th harmonic, that is, when

\[ \kappa_q \ll 1, \tag{12} \]

Eq. (10) can be reduced to

\[ I_q(\theta, \rho) = 2\pi \kappa_q \rho_0 |P_{q\theta}(\theta, \rho)|^2 [\Delta \kappa_q^2(\theta)^2 + (\kappa_q)^2]^{-1}. \tag{13} \]

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

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

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 \( \eta \) of high-order harmonics of 790-nm fundamental radiation in a 150-\( \mu \)m-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 \( \text{EH}_{11} \) waveguide mode. Keeping in mind a considerable absorption of rare gases in the short-wavelength spectral range, we assumed that condition (12) is satisfied (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-\( \mu \)m-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 \( \text{EH}_{11} \) waveguide mode.

![Fig. 1. The enhancement \( \eta \) 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-\( \mu \)m-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 \( \text{EH}_{11} \) waveguide mode.](image)
important role leading edge of the pulse with an inner diameter of 150 μm. Harmonics of 790-nm fundamental radiation generated in a hollow fiber achieved at the pressure corresponding to the phase-matching energy of the 41st harmonic in the absence of ionization is shown in Fig. 2. The phase mismatch calculated with the use of Eqs. (10) and (11) as a function of the gas pressure in the absence of ionization is shown in 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.

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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Ionization and absorption effects in high-order harmonic generation