 

PACS numbers: 42.65.Ky; 42.79.Gn
DOI: 10.1070/QE2001v031n02ABEH001912

# Third-harmonic generation in the field of ultrashort laser pulses in leaky modes of a gas-filled hollow fibre

O A Kolevatova, A N Naumov, A M Zheltikov

***Abstract*. The influence of phase mismatch, group delay, and optical losses of waveguide modes on third-harmonic generation in a hollow fibre is investigated. An analytical solution to the equation governing the slowly varying envelope of the third-harmonic pulse in a leaky mode of a gas-filled hollow fibre is derived. Optimal conditions for third-harmonic generation in such a fibre are determined for different pump pulse durations and various rare gases.**

***Keywords*:** *ultrashort laser pulses, third-harmonic generation, hollow fibre.*

## 1. Introduction

Several significant achievements of nonlinear optics and optics of ultrashort laser pulses within the last five years were due to the use of gas-filled hollow dielectric waveguides. In particular, the self-phase modulation of laser pulses in such fibres allowed the authors of Ref. [1] to produce unprecedentedly short light pulses with a duration of 4.5 fs. Since the threshold of optical breakdown for a gas filling a hollow fibre is typically much higher than the breakdown thresholds characteristic of conventional optical fibres, high-power ultrashort light pulses consisting of a few field cycles can be generated with the use of hollow fibres. Due to high breakdown thresholds of gases filling a hollow fibre, such waveguides also offer much promise for nonlinear-optical frequency conversion by means of parametric wave mixing and harmonic generation.

Experiments performed in Ref. [2] have demonstrated that the use of hollow fibres provides an opportunity to achieve high efficiencies of optical frequency conversion in third-harmonic generation (up to 0.2 %) and parametric four-wave mixing (up to 13 %). The authors of [2] have also experimentally demonstrated that the phase mismatch due to waveguide dispersion in hollow optical fibres can compensate for the phase mismatch related to gas dispersion. The efficiency of nonlinear-optical interactions substantially increases under these conditions, opening new ways of increasing the energy of high-order harmonics and controlling harmonic parameters by varying characteristics of pump radiation [3, 4].

Subsequent extensive studies of nonlinear-optical processes in gas-filled hollow fibres [5–8] have shown that phase-matching abilities of such fibres allow the efficiency of frequency conversion in the generation of optical harmonics up to the 45th order to be improved by a factor of 100–1000 with respect to the efficiencies of high-order harmonic generation attainable in experiments with gas jets [9–11]. Recent experiments [8] have demonstrated the possibility of using hollow fibres for the generation of sub-10-fs light pulses in the UV range through parametric four-wave mixing. The efficiency of frequency conversion achieved in these experiments exceeded 20 %.

Optical losses inherent in leaky modes of hollow fibres [12, 13] is an important factor that has a considerable influence on the efficiency of nonlinear-optical processes in such waveguides [14]. Analysis of such losses is, therefore, of considerable importance for practical applications of hollow fibres in nonlinear and ultrafast optics. In what follows, we shall dwell upon this issue.

## 2. Formulation of the problem

Consider third-harmonic generation in a hollow optical fibre that consists of a core with a radius $a$, filled with a gas medium having a refractive index $n_1$, and a cladding with a real dielectric constant $\varepsilon_2$ (we assume that $\varepsilon_2 > n_1^2$). To analyse this process, we will employ the approximation of slowly varying envelopes, assuming that the duration of light pulses is still large as compared with the cycle of the optical field.

Suppose that the transverse intensity distribution of the fundamental (pump) beam corresponds to the $EH_{n'n}$ mode of a hollow waveguide and consider the generation of the $EH_{m'm}$ hollow-waveguide mode of the third harmonic. Let us represent the pump and third-harmonic beams propagating along the $z$-axis in a hollow fibre filled with a gas possessing a third-order nonlinearity in the following form:

$$\boldsymbol{E}_{\rm p} = \frac{1}{2} f_{\rm p}^{(n'n)}(\boldsymbol{\rho}) \boldsymbol{e}_{\rm p} A^{(n'n)}(z,t) \times \exp\left[-{\rm i}\omega_{\rm p}t + \left({\rm i}K_{\rm p}^{(n'n)} - \frac{\alpha^{(n'n)}}{2}\right)z\right] + {\rm c.c.}, \quad (1)$$

$$\boldsymbol{E}_{\rm h} = \frac{1}{2} f_{\rm h}^{(m'm)}(\boldsymbol{\rho}) \boldsymbol{e}_{\rm h} B^{(m'm)}(z,t) \exp\left(-{\rm i}\omega_{\rm h}t + {\rm i}K_{\rm h}^{(m'm)}z\right) + {\rm c.c.}, \quad (2)$$

**O A Kolevatova, A N Naumov, A M Zheltikov** Department of Physics, M V Lomonosov Moscow State University, Vorob'evy gory, 119899 Moscow, Russia

Received 11 September 2000
*Kvantovaya Elektronika* **31** (2) 173–178 (2001)
Translated by A M Zheltikov

where the subscripts p and h are related to the parameters of the pump and third harmonic, respectively; $\omega_{\rm p}$ is the central frequency of pump radiation; $\omega_{\rm h} = 3\omega_{\rm p}$; $f_{\rm p}^{(n'n)}(\boldsymbol{\rho})$ and $f_{\rm h}^{(m'm)}(\boldsymbol{\rho})$ are the transverse field distributions of fundamental radiation and the third harmonic in the hollow fibre, respectively; $A^{(n'n)}(z,t)$ and $B^{(m'm)}(z,t)$ are the slowly varying envelopes of the pump and third-harmonic pulses, respectively; $K_{\rm p}^{(n'n)}$ and $K_{\rm h}^{(m'm)}$ are the propagation constants of the pump and third-harmonic pulses; $\alpha_{\rm p}^{(n'n)}$ is the attenuation coefficient of the pump pulse; and $\boldsymbol{e}_{\rm p}$ and $\boldsymbol{e}_{\rm h}$ are the polarisation unit vectors of pump radiation and the third harmonic, respectively.

We assume that both pump radiation and the third harmonic have wavelengths much less than the inner radius of the hollow fibre $a$ and sufficiently small attenuation coefficients [12, 13]:

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

$$\left| \frac{K_l^{(m'm)} c}{\omega_l n_1(\omega_l)} - 1 \right| \ll 1, \tag{4}$$

where $l = {\rm p, h}$; and $n_1(\omega_l)$ is the refractive index of the gas filling the fibre at the frequency $\omega_l$. Then, 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

$$f_l^{(1m)}(\boldsymbol{\rho}) \equiv f_l^{(m)}(r) = J_0\left(\frac{u_l^{(m)} r}{a}\right). \tag{5}$$

where $J_0(x)$ is the zeroth-order Bessel function and $u_l^{(m)}$ is the eigenvalue for the $EH_{1m}$ mode. The relevant propagation constants and attenuation coefficients are given by [13]

$$K_l^{(1m)} \equiv K_l^{(m)} \approx \frac{\omega^l n_1(\omega^l)}{c}\left\{1 - \frac{1}{2}\left[\frac{u_l^{(m)} c}{a\omega_l n_1(\omega_l)}\right]^2\right\}, \tag{6}$$

$$\alpha_l^{(1m)} \equiv \alpha_l^{(m)} \approx \frac{2}{a n_1(\omega_l)}\left(\frac{u_l^{(m)} c}{a\omega_l}\right)^2 \frac{\left[\varepsilon_2(\omega_l) + n_1^2(\omega_l)\right]}{2n_1^2(\omega_l)\left[\varepsilon_2(\omega_l) - n_1^2(\omega_l)\right]^{1/2}}. \tag{7}$$

Using an approach similar to that described in [15], we arrive at the following equation for the slowly varying envelope of the third harmonic $B^{(1m)}(z,t) \equiv B^{(m)}(z,t)$ in a lossy hollow fibre:

$$\left(\frac{\partial}{\partial z} + \frac{1}{\upsilon_{\rm h}^{(m)}}\frac{\partial}{\partial t}\right) B^{(m)}(z,t) + \frac{\alpha_{\rm h}^{(m)}}{2} B^{(m)}(z,t) = {\rm i}\beta^{(mn)}\left(A^{(n)}(z,t)\right)^3 \exp\left(-{\rm i}\Delta k^{(mn)} z\right). \tag{8}$$

Here, $A^{(1n)}(z,t) \equiv A^{(n)}(z,t)$; $\upsilon_{\rm h}^{(m)}$ and $\alpha_{\rm h}^{(m)}$ are the group velocity and the attenuation coefficient of the third-harmonic pulse, respectively; and

$$\Delta k^{(mn)} = K_{\rm h}^{(m)} - 3K_{\rm p}^{(n)} \approx \Delta k_0 + \Delta K_{\rm w}^{(mn)} \tag{9}$$

is the phase mismatch for third-harmonic generation including the waveguide dispersion, with

$$\Delta k_0 = \frac{\omega_{\rm h}}{c}\left[n_1(\omega_{\rm h}) - n_1(\omega_{\rm p})\right], \tag{10}$$

and

$$\Delta k_{\rm w}^{(mn)} = \frac{c}{2\omega_{\rm p}}\left[3\left(\frac{u_{\rm p}^{(n)}}{a}\right)^2 - \frac{1}{3}\left(\frac{u_{\rm h}^{(m)}}{a}\right)^2\right] \tag{11}$$

being the components of the phase mismatch due to gas and waveguide dispersion, respectively. The total phase mismatch can be represented as a sum of these two components in the case when the inequality $n_1(\omega_l) - 1 \ll 1$ is satisfied. The nonlinear coefficient $\beta^{(mn)}$ in Eqn (8) can be expressed in terms of the relevant nonlinear-optical cubic susceptibility:

$$\beta^{(mn)} = \frac{9\pi\omega_{\rm p}^2}{2K_{\rm h}^{(m)} c^2}\boldsymbol{e}_{\rm h}^* \hat{\chi}^{(3)}(\omega_{\rm h};\omega_{\rm p},\omega_{\rm p},\omega_{\rm p})\boldsymbol{e}_{\rm p}\boldsymbol{e}_{\rm p}\boldsymbol{e}_{\rm p} \times \frac{\iint f_{\rm h}^{(m)}(\rho)\left[f_{\rm p}^{(n)}(\rho)\right]^3 \rho\,{\rm d}\rho\,{\rm d}\theta}{\iint\left[f_{\rm p}^{(m)}(\rho)\right]^2 \rho\,{\rm d}\rho\,{\rm d}\theta}. \tag{12}$$

Equation (8) is similar to equations that describe third-harmonic generation in a gas medium with allowance for phase-mismatch, group-delay, and attenuation effects within the framework of plane-wave approximation. The right-hand side of Eqn (8) describes the third-order nonlinear polarisation of the medium responsible for third-harmonic generation. However, in contrast to the plane-wave approximation, Eqn (8) takes into account the influence of a waveguide through propagation constants (6), group velocities of the pump and third-harmonic pulses, and the nonlinear coefficient (12), which includes the transverse distributions of the pump and third-harmonic fields for the relevant hollow-fibre waveguide modes. In particular, the phase mismatch, which is involved in Eqn (8) and which determines the efficiency of third-harmonic generation, depends not only on the gas dispersion, but also on the dispersion of waveguide modes. As pointed out in [2], this circumstance provides an opportunity to improve phase-matching conditions for a certain pair of waveguide modes of pump radiation and the third harmonic.

Introducing the frame of reference running with the pulse of the third harmonic and replacing variables $t$ and $z$ by $\eta_{\rm h}^{(m)}$ and $z$, where $\eta_{\rm h}^{(m)} = (t - z/\upsilon_{\rm h}^{(m)})/\tau$ is the so-called running time normalised to the duration $\tau$ of the incident light pulse, we can rewrite Eqn (8) for the slowly varying envelope of the third harmonic as

$$\frac{\partial}{\partial z} B^{(m)}\left(z,\eta_{\rm h}^{(m)}\right) + \frac{\alpha_{\rm h}^{(m)}}{2} B^{(m)}\left(z,\eta_{\rm h}^{(m)}\right) = {\rm i}\beta^{(mn)}\left[A^{(n)}\left(z,\eta_{\rm h}^{(m)}\right)\right]^3 \exp\left(-{\rm i}\Delta k^{(mn)} z\right). \tag{13}$$

Integrating Eqn (13), we arrive at

$$B^{(m)}\left(\eta_{\rm h}^{(m)}\right) = {\rm i}\beta^{(mn)}\int_0^L \left[A_0^{(n)}\left(\eta_{\rm h}^{(m)} + \zeta^{(mn)} z\right)\right]^3 \times \exp\left[\left(\frac{\alpha_{\rm h}^{(m)} - 3\alpha_{\rm p}^{(n)}}{2} - {\rm i}\Delta k^{(mn)}\right) z - \frac{\alpha_{\rm h}^{(m)}}{2} L\right] {\rm d}z. \tag{14}$$

where $\zeta^{(mn)} = (1/\upsilon_{\rm h}^{(m)} - 1/\upsilon_{\rm p}^{(n)})/\tau$ is the group-delay coefficient and $L$ is the fibre length.

Similar to Eqn (8), Eqn (14) for the envelope of the third-harmonic pulse is formally analogous to expressions derived in the plane-wave approximation. However, in contrast to the formulas of the plane-wave approximation, Eqn (14) includes waveguide dispersion and transverse intensity distributions in the pump and third-harmonic beams.

## 3. Estimates on characteristic spatial scales

The efficiency of third-harmonic generation in the field of ultrashort laser pulses may be severely limited, as can be seen from Eqn (14), due to the phase mismatch, group delay, and attenuation of light pulses propagating in a hollow fibre. One can see from Eqns (6), (7), and (9)–(11), the phase mismatch, group delay, and attenuation coefficients of light pulses involved in third-harmonic generation in a hollow fibre can be varied by choosing waveguide modes, the inner diameter of a hollow fibre, and the pressure of the gas filling the fibre.

Table 1 presents estimates for the spatial scales typical of dispersion spreading, $l_{\rm d} = \tau^2(\partial^2 K_l^{(m)}/\partial\omega_l^2)^{-1}$; phase mismatch, $l_{\rm ph}^{(mn)} = |\Delta k^{(mn)}|^{-1}$; group delay, $l_{\rm w}^{(mn)} = (\zeta^{(mn)})^{-1} = \tau(|1/v_{\rm h}^{(m)} - 1/v_{\rm p}^{(n)}|)^{-1}$; and attenuation, $l_{\rm a} = (\alpha_l^{(m)})^{-1}$, characterising the influence of these effects on third-harmonic generation in a gas-filled hollow fibre and a free gas. These numerical estimates were obtained with the use of the data for the dispersion of rare gases taken from [16].

**Table 1.** Characteristic spatial scales for third-harmonic generation in the $EH_{1m}$ modes of a lossy argon-filled hollow fibre with an inner radius $a = 75$ μm and in an argon cell with 0.8-μm 20-fs pump pulses at the pressure of argon equal to 0.5 atm.

| $m$ | $l_{\rm d}$/m | $l_{\rm d0}$/m | $l_{\rm ph}^{(mn)}$/cm | $l_{\rm ph0}^{(1m)}$/cm | $l_{\rm w}^{(1m)}$/cm | $l_{\rm w0}^{(1m)}$/cm | $l_{\rm a}$/m |
|---|---|---|---|---|---|---|---|
| 1 | 10.9 | 10.6 | 0.24 | 0.42 | 24.4 | 18.9 | 14 |
| 2 | 12.1 | 10.6 | 0.32 | 0.42 | 21.3 | 18.9 | 2.6 |
| 3 | 15.1 | 10.6 | 0.67 | 0.42 | 16.9 | 18.9 | 1.1 |

Notation: $l_{\rm d}$ and $l_{\rm d0}$ are the lengths of dispersion spreading in a hollow fibre and a free gas, respectively; $l_{\rm ph}^{(1m)}$ and $l_{\rm ph0}^{(1m)}$ are the phase-matching lengths in a hollow fibre and a free gas, respectively; $l_{\rm w}^{(1m)}$ and $l_{\rm w0}^{(1m)}$ are the walk-off lengths in a hollow fibre and a free gas, respectively; and $l_{\rm a}$ is the attenuation length of pump radiation in the hollow fibre.

One can see from the estimates presented in Table 1 that the dispersion length of the pump pulse for the considered conditions substantially exceeds the characteristic spatial scales of phase mismatch, group delay, and attenuation. Dispersion lengths under these conditions are also typically much larger than the characteristic lengths of hollow fibres conventionally employed in harmonic-generation experiments (see [5–8]). Thus, our estimates show that the dispersion spreading of light pulses usually plays a negligible role in third-harmonic generation in gas-filled hollow fibres under standard experimental conditions. The estimates presented in Table 1 also demonstrate that, among the effects under consideration, phase mismatch is typically characterised by the shortest spatial scale, thus being the main factor limiting the efficiency of third-harmonic generation in gas-filled hollow fibres.

## 4. Numerical simulations

To investigate third-harmonic generation in a gas-filled hollow fibre in the field of a short pump pulse with a constant intensity including phase-mismatch, group-delay, and attenuation effects, we performed numerical simulations with the use of Eqns (7), (9)–(11), and (14). We assumed that the pump pulse has a Gaussian envelope with a constant amplitude, which will be set equal to unity:

$$A^{(1)}(z,t) = A_0\left(t - z/v_{\rm p}^{(1)}\right) = \exp\left[-\frac{1}{2\tau^2}\left(t - z/v_{\rm p}^{(1)}\right)^2\right]. \quad (15)$$

Then, Eqn (14) yields the following expression for the amplitude of the third-harmonic pulse:

$$B^{(m)}\left(\eta_{\rm h}^{(m)}\right) = {\rm i}\beta^{(mn)}\int_0^L \exp\left[-\frac{3}{2}\left(\eta_{\rm h}^{(m)} + \zeta^{(mn)}z\right)^3 + \frac{\alpha_{\rm h}^{(m)} - 3\alpha_{\rm p}^{(n)}}{2}z - {\rm i}\Delta k^{(mn)}z - \frac{\alpha_{\rm h}^{(m)}}{2}L\right]{\rm d}z. \quad (16)$$

We simulated third-harmonic generation in low-order modes $EH_{11}$, $EH_{12}$, and $EH_{13}$ of a gas-filled hollow fibre, whose eigenvalues are equal to $u_l^{(1)} \approx 2.41$, $u_l^{(2)} \approx 5.52$, and $u_l^{(3)} \approx 8.65$, respectively. As shown in [4], the energy of the third harmonic under these conditions is mainly concentrated in three lowest order modes, and effects related to the generation of higher order waveguide modes can be neglected. Since the phase mismatch in the case under study can be compensated, as is seen from Fig. 1, only for the $EH_{11}$ mode of pump radiation and the $EH_{13}$ mode of the third harmonic, we will consider below only the $EH_{11}$ mode of the pump beam and the $EH_{13}$ mode of the third harmonic.

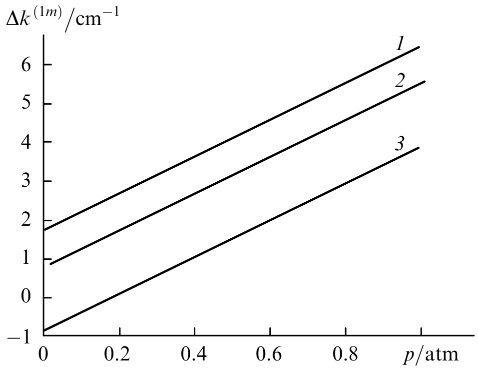


**Figure 1.** Phase mismatches for the $EH_{11}$ hollow-fibre mode of fundamental radiation and (*1*) $EH_{11}$, (*2*) $EH_{12}$, and (*3*) $EH_{13}$ hollow-fibre modes of the third harmonic as functions of the argon pressure for a hollow fibre with a length $L = 60$ cm and an inner radius $a = 75$ μm.

Our simulations were carried out for a hollow fibre with a length $L = 60$ cm filled with different rare gases (helium, neon, argon, krypton, and xenon). The wavelength of pump radiation was assumed to be equal to 800 nm. The dielectric constant of hollow-fibre walls was set equal to $\varepsilon_2 = 2.25$. We have employed the data for dispersion properties of rare gases taken from [16]. The refractive index of a rare gas filling the fibre at the frequency $\omega_l$ was assumed to be proportional to the gas pressure.

One can see from Eqns (6), (7) and (9)–(11) that the phase mismatch, group delay, and attenuation coefficient depend on the inner radius of a hollow fibre, the sort of the gas filling the fibre, the pressure $p$ of this gas, and the pair of waveguide modes of pump radiation and the third harmonic coupled through the nonlinear process. Thus, varying the above-specified parameters, one can control the efficiency of frequency conversion and optimise conditions for the generation of the third harmonic of ultrashort laser pulses in a gas-filled hollow fibre.

## 5. Results and discussion

In this section, we will discuss the results of numerical simulations performed for third-harmonic generation in hollow fibres filled with different rare gases under conditions specified above. Assuming that the intensity of a short pump pulse is fixed, we will examine the efficiency of third-harmonic generation as a function of the duration of the pump pulse and parameters of the fibre and the gas filling the fibre.

Our estimates of the characteristic spatial scales of the process under study have shown (see Table 1) that phase mismatch is the main physical factor limiting the efficiency of nonlinear-optical frequency conversion in the considered case. Therefore, we start our discussion with phase-matching effects in third-harmonic generation in a hollow fibre. Third-harmonic generation in a hollow fibre is perfectly phase-matched, as can seen from Eqns (9)–(11), if the gas pressure $p$ and the inner radius $a$ of a hollow fibre meet the following relationship:

$$p(a) = \frac{p_0 c}{2\Delta k_{01}\omega_{\mathrm{p}}}\left[\frac{1}{3}\left(\frac{u_{\mathrm{h}}^{(m)}}{a}\right)^2 - 3\left(\frac{u_{\mathrm{p}}^{(n)}}{a}\right)^2\right], \tag{17}$$

where $\Delta k_{01}$ is the component of the phase mismatch due to the gas dispersion (10) at the gas pressure $p_0$.

Fig. 2 displays the argon pressure phase-matching, in accordance with Eqn (17), the $EH_{11}$ mode of fundamental radiation and the $EH_{13}$ mode of the third harmonic as a function of the inner radius of the hollow fibre $a$. The energy of the third-harmonic pulse emerging from a hollow fibre filled with argon, $W_{\mathrm{h}} = \int P_{\mathrm{h}}(\eta_{\mathrm{h}}^{(3)})\,\mathrm{d}\eta_{\mathrm{h}}^{(3)}$ [where $P_{\mathrm{h}}(\eta_{\mathrm{h}}^{(3)}) = |B^{(3)}(L,\eta_{\mathrm{h}}^{(3)})| \iint |f_{\mathrm{h}}^{(3)}(\rho)|^2 \rho\,\mathrm{d}\varphi\,\mathrm{d}\rho$ is the power of the third-harmonic pulse], calculated as a function of the inner radius $a$ of the fibre and the argon pressure $p$ is presented in Fig. 3. A similar dependence for helium is shown in Fig. 4.

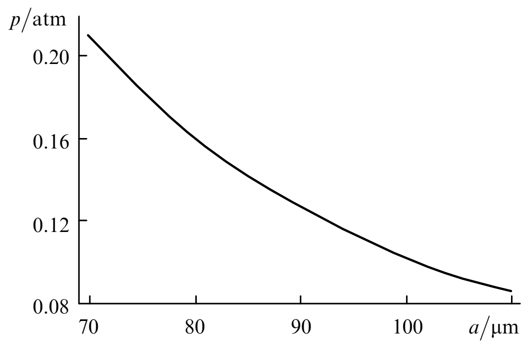


**Figure 2.** The argon pressure $p$ corresponding to a perfect phase matching of the $EH_{11}$ mode of pump radiation and the $EH_{13}$ mode of the third harmonic in a hollow fibre as a function of the inner radius of the fibre $a$.

Comparing Figs. 2 and 3, one can easily verify that the parameters $a$ and $p$ corresponding to the maximum energy of the third harmonic in Fig. 3 satisfy the phase-matching

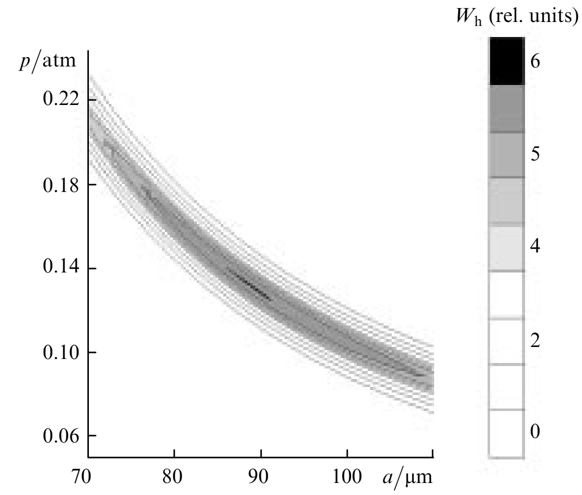


**Figure 3.** The energy $W_{\mathrm{h}}$ (levels of grey scale) of the third-harmonic pulse emerging from an argon-filled hollow fibre with a length $L = 60$ cm as a function of the inner radius of the fibre $a$ and the argon pressure $p$ for the pump pulse duration $\tau = 20$ fs.

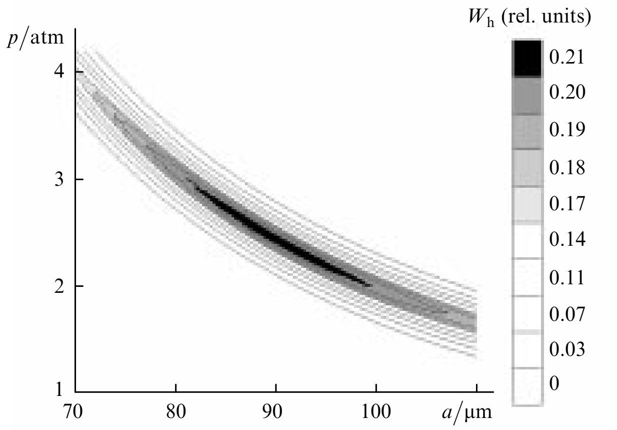


**Figure 4.** The energy $W_{\mathrm{h}}$ (levels of grey scale) of the third-harmonic pulse emerging from a helium-filled hollow fibre with a length $L = 60$ cm as a function of the inner radius of the fibre $a$ and the argon pressure $p$ for the pump pulse duration $\tau = 20$ fs.

relation illustrated in Fig. 2. This finding confirms that phase matching is a factor of crucial importance for the efficiency of third-harmonic generation in the field of ultrashort laser pulses propagating in lossy hollow fibres.

Since phase matching of third-harmonic generation is the necessary condition of efficient frequency tripling in gas-filled hollow fibres, we shall discuss below the dependence of the energy of the third-harmonic pulse on the parameters of the problem assuming that phase-matching conditions are satisfied. Fig. 5a displays the energy $W_{\mathrm{h}}$ of the third-harmonic pulse emerging from a hollow fibre filled with different rare gases (helium, neon, argon, krypton, and xenon) calculated as a function of the inner radius $a$ of the fibre. Third-harmonic generation was assumed to be phase-matched in these calculations, which implies that the radius $a$ and the gas pressure $p$ are not independent, but change jointly in accordance with Eqn (17). Dependences for different gases presented in Fig. 5a are very similar to each other, which indicates that the examined dependence is characteristic of the waveguiding regime of third-harmonic

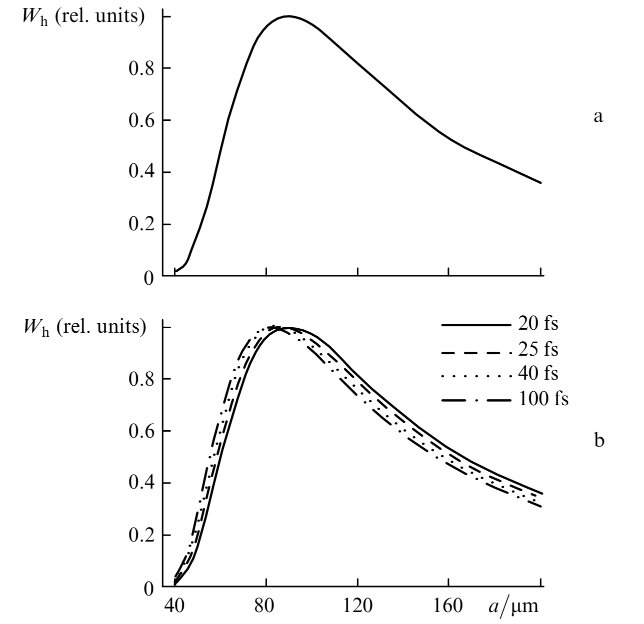


**Figure 5.** The energy $W_h$ of the third-harmonic pulse emerging from a gas-filled hollow fibre with a length $L = 60$ cm as a function of the inner radius of the fibre $a$ in the case of perfectly phase-matched third-harmonic generation (a) for different rare gases with the pump pulse duration $\tau = 20$ fs and (b) for argon with different $\tau$.

generation in a hollow fibre. We should note, however, that, according to Eqn (17), different pressures $p$ should be taken for different gases with a fixed radius $a$ to phase-match third-harmonic generation. Fig. 5b presents the energy of the third-harmonic pulse on the inner radius of a hollow fibre filled with argon for different durations of the pump pulse.

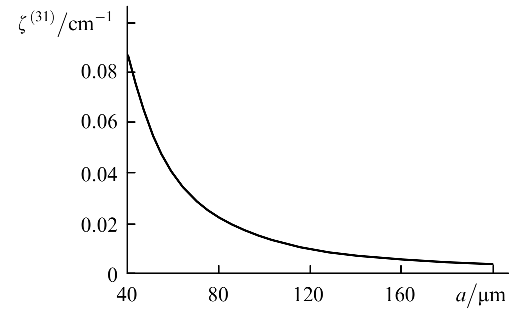


**Figure 6.** The group-delay coefficient $\zeta^{(31)}$ for the $EH_{11}$ mode of fundamental radiation and the $EH_{13}$ mode of the third harmonic as a function of the inner radius of the fibre $a$ for the pump pulse duration $\tau = 20$ fs.

One can see from Figs 5a and 5b that the dependence of the third-harmonic energy on the inner radius of a fibre (and, consequently, the dependence of the third-harmonic energy on the gas pressure in the fibre) displays a clearly pronounced maximum, which arises as a result of opposite tendencies observed in the dependences of the group delay and attenuation coefficients of the pump and third-harmonic pulses (Figs 6 and 7) and the nonlinear coefficient $\beta^{(31)}$ on the inner radius of the fibre. The third-harmonic energy first increases with the growth in the inner radius $a$ of

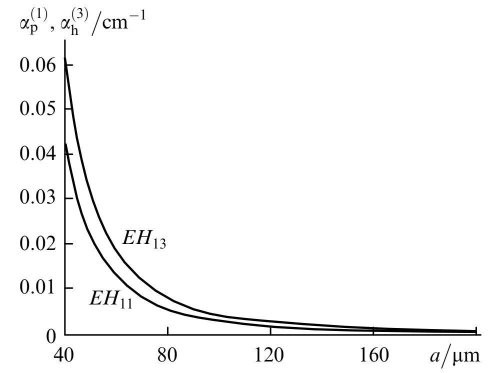


**Figure 7.** Attenuation coefficients for the $EH_{11}$ hollow-fibre mode of fundamental radiation ($\alpha_p^{(1)}$) and the $EH_{13}$ hollow-fibre mode of the third harmonic ($\alpha_h^{(3)}$) as functions of the inner radius of the fibre $a$ for the pump pulse duration $\tau = 20$ fs.

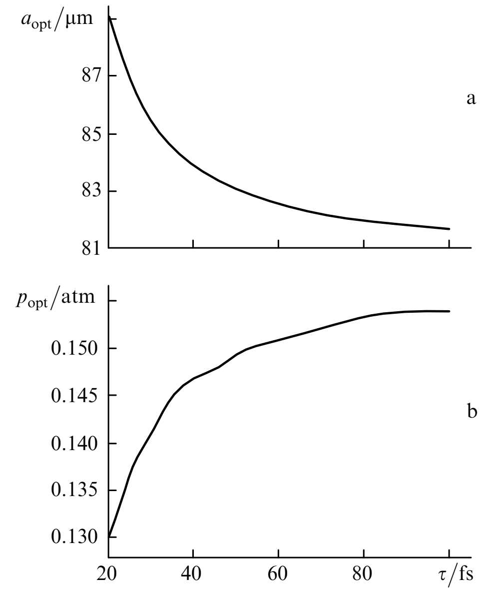


**Figure 8.** The optimal radius of a hollow fibre $a_{opt}$ (a) and the optimal argon pressure in the hollow fibre $p_{opt}$ (b) as functions of the pump pulse duration $\tau$ in a hollow fibre with the length $L = 60$ cm.

the fibre (and, consequently, with the lowering of the pressure $p$) because of the decrease in the attenuation coefficients (Fig. 7) and the group delay (Fig. 6) of the pump and third-harmonic pulses in the hollow fibre. When the attenuation and delay lengths of the pump and third-harmonic pulses become comparable with the fibre length, a further increase in the inner radius $a$ (and the corresponding lowering of the pressure $p$) reduces the third-harmonic energy due to a decrease in the effective nonlinearity $\beta^{(31)}$ of the gas filling the fibre, which is proportional to the pressure $p$.

Figs 8a and 8b present the optimal values of the inner radius of a hollow fibre and the argon pressure corresponding to the maximum energy of the third-harmonic pulse as functions of the pump pulse duration. The variation ranges of the optimal values of the inner radius of the fibre and the gas pressure in the fibre as functions of the fibre radius are not very broad, indicating that the attenuation of light pulses in hollow fibres with small inner diameters may be the main factor limiting the efficiency of phase-matched third-harmonic generation.

## 6. Estimates on the efficiency of nonlinear frequency conversion

Let us estimate the efficiency of phase-matched third-harmonic generation in a hollow fibre filled with argon for optimal parameters of the fibre and the gas. For this purpose, we will employ the estimate for the third-order nonlinear-optical susceptibility per single argon atom presented in [17]: $\chi^{(3)}(\omega_{\rm h};\omega_{\rm p},\omega_{\rm p},\omega_{\rm p}) \approx 124\times10^{-39}$ CGSE. Then, assuming that the $EH_{11}$ waveguide mode of fundamental radiation is perfectly phase-matched with the $EH_{13}$ mode of the third harmonic ($\Delta k^{(31)} = K_{\rm h}^{(3)} - 3K_{\rm p}^{(1)} = 0$), the gas pressure is $p = 98$ Torr, and the inner diameter of the hollow fibre is 150 μm (which corresponds to the optimal conditions for a pump pulse with a duration $\tau = 35$ fs), we can apply Eqn (12) to find the following estimate for the relevant nonlinear coefficient: $\beta^{(31)} \approx 1.2\times10^{-14}$ cm$^{-4}$.

The energy of the third-harmonic pulse at the output of a hollow fibre under conditions of perfect phase matching is then given by

$$W_{\rm h} = \frac{c}{8\pi}\int {\rm d}\eta_{\rm h}^{(3)}\left|B^{(3)}\left(\eta_{\rm h}^{(3)}\right)\right|^2\int {\rm d}\varphi\int\rho\,{\rm d}\rho\left|f_{\rm h}^{(3)}(\rho)\right|^2. \quad (18)$$

If the energy of fundamental radiation coupled into the fibre is equal to 0.8 mJ and the length of the fibre is $L = 60$ cm, Eqns (15) and (16) yield the following estimate for the energy of the third-harmonic pulse (with $\Delta k^{(31)} = 0$): $W_{\rm h} \approx 23$ μJ. Such an energy of the third harmonic would correspond to the efficiency of frequency tripling of $\sim 3$ %, which is more than an order of magnitude higher than the efficiency achieved in experiments [2].

Thus, our estimates demonstrate that, in principle, the efficiency of third-harmonic generation in gas-filled hollow fibres can be further improved with respect to the efficiencies that have been already achieved in experiments. However, this conclusion should be carefully tested beyond the framework of slowly varying envelope approximation with the use of models including accompanying nonlinear and intermode-exchange effects. Apparently, such an analysis can be performed only by means of numerical simulations. Our estimate for the efficiency of third-harmonic generation also indicates that pump-depletion effects can be ignored even for optimal conditions, when the maximum efficiency of frequency conversion is achieved. Indeed, our calculations have shown that up to 3 % of the energy of pump radiation may be converted into the third harmonic. The amplitude of the pump field is reduced by approximately 1.5 % under these conditions.

## 7. Conclusions

Analysis of the solution to the equation for the slowly varying envelope of the third-harmonic pulse in a gas-filled hollow fibre shows that attenuation and the group delay of pump and third-harmonic pulses may have a considerable influence on the efficiency of frequency conversion through third-harmonic generation in hollow fibres. Phase matching of third-harmonic generation, which is the necessary condition for efficient frequency tripling, can be achieved in gas-filled hollow fibres by choosing gases with suitable dispersion properties and through the excitation of appropriate waveguide modes. In contrast to the plane-wave approximation, the solution to the equation for the amplitude of the third harmonic derived in this paper allows waveguide effects to be taken into consideration through waveguide propagation constants, group velocities, attenuation constants, and the nonlinear coefficients normalised to include the transverse distributions of the pump and third-harmonic fields in waveguide modes of a hollow fibre. Optimal conditions are shown to exist for third-harmonic generation in leaky modes of a hollow fibre for different pulse durations and various rare gases filling the fibre. The maximum efficiency of perfectly phase-matched third-harmonic generation in an argon-filled hollow fibre with a length of 60 cm is estimated as approximately 3 %.

***Acknowledgements.*** This study was supported by the President of Russian Federation Grant No. 00-15-99304, the Russian Foundation for Basic Research (Grant No. 00-02-17567), and INTAS (Grant No. 97-0369).

## References

1. Nisoli M, De Silvestri S, Svelto O, Szipocs R, Ferencz K, Spielmann Ch, Sartania S, Krausz F *Opt. Lett.* **22** 522 (1997)
2. Durfee C G III, Backus S, Murnane M M, Kapteyn H C *Opt. Lett.* **22** 1565 (1997)
3. Koroteev N I, Zheltikov A M *Appl. Phys. B* **67** 53 (1998)
4. Zheltikov A M, Koroteev N I, Naumov A N *Zh. Eksp. Teor. Fiz.* **113** 1561 (1999) [*JETP* **88** 857 (1999)]
5. Rundquist A, Durfee C G III, Chang Z, Herne C, Backus S, Murnane M M, Kapteyn H C *Science* **5368** 1412 (1998)
6. Tamaki Y, Midorikawa K, Obara M *Appl. Phys. B* **67** 59 (1998)
7. Constant E, Garzella D, Breger P, Mevel E, Dorrer Ch, Le Blanc C, Salin F, Agostini P *Phys. Rev. Lett.* **82** 1668 (1999)
8. Durfee C G III, Backus S, Kapteyn H C, Murnane M M *Opt. Lett.* **24** 697 (1999)
9. Li X F, L'Huillier A, Ferray M, Lompre L A, Mainfray G *Phys. Rev. A* **39** 5751 (1989)
10. Balcou Ph, Gomes A S L, Coruaggia C, Lompre L A, L'Huillier A *J. Phys. B* **25** 4467 (1992)
11. Macklin J J, Kmetec J D, Gordon C L III *Phys. Rev. Lett.* **70** 766 (1993)
12. Marcatili E A J, Schmeltzer R A *Bell Syst. Tech. J.* **43** 1783 (1964)
13. Adams M *An Introduction to Optical Waveguides* (New York: Wiley, 1981)
14. Miles R B, Laufer G, Bjorklund G C *Appl. Phys. Lett.* **30** 417 (1977)
15. Shen Y R *The Principles of Nonlinear Optics* (New York: Wiley, 1984)
16. *Landolt Börnshtein Physikalisch-Chemische Tabellen, ed. by Roth W A, Schell K* (Berlin: Verlag von Julius Springer, 1931, 1935), vols. II, III.
17. Reintjes J *Nonlinear Optical Parametric Processes in Liquids and Gases* (New York: Academic, 1984)