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Successive high-order harmonic generation in hollow fibres

A M Zheltikov, A N Naumov

Abstract. The method for phase matching of successive high-order harmonic generation in gas-filled hollow fibres is proposed. Successive harmonic generation allows the energy of optical harmonics to be increased as compared with the direct harmonic-generation process. With an appropriate choice of the type and the pressure of the gas filling the fibre, parameters of the fibre, wavelength of pump radiation, and waveguide modes, the phase-matching conditions can be simultaneously satisfied for all the steps of successive nonlinear optical high-order harmonic generation. Under these conditions, a gas-filled hollow fibre may serve as an efficient two-colour source of coherent short-wavelength radiation.

1. Introduction

The fact that successive processes may play an important role in nonlinear optical frequency conversion is known from classical works on nonlinear optics [1]. Currently, much attention is focused on the possibility of using sequential three-wave mixing in media with quadratic nonlinearities in the presence of a common pump field for the enhancement of the efficiency of quasi-phase-matched frequency conversion [2 – 5].

In this paper, we will demonstrate that the idea of simultaneously phase matching two or more nonlinear optical processes in order to enhance the efficiency of frequency conversion and simultaneously to generate several new frequency components of optical radiation offers much promise not only for three-wave mixing, but also for higher order nonlinear processes. In particular it is of special interest to extend this approach to high-order harmonic generation in gas-filled hollow fibres.

The possibility of increasing the length of nonlinear optical interactions in gas media by using hollow dielectric waveguides was demonstrated by Miles et al. [6]. Important theoretical aspects of four-wave mixing in gas-filled hollow fibres were analysed in Ref. [7]. Nisoli et al. [8, 9] have opened a new phase in the nonlinear optics of hollow fibres by demonstrating that such fibres allow the efficient spectral broadening of ultrashort laser pulses to be achieved by self-phase modulation. This effect allowed extremely short pulses to be produced. Currently, hollow fibres are widely used for both efficient generation of short-wavelength radiation [10 – 13] and the formation of ultrashort pulses [9, 14].

2. Basic relations

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

$$E_1 = \tfrac{1}{2}\, f_1^n(\boldsymbol{\rho}) A^n(t,z) \exp[-\mathrm{i}(\omega t - K_1^n z)] + \text{c.c.}\,, \tag{1}$$

$$E_q = \tfrac{1}{2} f_q^m(\boldsymbol{\rho}) B_q^m(tz) \exp[-\mathrm{i}(q\omega t - K_q^m z)] + \text{c.c.}\,, \tag{2}$$

where ω is the frequency of fundamental radiation; q is the harmonic order; $f_1^n(\boldsymbol{\rho})$ and $f_1^m(\boldsymbol{\rho})$ are the transverse field distributions in fundamental radiation and the qth harmonic corresponding to the EH_{1n} and EH_{1m} modes of the hollow waveguide, respectively; K_1^n and K_q^m are the propagation constants of the pump and harmonic beams corresponding to the eigenmodes of the hollow fibre; $A^n(t,z)$ and $B_q^m(t,z)$ are the slowly varying envelopes of the pump and harmonic pulses, respectively.

Provided that the inequalities

$$\frac{\omega_i a}{c} \gg 1\,, \tag{3}$$

$$\left|\frac{K_i^m c}{\omega_i n_1(\omega_i)}\right| \ll 1, \tag{4}$$

are satisfied, where a is the inner radius of the hollow fibre and $n_1(\omega_i)$ is the refractive index of the gas filling the fibre at the frequency $\omega_i = i\omega$, $i = 1, 3, q$, we can employ approximate analytical solutions for the transverse distribution of the electric field and propagation constants of light beams in a hollow fibre. In particular, for EH_{1m} modes of a hollow fibre, we can write [15]

$$f_i{}^m(\boldsymbol{\rho}) = J_0\left(\frac{u^m \rho}{a}\right) \tag{5}$$

where $J_0(x)$ is the zeroth-order Bessel function and u^m is the eigenvalue of the EH_{1m} mode. The propagation constants are given by

$$K^m \approx \frac{\omega_i n_1(\omega_i)}{c}\left[1 - \left(\frac{u^m c}{a\omega_i n_1(\omega_i)}\right)^2 \left(\frac{1}{2} + \frac{\mathrm{Im}\mu(\omega_1)}{a\omega_i}\right)\right], \tag{6}$$

where

$$\mu(\omega_i) = \frac{\varepsilon_2(\omega_i) + n_1^2(\omega_i)}{2n_1^2(\omega_i)\left[\varepsilon_2(\omega_i) - n_1^2(\omega_i)\right]^{1/2}}$$

for EH modes and $\varepsilon_2(\omega_i)$ is the dielectric constant of the fibre cladding at the frequency ω_i.

A M Zheltikov, A N Naumov Physics Faculty, M V Lomonosov Moscow State University, Vorob'evy gory, 119899 Moscow, Russia

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We will consider successive generation of the qth harmonic

$$\omega_3 = \omega + \omega + \omega, \quad \omega_q = (q - 3l)\omega + l\omega_3 \ , \tag{7}$$

where l is an integer. The field with the frequency ω_q is generated in two steps in process (7). In the first step, the third harmonic is generated, whereas the second step involves the qth-harmonic generation through the nonlinearity of the $(q - 2l)$th order.

In our analysis we will ignore pump depletion and assume that the transverse distribution of the pump intensity corresponds to the EH_{1n} waveguide mode. Then the expression for the amplitude of the EH_{1m} mode of the third harmonic is written as

$$\frac{\partial B_3^m}{\partial z} = \mathrm{i}\beta_3^{nm}(A_1^n)\exp(-\mathrm{i}\Delta k_3^{nm} z) \ , \tag{8}$$

where $\Delta k_3^{nm} = K_3^m - 3K_1^n$ is the phase mismatch that includes the waveguide dispersion and

$$\beta_3^{nm}(A^n) = \frac{2\pi}{K_3^m}\left(\frac{\omega_3}{c}\right)^2 \frac{\int\int f_3^m(\boldsymbol{\rho})P_3^{\mathrm{NL}}\mathrm{d}\boldsymbol{\rho}}{\int\int\left[f_3^m(\boldsymbol{\rho})\right]^2\mathrm{d}\boldsymbol{\rho}} \tag{9}$$

is the nonlinear coefficient responsible for third-harmonic generation. This coefficient depends on the amplitude of the pump field via the amplitude P_3^{NL} of the nonlinear polarisation of the medium and includes the transverse distributions of the third-harmonic and pump fields in the relevant waveguide modes.

If the transverse intensity distribution in the third harmonic corresponds to the EH_{1m} waveguide mode, then the equation for the amplitude of the qth harmonic produced through the successive process (7) in the EH_{1h} waveguide mode can be represented as

$$\frac{\partial B_q^h}{\partial z} + \varkappa_q B_q^h = \mathrm{i}\beta_q^{nh}(A^n)\exp\left(-\mathrm{i}\Delta k_q^{nh} z\right) + \sum_{l\neq 0}\mathrm{i}\gamma_{ql}^{nmh}(A^n, B_3^m)\exp\left(-\mathrm{i}\Delta k_{ql}^{nmh} z\right). \tag{10}$$

Here $2\varkappa_q$ is the absorption coefficient at the frequency of the qth harmonic; $\Delta k_q^{nh} = K_q^h - qK_1^n$ is the phase mismatch for the direct process of qth-harmonic generation; $\Delta k_{ql}^{nmh} = K_q^h - (q - 3l)K_1^n - lK_3^m = \Delta k_q^{nh} - l\Delta k_3^{nm}$ is the phase mismatch for the step process of qth-harmonic generation involving l quanta of the third harmonic;

$$\beta_q^{nh}(A^n) = \frac{2\pi}{K_q^h}\left(\frac{\omega_q}{c}\right)^2 \frac{\int\int f_q^h(\boldsymbol{\rho})P_q^{\mathrm{NL}}\mathrm{d}\boldsymbol{\rho}}{\int\int\left[f_q^h(\boldsymbol{\rho})\right]^2\mathrm{d}\boldsymbol{\rho}} \tag{11}$$

is the nonlinear coefficient responsible for the direct process of qth-harmonic generation, which depends on the pump amplitude via the amplitude P_q^{NL} of the relevant nonlinear polarisation; and

$$\gamma_{ql}^{nmh}(A^n, B_3^m) = \frac{2\pi}{K_q^h}\left(\frac{\omega_q}{c}\right)^2 \frac{\int\int f_q^h(\boldsymbol{\rho})P_{ql}^{\mathrm{NL}}\mathrm{d}\boldsymbol{\rho}}{\int\int\left[f_q^h(\boldsymbol{\rho})\right]^2\mathrm{d}\boldsymbol{\rho}} \tag{12}$$

is the nonlinear coefficient responsible for qth-harmonic generation through the step process (7), which depends on the pump and third-harmonic amplitudes via the amplitude P_{ql}^{NL} of the relevant nonlinear polarisation.

3. Results and discussion

As can be seen from expressions (10)–(12), the efficiency of successive qth-harmonic generation through the step process (7) depends not only on the pump field amplitude A^n, but also on the amplitude B_3^m of the third harmonic. If third-harmonic generation is phase matched, then the amplitude B_3^m increases as a function of z, opening up an additional channel of energy transfer to the qth harmonic. This circumstance allows the intensity of a high-order harmonic produced through a phase matched step process (7) to be increased as compared with the intensity of the same harmonic produced through a direct process.

Fig. 1 displays the effective refractive index $n_{\mathrm{ef}} = K_q^m c/\omega$ — calculated for the EH_{11}, EH_{12}, EH_{13}, and EH_{14} modes of a hollow fibre with the inner diameter $a = 85$ μm filled with helium at the pressure $p = 0.018$ atm — as a function of the wavelength. In these calculations, we employed the data on the frequency dependences of the absorption coefficient and the refractive index of helium taken from Refs [16–19]. As can be seen from the results of calculations presented in Fig. 1, the effective refractive index $K_i^m c/\omega_i$ for the EH_{11} mode of radiation with the wavelength $\lambda_0 = 252.9$ nm is equal to the effective refractive index of the third harmonic of this radiation at the wavelength $\lambda_0/3 = 84.3$ nm for the EH_{13} mode. Both of these refractive indices are equal to unity, which implies that any step process of harmonic generation of a sufficiently high order is asymptotically phase matched, because the effective refractive indices for high-order harmonics are also close to unity (see Fig. 1).

Fig. 2 presents the phase mismatches calculated for the direct process of 13th-harmonic generation, step process (7) of 13th-harmonic generation involving third-harmonic generation, and third-harmonic generation, as well as the absorption coefficient for the 13th harmonic as functions of the pressure p of helium filling a hollow fibre with the inner radius $a = 75$ μm for the wavelength of pump radiation equal to 0.24 μm and the mode indices $n = 1$, $m = 3$, and $h = 1$. As can be seen from the results of these calculations, direct and successive 13th-harmonic generation and third-harmonic generation in the EH_{13} waveguide mode can be simultaneously phase-matched at the helium pressure $p = 0.017$ atm. Under these conditions a gas-filled hollow fibre may serve

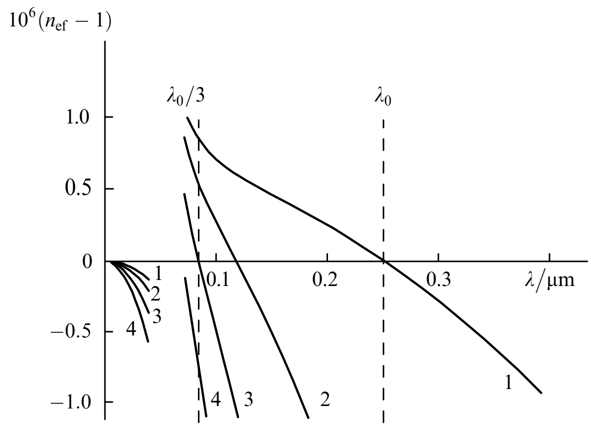

Figure 1. Effective refractive index $n_{\mathrm{ef}} = K_q^m c/\omega$ for (1) the EH_{11}, (2) EH_{12}, (3) EH_{13}, and (4) EH_{14} modes of a hollow fibre with an inner diameter $a = 85$ μm filled with helium at the pressure $p = 0.018$ atm calculated as a function of the radiation wavelength.

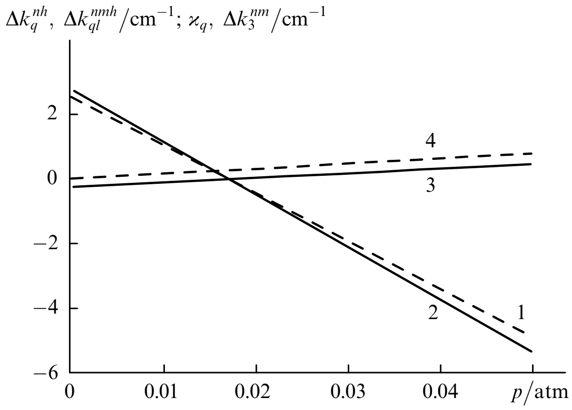

Figure 2. Phase mismatches for (1) direct process of 13th-harmonic generation (Δk_q^{nh}), (2) step process of 13th-harmonic generation involving third-harmonic generation with $l = 1$ (Δk_{ql}^{nmh}) and, (3) third-harmonic generation (Δk_3^{nm}) and (4) absorption coefficient for the 13th harmonic calculated as functions of the pressure of helium filling a hollow fibre with an inner radius $a = 75$ μm. The wavelength of pump radiation is 0.24 μm, and the mode indices are $n = 1$, $m = 33$, and $h = 1$.

as a source of coherent radiation generating short-wavelength light at two frequencies simultaneously.

The results of calculations presented in Fig. 2 also show that absorption effects play an important role in high-order harmonic generation in hollow fibres (curve 4 in Fig. 2). Absorption at the frequency of the third harmonic is much less important, which allows the efficiency of high-order harmonic generation to be improved through the use of step processes due to the energy transferred from the third harmonic. The amplitude of the third harmonic continues to grow as a function of the propagation coordinate when the direct process of high-order harmonic generation saturates due to absorption effects [20].

The physics of successive high-order harmonic generation in gas-filled hollow fibres involves several substantially new aspects as compared with nonlinear optical processes in quasi-phase-matched structures. While frequency conversion in quasi-phase matched crystals employs second-order nonlinearities [21, 22], allowing, as mentioned above, quasi-phase-matched three-wave mixing to be implemented [2 – 5], gases filling hollow fibres possess only odd-order nonlinearities. However, since the thresholds of optical breakdown of gases filling hollow fibres are typically much higher than the thresholds of optical breakdown in solids, laser intensities that can be coupled into a gas-filled fibre are typically much higher than the intensities that can be coupled into quasi-phase-matched structures. Therefore, nonlinearities of very high orders can be involved in step processes in gas-filled hollow fibres, allowing radiation with very short wavelengths to be produced.

We should note also that the use of a gas filling a hollow fibre as a nonlinear optical medium allows the broadening of short laser pulses due to group-velocity dispersion to be avoided in nonlinear optical processes [23, 24]. Finally, keeping in mind high efficiencies of second-harmonic generation and parametric wave mixing attainable in quasi-phase-matched structures [21, 22], it would be of interest also to extend the method of phase matching proposed in this paper to nonlinear optical interactions in quasi-phase-matched waveguides.

4. Conclusions

Thus, theoretical analysis performed in this paper demonstrates that successive nonlinear optical interactions open up new ways of improving the efficiency of high-order harmonic generation in gas-filled hollow fibres and controlling such processes. Analysis of propagation effects in nonlinear optical interactions in gas-filled hollow fibres shows that successive harmonic generation allows the energy of harmonic radiation to be increased as compared with harmonic radiation generated through a direct process.

With an appropriate choice of the sort and the pressure of the gas filling the fibre, parameters of the fibre, the wavelength of pump radiation, and the waveguide modes involved in the interaction, phase-matching conditions can be simultaneously satisfied for each of the steps of successive high-order harmonic generation. Under these conditions, a gas-filled hollow fibre may serve as an efficient two-colour source of coherent short-wavelength radiation.

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